Non-Linear Spectroscopy of Rubidium in Hollow-Core Fibres

Christopher Perrella B.Sc Hons.

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The University of Western Australia

Supervisors:
Prof. Andre N. Luiten
Dr. Philip S. Light
Abstract

This thesis concerns a new photonic technology based on hollow-core photonic-crystal fibre (HC-PCF) that has been loaded with an atomic vapour. It was demonstrated that this technology may have important applications in the fields of metrology and quantum optics. Furthermore, the small size and robustness of HC-PCF allows these devices to potentially be miniaturised for possible commercial and industrial applications.

By filling the HC-PCF with an atomic vapour, excellent light-atom coupling can be attained as both the atoms and light are confined within the same volume. The small transverse dimensions of the fibre’s optical mode leads to high intensities at low input powers, enabling efficient driving of non-linear transitions which would otherwise be difficult with free-space optical systems. Additionally, long interaction lengths, obtained through an arbitrary length of HC-PCF, lead to large optical depths on these typically weak non-linear transitions. The non-linear transition employed here is the $5S_{1/2} \rightarrow 5D_{5/2}$ rubidium (Rb) two-photon transition. This system is of great interest due to its non-linearity, long excited state lifetime, efficient excitation and large optical depth attained within the HC-PCF.

A number of methods for loading a HC-PCF with Rb are discussed, along with proposed ideas for making this process more efficient. Spectroscopic techniques were used to investigate the effect of the fibre’s confined geometry upon the atomic spectra. Extensive theoretical modelling was employed to confirm the observed spectra and give insight to the mechanisms behind the observations. Using this experimental and theoretical knowledge, two applications were targeted: optical atomic frequency standards; and cross phase modulation for quantum logic gates.

The long atomic lifetime for the Rb $5D_{5/2}$ energy level makes it an ideal basis for optical atomic frequency standards. Here two different types of frequency standards are demonstrated based on the $5S_{1/2} \rightarrow 5D_{5/2}$ Rb two-photon transition. Both standards are potentially compact and robust with applications in commercial and industrial environments. The first frequency standard is based on Rb vapour loaded within a HC-PCF. The $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition was excited with two lasers at 780 nm and 776 nm. The sum frequency of these lasers was stabilised to this transition using modulation-transfer spectroscopy, resulting in a fractional frequency stability of $9.8 \times 10^{-12}$ at 1 s. The second scheme used Rb trapped within a traditional bulk cell. Here, modulation-transfer spectroscopy could not be employed...
due to the weak absorption within the cell, hence detection of atomic fluorescence was employed. A fractional frequency stability of $8 \times 10^{-13}$ at 1 s was measured by comparison to an optical frequency comb. Current performance limitations for both standards are presented, along with a clear pathway to improving the performance of both standards by an order of magnitude.

Another research field which could benefit from an efficiently driven two-photon transition within a HC-PCF fibre is that of quantum optics, where strong cross-Kerr non-linearities have been long sought after for quantum information applications. A detailed systematic study of cross-Kerr non-linearities, and absorption, is presented based around the two-photon transition excited within a HC-PCF. Phase modulations of up to $\pi$ rad with a signal power of 45 $\mu$W were observed which correspond to a non-linear Kerr coefficient, $n_2$, of $1.4 \times 10^{-6}$ rad cm$^2$/W, or $0.7 \times 10^{-6}$ rad per photon.
Statement of Candidate Contribution

The papers generated from the work contained in this thesis, and the author contribution to each, are outlined below:

Published


  The entirety of the experiment and data analysis was carried out by the author, under the supervision and assistance of Prof. Luiten and Dr. Light. Much theoretical assistance was provided by Dr. Stace. This paper is included in chapter 7.


  Vital experimental equipment was delivered and set up at Swinburne University of Technology by Mr. Truong and the author. This paper is included in chapter 11.


  The entirety of the experiment and data analysis was carried out by the author, under the supervision and assistance of Prof. Luiten, Dr. Light and Dr. Anstie. Theoretical assistance was provided by Dr. Stace. This paper is included in chapter 8.


  The entirety of the experiment and data analysis was carried out by the author, under the supervision and assistance of Prof. Luiten, Dr. Light, Dr. Anstie and Dr. Baynes. This paper is included in chapter 9.

The entirety of the experiment and data analysis was carried out by the author, under the supervision and assistance of Prof. Luiten, Dr. Light and Dr. Anstie. Theoretical assistance was provided by Dr. Stace and Prof. White. This paper is included in chapter 10.


The experiment and data analysis was carried out by Dr. Light and the author, under the supervision and assistance of Prof. Luiten, Dr. Anstie and Dr. Baynes. This paper is included in chapter B.


Vital experimental equipment was built and provided by the author for use in this experiment. This paper is not included in this thesis.

Accepted


Vital experimental equipment was delivered and set up at Swinburne University of Technology by Mr. Truong and the author. This paper is not included in this thesis.

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The entirety of the experiment and data analysis was carried out by the author, under the supervision and assistance of Prof. Luiten and Dr. Light. Theoretical assistance was provided by Dr. Stace and Mr. Milburn. This paper is not included in this thesis.
Signed: Candidate: 

Signed: Supervisor: 

v
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Foreword

The demand for portable high-stability timing signals in today’s technological society and the continual quest for practical quantum-logic technology are very active and exciting research areas. Both of these fields stand to benefit from the technology of hollow-core photonic-crystal fibres loaded with rubidium vapour. This technology promotes extremely strong light-atom interactions, providing efficient driving of a non-linear two-photon transition within the rubidium vapour. Properties of the two-photon transition made it an ideal base for both a frequency standard for high-stability timing, as well as mediating photon-photon interaction, essential for optical quantum logic gates and quantum measurement.

An introduction to the research areas of frequency standards and quantum optics is presented in chapter 1. Highlighted are the benefits of a hollow-core photonic-crystal fibre loaded with rubidium vapour to these areas of research. A brief introduction to hollow-core fibres is presented in chapter 2, before the particular hollow-core fibres used to produce this body work are discussed. Finally, the physical and quantum properties of rubidium are discussed in chapter 3 with particular reference to the atomic transitions excited within the hollow-core fibre.

Detailed mathematical modelling was conducted to understand the atomic mechanisms which give rise to the observed atomic transition spectra within the fibre. Chapter 4 introduces density matrix theory which forms the basis of the modelling presented here and forms a connection between this mathematical model and experimental observations. Chapter 5 draws from this basis to model atomic absorption within the confined geometry of the fibre in an effort to understand spectral broadening effects such as: transit-time, power broadening, light shifts, Rabi splitting, magnetic field broadening, collisional broadening and population pumping. Finally, predictions for experimental observations are generated which include all of these effects.

The experimental set-up which was used is discussed in chapter 6. Specifically, the optical components which were used to interrogate the vapour filled hollow-core fibre are discussed. Low optical powers used within the fibre presented challenges for optical detection methods which are discussed. The apparatus and methods used to load a rubidium vapour within a hollow-core fibre are also presented. Finally, a ECDL was designed and manufactured to interrogate the loaded fibre with a narrow linewidth laser source.
Initial characterisation of the rubidium loaded fibre was conducted using hole-burning spectroscopic techniques. This used the well studied $D_1$ and $D_2$ transitions within rubidium to experimentally measure the broadening mechanisms within the fibre and compare them to theory. Results from these experiments are presented in the peer-reviewed publication in chapter 7. An extension to this work was made to the two-photon transition in chapter 8. This chapter presents a detailed characterisation of this transition excited within the hollow-core fibre, exploring: detuning from an intermediate state, pump power dependence, and broadening mechanisms.

Using the knowledge provided from chapters 7 and 8, the above mentioned applications were targeted. Chapter 9 presents a frequency standard based on the two-photon transition excited within a hollow-core fibre. A fractional frequency stability of $9.8 \times 10^{-12}$ integrated over 1 s was demonstrated with avenues for improvement identified. Furthermore, a pathway towards a compact frequency standard capable of producing high-stability timing signals is presented.

The strong light-atom coupling produced within the hollow-core fibre is advantageous in the field of quantum optics where photon-photon interactions are sought after for use in quantum optical logic gates. Chapter 10 presents a detailed characterisation of how the two-photon rubidium transition excited within a hollow-core fibre produces efficient, deterministic photon-photon interactions. Photon loss mechanism are also investigated to identify the viability of this technology for non-demolition logic gate and quantum measurement applications. A maximum phase-shift of $0.7 \times 10^{-6}$ rad per photon was demonstrated. Furthermore, chapter 11, presents preliminary work for possibly extending the technology to become a single-photon source at convenient wavelengths for demonstrated schemes for optical quantum memories.

Finally, chapter 12 summarises the work presented in this thesis. It also outlines future pathways that could be taken to improve the results presented here, and extend the application base of this technology into other areas.
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<tr>
<td>APD</td>
<td>Avalanche photodiode</td>
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<tr>
<td>cgs</td>
<td>Centimetre - gram - second system of units</td>
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<tr>
<td>ECDL</td>
<td>Extended cavity diode laser</td>
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<tr>
<td>EIT</td>
<td>Electromagnetically induced transparency</td>
<td></td>
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<tr>
<td>FSR</td>
<td>Free spectral range</td>
<td></td>
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<tr>
<td>FWHM</td>
<td>Full-width at half-maximum</td>
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<tr>
<td>HC-PBF</td>
<td>Hollow-core photonic-bandgap fibre</td>
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<td>HC-PCF</td>
<td>Hollow-core photonic-crystal fibre</td>
<td></td>
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<tr>
<td>MBR</td>
<td>Monolithic block resonator</td>
<td></td>
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<tr>
<td>PBF</td>
<td>Photonic-bandgap fibre</td>
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</tr>
<tr>
<td>PBS</td>
<td>Polarising beam splitter</td>
<td></td>
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<tr>
<td>PCF</td>
<td>Photonic-crystal fibre</td>
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<tr>
<td>PD</td>
<td>Photodiode</td>
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<tr>
<td>PDF</td>
<td>Probability density function</td>
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<tr>
<td>PMT</td>
<td>Photomultiplier tube</td>
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<tr>
<td>PSD</td>
<td>Power spectral density</td>
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<tr>
<td>RF</td>
<td>Radio frequency</td>
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<tr>
<td>RWA</td>
<td>Rotating wave approximation</td>
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<tr>
<td>SAS</td>
<td>Saturated absorption spectroscopy</td>
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<tr>
<td>SEM</td>
<td>Scanning electron microscope</td>
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<tr>
<td>SI</td>
<td>International System of Units</td>
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<tr>
<td>TEC</td>
<td>Thermoelectric cooler</td>
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<td>Ti:S</td>
<td>Titanium Sapphire</td>
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List of Symbols

\( \alpha \) Absorption coefficient  
\( c \) Speed of light  
\( d \) Distance  
\( \Delta f \) Linewidth/bandwidth in linear frequency units  
\( \Delta \omega \) Linewidth/bandwidth in angular frequency units  
\( \Delta \phi \) Phase shift  
\( E \) Electric field amplitude  
\( \varepsilon \) Energy  
\( f \) Frequency of light  
\( \Im \) Imaginary part of a complex number  
\( k \) Wave vector  
\( k_B \) Boltzmann constant  
\( \lambda \) Wavelength of light  
\( m \) Molar mass  
\( M \) Atom number  
\( \mu \) Atomic dipole moment  
\( \bar{n} \) Mean value  
\( n, \tilde{n} \) Refractive index  
\( N_A \) Avogadro’s number  
\( \mathcal{N} \) Photon number  
\( NA \) Numerical Aperture  
\( \phi \) Angle  
\( Q \) Quality Factor  
\( r \) Radial axis in a Cylindrical/Polar co-ordinate system  
\( R \) Radius of a hollow-core fibre  
\( \Re \) Real part of a complex number  
\( S/N \) Signal-to-noise ratio  
\( \sigma \) Standard deviation  
\( \sigma_y \) Fractional frequency stability  
\( T \) Temperature  
\( \tau \) Frequency standard integration time  
\( \tau \) Transit time  
\( \tau \) Energy level lifetime  
\( \theta \) Azimuthal angle  
\( v_0 \) Most probable velocity  
\( v_t \) Transverse velocity  
\( V \) Potential energy  
\( \omega \) Angular frequency of light  
\( x \) \( x \)-axis in a Cartesian co-ordinate system  
\( y \) \( y \)-axis in a Cartesian co-ordinate system  
\( z \) \( z \)-axis in a Cartesian co-ordinate system
Part I

Background
This chapter presents an overview of all the work presented in this document, as well as several applications that have the potential to benefit from this work. In particular, a brief description of the complete thesis is given in section 1.1. Following this, a background for two potential applications of this technology is presented: the first is towards an atomic frequency standards in section 1.2, and then quantum optics applications in section 1.3.

1.1 Overview

This thesis presents work based on hollow-core photonic-crystal fibre that has been filled with a dilute atomic vapour. An optical field is guided in the core and can strongly interact with atomic resonances of the vapour. This technology has powerful applications in the fields of metrology and quantum optics.

Hollow-core fibre guides an optical mode within a hollow core rather than a solid glass-core as does traditional optical fibre. It is this unique feature that allows intimate light-atom interaction that is necessary for high-performance frequency standards and quantum optics. The small transverse dimensions of the optical mode magnify the light-atom coupling strength as high optical intensities can be produced for low optical powers. Additionally, the fibre’s arbitrary length provides extended light-atom interaction lengths. This allows efficient driving of non-linear transitions within the gas that would otherwise be difficult with free-space optical systems. The structure of these fibres is discussed in more detail in chapter 2.

One such non-linear transition is the two-photon transition in a rubidium vapour which is discussed in detail in chapter 3. This transition allows access to a long lived excited state within rubidium. This, in conjunction with the inherent non-linearity of the transition, is of great benefit to a number of research fields as explained below.
CHAPTER 1. INTRODUCTION

For atomic frequency standards, energy levels with long lifetimes are highly desirable. The stability of atomic frequency standards is inversely proportional to both the lifetime of the atomic transition, and the signal strength of the detected transition, as discussed in section 1.2. Hence the ability to drive the two-photon transition efficiently to a long lived energy level is of great benefit. A history of atomic frequency standards is discussed in section 1.2 along with experimental progress towards making these devices compact and robust.

Another research field which could benefit from an efficiently driven two-photon transition within a hollow-core fibre is that of quantum optics. Photons are an attractive carrier of quantum information as they interact very weakly with their environment, and with each other, leading to long lived quantum states. Unfortunately, this makes photon-photon interactions exceedingly difficult to engineer for quantum information logic operations. As a result, platforms that can provide efficient photon-photon interactions are keenly sought in the field of quantum optics. An overview of the current progress of quantum optics is discussed in chapter 1.3 along with the specific applications that we have demonstrated in this field.

1.2 Atomic Frequency Standards

Frequency standards play an important role in technological society, especially as our world becomes more fast-paced and information transfer becomes increasingly limited by accurate timing in applications such as mobile networks [1, 2] and high-speed stock trading [3]. The most prominent commercial use of frequency standards is in global positioning systems [4–8] where accurate timing is required to pinpoint a location on the Earth’s surface. Many more applications are found in: telecommunications and wavelength division multiplexing (DWDM) systems [9–11], optical fibre sensing [11], and high-accuracy optical measurement systems. Beyond everyday life, fundamental physics uses ultra-high precision frequency standards to: test general relativity [12, 13], investigate the temporal stability of fundamental constants [14–16], explore atomic physics [17–21], study Newtonian gravity [22], measure optical wavelengths with high-accuracy, make very-long baseline interferometry (VLBI) measurements in radio astronomy [23], and track probes used for deep space exploration [23].

These frequency standards are based on the stabilisation of an electromagnetic oscillator to a transition frequency between two well defined energy states of an atom. The guiding principle here is that atomic energy levels are discretely quantised with their energy state determined by fundamental constants, hence are exactly the same
1.2. ATOMIC FREQUENCY STANDARDS

for all atoms of a particular species (assuming they are unperturbed). This makes atomic transitions between energy levels an excellent basis for frequency standards.

There are two key performance requirements for a frequency standard: accuracy and precision. An accurate frequency standard can be directly related to the SI second. In turn, the SI second is defined in terms of a transition in a caesium atom, as discussed in the next section. Thus, measurements of frequency standard accuracy are made via comparison to a caesium frequency standard, and hence accuracy measurements are limited to the best caesium frequency standards. By taking care to remove systematic shifts in the atom’s transition frequency, a frequency standard’s accuracy can exceed that of a caesium standard. The best modern frequency standards have demonstrated this by comparing two separate clock realisations and measuring their relative uncertainty. In these cases systematic uncertainties are given as the representation of the potential for accuracy of the frequency standards were the SI definition to change. The precision of a frequency standard is measured through its frequency stability, which can be measured against any other frequency standard.

The stability of atomic frequency standard, \( \sigma_y(\tau) \), over \( \tau \) seconds of integration can be estimated to be [23]:

\[
\sigma_y(\tau) = \frac{1}{Q(S/N)} \tau^{-1/2},
\]

(1.1)

Here, \( Q \) is the quality factor of the transition, defined to be \( Q = \Delta f / f \) where \( \Delta f \) is the spectral linewidth of the transition, centred at a transition frequency \( f \) which usually falls in the microwave or optical frequency domain for modern frequency standards. There are two routes to attaining a better \( Q \) and hence stability: choosing a transition with a higher transition frequency \( f \), or by choosing an excited state with a long lifetime \( \tau_l \). This can be seen because as the natural linewidth is given by \( \Delta f = 1/(2\pi\tau_l) \). In modern frequency standards, the transition lifetime is so long that the transition’s linewidth is usually limited by the coherent light-atom interaction time rather than the lifetime of the excited state.

Additionally, the stability of a frequency standard is inversely proportional to the quality of the signal used for stabilisation, which is typically quantified using the signal-to-noise ratio \( (S/N) \). Maximising the signal is achieved by using the highest number of absorbers (atoms), while interrogating with the largest number of photons. Noise on top of the signal can originate from technical sources, such as electronics, or fundamental noise such as photon (or atomic) shot-noise, which scales with the square root of the number of photons (atoms) detected. This is discussed in...
more detail in section 6.1.5. There is a compromise that must be made between the detected signal-to-noise ratio and the potential for unwanted frequency shifts that compromise the accuracy of the standard. For example, increasing the probing laser power increases the signal-to-noise ratio, but can also induce frequency shifts of the atomic transition, as discussed in chapter 5.

All of these parameters discussed above, which contribute towards a frequency standard’s stability $\sigma_y(\tau)$, as defined in equation 1.1, have been explored throughout the history of atomic clocks to generate more stable frequency standards.

### 1.2.1 History of Frequency Standard

The first practical frequency standards were based on microwave transitions in atomic and molecular vapours. In the 1950s the caesium atomic clock became the favoured frequency standard reaching stabilities of $1 \times 10^{-9}$ at integration times of $\tau = 1\text{ s}$ [24]. This clock was based on a microwave transition between two ground states of a caesium atom. The stability of atomic frequency standards progressed so far that in 1967 the unit of the second was redefined within the International System of Units (SI) as “the duration of 9192,631,770 periods of the radiation corresponding to the transition between the two hyperfine levels of the ground state of the caesium- 133 atom” by the International Bureau of Weights and Measures (BIPM) [25]. Further development led to the creation of caesium fountains which have demonstrated frequency stabilities of $1.6 \times 10^{-14}\tau^{-1/2}$ [26]. This breakthrough was achieved by increasing the coherent interaction time between the probing radiation and the atoms, leading to a reduced linewidth and improved frequency stability as predicted by equation 1.1.

There are now other competing modern microwave frequency standards such as: rubidium fountains, reaching stabilities of $2 \times 10^{-13}\tau^{-1/2}$ [28]; and compact caesium and rubidium clocks [7, 29–31] for possible use in global positioning systems that achieve $3 \times 10^{-13}\tau^{-1/2}$ [6] and $4 \times 10^{-13}\tau^{-1/2}$ [30].

In the 1980s, competitive optical frequency standards started to be developed, based on atomic and molecular transitions in the optical domain. The higher transition frequency potentially leads to higher quality factors $Q$, and hence better frequency stability as highlighted by equation 1.1. Initially these optical frequency standards were based on simple atomic or molecular spectroscopy techniques, with the first optical clock being a He-Ne laser stabilised to iodine vapour [32]. Rapid increases in stability were achieved in the latter part of the 20th century, as seen in figure 1.1, achieved through the use of laser cooled ions and neutral atoms. This saw
1.2. ATOMIC FREQUENCY STANDARDS

![Fractional frequency stability of various frequency standards since 1955. The stability is expressed over 1000 s integration time. Microwave caesium standards (blue circles) are shown along with neutral atom (green squares) and trapped ion standards (red triangles). The majority of this data was taken from Ref. [23]. The most recent and stable results presented were sourced from: Al$^+$, Ref. [16]; Sr$^+$, Ref. [21]; Sr, Ref. [27].](image_url)

**Figure 1.1:** Fractional frequency stability of various frequency standards since 1955. The stability is expressed over 1000 s integration time. Microwave caesium standards (blue circles) are shown along with neutral atom (green squares) and trapped ion standards (red triangles). The majority of this data was taken from Ref. [23]. The most recent and stable results presented were sourced from: Al$^+$, Ref. [16]; Sr$^+$, Ref. [21]; Sr, Ref. [27].

optical frequency standards surpass the stability of the older microwave standards in the last few years.

Laser cooled ion clocks utilise a single ionised atom, with examples seen in Al$^+$ [15], Ca$^+$ [33], Sr$^+$ [34], In$^+$ [35, 36], Yb$^+$ [37, 38], Hg$^+$ [39], which are laser cooled and trapped in a radio frequency (RF) electric field trap inside a vacuum chamber [23]. This tight trapping technique removes many effects that can perturb the atom’s energy levels, including; first and second order Doppler shifts and atomic collisions [23] while also allowing an indefinite light-atom interaction time. As a result, laser cooled ion clocks have reached the $4 \times 10^{-15} \tau^{-1/2}$ level of stability and a systematic uncertainties of $7 \times 10^{-18}$ [16].

Neutral atom clocks have also been intensively developed over the last couple of decades. Modern standards take neutral atoms that are laser cooled and place them into an optical lattice creating trapped and spatially isolated atoms [19, 40, 41]. The conceptual realisation of the *magic wavelength*, where the trapping light is chosen so that no light shifts are created [42], has resulted in stabilities of below $4.4 \times 10^{-16} \tau^{-1/2}$ [27]. This eliminates many of the effects that plagued the standards beforehand and in addition posses a major advantage, in that they are able to trap more than one atom. With an occupancy of one atom per lattice node, a typical optical lattice can hold $10^4$ atoms [43]. The signal-to-noise ratio scales as the square root of the number of atoms (assuming the measurement it photon shot-noise limited [44]),
hence an increase of two orders-of-magnitude is possible compared to a single ion standard. This hence improves the stability by two-orders of magnitude according to equation 1.1. This is a major advantage for high stability clocks as it allows good stability in the short-term. On the other hand, collisional shifts and light shifts due to black body thermal radiation are typically more severe within neutral atom standards compared to ion clocks. A Sr lattice clock has achieved stabilities of $4.4 \times 10^{-16} \tau^{-1/2}$, reaching $1 \times 10^{-17}$ stability after 1000s of integration, with a systematic uncertainty due to atomic collisions of $3 \times 10^{-17}$[27].

1.2.2 Deployable Frequency Standards

Frequency standards based on neutral atoms trapped in optical lattices or laser cooled ions are very delicate and complex devices. Typically these standards occupy an entire laboratory and hence cannot be moved into the field. This is not convenient if a stable timing signal is required anywhere else for applications such as: high performance timing, navigation [4–8], telecommunications [9], or frequency measurement. Two approaches have been taken to overcome this: frequency dissemination of stable signals [45, 46], or development of compact, portable, robust and efficient frequency standards. This thesis focusses on the second approach.

Currently the hydrogen maser is the most stable commercially available frequency standard operating at the $\approx 10^{-13} \tau^{-1/2}$ level of stability [47]. Although stable, this device is not accurate, is expensive ($>\$200,000) and occupies a physical volume of $\approx 400 \ell$. More accurate caesium and rubidium frequency standards are commercially available which are substantially more compact than the hydrogen maser, occupying $\approx 0.7 \ell$ and $\approx 30 \ell$ respectively. Although smaller than the hydrogen maser, the frequency stability of these standards suffers, achieving only $\approx 2.7 \times 10^{-11} \tau^{-1/2}$ [48, 49]. The most compact, commercially available frequency standards are chip-scale designs based on coherent population trapping in a caesium vapour [50]. This device is 16 m$\ell$ in volume and achieves stabilities of $1.5 \times 10^{-10} \tau^{-1/2}$ [50]. The frequency stability of these commercial devices are compared in figure 1.2.

Other compact solutions, that are currently under development, though not yet commercially available, include miniaturised caesium [6], and rubidium [7, 30, 31], frequency standards achieving stabilities in the low $10^{-13} \tau^{-1/2}$ range. These devices display better frequency stability than the commercially available caesium and rubidium standards as shown in figure 1.2. It should be noted that all of the compact devices discussed so far, both commercial and lab based, are based on microwave hyperfine ground state transitions.
A completely separate solution was proposed and implemented based on an optical transition within a rubidium vapour, for possible applications in the telecommunications sector [56–59]. This is of particular interest to the work presented here as these frequency standards were based on the rubidium $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition, which is discussed in detail in chapter 3.1.2. These standards used either a doubled 1556 nm or 778 nm laser tuned to half the energy difference between the $5S_{1/2}$ and $5D_{5/2}$ states. A frequency stability of $2 \times 10^{-13} \tau^{-1/2}$ has been demonstrated [56–59] making these standards the most stable optical atomic frequency standards based on a room temperature effusive vapour. To generate these frequency stabilities, the rubidium two-photon transition rate was enhanced using an optical cavity to increase the optical power interrogating the transition [56–59]. However, this approach made these clocks complex and fragile devices.

A possible solution was introduced when frequency standards made use of molecular vapours confined within a hollow-core optical fibre [54]. This approach has the advantage of potentially being very compact and robust, and was able to achieve stabilities of $1.2 \times 10^{-11}$ and $2.3 \times 10^{-12}$ at an integration time of $\tau = 1\, \text{s}$ for an
CHAPTER 1. INTRODUCTION

Acetylene [54] and iodine [55] vapours respectively. These frequency standards are compared with the previously discussed standards in figure 1.2. Furthermore, the iodine standard has been projected to operate with stabilities of $10^{-13} \tau^{-1/2}$ with further development [55].

This thesis presents work aimed at exploiting the two-photon rubidium resonance within a hollow-core fibre to produce an efficiently driven, strong, two-photon transition. A frequency standard based on this technology is predicted to potentially reach a frequency stability of $10^{-13} \tau^{-1/2}$. The two-photon transition to be used in this frequency standard is discussed in section 3.1.2 and the hollow-core fibre used discussed in chapter 2. The experimental setup is discussed in chapter 6, while experimental results of the two-photon transition within the hollow-core fibre are discussed in chapter 8. Finally the frequency stability measurements are presented in chapter 9.

1.3 Quantum Optics

Quantum computers are a fundamentally different approach to computing, which has become the focus of much interest over the last 20 years. Such devices are predicted to execute certain computation tasks on timescales orders of magnitude faster than traditional computers, and potentially execute some tasks that cannot be solved with today’s computers [60]. This is due to the way information is stored and transformed in the quantum world.

In traditional computers, information is stored in a bit with the discrete values of 1 or 0. However, a quantum computer stores information in a qubit which takes any value in a two dimensional Hilbert space of states parametrised by $\theta$ and $\phi$. The state of a qubit can be expressed as [60, 61]:

$$|\psi\rangle = \cos(\theta)|0\rangle + \exp(i\phi) \sin(\theta)|1\rangle$$  \hspace{1cm} (1.2)

This enables an individual qubit (labeled $a$) to take on a superposition of two states, e.g. $\frac{1}{\sqrt{2}}(|0\rangle_a + |1\rangle_a)$ [60]. Moreover, superpositions of multiple qubits (labeled $a$ and $b$) can exist, e.g. $\frac{1}{\sqrt{2}}(|0\rangle_a|0\rangle_b + |0\rangle_a|1\rangle_b) = \frac{1}{\sqrt{2}}|0\rangle_a(|0\rangle_b + |1\rangle_b)$, as well as entanglement of multiple qubits, e.g. $\frac{1}{\sqrt{2}}(|1\rangle_a|0\rangle_b + |0\rangle_a|1\rangle_b)$ [60, 61]. By using the superposition and entanglement properties of qubits and quantum logical operations, computational tasks such as: factorising numbers (Peter Shor’s factoring algorithm [62]), searching databases (Grover’s search algorithm [63]), simulations of quantum systems [64], and travelling salesman problems [60, 63], are all predicted to be solvable in a time
scale orders of magnitude faster than that capable with traditional computers.

There are various competing proposals for the implementation of a quantum computer including: trapped ions [65, 66], quantum dots [67], quantum optics [68], and nuclear magnetic resonance spectroscopy (NMR) [69]. Any particular scheme needs five key properties [61]: well characterised, and scalable qubits; ability to initialize states; long de-coherence times; universal set of quantum gates; and the ability to measure the qubit’s state.

One implementation of interest in this thesis is optical quantum computing. This approach has a key advantage over the other approaches in that the optical qubit, the photon, is extremely long lived, as it interacts very weakly with its environment and other photons. Furthermore, photons have flexibility in their representation of quantum information, where common encodings makes use of polarisation, or spatial position [70]. Additionally, it is possible to transform from polarisation encoding to position encoding and vice-versa, simply using polarising beam splitters [68]. Unfortunately optical quantum computing suffers from a big disadvantage that other approaches can provide. The weakness of the photon’s interaction makes implementation of a universal set of quantum gates very difficult. This thesis attempts to address this problem by enhancing the interaction of two photons using the hollow-core fibre platform.

Much progress has been made in the development of optical quantum computers. Discussed below are some specific highlights of research conducted in this area; it is by no means a comprehensive list.

### 1.3.1 Single-Photon Sources

To implement optical quantum logic operations a reliable source of single-photons is required. Ideally a single-photon source could produce single indistinguishable photons on-demand, at high bandwidth, with low error and high efficiency and be operational at room temperatures [71]. Furthermore, it would be ideal if the source was of narrow linewidth, and tunable over a broad wavelength range [71]. Many different implementations have been investigated which fulfill some of these characteristics while not all. These implementations include: thermal atom sources [72], trapped ions [73], trapped neutral atoms [74], diamond defects [75], parametric down conversion [76], and quantum dots [71, 77–79].
1.3.2 Qubit Storage

For complex quantum computational tasks, storage of qubits is required. There are many different approaches to storing photon qubits including: gradient echo memory (GEM) [80, 81], electromagnetically induced transparency (EIT) [82], atomic frequency combs (AFC) [83], off-resonant Raman interactions [84], controlled reversible inhomogeneous broadening [85], and spin polarisation [86]. It should be noted that the majority of these memories use a cesium or rubidium vapour as the storage media.

1.3.3 Logic Gates

Quantum computers cannot exist without logical operation gates which have been the focus of much research both theoretical and experimental. A major breakthrough in efficient quantum computation with linear optics was the generation of the Knill, Laflamme, Milburn (KLM) protocols [68]. Since then a number of schemes and protocols based around the KLM protocols have been suggested [70, 87, 88] which are less resource-intensive and hence more easily implemented and scalable.

Experimentally these quantum logic gates have been demonstrated to various extents [89–92]. A controlled-NOT gate (CNOT) based on the KLM method has been demonstrated using an array of precisely aligned beam splitters and waveplates [89]. Furthermore, a universal set of single-qubit and non-trivial two-qubit quantum logic gates has been demonstrated using an entangled four-qubit cluster state in a one-way quantum computer [91]. Miniaturisation and simplification has been made by writing these quantum circuits onto silica wafers to minimise alignment constraints [93, 94]. Furthermore, by adding dynamic elements these circuits can be made to be reconfigurable [95].

The downfall of the above mentioned quantum logic gates is that they are non-deterministic. This does not lend itself towards scalability, and hence deterministic quantum logic gates are sought after. Techniques such as the cross-Kerr effect are avenues towards deterministic quantum logic gates, it is this approach which is utilised in this thesis.

1.3.4 Measurement

Efficient single-photon detectors have also been developed for readout of optical quantum logic gates [96, 97]. However, non-demolition measurements are desired as they do not add back-action noise into the quantum system under study, which is
inherent to the process of measurement in the quantum regime [98–100]. Systems working towards making non-demolition measurements are typically based on the cross-Kerr effect, which induces cross phase modulation [99–102]. Unfortunately, recent theoretical [103, 104] and experimental [105] studies have shown that cross-phase nonlinearities in a single-pass configuration are not feasible. It has been shown that for travelling waves, single-photon-induced phase shifts are always too small to be reliably resolved shot-to-shot [104] due to the interplay between quantum noise and the intrinsic saturation of the nonlinear medium. A proposed solution is the use of an optical cavity to enhance the cross-phase nonlinearity [88] and potentially resolve shot-to-shot measurements.

### 1.3.5 Enhancing Photon-Photon Interactions

The property of photons that make them so attractive; namely their weak interaction with their environment and each other, is also a hindrance. As photons do not interact with each other, it is very difficult to engineer photon-photon interactions required to produce a universal set of quantum logic gates. Typically beam splitters and auxiliary photons are used to produce photon-photon interactions by exploiting quantum interference at the single photon level using beam splitters [68]. This produces large, complicated, and very delicate optical networks which are not scalable and hence not viable for a realistic optical quantum computer.

If a photon-photon interaction could be engineered within an efficient, compact and robust device, this would be of great interest to this field of research. One possible approach is to use a two-photon atomic transition, although non-linear transitions of this type are typically driven with high optical intensities and are often very weak. Using hollow-core fibre to confine the optical mode, and therefore producing high intensities for low input powers, a two-photon transition could be driven much more efficiently and hence an efficient photon-photon interaction may be achievable. This technology could be used in quantum optics logic gates where photon-photon interactions are required for conditional logic operations, or as a non-demolition photon number detector. Furthermore, a two-photon atomic transition within a hollow-core fibre could also have applications in production of single-photons using four-wave mixing techniques.

The two-photon transition to be used in this approach is discussed in section 3.1.2 and the hollow-core fibre used discussed in both chapters 2 and 6. The experimental set-up is discussed in chapter 6, while experimental results of the two-photon transition within the hollow-core fibre is discussed in chapter 8. Experimental mea-
surements of the effective photon-photon interaction efficiency presented in chapter 10, while preliminary work regarding single-photon generation is presented in chapter 11.
Hollow-Core Photonic-Crystal Fibre

This chapter gives some background on a key element of the experimental work discussed in this thesis: hollow-core photonic-crystal fibre (HC-PCF). A brief history into the development of HC-PCF is presented in section 2.1. Following this, two types of HC-PCF are discussed, which are used in the experimental work presented in later chapters. In particular, hollow-core photonic-bandgap fibre is discussed in section 2.2, while kagomé style hollow-core photonic-crystal fibre is discussed in section 2.3. The final section presents a characterisation of the particular fibres that were used in the experiments presented in this thesis.

2.1 History and Development

The theoretical [106, 107], and experimental [108, 109], development of single-mode optical wave-guides in the 1970s, was an enormous technological step which has led to the revolution of high-speed world-wide communication. Although single-mode fibre (SMF) has excelled in information transfer over vast distances, there is a much wider range of potential applications for hollow-core fibres. As an example, optical fibre with more complex optical-guiding structures, such as transverse Bragg wave-guides, were theorised and demonstrated in the late 1970s [110, 111]. In the early 1990s the idea took form that light could be guided within a hollow core, surrounded by a periodic lattice structure of microscopic holes within the cladding glass [112], this was the beginning of hollow-core photonic-crystal fibre. The designs of these fibres allowed for applications that standard SMF could not accomplish, furthermore, it has been suggested that the losses of this new fibre could potentially be below that of conventional SMF and could replace it in the telecommunications sector [112, 113]. It was not until 1999 that the first HC-PCF was demonstrated [114], and since then there has been enormous effort in improving manufacturing (producing lower loss...
and broad guidance fibres [115]) and applications of these fibres [116, 117]. Since their development, these fibres have proven useful in: optical sensing, transmitting high optical powers, and broadband guidance [112], whilst exhibiting many useful properties including: high non-linearities, high birefringence, engineerable dispersion and better hosts for doping materials [112]. A more in-depth history of both PCF and HC-PCF can be found in references [112, 118, 119], while references [117, 119] provide a good overview of the many applications of HC-PCF.

2.2 Hollow Core Photonic Bandgap Fibre

The first type of hollow-core fibre to be considered here is hollow-core photonic-bandgap fibre (HC-PBF). A photonic-bandgap is a phenomenon that can occur within a dielectric material with a periodically changing refractive index [120]. Certain bands of optical frequency cannot propagate in certain directions within this periodic structure due to Bragg reflections; these bands are known as stop-bands or bandgaps of the structure [121, 122]. A material showing such features is termed a photonic-crystal, and an example this is shown in figure 2.1 where a fibre is constructed from a photonic-crystal.

A HC-PBF uses a two-dimensional photonic-crystal in the transverse plane (i.e perpendicular to the propagation direction), extended along the fibres length to create a photonic-bandgap, in which the core of the fibre is a defect of the photonic-crystal. For frequencies of light within the photonic-crystal’s bandgap, the light is confined within the core of the fibre as there are no available transverse modes of propagation within the photonic-crystal [114, 121, 122]. Hence, it is this photonic-bandgap which guides the light within the core of a HC-PBF. One example of a
HC-PBF design has a photonic-bandgap of $\approx 60$ nm, centred around 800 nm, as shown in figure 2.1. The photonic-bandgaps of each of the fibres used in this thesis are shown in figure 2.3.

The physical scale of the photonic-crystal structure is restricted by the required Bragg reflections within the cladding to create the photonic-bandgap. Although guidance within the core depends on many factors such as: refractive index of the cladding materials, inter-hole spacing $\Lambda$, (usually refered to as pitch); and hole diameter within the cladding [120], a crude estimate suggests that the pitch is on the order of a wavelength of the guided light [121, 122]. A more rigorous description of this can be found in reference [120].

Fibres can be designed to guide a low number of optical modes which can be selected through input mode matching. Such designs place restrictions on the geometry of the hollow-core, although solutions to such design requirements cannot be found analytically as they depend on the physical geometry of the cladding structure in a very complex fashion [120]. However, to maintain the photonic-crystals periodic structure, the core defect is usually created by simply removing unit cells of the photonic-crystal; hence terms like 7-cell defect HC-PBF refers a core defect created by removing 7 photonic-crystal cladding cells. This leads to a loose relationship between the core’s diameter and the cladding’s pitch, hence guiding wavelength. An example, a 7-cell defect HC-PBF designed to guide predominantly in a single mode at 1500 nm has a core diameter of $\approx 10 - 15 \mu$m [123], whereas a 7-cell defect HC-PBF designed to guide at $\approx 610$ nm has a core diameter of $\approx 5 - 10 \mu$m [124]. More thorough descriptions regarding guidance mechanisms within HC-PBF can be found in literature or text books such as reference [120]; as this was not the focus of this thesis, this level of detail will be omitted.

### 2.3 Kagomé Photonic Crystal Fibre

A different type of hollow-core fibre uses a kagomé style cladding, shown in the scanning electron microscope images in figure 2.2. The change in cladding structure from HC-PBF results in a different form of optical guidance. It has been suggested that the guidance mechanism in kagomé style fibre is due to the existence of modes analogous to Von Neumann-Wigner bound states [125].

When compared with HC-PBF, the physical dimensions of kagomé fibre can be much larger with a cladding pitch of typically more than $10 \mu$m due to the different guidance mechanism. As a result, the core sizes of these fibres typically range from $15 \mu$m to $70 \mu$m enabling guidance of a variety of high-order transverse optical
modes; however, single-mode guidance is still attainable [115]. Large core sizes offer advantages when the fibre is to be loaded with gases for gas-sensing applications, due to the higher potential gas throughput, as discussed in chapter 6.3.

The kagomé guidance mechanism also results in a very large window of transmitted frequencies, much larger than available from HC-PBF, for a comparison see figure 2.3. In some cases the transmission window stretches over the entire visible spectrum and into the infra-red (i.e 400 – 1700 nm) with possibly only an island of reduced transmission at 1390 nm due to OH absorption within the fibre [115, 126] which can be removed with careful manufacturing. Such large transmission windows are excellent for super-continuum guidance [126] and interrogation of molecular or atomic gases loaded into the fibre’s core that have resonant transitions at many different wavelengths.

2.4 HC-PCF Fabrication and Characterisation

All the fibres used in the experiments presented in the following chapters were fabricated at the Centre for Photonics and Photonic Materials, located at the University of Bath in the United Kingdom by a research group headed by Fetah Benabid. These fibres are made of pure silica and manufactured using the stack-and-draw technique [127], whereby slender glass capillaries are stacked together to produce the desired fibre structure. The stacked capillaries, measuring roughly 25 mm in diameter, are drawn down in two stages using a fibre drawing tower. There are three major parameters to control during a draw: furnace temperature; draw speed of both the stack in and fibre out; and gas pressure inserted into the cane/fibre (if any), which
can drastically change the final fibre produced. Firstly the capillaries are drawn down to a cane of $\approx 3$ mm diameter. This first draw fuses the capillaries together and prepares the cane for the draw which produces the final fibre with a diameter of $\approx 125 \mu m$. The diameter of the final fibre can be varied, using the above mentioned parameters, to tailor the fibre’s guidance wavelength.

The two fibres predominantly used in the following experiments were a 7-cell HC-PBF and single-cell kagomé HC-PCF. Scanning electron microscope images of these fibres are shown in figure 2.3 (a) and (b) respectively, along with their measured transmission bands. The core diameter of the HC-PBF is $5 \mu m$, whereas the kagomé HC-PCF core measures $45 \mu m$ and $51 \mu m$ for the short- and long-axis respectively. This results in an optical mode with $1/e^2$ intensity diameters of $\approx 4 \mu m$ and $\approx 36 \mu m$.

The divergence of the optical mode emitted from the fibre defines a numerical aperture or NA, for the fibre. The NA is defined as:

$$NA = n \sin(\theta)$$  \hspace{1cm} (2.1)

where $n$ is the refractive index of the material the light is propagating in, for air $n \approx 1$, and $\theta$ is the divergence angle. A typical NA value for a bandgap fibre is $\approx 0.1$ which places constraints on the distance between the fibre and the focusing optics used to couple a laser into the fibre for a given laser beam diameter. On the other hand, kagomé fibres can have a NA of $\approx 0.01$ due to their large core diameters, hence it is possible to couple efficiently into the fibre from a coupling lens far from the fibre.

All fibres used here have low loss guidance from 750 nm to 800 nm, which is required for probing the rubidium atomic transitions described in chapter 3. It can be seen from the transmission spectra in figure 2.3 (b), (c) and (d) that the kagomé HC-PCF has a much broader transmission window when compared to the HC-PBF. Unfortunately, neither of the two fibres predominantly used (figure 2.3 (a) and (b)) guide the 420 nm fluorescence produced by a decay route in rubidium, however, for future work the Centre for Photonics and Photonic Materials has recently produced a fibre which potentially guides at this wavelength, shown in figure 2.3 (c).

A number of other suitable fibres were provided for this project, and scanning electron microscope images of these are shown in figure 2.3 (c) to (e). The transmission or loss spectra provided by the Centre for Photonics and Photonic Materials for each fibre is also shown in Fig (c) through to (e). The loss of a fibre is measured using the cut-back technique where the fibre’s transmission is measured at two different lengths which eliminates in-coupling losses from the measurement. Typically
the length of fibre required to make a cut-back loss measurement is 10 meters for low-loss fibre in order for high-order transverse optical modes to be sufficiently attenuated. Loss spectra of each fibre in these experiments could not be measured because the total length was not sufficient to make cut-back loss measurements. In those cases transmission data is presented.
Figure 2.3: Scanning electron microscope images of the fibres provided by the Centre for Photonics and Photonic Materials (left) and their transmission (red) and loss (blue) spectra (right). The 7-cell Bandgap (a) and Single-cell kagomé (b) were predominantly used, the 7-cell kagomé (c) possibly guides 420 nm light while 19-cell kagomé (d) and 19-cell bandgap (e) are designed to guide at 780 nm light.
Rubidium

The main focus of this thesis is the detailed spectroscopy, and applications of the element Rubidium (Rb). The following chapter summarises the quantum mechanical, physical and chemical characteristics of Rb, which are referred to in the following chapters. The energy level structure and transitions used in this thesis are discussed in section 3.1. Section 3.2 discusses the physical and chemical properties of Rb that affected the procedures used throughout this body of work.

3.1 Energy Level Structure

The energy level structure of Rb is complex due to its high atomic number. However, this complexity can be reduced by considering couplings of the angular momentum and spin of both the valence electron and nucleus. An atom’s energy state can be characterised by its energy level configuration and angular momentum state. The most conventional way of writing this is in the form known as spectroscopic notation which is briefly summarised below.

To fully describe the electronic configuration of an atom, each of the electronic states need to be defined [128]. The gross structure of the atom’s energy levels are represented using the principal quantum number \( n \), which loosely describes size and energy of an electronic shell. Within each electron shell there are sub-shells that are represented by the orbital quantum number \( l \), which describes the shape of the sub-shell’s wave function. The principal quantum numbers are integer values whereas, for historical reasons, the orbital quantum numbers take the letters shown in table 3.1.

| Table 3.1: The code values for an atom’s orbital quantum number state, \( l \). |
|---|---|---|---|---|---|---|---|---|---|---|
| \( l \) | 0 | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | ... |
| s | p | d | f | g | h | i | k | l | m | ... |

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The orbital quantum number can take integer values \(0 \leq l \leq n - 1\) for a given principal quantum number, hence there are \(n\) sub-shells within each electronic shell. Within each sub-shell there are a number of orbitals which describe the orientation of the electron’s wave-function around the nucleus. These are enumerated by the magnetic quantum number \(m_l\). For a given orbital quantum number, the magnetic quantum number can take the values \(-l \leq m_l \leq l\), hence there are \(2l + 1\) orbitals within each electron’s sub-shell. Furthermore, each orbital can also hold two electrons.

Using all this, a full electronic configuration of a ground state atom can be made using the notation \(n l^e\) where, \(e\) denoted the number of electrons within the corresponding sub-shell [128]. For Rb the ground state energy level configuration is \(1s^22s^22p^63s^23p^63d^{10}4s^24p^65s^1\).

**Figure 3.1:** Energy level diagram of Rb showing the fine structure energy level splitting and transitions of concern within this document. Lines connecting energy levels indicate decay/excitation routes with their resonant wavelength marked.

In Rb, and other “hydrogen-like” atoms, the strongest electromagnetic interaction is with the valence shell electron. To fully specify the state of the atom’s valence electron the principal quantum number \(n\), *total electronic* orbital angular momentum \(L\), *total electronic* spin \(S\), and *total electronic* angular momentum number \(J\) are required. The total electronic angular momentum number is a product of the interaction of the electron’s spin and total orbital angular momentum, which is described in more detail in appendix C. The notation most commonly used for con-
3.1. ENERGY LEVEL STRUCTURE

veying the state of the valence electron is $n^2S + 1L_J$ [129], where the letter notation for $L$ is an upper-case version of $l$ discussed previously and shown in table 3.1. For Rb the ground state of the valence electron is $5^2S_{1/2}$ with the first set of excited states being $5^2P_{1/2}$ and $5^2P_{3/2}$. These energy levels are shown in figure 3.1, along with other levels discussed in the following chapters.

In addition to the atomic fine structure described by the total electronic angular momentum $J$, further hyper-fine structure is produced through coupling between the total electronic angular momentum $J$, and total nuclear angular momentum $I$. This produces the total angular momentum quantum number $F$, which is described in more detail in appendix C. Frequency spacings between hyper-fine energy levels range from 1MHz to 10GHz for the atomic transitions considered here, hence are easily resolvable with a narrow line-width laser source. A final source of level splitting is revealed when a magnetic field is applied to the atoms, causing Zeeman splitting of the hyper-fine levels into $2F + 1$ magnetic sub-levels. These levels are labelled with the integer value $m_F$ which ranges from $-F \leq m_F \leq F$. In the absence of an external magnetic field, these magnetic sub-levels are degenerate.

3.1.1 The $D_1$ and $D_2$ Transitions

Two commonly used transitions within Rb are the $5^2S_{1/2} \rightarrow 5^2P_{1/2}$ and $5^2S_{1/2} \rightarrow 5^2P_{3/2}$ transitions which are historically named the $D_1$ and $D_2$ transitions respectively. As shown in figure 3.1 these are transitions to the first excited states of a Rb atom. The $D_1$ transition has a wavelength of 795nm while the $D_2$ transition is resonant at a wavelength of 780nm [130, 131].

For an atomic transition $g \rightarrow e$, the transition strength, characterised by the dipole moment $\mu_{g,e}$, is related directly to the lifetime of the excited energy level $\tau_{e,g}$, through the equation [130, 132, 133]:

$$\mu_{g,e} = \sqrt{\frac{2J_e + 1}{2J_g + 1} \frac{3\pi \epsilon_0 \hbar c^3}{\tau_{e,g} \omega_{ik}}}$$

(3.1)

where $J_e$ and $J_g$ are the total electronic angular momentum quantum numbers for the excited and ground states respectively, and $\omega_{ik}$ is the angular transition frequency. The lifetime of the excited $5^2P_{1/2}$ and $5^2P_{3/2}$ energy levels are 27.679(27)ns and 26.2348(77)ns respectively [130]. This results in a strong dipole moment of $2.9931(20)e\alpha_0$ $(2.5377(17) \times 10^{-29}$ C.m) and $4.22753(87)e\alpha_0$ $(3.58425(74) \times 10^{-29}$ C.m) for the $D_1$ and $D_2$ transitions respectively, where $e$ is the charge of an electron and $\alpha_0$ is a Bohr radius. These transitions strengths result in a $\approx 50\%$ absorption of a
Typical experimental saturated absorption spectra (described in the text) of the $D_1$ (top left) and $D_2$ (bottom left) transitions with corresponding energy level diagrams and atomic transitions (right). Colours are used to match the spectra with atomic transitions.

The short lifetime of these excited states results in relatively broad transition line-widths of 5.7500(56) MHz and 6.0666(18) MHz for the $D_1$ and $D_2$ transitions respectively [130]. Atomic motion within the vapour Doppler shifts the light frequency as observed by the atom, hence the transitions linewidth are Doppler broadened. This is a Gaussian broadening mechanism resulting in a $1/e$ full width of [134]:

$$\Delta \omega_{1/e} = \frac{\omega_{b}}{c} \sqrt{\frac{8 \ln(2) k_B T}{m}} \quad (3.2)$$

where $k_B$ is the Boltzmann-constant, $T$ is the temperature of the gas and $m$ is the atomic mass of Rb. At room temperature ($T = 295$ K) the $1/e$ full width of a Doppler broadened absorption for $^{85}$Rb and $^{87}$Rb is 616 MHz and 609 MHz respectively.

Doppler broadening masks the underlying hyper-fine structure of the excited state. To resolve this structure, and the natural line-width of the $D_1$ and $D_2$ transi-
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tions, a spectroscopic technique known as saturated absorption spectroscopy (SAS) can be employed [133–136]. In this technique, a weak probe beam is passed through a Rb vapour coaxial with a strong, counter-propagating pump beam delivered from the same laser source. The pump and probe beams can interact with the same atoms if they are at rest in the reference frame relative to the directions of the laser beams. This causes optical pumping which reduces the absorption for this specific velocity class of atoms, which reveals both the natural linewidth and the atomic hyperfine structure within the Doppler broadened absorption. A typical saturated absorption spectra is shown in figure 3.2 for both the $D_1$ and $D_2$ transitions. Additional narrow cross-over features are produced as a by-product of this observational technique [134].

3.1.2 The Two-Photon Transition

A two-photon transition is an atomic transition that requires the absorption of two photons to satisfy energy and angular momentum conservation. That is, a transition whereby the valence electron’s orbital angular momentum, $L$, changes by two units. This can only be achieved with absorption of two photons bringing one unit of angular momentum each. In this particular case, the two-photon transition of interest is the $5S_{1/2} \rightarrow 5D_{5/2}$ absorption in Rb. This excited state of Rb has a number of excitation and decay pathways that are shown in figure 3.1. This adds another level of complexity over and above that experienced by the $D_1$ and $D_2$ transitions. These include: multiple possible excitation mechanisms, multiple excitation configurations, and many possible decay routes.

The $5S_{1/2} \rightarrow 5D_{5/2}$ transition can be excited with two photons of 778 nm light, originating from the same laser, which together satisfy energy and angular momentum conservation. Once in the excited state there are a number of decay paths which can be taken, as shown in figure 3.1. The $5D_{5/2} \rightarrow 6P_{1/2} \rightarrow 5S_{1/2}$ decay route produces 420 nm fluorescence, which is commonly used to measure the populations in the $5D$ states. This is because the two-photon transition is typically very weak and thus the direct absorption of the 778 nm beam is minimal. In principle, if the two-photon transition could driven strongly, absorption of the 778 nm beam could be observed.

There are two experimental configurations which can be used to excite the transition: namely the co- and counter-propagating setups shown in figure 3.3. In a co-propagating configuration, Doppler shifts caused by atomic motion produce a
Figure 3.3: Two possible configurations for exciting the two-photon transition (left) and resulting fluorescence spectra (right). Co-propagating (top) is sensitive to the atomic velocity whereas counter-propagating (bottom) is insensitive. This is seen in the corresponding Doppler (red) and Doppler-free (blue) 420 nm fluorescence spectra from the $5S_{1/2} \rightarrow 5D_{5/2}$ transition (right). Note the Doppler-free signal is factor of 100 stronger than the Doppler broadened signal.

Doppler broadened absorption profile, as described for the $D_1$ and $D_2$ transitions above. For the two-photon transition, the $1/e$ Doppler full width is 1.23 GHz, approximately twice that of the $D_1$ and $D_2$ transitions due to the almost doubling in transition frequency. On the other hand, an atom excited by two 778 nm photons from opposite directions demonstrates no velocity dependence. This is seen by considering the frequency of the photons in the atom’s reference frame:

$$\left(\omega_0 + \delta\omega\right) + k_1 \cdot v + \left(\omega_0 + \delta\omega\right) + k_2 \cdot v = 2\omega_0 + 2\delta\omega$$  \hspace{1cm} (3.3)$$

since $k_2 = -k_1$, where $2\omega_0$ is the rest-frame transition frequency, $k$ is the light’s wave-vector, $v$ is the atom’s velocity vector, and $\delta\omega$ is the laser frequency detuning. This shows that there is no longer any velocity dependence, hence all atoms can absorb the counter-propagating beams, unlike the co-propagating configuration, where only a certain velocity class can absorb light, dependent on their velocity. A large increase in absorption strength is observed since all the atoms contribute towards the absorption at a single frequency. In these circumstances, the line width of the transition is limited by the decay rate of the excited state which is $238.5(2.3)$ ns and $246.3(1.6)$ ns for the $5D_{5/2}$ and $5D_{3/2}$ respectively [137], resulting in linewidths of $667.3(6.4)$ kHz and $646.2(4.2)$ kHz.

Instead of using two photons at 778 nm, photons at different wavelengths could be used to excite to the $5D$ state. This potentially delivers a large enhancement through reducing the detuning from an intermediate level, discussed in detail in section 4.4.2. Taking this to the extreme, the two-photon transition can be excited
in a method called step-wise absorption. In this case the atom passes through an intermediate energy level before reaching the two-photon excited state. When exciting in a step-wise configuration, properties of the intermediate state, such as line-width and absorption strength are mapped into the final two-photon spectra.

Unfortunately, using two different wavelengths to excite the two-photon transition compromises the velocity insensitivity described above. Residual Doppler broadening is introduced as the wave-vectors no longer exactly cancel, as shown in equation 3.3. By evaluating equation 3.3 using $k_1 = \omega_1 / c$ and $k_2 = -\omega_2 / c$, the sign is the second term arises from the counter-propagating configuration, the following residual Doppler width is obtained [134, 138]:

$$\Delta \omega_{1/e} = \frac{|\omega_1 - \omega_2|}{c} \sqrt{\frac{8 \ln(2) k_B T}{m}}$$  \hspace{1cm} (3.4)

This is a Gaussian spectra where the $1/e$ full width scales by the fractional frequency difference of the two exciting beams. In the experiments conducted here using 776 nm and 780 nm lasers, equation 3.4 predicts a residual Doppler broadening of $\Delta \omega_{1/e} = 3.3 \text{ MHz}$ for a vapour at $T = 20^\circ \text{C}$.

### 3.2 Physical Properties

Rubidium is an alkali metal that shares many of the physical properties of other elements in this group. These characteristics arise from the physical size of alkali metal atoms which are the largest atoms in their period within the periodic table.
due to their single valence electron being relatively far from the nucleus [128]. The radius of a Rb atom is 248 pm, which is the second largest measured [128], results in a light weight, low density metal. Furthermore, this causes metallic bonds within the metallic lattice to be weak and hence easily deformed resulting in a malleable metal with a low melting point [128]. Rb melts at 39.3°C and boils at 668°C [130], however at room temperature it is a silver solid metal.

There are many isotopes of Rb that can be manufactured, however there are only two which can readily be found in nature, being $^{85}$Rb and $^{87}$Rb. The $^{85}$Rb isotope is stable with 37 protons and 48 neutrons, whereas $^{87}$Rb has 50 neutrons and is technically unstable, having a lifetime of more than $10^{10}$ years. Abundance ratios for these two isotopes are 72.17(2)% and 27.83(2)% for $^{85}$Rb and $^{87}$Rb respectively [130].

### 3.2.1 Reactivity

The valence electron of Rb is weakly bound to the atom, resulting in the atom’s large size and producing the metal’s low melting point, low density and malleability [128]. Such a weak bond makes Rb extremely reactive and a powerful reducing agent, a trait common to all alkali metals. Below are a number of reactions that occur when Rb is exposed to air or the remaining gases within a vacuum system. As a result these reactions need to be considered when attempting to load a HC-PCF with Rb as discussed in chapter 6.3.

Ionic solids are formed when alkali metals reduce the Halogens (F, Cl, Br, I, At) and, in the process, vast quantities of heat are released. When exposed to water, Rb reduces the hydrogen in water to form rubidium hydroxide through the reaction:

$$2\text{Rb} + 2\text{H}_2\text{O} \rightarrow \text{H}_2 + 2\text{RbOH} + \text{heat} \quad (3.5)$$

This reaction is explosive due to the amount of heat and Hydrogen gas produced. A superoxide is formed when Rb reduces Oxygen:

$$\text{Rb} + \text{O}_2 \rightarrow \text{RbO}_2 \quad (3.6)$$

Furthermore, Rb reduces Hydrogen gas to form a hydride:

$$2\text{Rb} + \text{H}_2 \rightarrow 2\text{RbH} \quad (3.7)$$
3.2. PHYSICAL PROPERTIES

Figure 3.5: Vapour pressure of Rb as a function of temperature above and below melting point, indicated by the dashed vertical line.

3.2.2 Vapour Pressure

At room temperature, Rb is a solid metal, however the weak metallic bonds within the metallic lattice allow surface Rb atoms to be ejected from the solid and produce a Rb vapour. The pressure of effusive Rb vapour has been well researched and documented [139–141] through the last seven decades. Empirical models have been developed to characterise the temperature dependence of the Rb vapour pressure. One such model gives the vapour pressure of Rb when in the solid and liquid phase to be [139]:

\[
\begin{align*}
\log_{10}(P) &= 2.881 + 4.857 - \frac{4215}{T} \quad \text{(solid phase)} \\
\log_{10}(P) &= 2.881 + 4.312 - \frac{4040}{T} \quad \text{(liquid phase)}
\end{align*}
\]

which are plotted in figure 3.5. This is the vapour pressure of a naturally occurring sample of Rb, that is 72.17(2)% and 27.83(2)% contribution by weight from $^{85}$Rb and $^{87}$Rb respectively.

An understanding of the temporal evolution of a Rb vapour to a steady-state is required when loading a HC-PCF with Rb, as discussed in section 6.3.2. A Rb vapour within a chemically inert container is in steady-state with its surroundings when the flux of atoms being ejected from, and sticking to, the walls of the containing vessel does not evolve with time [142]. This strongly suggests that each section of the vessel must be coated with Rb before these steady state conditions are attained. Hence, it is expected, and observed, that there is time taken to form a steady-state Rb vapour pressure after insertion of a solid pellet of Rb within a clean,
evacuated vessel. Furthermore, if the vessel’s walls are contaminated with elements or compounds that react with Rb, more time may be taken to coat the walls, and hence form a steady-state Rb vapour due to chemical reactions. An example of such a situation would be a vacuum chamber with residual water and hydrogen within it. Both of these situations may seem trivial, however these are important issues to consider when loading a HC-PCF with Rb, as is shown in section 6.3.2.
Part II

Theory
This chapter introduces the mathematical constructs used here to model how light interacts with atoms. Also introduced is the formalisms used to move from this mathematical basis to the experimental results. The first section of this chapter introduces the density matrix and the formalism which governs its temporal evolution. The second section makes a connection between the density matrix and observable quantities. Various general density matrix models are introduced in section 4.3 which are then extended to the Rb transitions in section 4.4. Finally sections 4.5 and 4.6 respectively introduce methods to numerically solve the density matrix equations and calculate the observed spectra from a Gaussian beam interrogating an effusive vapour. The formalism derived in this chapter is used throughout the remainder of this body of work. The work below closely follows and extends upon that presented in references [143–148].

4.1 The Density Matrix

The density matrix is a mathematical construct used for calculating the populations of, and coherences between, atomic energy levels in an optically driven atomic system. More broadly, it is not constrained to a single light-atom system but is capable of handling an ensemble of atoms being excited by a laser. Using this construct observable quantities can be calculated, such as: absorption spectra; dipole moments; and energy level populations. From this, phenomena such as: power broadening; electromagnetically induced transparency; Doppler broadening; and transit-time broadening can be modelled and compared to experimental results.

The density matrix, typically labelled $\rho$, is defined as a matrix where the atomic energy level populations appear on the diagonal matrix elements, with the off diagonal elements representing coherences between energy levels [143–148]. Labelling
convention has the population of the state $|j\rangle$ level labelled as $\rho_{j,j}$, and the coherence between energy levels $|j\rangle$ and $|k\rangle$ written as $\rho_{j,k}$. The density matrix for a two-level system is hence:

$$\rho = \begin{pmatrix} \rho_{j,j} & \rho_{j,k} \\ \rho_{k,j} & \rho_{k,k} \end{pmatrix} \quad (4.1)$$

The dynamics of an atom’s energy level populations and coherences under the influence of light is governed by an interaction Hamiltonian $H$ [143–148]. This is the sum of the unperturbed atomic Hamiltonian $H_0$, and the electromagnetic interaction Hamiltonian $H_V(t)$:

$$H = H_0 + H_V(t) \quad (4.2)$$

The equations of motion for the density matrix are defined through the commutation of the interaction Hamiltonian with the density matrix

$$\dot{\rho} = -\frac{i}{\hbar} [H, \rho] \quad (4.3)$$

where the commutator $[A, B]$ is defined as $[A, B] = A.B - B.A$. Equation 4.3 is derived from Schrödinger’s equation, see [143–147].

Spontaneous decay from any energy level is not included in equation 4.3, to include this the equation is modified to [149]:

$$\dot{\rho} = -\frac{i}{\hbar} [H, \rho] + \sum_{j,k} \gamma_{j,k} D(|j\rangle\langle k|)\rho \quad (4.4)$$

where $\gamma_{j,k}$ is the decay rate linking state $|j\rangle$ to $|k\rangle$, mathematically implemented via the operator $D$ which is defined as $D[b]\rho = bpb\dagger - (b\dagger bp + \rho b\dagger b)/2$ for the atomic state $b$ [149].

### 4.2 From Density Matrix to Experiment

The density matrix models presented in the following chapters are either analytically or numerically solved under a variety of different circumstances. To relate these solutions to physically observable quantities requires further formalism to be derived. The observable quantity which will be focused on here is the absorption depth which, once related to the density matrix formalism, can be used to model many spectroscopic features including: transition linewidths, optical pumping, and electromagnetically induced transparency, which are all discussed later. Two sepa-
rate derivations are presented. The first connects the density matrix to the electric susceptibility, refractive index and finally the absorption coefficient. The second simply counts the photons being scattered from the laser and the corresponding atom dynamics.

4.2.1 Refractive Index Derivation

The absorption coefficient $\alpha(\omega)$ and phase shift $\phi(\omega)$ of a laser beam interrogating an atomic transition from ground $|g\rangle$ to the excited state $|e\rangle$ are defined as [143–147]:

$$\alpha(\omega) = 2\frac{\omega}{c} \Im [\tilde{n}(\omega)]$$  
$$\phi(\omega) = \frac{\omega}{c} (\Re [\tilde{n}(\omega)] - 1)$$

where $\omega$ is the angular frequency of the laser, $c$ is the speed of light, and $\tilde{n}(\omega)$ is the complex refractive index of the material. To be able to convert between theory and observation easily, an expression for the absorption length in terms of the density matrix components is required. To begin with, the refractive index is related to the complex electric susceptibility, $\chi^{(e)}$, via:

$$\tilde{n}(\omega) = \sqrt{1 + \chi^{(e)}(\omega)}$$

The electric susceptibility is in turn related to the atomic polarisability, $\mathcal{P}$ via:

$$\mathcal{P} = \epsilon_0 E_0 \chi^{(e)}(\omega)$$

where $\epsilon_0$ is the vacuum permittivity and $E_0$ is the interrogating laser’s electric field amplitude [143–148]. Once positive and negative frequency components are taken into account, the following relation is attained [143–146]:

$$\mathcal{P} \equiv \frac{\epsilon_0 E_0}{2} \left( \chi^{(e)}(\omega)e^{-i\omega t} + \chi^{(e)}(-\omega)e^{i\omega t} \right)$$

The atomic polarisability is also defined in relation to the expectation value of the induced dipole moment $\langle \mu \rangle$. By noting that an operator $O$ has an expectation value defined as $\langle O \rangle = \text{Tr} [\rho O]$, the atomic polarisability can be expressed as [143–146]:

$$\mathcal{P} \equiv N \langle \mu \rangle$$

$$= N (\tilde{\rho}_{g,e}(\omega) \mu_{e,g} + \tilde{\rho}_{e,g}(\omega) \mu_{g,e})$$

where $N$ is the number density of the atoms and $\tilde{\rho}_{g,e}$ and $\tilde{\rho}_{e,g}$ are coherences between the ground and excited state.

By finally noting that the coherence terms can be separated into rotating and
CHAPTER 4. LIGHT-ATOM INTERACTIONS

stationary terms, $\rho_{e,g}(\omega) = \tilde{\rho}_{e,g}(\omega)e^{i\omega t}$ (described in more detail in section 4.3.1), and that $\mu_{e,g} = \mu_{g,e}$, equations 4.8 and 4.9 can be equated:

$$N\mu_{g,e}(\tilde{\rho}_{g,e}(\omega)e^{-i\omega t} + \tilde{\rho}_{e,g}(\omega)e^{i\omega t}) = \frac{\epsilon_0 E_0}{2} \left( \chi^{(e)}(\omega)e^{-i\omega t} + \chi^{(e)}(-\omega)e^{i\omega t} \right)$$  \hspace{1cm} (4.10)

This leads to an expression for the electric susceptibility in terms of the density matrix coherence terms [143–146]:

$$\chi^{(e)}(\omega) = \frac{2N\mu_{g,e}}{\epsilon_0 E_0} \tilde{\rho}_{g,e}(\omega)$$ \hspace{1cm} (4.11a)

$$= \frac{2N\mu_{g,e}^2}{\hbar \epsilon_0 \Omega} \tilde{\rho}_{g,e}(\omega)$$ \hspace{1cm} (4.11b)

$$= \frac{Nc\hbar\Omega}{I} \tilde{\rho}_{g,e}(\omega)$$ \hspace{1cm} (4.11c)

where substitutions were made for the Rabi frequency $\Omega$ and optical intensity $I$, assuming $n \approx 1$, via [143–148]:

$$\Omega \equiv \frac{\mu E_0}{\hbar}$$ \hspace{1cm} (4.12)

$$I \equiv \frac{c\epsilon_0 n E_0^2}{2}$$ \hspace{1cm} (4.13)

To arrive at an expression for the absorption coefficient in terms of density matrix coherences, the complex refractive index can be simplified by assuming $\chi^{(e)}(\omega)$ is small (i.e. for a dilute vapour). This enables the refractive index to be simplified to $\tilde{n}(\omega) = \sqrt{1 + \chi^{(e)}(\omega)} \approx 1 + \chi^{(e)}(\omega)/2$. Hence, from equation 4.5 the absorption coefficient of the atomic vapour can be written as:

$$\Rightarrow \alpha(\omega) = 2\frac{\omega}{c} \Re [\tilde{n}(\omega)]$$ \hspace{1cm} (4.14a)

$$= \frac{\omega}{c} \Re \left[ \chi^{(e)}(\omega) \right]$$ \hspace{1cm} (4.14b)

$$= N\omega \hbar I \Re \left[ \tilde{\rho}_{g,e}(\omega) \right]$$ \hspace{1cm} (4.14c)

A similar definition can be found for the phase-shift induced by the atomic transition:

$$\Delta \phi(\omega) = \frac{\omega}{c} \left( \Re [\tilde{n}(\omega)] - 1 \right)$$ \hspace{1cm} (4.15a)

$$= \frac{\omega}{c} \Re \left[ \chi^{(e)}(\omega) \right]$$ \hspace{1cm} (4.15b)

$$= N\omega \hbar I \Re \left[ \tilde{\rho}_{g,e}(\omega) \right]$$ \hspace{1cm} (4.15c)
The observed absorption of the laser is finally related to the absorption coefficient through the Beer-Lambert law [143–148]:

\[ I(\omega, z) = I(\omega, 0)e^{-\alpha(\omega)z} \]  

(4.16)

where \( z \) is the absorption path length.

### 4.2.2 Photon Number Derivation

An alternative derivation can be made by tracking the changes of the laser beam’s photon number per second \( N \), and comparing this to the changes of atomic ground state population [149]. First, let’s consider the rate of scattering of the laser beam’s photons \( N_{\text{Scat}} \). Photons are lost from the beam through atomic absorption from the ground state to the excited state, \( |g\rangle \to |e\rangle \), at a rate of \( N_{\text{Abs}}(g \to e) \). Stimulated fluorescence from \( |e\rangle \to |g\rangle \) re-emits photons back into the beam at a rate of \( N_{\text{Stim}}(e \to g) \). This results in the rate of change in the laser beams photon number as:

\[ N_{\text{Scat}} = N_{\text{Abs}}(g \to e) - N_{\text{Stim}}(e \to g) \]  

(4.17)

Accompanying the absorption and scattering of laser photons are population changes in the atomic ensemble. The number of atoms in the ground state \( M_{g,g} \) experiences a loss of population due to excitation from \( |g\rangle \) to \( |e\rangle \) through photon absorption, occurring at a rate of \( m_{\text{Abs}}(g \to e) \). Atomic ground state population is increased through either decay from \( |e\rangle \to |g\rangle \) at a rate of \( m_{\text{Scat}}(e \to g) \); or though undergoing stimulated fluorescence occurring at a rate of \( m_{\text{Stim}}(e \to g) \). It should be noted that the units of \( M_{g,g} \) and \( m \) are population and population per second respectively, i.e \( m \) is the rate of population change. This results in overall rate change of ground state population given by:

\[ \frac{dM_{g,g}}{dt} = m_{\text{Stim}}(e \to g) + m_{\text{Scat}}(e \to g) - m_{\text{Abs}}(g \to e) \]  

(4.18)

The rate of atoms decaying from \( |e\rangle \to |g\rangle \) scales with the total decay rate, \( \Gamma_e^{(\text{Tot})} \), of the excited state [143–148]:

\[ m_{\text{Scat}}(e \to g) = (NA \, dz) \, \Gamma_e^{(\text{Tot})} \tilde{\rho}_{e,e} \]  

(4.19)

where \((NA \, dz)\) is the number of atoms within a laser beam with an area of \( A \) and length \( dz \) for an atomic vapour of density \( N \). The density matrix element \( \tilde{\rho}_{e,e} \) represents the probability of an atom being in the excited state. Inserting this into...
equation 4.18, noting that $\mathcal{M}_{g,g} = (NA dz) \tilde{\rho}_{g,g}$, results in the following:

$$NA dz \left( \frac{d\tilde{\rho}_{g,g}}{dt} - \Gamma_e^{(\text{Tot})} \tilde{\rho}_{e,e} \right) = m_{\text{Stim}}(e \rightarrow g) - m_{\text{Abs}}(g \rightarrow e)$$ \hspace{1cm} (4.20)

Furthermore, evaluating equation 4.4 with the Hamiltonian described in the next section, equation 4.33, yields the following expression:

$$\frac{d\tilde{\rho}_{g,g}}{dt} - \Gamma_e^{(\text{Tot})} \tilde{\rho}_{e,e} = - \frac{i}{2} \Omega (\tilde{\rho}_{g,e} - \tilde{\rho}_{e,g})$$ \hspace{1cm} (4.21a)

$$= \Omega \Im \{\tilde{\rho}_{g,e}\}$$ \hspace{1cm} (4.21b)

where $\Omega$ is the Rabi frequency of the probing laser. By combining equations 4.20 and 4.21b, the following is attained:

$$m_{\text{Stim}}(e \rightarrow g) - m_{\text{Abs}}(g \rightarrow e) = NA \Omega dz \Im \{\tilde{\rho}_{g,e}\}$$ \hspace{1cm} (4.22)

To relate this to the change in the laser’s photon number, the connection that atoms are only excited by the laser’s photons is required, hence $\mathcal{N}_{\text{Abs}}(g \rightarrow e) \equiv m_{\text{Abs}}(g \rightarrow e)$. Also the number of photons scattered back into the beam is only caused by atoms undergoing stimulate fluorescence $\mathcal{N}_{\text{Stim}}(e \rightarrow g) \equiv m_{\text{Stim}}(e \rightarrow g)$. Again, it is important to note that $\mathcal{N}$ and $m$ are both rates of change of the photon number and atomic population respectively. This allows the rate of change of the photon number in the laser beam to be defined as:

$$\mathcal{N}_{\text{Scat}} = -(m_{\text{Stim}}(e \rightarrow g) - m_{\text{Abs}}(g \rightarrow e))$$ \hspace{1cm} (4.23a)

$$= -NA \Omega dz \Im \{\tilde{\rho}_{g,e}\}$$ \hspace{1cm} (4.23b)

Hence, the fraction of photons scattered from a laser beam with photon flux of $\mathcal{N}_{\text{Tot}} = P/(\hbar \omega)$, where $P$ is the laser’s power and $\hbar \omega$ is the photon energy, is:

$$\frac{\mathcal{N}_{\text{Scat}}}{\mathcal{N}_{\text{Tot}}} = - \frac{NA \Omega dz}{P/(\hbar \omega)} \Im \{\tilde{\rho}_{g,e}\}$$ \hspace{1cm} (4.24a)

$$= -N \omega \hbar dz \frac{\Omega}{I} \Im \{\tilde{\rho}_{g,e}(\omega)\}$$ \hspace{1cm} (4.24b)

$$\Rightarrow \frac{\mathcal{N}_{\text{Scat}}}{\mathcal{N}_{\text{Tot}}} = -\alpha(\omega) dz$$ \hspace{1cm} (4.24c)

since $I = P/A$. To account for attenuation through a length of vapour $z$, equation 4.24c is rewritten so that $\mathcal{N}_{\text{Scat}} \rightarrow d\mathcal{N}(z)$ and $\mathcal{N}_{\text{Tot}} \rightarrow \mathcal{N}(z)$. From here
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integration is used to derive the photon number after a vapour length of $z$:

$$\frac{d\mathcal{N}(z)}{dz} = -\alpha(\omega)\mathcal{N}(z) \tag{4.25a}$$

$$\mathcal{N}(\omega, z) = \mathcal{N}(\omega, 0)e^{-\alpha(\omega)z} \tag{4.25b}$$

$$\Rightarrow I(\omega, z) = I(\omega, 0)e^{-\alpha(\omega)z} \tag{4.25c}$$

The result in equation 4.25c is better known as the Beer-Lambert law [143–148].

4.3 Various Generic Models

This section discusses the general two and three-level density matrix models, which are the simplest representation of real atomic systems. A number of simple observable effects are derived and limitations of these models are discussed. Both of these representations are the basis for the Rb models presented in section 4.4 and extensively studied in chapter 5.

4.3.1 Two-Level Model

The simplest non-trivial atom-light system is a two-level atom interacting with a laser. Much insight can be gained through solving this system in its steady state form. The model consists of a ground state and excited state, labelled $|g\rangle$ and $|e\rangle$ respectively, which, when unperturbed, are separated by an energy $\delta = \hbar\omega$. A laser with detuning $\Delta$ and Rabi frequency $\Omega$ drives the $|g\rangle \rightarrow |e\rangle$ transition. The atom decays from $|e\rangle \rightarrow |g\rangle$ at a rate $\gamma$ as shown in figure 4.1. The density matrix corresponding to this system is:

$$\rho = \begin{pmatrix} \rho_{g,g} & \rho_{g,e} \\ \rho_{e,g} & \rho_{e,e} \end{pmatrix} \tag{4.26}$$

The electromagnetic interaction Hamiltonian, $H_V(t)$, is defined by the laser light that the atom experiences. An electromagnetic wave of frequency $\omega$ and amplitude $E_0$ is defined as $E(t) = E_0 \cos(\omega t) = E_0/2(e^{i\omega t} + e^{-i\omega t})$, which induces a potential of the form $V(t) = \mu E(t)$ where $\mu$ is the dipole moment of the atomic transition [145]. This results in an interaction potential of the form:

$$V(t) = \frac{\mu E_0}{2}e^{i\omega t} + \frac{(\mu E_0)^*}{2}e^{-i\omega t} \tag{4.27a}$$

$$= \frac{\hbar\Omega}{2}e^{i\omega t} + \frac{\hbar\Omega^*}{2}e^{-i\omega t} \tag{4.27b}$$
where $\Omega$ is the Rabi frequency of the incident light defined by equation 4.12. The interaction Hamiltonian is defined using equation 4.3 to be [145]:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_V(t)$$

$$= \begin{pmatrix} 0 & V(t) \\ \hbar\omega_e & 0 \end{pmatrix} - \begin{pmatrix} 0 & V(t)^* \\ \hbar\omega_e & 0 \end{pmatrix}$$

$$= -\frac{\hbar}{2} \begin{pmatrix} 0 & \Omega e^{i\omega t} + \Omega^* e^{-i\omega t} \\ 0 & \Omega^* e^{-i\omega t} - \Omega e^{i\omega t} \end{pmatrix}$$

Before the equations of motion can be solved (equation 4.3) a number of simplifications can be made.

The first approximation to be made is the rotating wave approximation (RWA), in which rapidly rotating parts of the Hamiltonian are ignored. Specifically, components rotating at frequencies on the order of $\omega$ are ignored. The physical interpretation is that rapidly oscillating components of the atom are associated with far off-resonance dynamics [130, 147, 150] which are not of concern here. To apply this approximation, it is convenient to change into a basis which is rotating with the induced atomic dipole moment. This is achieved through transforming both the Hamiltonian and density matrix through a unitary transformation matrix [147]:

$$U = \begin{pmatrix} 1 & 0 \\ 0 & e^{-i\omega t} \end{pmatrix}$$

The density matrix in this new basis $\tilde{\rho}$ is related back to the original via [145, 147]:

$$\rho = U \tilde{\rho} U^\dagger = U \begin{pmatrix} \tilde{\rho}_{g,g} & \tilde{\rho}_{g,e} \\ \tilde{\rho}_{e,g} & \tilde{\rho}_{e,e} \end{pmatrix} U^\dagger = \begin{pmatrix} \tilde{\rho}_{g,g} e^{i\omega t} & \tilde{\rho}_{g,e} \\ \tilde{\rho}_{e,g} e^{-i\omega t} & \tilde{\rho}_{e,e} \end{pmatrix}$$

where it can be seen that the coherence terms are split into fast rotating ($e^{i\omega t}$) and slowly varying terms $\tilde{\rho}$. 

---

**Figure 4.1:** Energy level diagram of a two-level atom with a driving laser of strength $\Omega$, frequency detuned from resonance by $\Delta$. The excited energy level $|e\rangle$ has a decay rate of $\gamma$ to the ground state $|g\rangle$. 

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CHAPTER 4. LIGHT-ATOM INTERACTIONS
4.3. VARIOUS GENERIC MODELS

To satisfy the Schrödinger equation, the Hamiltonian in the rotating basis \( \tilde{H} \) takes the form [147]:

\[
\tilde{H} = U^\dagger H U - i \hbar U^\dagger \frac{\partial U}{\partial t} \tag{4.31}
\]

while an operator is typically transformed as \( \tilde{O} = U^\dagger O U \). Applying this transformation to \( H \) (as defined in equation 4.28c) gives the Hamiltonian in the rotating basis as:

\[
\tilde{H} = \begin{cases} 
0 & -\hbar \Omega (1 - e^{-2i\omega t}) / 2 \\
-\hbar \Omega (1 - e^{2i\omega t}) / 2 & \hbar \Delta 
\end{cases} \tag{4.32}
\]

where \( \Delta \equiv \omega - \omega_e \) is the optical detuning from atomic resonance. It can now clearly be seen that there are components rapidly rotating at a frequency of \( 2\omega \). At this point the RWA can be applied by setting \( e^{\pm 2i\omega t} \to 0 \) which gives the following Hamiltonian [147]:

\[
\tilde{H} = \begin{cases} 
0 & -\hbar \Omega / 2 \\
-\hbar \Omega / 2 & \hbar \Delta 
\end{cases} \tag{4.33}
\]

Using this simplified Hamiltonian, equation 4.3 gives the following set of first order differential equations:

\[
\begin{pmatrix}
\dot{\tilde{\rho}}_{g,g}(t) \\
\dot{\tilde{\rho}}_{g,e}(t) \\
\dot{\tilde{\rho}}_{e,g}(t) \\
\dot{\tilde{\rho}}_{e,e}(t)
\end{pmatrix} =
\begin{pmatrix}
0 & -\frac{1}{2}i\Omega & +\frac{1}{2}i\Omega & \gamma \\
-\frac{1}{2}i\Omega & -\frac{1}{2}\gamma + i\Delta & 0 & +\frac{1}{2}i\Omega \\
+\frac{1}{2}i\Omega & 0 & -\frac{1}{2}\gamma - i\Delta & -\frac{1}{2}i\Omega \\
0 & +\frac{1}{2}i\Omega & -\frac{1}{2}i\Omega & -\gamma
\end{pmatrix}
\begin{pmatrix}
\tilde{\rho}_{g,g}(t) \\
\tilde{\rho}_{e,e}(t) \\
\tilde{\rho}_{e,g}(t) \\
\tilde{\rho}_{e,e}(t)
\end{pmatrix} \tag{4.34}
\]

The steady state solution, \( \tilde{\rho}(t) = 0 \), to the above equations gives an insight to many physically observable quantities. Recognising that population must be conserved within the atomic system \( \tilde{\rho}_{g,g} + \tilde{\rho}_{e,e} = 1 \), the solutions to the steady state equations are:

\[
\tilde{\rho}_{g,g} = \frac{2\gamma (\Gamma^2 + \Delta^2) + \Gamma \Omega^2}{2\gamma (\Gamma^2 + \Delta^2) + 2\Gamma \Omega^2} \tag{4.35a}
\]

\[
\tilde{\rho}_{e,e} = \frac{\Gamma \Omega^2}{2\gamma (\Gamma^2 + \Delta^2) + 2\Gamma \Omega^2} \tag{4.35b}
\]

\[
\tilde{\rho}_{g,e} = -\frac{i\Omega (\tilde{\rho}_{g,e} - \tilde{\rho}_{e,e})}{2(\Gamma - i\Delta)} \tag{4.35c}
\]

\[
= \frac{\gamma \Omega (\Delta - i\Gamma)}{2\gamma (\Gamma^2 + \Delta^2) + 2\Gamma \Omega^2} \tag{4.35d}
\]

where the density matrix elements are now time independent and the coherence decay rate \( \Gamma = \gamma/2 \) was introduced. A separate variable is assigned to the coherent
and population decay rates as these are not necessarily related. For example, atomic collisions can cause a loss of coherence with no population perturbation, hence increasing the coherence decay rate and not the population decay rate. The above solutions can now be used to extract measurable quantities, such as the intensity at which the absorption of the atomic vapour saturates, or the linewidth.

At a specific intensity the atomic populations will be perturbed so much that the resonant absorption coefficient of the incident laser will drop to \(1/2\) its low power absorption level. The optical intensity required to reach this point is referred to as the saturation intensity \(I_{\text{sat}}\) of the atom [145, 146]. To explore this condition, a ratio of the absorption coefficient as a function of intensity \(\alpha(I)\) is made to the zero intensity absorption coefficient. Making use of equation 4.35c yields the following result:

\[
\frac{\alpha(I, \Delta)}{[\alpha(I, \Delta)]_{E \to 0}} = \frac{\frac{\Omega}{2} \text{Im} \left[ \hat{\rho}_{e,g}(I) \right]}{\left[ \frac{\Omega}{2} \text{Im} \left[ \hat{\rho}_{e,g}(I) \right] \right]_{E \to 0}}
= \frac{\Omega^2}{I} \frac{\hat{\rho}_{g,e} - \hat{\rho}_{g,g}}{2(\Gamma^2 + \Delta^2)}
= \rho_{g,g} - \rho_{e,e}
= \frac{1}{1 + \frac{\Omega^2}{g(\Gamma^2 + \Delta^2)}}
= \frac{1}{1 + I/I_{\text{sat}}(\Delta)}
\]

where simplification was made by substituting the Rabi frequency and intensity (equations 4.12 and 4.13) into the above equations. The final substitution was made by noticing that \(\Omega^2 \propto I\) as seen in equations 4.12 and 4.13 and all the remaining coefficients were placed into the function \(I_{\text{sat}}(\Delta)\). It can be seen that in the limit that \(I \to I_{\text{sat}}(\Delta)\) then \(\alpha(I_{\text{sat}}(\Delta)) = \alpha(0)/2\), which satisfies the definition of \(I_{\text{sat}}\).

By comparing equations 4.36d and 4.36e an analytic expression for \(I_{\text{sat}}\) is gained:

\[
I_{\text{sat}}(\Delta) = \frac{c(\Gamma^2 + \Delta^2) \epsilon_0 \hbar^2}{\mu^2}
\]

Typically the saturation intensity is quoted for an on-resonance laser \((\Delta = 0)\), which results in the following, on-resonance, definition of \(I_{\text{sat}}\) [130, 145]:

\[
I_{\text{sat}}(0) = \frac{c \Gamma^2 \epsilon_0 \hbar^2}{\mu^2}
\]

The absorption behaviour related to this saturation is shown in figure 4.2.
are two clear limits: The first is where $I \ll I_{\text{sat}}(\Delta)$, where the absorption is constant as the atomic populations are not substantially perturbed; for $I \gg I_{\text{sat}}(\Delta)$, the fractional absorption decreases due to substantial atomic population being pushed to the excited state, reducing the number of atoms available to absorb and increasing stimulated emission. Saturation in this manner is easily observable and measurements of this type are presented in chapter 7 for both a Rb vapour in a bulk cell and loaded within the HC-PCF.

Another relationship can be extracted from equations 4.36d and 4.36e to express the ratio $I/I_{\text{sat}}$ in relation to the Rabi frequency and natural atomic decay. On resonance ($\Delta = 0$) these equations can be arranged to give:

$$\frac{I}{I_{\text{sat}}(0)} = 2 \left( \frac{\Omega}{\gamma} \right)^2$$

(4.39)

This is a direct relationship between the optical intensity as a fraction of the saturation intensity, and the Rabi frequency induced in the atom. Equation 4.39 is very useful for relating experimental and theoretical parameters and will be used frequently in this work.

This simple two-level model can still reveal more interesting phenomenon. Specifically, the linewidth of the absorption spectra for an atom can be easily determined. Through rearranging the normalised absorption coefficient given by equation 4.36 the following absorption spectrum can be extracted

$$\frac{\alpha(I, \Delta)}{[\alpha(I, 0)]_{E \rightarrow 0}} = \frac{\gamma^2}{\gamma^2 + 2\Omega^2 + 4\Delta^2}$$

(4.40)
This absorption spectra takes the form of a Lorentzian profile with an amplitude of the form described in equation 4.36e.

The full width at half maximum (FWHM) of the Lorentzian profile is given by:

$$\Delta \omega_{\text{FWHM}} = \gamma \sqrt{1 + 2 \left( \frac{\Omega}{\gamma} \right)^2}$$  \hspace{1cm} (4.41)

$$= \gamma \sqrt{1 + \frac{I}{I_{\text{sat}}}}$$  \hspace{1cm} (4.42)

where the last line uses the relationship in equation 4.39. The width of the spectral absorption is dependent on the incident laser power, hence broadening of this nature is termed power broadening. As the optical power tends towards zero the spectral FWHM tends towards $\gamma$, where the absorption linewidth is limited by the lifetime of the excited state. Figure 4.3 shows example spectra for a variety of input laser intensities to demonstrating the effect of power broadening.

4.3.2 Three-Level Model

The three-level model is important to consider as it is often used for modelling population dynamics within a Rb vapour for which a two-level model is inadequate. To properly represent the population dynamic, both ground states (discussed in chapter 3) and an excited state of Rb are required. The two ground state energy levels are labelled $|g_1\rangle$ and $|g_2\rangle$ which are separated by an energy $\ell$ as shown in figure 4.4. The upper state, $|e\rangle$, has separate decay paths to each ground state $\gamma_{e,g1}$, $\gamma_{e,g2}$. A laser field can connect either the $|g_1\rangle \rightarrow |e\rangle$ or $|g_2\rangle \rightarrow |e\rangle$ transition, for this
4.3. VARIOUS GENERIC MODELS

example, the former is used.

\[ \dot{\rho} = -\frac{i}{\hbar} \left[ \hat{H}, \rho \right] + \gamma_{e,g_1} \mathcal{D}[|e\rangle \langle g_1|] \rho + \gamma_{e,g_2} \mathcal{D}[|e\rangle \langle g_2|] \rho \]  

\[ \hat{H}_I = \begin{pmatrix} 0 & 0 & -\hbar \Omega/2 \\ 0 & \mathcal{E} & 0 \\ -\hbar \Omega/2 & 0 & \hbar \Delta \end{pmatrix} \] (4.43)

\[ \dot{\rho} = -\frac{i}{\hbar} \left[ \hat{H}, \rho \right] + \gamma_{e,g_1} \mathcal{D}[|e\rangle \langle g_1|] \rho + \gamma_{e,g_2} \mathcal{D}[|e\rangle \langle g_2|] \rho \] (4.44)

The steady state solution for this set of equations is trivial. This is due to the fact that there are two decay paths from the excited state. All the population from the probed ground state, \( |g_1\rangle \), will eventually decay from the excited state, \( |e\rangle \), to the other ground state \( |g_2\rangle \). Since there is no population transfer between ground states through decay mechanism the population pools in \( |g_2\rangle \) leading to a steady state solution of \( \dot{\rho}_{g_1,g_1} = \dot{\rho}_{e,e} = 0 \) and \( \dot{\rho}_{g_2,g_2} = 1 \) for all detunings \( \Delta \) resulting in no absorption of the laser. Hence, in steady state no information can be gained about absorption processes in the system.

This behaviour is called optical pumping as all the population is pumped from \( |g_1\rangle \) to \( |g_2\rangle \). In a thermal Rb vapour, optical pumping is continually occurring which makes the vapour transparent to the laser. However, the rapid motion of atoms through the laser beam continually replenishes the pumped atoms generating substantial absorption in steady state. Such a steady state solution can be obtained by including extra re-population rates into the ground state populations [145].

On the other hand, information can be gained from this model by considering the time dependent solution discussed in section 4.5. Analytic and numerical solutions using the three-level model are used to model transit-time broadening of Rb vapour in chapter 5.1.1.

Figure 4.4: Energy level diagram of a three-level atom with a driving laser of strength \( \Omega \), detuned from resonance by \( \Delta \). The excited energy level \( |e\rangle \) has decay rates of \( \gamma_{e,g_1} \) and \( \gamma_{e,g_2} \) to the ground states \( |g_1\rangle \) and \( |g_2\rangle \) respectively.
CHAPTER 4. LIGHT-ATOM INTERACTIONS

4.4 Models of Rubidium

To verify experimental observations, a number of atom-light models were created, that were tailored to the specific experimental configurations. These models, some simple, some more complex, gave great insight into the atomic behaviour that was observed. As such, these specific models are worth discussing in more detail.

4.4.1 Hole Burn Experimental Model

One of the major experiments conducted to determine spectral broadening mechanisms within the HC-PCF was to observe Doppler free features on the single-photon $D_1$ and $D_2$ transitions. An energy level diagram of this setup is shown in figure 4.5. A velocity class of atoms, within the Doppler velocity distribution, was optically pumped from the $|g_2\rangle$ to the $|g_1\rangle$ ground state with a fixed laser on the $D_1$ transition. The effect of this was to create a spectral hole within the Doppler broadened $D_2$ transition due to an absence of atoms at this velocity when a probe laser scanned across the $|g_2\rangle \rightarrow |b\rangle$. Accompanying these spectral holes are features that exhibit stronger absorption than normal when observing the $|g_1\rangle \rightarrow |b\rangle$ transition. This is due to an increase in atomic population in the $|g_1\rangle$ ground state due to optical pumping. The theoretical aspects of this experiment are discussed here without need of the experimental details which are discussed in further depth in chapter 7.

To adequately capture the dynamics of this system, a four level density matrix is required with two coupling lasers. A strong laser couples the ground state $|g_2\rangle$ to the excited state $|a_i\rangle$, this transition is the optically pumped transition. Upper state hyperfine levels in Rb are labeled through the subscript $i$, however, in this modeling only one upper state level is considered, hence, $|a_i\rangle$ will be written as $|a\rangle$. A weak laser probes the velocity distribution of atoms on the $|g_2\rangle \rightarrow |b_j\rangle$ or $|g_1\rangle \rightarrow |b_j\rangle$ transitions. Again the subscript $j$ labels upper state hyperfine levels, here only one level will be considered hence $|b_j\rangle$ will be written as $|b\rangle$.

The Hamiltonians that describe this energy level and coupling laser configuration are defined as:

$$\mathcal{H}_0 = |a\rangle \langle a| \Delta_p + |b\rangle \langle b| \Delta_s$$
$$\mathcal{H}_V = \frac{\Omega_p}{2} (|a\rangle \langle g_2| + |g_2\rangle \langle a|) + \frac{\Omega_s}{2} (|b\rangle \langle g_1 + g_2| + |g_1 + g_2\rangle \langle b|)$$

where $\Delta_p$ and $\Delta_s$ are, respectively, the pump and signal/probe detuning from their transition frequencies and $\Omega_p$ and $\Omega_s$ are the pump and signal/probe Rabi frequen-
4.4. MODELS OF RUBIDIUM

Figure 4.5: a) Transmission of a probe laser interrogating the $D_2$ transition. Spectral holes are visible due to optical pumping on the $D_1$ transition. b) Energy level diagram showing the $D_1$ and $D_2$ transitions with coupling pump and probe lasers.

Equation 4.45b is written so that the signal laser can be scanned across both ground state levels, hence the appearance of $|g_1 + g_2\rangle$. From these Hamiltonians the equations of motion for this system is written as:

$$\dot{\rho} = -\frac{i}{\hbar} [\mathcal{H}_0 + \mathcal{H}_V, \rho] + \gamma_{a,g_1} \mathcal{D} [a] \langle g_1 | \rho + \gamma_{a,g_2} \mathcal{D} [a] \langle g_2 | \rho$$

$$+ \gamma_{b,g_1} \mathcal{D} [b] \langle g_1 | \rho + \gamma_{b,g_2} \mathcal{D} [b] \langle g_2 | \rho$$

where $\gamma_{i,j}$ are the decay rates between state $|i\rangle$ and $|j\rangle$.

Solutions to equation 4.46 were sought for atoms passing through a beam diameter that resembles both the fibre and cell, as well as for a wide range of input pump powers. In the case of the fibre, typical simplifications such as: the RWA, steady state populations, slowly evolving coherences [145], or negligible population changes could not be made. This was due to the high optical intensities produced within the fibre capable of optical pumping and saturation. Furthermore, the small beam diameter rules out slow, time dependent approximations. As a result the problem was solved numerically using methods described in Ref [149], and discussed in chapters 5 and 7.

4.4.2 Two Photon Experimental Model

The other major type of experiment conducted within the HC-PCF was spectroscopy of the two-photon transition that can be excited in both the step-wise and direct two-photon processes as shown in figure 4.6. In this model, the two ground states
are treated as one for simplicity. This is justified, as no lasers interact with the other ground state. Furthermore, in the fibre there is little or no time for population to be optically pumped between ground states due to the long lifetime of the $5D_{5/2}$ state. Intermediate states, such as the $5P_{1/2}$ state, have also been dropped as there are no lasers probing these levels. Furthermore, they do not contribute substantially to the strength of the two-photon transition when compared to the near resonant $5P_{3/2}$ energy level. The $6P_{3/2}$ decay route is included so that the blue fluorescence can be modeled. As a result of these simplifications, the energy level structure is as shown in figure 4.6. Here $|g\rangle$ is the ground state, $|a\rangle$ is the intermediate state, $|b\rangle$ is the two-photon state and $|c\rangle$ is the blue fluorescence decay route. Decay rates between energy levels are noted as $\gamma_{i,j}$ for decay between energy levels $|i\rangle$ and $|j\rangle$. Laser fields of frequency $\omega_{g,a}$ and $\omega_{a,b}$ and Rabi frequency $\Omega_{g,a}$ and $\Omega_{a,b}$ couple the $|g\rangle \rightarrow |a\rangle$ and $|a\rangle \rightarrow |b\rangle$ transitions respectively as shown in figure 4.6.

The RWA approximation is again employed to greatly simplify the Hamiltonian governing the two-photon interaction. For the situation where the two exciting lasers are arranged in a counter-propagating configuration, the unitary transformation matrix takes the form:

$$ U = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & e^{-i\omega_{g,a}t} & 0 & 0 \\ 0 & 0 & e^{-i(\omega_{g,a}+\omega_{a,b})t} & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} $$  \hspace{1cm} (4.47) $$

Using the same Hamiltonian transformations described by equation 4.31, the
4.4. MODELS OF RUBIDIUM

Hamiltonians that describes this energy level are defined as:

\[ \tilde{H}_0 = |a\rangle \langle a| \Delta_a + |b\rangle \langle b| \Delta_b \]

\[ \tilde{H}_V = \frac{\Omega_{g,a}}{2} (|a\rangle \langle g| + |g\rangle \langle a|) + \frac{\Omega_{a,b}}{2} (|b\rangle \langle a| + |a\rangle \langle b|) \]

where \( \Delta_a = (E_{g,a} - \hbar \omega_{g,a})/\hbar \) and \( \Delta_b = (E_{a,b} - \hbar \omega_{g,a} - \hbar \omega_{a,b})/\hbar \) are detunings from the intermediate and excited state energy levels respectively. From here the equations of motion, described by equation 4.4, under the RWA give the following:

\[
\dot{\tilde{\rho}} = -\frac{i}{\hbar} [\tilde{H}_0 + \tilde{H}_V, \tilde{\rho}] + \gamma_{a,g} \mathcal{D}[|a\rangle \langle g|] \tilde{\rho} + \gamma_{b,a} \mathcal{D}[|b\rangle \langle a|] \tilde{\rho} + \gamma_{b,c} \mathcal{D}[|b\rangle \langle c|] \tilde{\rho} + \gamma_{c,a} \mathcal{D}[|c\rangle \langle a|] \tilde{\rho} \]

(4.49)

As was found in the hole burning model, general analytic solutions to these differential equations are not possible. In a certain set of circumstances, simplified solutions can be found, some of which are discussed below.

Insight to the spectral profile and absorption strength of the two-photon is gained by solving equation 4.49 in the steady state condition. This alone is still challenging as it results in the following equations describing the atomic coherences:

\[
0 = -i ((2\Delta_a + i\gamma_{ag}) \tilde{\rho}_{g,a}(t) + \Omega_{ab} \tilde{\rho}_{g,b}(t) + \Omega_{ga} (-\tilde{\rho}_{a,a}(t) + \tilde{\rho}_{g,g}(t))) \]

(4.50a)

\[
0 = i\Omega_{ga} \tilde{\rho}_{a,b}(t) - i\Omega_{ab} \tilde{\rho}_{g,a}(t) + (-2i\Delta_b + \gamma_{b,Tot}) \tilde{\rho}_{g,b}(t) \]

(4.50b)

\[
0 = (2i(\Delta_a - \Delta_b) + \gamma_{ag} + \gamma_{b,Tot}) \tilde{\rho}_{a,b}(t) - \Omega_{ab} (\tilde{\rho}_{a,a}(t) - \tilde{\rho}_{b,b}(t)) + i\Omega_{ga} \tilde{\rho}_{g,b}(t) \]

(4.50c)

where \( \gamma_{b,Tot} = \gamma_{b,a} + \gamma_{b,c} \). As the two-photon transition is weak, it can be assumed that the populations are not greatly perturbed (i.e \( \tilde{\rho}_{g,g} \approx 1 \) while \( \tilde{\rho}_{a,a} = \tilde{\rho}_{b,b} = \tilde{\rho}_{c,c} \approx 0 \)). This results in:

\[
\tilde{\rho}_{g,a} = i \frac{\Omega_{ga}}{\gamma_{ag} - 2i \Delta_a} + i \frac{\Omega_{ab} \tilde{\rho}_{g,b}}{\gamma_{ag} - 2i \Delta_a} \]

(4.51a)

\[
\tilde{\rho}_{g,b} = i \frac{\Omega_{ab} \tilde{\rho}_{g,a} - \Omega_{ga} \tilde{\rho}_{g,b}}{\gamma_{b,Tot} - 2i \Delta_b} \]

(4.51b)

\[
\tilde{\rho}_{a,b} = -i \frac{\Omega_{ga} \tilde{\rho}_{g,b}}{(\gamma_{ag} + \gamma_{b,Tot}) - 2i(\Delta_b - \Delta_a)} \]

(4.51c)

It can be seen in the first of these equations that the coherence between the \( |g\rangle \) and \( |a\rangle \) states has two components: The first is the term corresponding to single-photon absorption, as seen in the two-level system (equation 4.35c by setting \( \tilde{\rho}_{g,g} \approx 1 \) while \( \tilde{\rho}_{c,c} = 0 \)); while the second term corresponds to two-photon absorption. A further simplification can be made by recognising that the single-photon transition is much
stronger than the two-photon transition, hence the two-photon absorption term can be dropped, resulting in the familiar two-level transition coherence:

\[ \tilde{\rho}_{g,a} = i \frac{\Omega_{ga}}{\gamma_{ag} - 2i\Delta_a} \]  

(4.52)

From here, the rest of the coherences can be solved. To derive the absorption strength and linewidth of the two-photon transition, the absorption of the \( |a\rangle \rightarrow |b\rangle \) laser is considered. This is determined from the \( \tilde{\rho}_{a,b} \) coherence term which is:

\[ \tilde{\rho}_{a,b} = i \frac{\Omega_{ab}\Omega_{ga}^2}{4\Delta_a^2 + \gamma_{ag}^2} \left( \frac{2i\Delta_b + \gamma_{b,Tot}}{4\Delta_b^2 + \gamma_{b,Tot}^2} - \frac{\gamma_{ag} + \gamma_{b,Tot} - 2i(\Delta_a - \Delta_b)}{4(\Delta_a - \Delta_b)^2 + (\gamma_{ag} + \gamma_{b,Tot})^2} \right) \]  

(4.53)

From this the complex refractive index of the atomic vapour, and hence the two-photon absorption strength, is given by equation 4.14 to be:

\[ \tilde{n}_{a,b} = 1 + N c \hbar \left( \frac{i\Omega_{ab}\Omega_{ga}^2}{2I_{a,b} \left( 4\Delta_a^2 + \gamma_{ag}^2 \right)} \left( \frac{2i\Delta_b + \gamma_{b,Tot}}{4\Delta_b^2 + \gamma_{b,Tot}^2} - \frac{\gamma_{ag} + \gamma_{b,Tot} - 2i(\Delta_a - \Delta_b)}{4(\Delta_a - \Delta_b)^2 + (\gamma_{ag} + \gamma_{b,Tot})^2} \right) \right) \]  

(4.54)

and:

\[ \alpha_{a,b} = N \omega_{ab} \hbar \left( \frac{i\Omega_{ab}\Omega_{ga}^2}{I_{a,b} \left( 4\Delta_a^2 + \gamma_{ag}^2 \right)} \left( \frac{\gamma_{b,Tot}}{4\Delta_b^2 + \gamma_{b,Tot}^2} - \frac{\gamma_{ag} + \gamma_{b,Tot}}{4(\Delta_a - \Delta_b)^2 + (\gamma_{ag} + \gamma_{b,Tot})^2} \right) \right) \]  

(4.55)

This absorption strength comprises of three Lorentzian functions. The pre-factor Lorentzian is centred on the single-photon transition, \( \Delta_a = 0 \), and has a width governed by the decay rate of state \( |a\rangle \). The first of the bracketed Lorentzian components represents the two-photon transition, which is centred at \( \Delta_b = 0 \), and has a FWHM of \( \gamma_{b,Tot} \). The final Lorentzian is also associated with the single-photon resonance. This can be seen by making the substitution \( \Delta_a = \Delta_{\gamma 80} \) and \( \Delta_b = \Delta_{\gamma 80} + \Delta_{\gamma 76} \) where the detunings are now written in terms of laser detunings from resonance. Now the final Lorentzian is centred on \( \Delta_{\gamma 76} = 0 \) and has a FWHM of the sum of the decay rates of levels \( |a\rangle \) and \( |b\rangle \) being \( \gamma_{ag} + \gamma_{b,Tot} \). From this analysis it can be seen that the strength of the two-photon transition falls off like a Lorentzian centred on the single-photon transition and, far from single-photon resonance, falls off like \( 1/\Delta_a^2 \). Hence this shows that there is a very large enhancement of the two-photon transition strength due to the intermediate level. These results agree well with that derived in Ref [138].
4.5 Numerically Evaluating the Density Matrix

It has been highlighted that generalised solutions to density matrix equations are difficult, if not impossible to derive for even basic models. Numerical methods are used to solve the density matrix equations for arbitrary: evolution times, intensity profiles, and optical detunings. The density matrix equations of motion do not change from that discussed earlier in section 4.1, specifically: the equations of motion are still given by equation 4.4.

To incorporate typical experimental conditions, a exciting laser beam with transverse Gaussian intensity profile is assumed which takes the form $\Omega(r) = \Omega_0 e^{-r^2/\sigma^2}$ with a 1/e half width of $\sigma$ and maximum Rabi frequency $\Omega_0$. Hence, the density matrices are solved over the parameter set of: the physical dimensions, $x$ and $y$; transverse velocity $v_t$; and optical detunings $\Delta$, this could be multiple detunings from multiple levels.

It is convenient to numerically solve over the physical dimensions of the exciting beam rather than evolution time, hence a change of parameters is made. Assuming the atoms are moving in the positive $y$ direction, the substitution follows from $y = v_t t$, hence $dt = dy/v_t$ resulting in the equations of motion:

$$v_t \frac{d\tilde{\rho}(x, y, v_t, \Delta)}{dy} = -\frac{i}{\hbar} \left[ \hat{H}, \rho(x, y, v_t, \Delta) \right] + \sum_{j,k} \gamma_{j,k} D[|j\rangle\langle k|] \tilde{\rho}(x, y, v_t, \Delta) \quad (4.56)$$

This change of variable has the physical interpretation that all atoms were propagating in the vertical direction through the optical beam as shown in figure 4.7. It should be noted that the RWA is implemented so that rapid oscillations are not present in the numerical solution as this becomes unfeasible to solve.

To simplify the computational task, the number of equations are reduced by noting that the coherence, off diagonal, elements of the density matrix obey $\rho_{a,b} = \rho_{b,a}^*$. Furthermore, equations of motion terms that result in $d\tilde{\rho}_{a,b}(x, y, v_t, \Delta)/dy = 0$ are dropped from the equations to be solved as they provide no information, take up memory and slow down the differential equation solver. To further reduce the computational task, all physical dimensions are normalised to the optical beam 1/e radius $\sigma$, and all time scales are normalised to an atomic decay rate $\gamma$. Finally, to numerically solve the density matrix equations, initial conditions are provided which take the form of the initial population distributions and setting all coherences to zero.

The numerical solutions were found over a range of the physical dimensions defined by $x \in (0, 4\sigma)$ and $y \in (-4\sigma, 4\sigma)$, where symmetry of a Gaussian beam
CHAPTER 4. LIGHT-ATOM INTERACTIONS

Figure 4.7: Physical interpretation of the numerical integration methods to solve the density matrix equations (left). All transverse velocity classes $v_t$ are propagated through the laser beam in a single direction. A three-level energy level diagram (right) used in the example solution.

was used to reduce the solution range in the $x$ dimension. The range of transverse velocities solved over were $v_t \in (v_0/25, 4v_0)$, where $v_0$ is the most probable velocity, and the range of detunings covered $\Delta \in (-\Delta_{\text{Max}}, \Delta_{\text{Max}})$, where $\Delta_{\text{Max}} = \text{Max}(5, 2\Omega_0)$. Hence, the solutions of the density matrices are found by numerically integrating the following:

$$\tilde{\rho}(x, y, v_t, \Delta) = \int_{-4\sigma}^{4\sigma} \frac{d\tilde{\rho}(x, y, v_t, \Delta)}{dy} dy \quad (4.57)$$

As an example, the density matrix equations for a three-level atom will be solved using the above methods. It was discussed in section 4.3.2 that the steady-state solution for a three-level atom, shown in figure 4.7, does not yield useful information as the solutions are $\tilde{\rho}_{g_1,g_1}(\Delta) = \tilde{\rho}_{e,e}(\Delta) = 0$ and $\tilde{\rho}_{g_2,g_2}(\Delta) = 1$. However, when numerically solving this system using the above methods, the transient behaviour of the system can be examined which are shown in figure 4.8. The system was started with the populations $\rho_{g_1,g_1} = 1$, $\rho_{g_2,g_2} = \tilde{\rho}_{e,e} = 0$ and coherences $\tilde{\rho}_{g_2,e} = \tilde{\rho}_{g_1,e} = 0$. As the atomic population moves through the optical beam, shown in figure 4.8, the population is optically pumped from $|g_1\rangle$ to $|g_2\rangle$ through the excited state $|e\rangle$.

The numerical solutions described here provides solutions for the density matrices at specific detunings, velocities and positions through the optical beam; this is also true for the steady state solutions derived earlier in this chapter. However, when a
4.6 Modelling Atoms Crossing a Gaussian Laser Beam

When considering absorption of a laser through a Rb vapour, density matrix solutions fall short of providing the entire picture. This is because these models only solve for a specific set of circumstances being either: sampled optical power, velocity, or trajectory through the laser beam. Hence, the absorption profile of each atom passing through the optical beam must be modelled so that an accurate profile is constructed. Each atom within the vapour takes a different trajectory through the beam with a velocity described by Maxwell-Boltzmann statistics.

There are two approaches to tackling this problem; A Monte-Carlo approach could be employed to randomly sample trajectories and velocities through the beam;
the other approach involves numerical integration over all possible trajectories and velocities through exploiting symmetries in the original problem. Both approaches were used to varying degrees to solve various problems associated with numerically modeling Rb within HC-PCF. As such, both approaches will briefly be discussed below.

### 4.6.1 Numerical Integration

For an observable or quantity $O$, it is possible to average over all atomic trajectories and velocities in a cylindrically symmetric laser beam using integration techniques [149]. It can be shown that there is a rotation for every possible atomic trajectory that transforms the velocity vector to be parallel to the $y$-axis as shown in figure 4.9. Hence, only the vertically moving trajectories require modelling as, due to cylindrical symmetry, the system is invariant under rotation.

Cylindrical co-ordinates are used to simplify integration over the beam, however as atoms trajectories are linear, the observable $O$ is expressed in Cartesian co-ordinates $O(x, y, z, v_t, v_z, \Delta)$. There is no time dependence in $O$ as this is contained within the transverse velocity term $v_t = v_x^2 + v_y^2$. The observable $O$ could also depend on the axial velocity $v_z$ and optical detuning from resonance $\Delta$. A simple transformation to cylindrical co-ordinates $O(r \cos(\theta), r \sin(\theta), z, v_t, v_z, \Delta)$ makes integration over all velocities and trajectories possible [149]:

$$
\langle O \rangle = \int_0^\infty \int_{-\pi}^\pi \int_0^\infty \int_{-\infty}^{\infty} \left( \frac{r}{2\pi} F_{v_0}^{(t)}(v_t) F_{v_0}^{(z)}(v_z) O \right) dv_z dv_t d\theta dr
$$

where for simplicity the dependencies of $O$ are dropped. The integration over $\theta$ rotates the observable $O$ through $2\pi$, covering all trajectories, while the integration over $v_z$ and $v_t$ accounts for all possible velocities as defined by Maxwell-Boltzmann statistics.

The functions $F_{v_0}^{(t)}(v_t)$ and $F_{v_0}^{(z)}(v_z)$ are the Maxwell-Boltzmann probability density functions (PDF) for the transverse and axial directions, respectively, which provide the correct weighting factors for the velocity classes being integrated over. These PDFs are defined as [149]:

$$
F_{v_0}^{(t)}(v_t) = \frac{2v_t}{v_0^2} e^{-\left(\frac{v_t}{v_0}\right)^2} \\
F_{v_0}^{(z)}(v_z) = \frac{1}{\sqrt{\pi} v_0} e^{-\left(\frac{v_z}{v_0}\right)^2}
$$
4.6. MODELLING ATOMS CROSSING A GAUSSIAN LASER BEAM

Figure 4.9: a) Monte-Carlo approach of randomly starting all the atoms at one point. b) Integration approach at launching all the atoms in a vertical direction. It can be seen that there is a rotation for each atom to transform between these two frames.

where the defining velocity $v_0$ is given by

$$v_0 = \sqrt{\frac{2k_BT N_A}{m}} \quad (4.61)$$

where $T$ is the average thermal temperature of the vapour, $N_A$ is Avagadro’s constant, $k_B$ is Boltzmann’s constant and $m$ is the molar mass of the atomic species.

Equation 4.58 was used to determine the absorption of a thermal Rb vapour in order to model the transit-time absorption profile. Here numerical methods, as described in the previous section were used to solve the density matrix equations, which were then numerically integrated using equation 4.58. In this case $O$ took the form:

$$O = O(r \cos(\theta), r \sin(\theta), v_t, v_z, \Delta)$$

$$O = N \omega h \Omega(r \cos(\theta), r \sin(\theta)) I(r \cos(\theta), r \sin(\theta)) \Re[\mathcal{P}_{g,e}(r \cos(\theta), r \sin(\theta), v_t, v_z, \Delta)] \quad (4.62)$$

where the probing beam Rabi frequency and intensity, $\Omega$ and $I$, and the atomic coherence $\rho_{g,e}$ are functions of position, transverse and axial velocities and optical detuning. It was convenient to define the density matrix is this way as it removes time dependence from the solution. The time dependence is replaced with transverse velocity, making combination with Maxwell-Boltzmann statistics simpler. In depth discussion regarding the above methodology can be found in Ref. [149].

To avoid transformation into cylindrical co-ordinates, a competing integration method was derived. As described above, the probability of an atom moving with a transverse speed of $v_t$ is given by $F^{(t)}_{w}(v_t)$. An atom travelling across a distance $d$ in time $\tau$ has a velocity of $v_t = d/\tau$, hence the two-dimensional speed PDF can be
redefined to give the probability to cross the fibre in time $\tau$:

$$F_{v_0}(v_t) \rightarrow F_{v_0}(\tau) \left| \frac{dv_t}{d\tau} \right|$$  \hspace{1cm} (4.63)

where the derivative $|dv_t/d\tau|$ is required for normalisation purposes. It should be noted that the time $\tau$ is the transit-time across the fibre. Assuming all atoms are moving parallel to the $y$-axis in the positive direction, the distance $d$ that an atom starting at position $x$ on the $x$-axis travels to cross a fibre of radius $R$ is given by $d = 2\sqrt{R^2 - x^2}$, as shown in figure 4.10.

By integrating over the $x$-axis, an integration over all possible trajectories is performed due to the rotational invariance of the geometry. The segment $S$ of the fibre integrated over is not uniform along the $x$-axis as shown in figure 4.10. As the segment section of fibre is constant in terms of angle $\theta$, a normalisation factor of $d\theta/dx$, where $\theta = \text{Asin}(x/r)$ is required. Combining these terms, the PDF for an atom to cross the fibre in time $\tau$, starting at point $x$ along the $x$-axis is given by:

$$F_{v_0}(\tau, x) = \frac{1}{\pi} F_{v_0}(\tau) \left| \frac{dv_t}{d\tau} \right| \left| \frac{d\theta}{dx} \right|$$  \hspace{1cm} (4.64a)

$$= \frac{8\sqrt{R^2 - x^2}}{\pi \tau^3 v_0^2} e^{-\left(\frac{2\sqrt{R^2 - x^2}}{\tau v_0}\right)^2}$$  \hspace{1cm} (4.64b)

where $v_0$ is defined in equation 4.61. A factor of $1/\pi$ is introduced to normalise the function $d\theta/dx$ over the range $x \in (-R, R)$.

In order to average an observable $O(\tau, x, \Delta)$ over all trajectories and velocities,
Figure 4.11: Transit-time probability distribution function (PDF) of atoms at 100°C crossing a fibre core with diameter of 36 μm.

Integration must be performed over transit-time, the x-axis and axial velocities:

$$\langle O \rangle = \frac{2}{\pi} \int_0^R \int_0^\infty \int_{-\infty}^\infty \tau \left( \frac{d}{\tau} \right) \left( \frac{dv_t}{d\tau} \right) \left( \frac{d\theta}{dx} \right) F_{v_0}^{(z)}(v_z) O(\tau, x, \Delta - v_x k) dv_z d\tau dx$$

(4.65)

where \( k \) is the wave-vector of the light and symmetry is employed to simplify the integration region from \( x \in (-R, R) \) to \( x \in (0, R) \) producing a pre-factor of 2. Doppler broadening effects are included though inclusion of a convolution over \( v_z \). This integral varies from equation 4.58 as one of the physical dimensions and transverse velocity are combined to remove a layer of integration.

The average transit-time across the fibre is calculated by computing the expectation value of the transit-time observable \( \mathcal{T}(\tau) = 1 \). Substituting \( O \rightarrow \mathcal{T}(\tau) \) yields an average transit-time of:

$$\langle \mathcal{T} \rangle = \frac{2}{\pi} \int_0^R \int_0^\infty \int_{-\infty}^\infty \tau \left( \frac{d}{\tau} \right) \left( \frac{dv_t}{d\tau} \right) \left( \frac{d\theta}{dx} \right) F_{v_0}^{(z)}(v_z) \mathcal{T}(\tau) dv_z d\tau dx$$

(4.66a)

$$= \frac{4 R}{v_0 \sqrt{\pi}}$$

(4.66b)

$$= \frac{2 R}{v_{t,\mu}}$$

(4.66c)

where \( v_{t,\mu} \) is the mean transverse speed, defined as \( v_0 \sqrt{\pi}/2 \). Interestingly this average transit time is simply the fibre diameter divided by the mean transverse speed. For Rb atoms at 100°C traversing a fibre of diameter 45 μm and optical beam diameter of 36 μm, an average transit time of 188 ns and 150 ns is calculated respectively.

Furthermore, to produce a PDF for the transit-time across the fibre the integration over \( \tau \) in equation 4.65 is dropped along with the factor of \( \tau \) within the integral, as this would produce an expectation value and not a PDF. Hence, the PDF \( \langle \mathcal{T}(\tau) \rangle \)
averaged over all trajectories and velocities is given by:

$$\langle \mathcal{T}(\tau) \rangle = \frac{2}{\pi} \int_0^R \int_{-\infty}^\infty \left( F^{(t)}_{v_0} \left( \frac{d}{\tau} \right) \left| \frac{dv}{d\tau} \right| d\theta \right) F^{(z)}_{v_0}(v_z) \mathcal{T}(\tau) dv_z \, dx$$

(4.67a)

$$= \frac{2}{\pi} \int_0^R 8\sqrt{r^2 - x^2} e^{-\left(\frac{2\sqrt{r^2 - x^2}}{v_0\tau}\right)^2} \, dx$$

(4.67b)

$$= \frac{2}{\tau} \alpha e^{-\alpha} (I_0(\alpha) - I_1(\alpha))$$

(4.67c)

where $I_0(\alpha)$ and $I_1(\alpha)$ are the modified Bessel function of the first kind which are related to the Bessel function, $J_\beta(x)$, through $I_\beta(x) = i^{-\beta} J_\beta(ix)$. The parameters $\alpha$ is defined as:

$$\alpha = \left( \frac{\sqrt{2R}}{v_0 \tau} \right)^2$$

(4.68)

This PDF, equation 4.67c, is displayed in figure 4.11.

### 4.6.2 Monte-Carlo Approach

A Monte-Carlo approach was employed to confirm the numerical integration approach through building up a statistical average of an observable $O$ within the HC-PCF. Valuable information was gained regarding the average atomic transit time and absorption spectra through the fibre’s optical mode. Statistics were built up through sampling a large number of atoms with randomly distributed trajectories and velocities, the methodology of which is discussed briefly below.

An initial assumption is made which assumes that the fibre is a cylinder instead of the complex shapes shown in figure 2.3. Cylindrically symmetry was used to simplify the simulation such that all atomic trajectories started at the same point on the fibre’s core wall as seen in figure 4.9 (a). The fibre was modelled in all three dimensions, hence random velocities were produced in each dimension governed by a Normal distribution with mean $\mu = 0$ and standard deviation $\sigma = v_0$, where $v_0$ is defined by equation 4.61. Generation of these velocities in this manner satisfied Maxwell-Boltzmann statistics. Using the velocities and a fixed starting point, the intersection point with the fibre wall was computed yielding a transit time across the fibre. Using this method a number of different scenarios could be calculated which included: simulating transit-times across the fibre’s optical mode (diameter 36 $\mu$m); from fibre wall-to-wall (diameter 45 $\mu$m); or the time taken for atoms to propagate from the centre of the fibre to the fibre’s wall.

Typically the number of atomic trajectories calculated was of the order $10^5$ to $10^6$. This resulted in average transit-times for atoms passing through the optical mode
4.6. MODELLING ATOMS CROSSING A GAUSSIAN LASER BEAM

![Graph showing PDF vs Flight Time (ns) for Monte-Carlo Simulation and Analytical Theory]

**Figure 4.12:** Results from a Monte-Carlo simulation of atoms crossing a fibre core with diameter of 36 μm compared to that calculated from equation 4.67c.

With 1/e intensity diameter of 36 μm to be 151 ns for a Rb vapour at 100°C. The average transit-time for atoms travelling from wall-to-wall across a fibre of diameter 45 μm is 189 ns. These results are shown in figure 4.12 to be in good agreement with numerical methods derived in equation 4.67c. In contrast, the average transit-time across an optical beam of diameter 1 mm is 8.5 μs.

Using the transit-times and trajectories modelled, information about how atoms sample the fibre’s optical mode can be extracted. The three values of importance here are: time spent within the optical mode, transit-time, and the highest optical intensity experienced in the Gaussian beam. Using these parameters the temporal evolution of the optical intensity experienced by the atom can be constructed. Using these extracted values, effects such as transit-time broadening and population pumping could be modelled and compared to experiments. Both of these effects are discussed in chapters 7 and 8.
This chapter gives a theoretical basis for broadening mechanisms of atomic spectra within the HC-PCFs used in the following chapters. This theory underpins experimental results shown in chapters 7 and 8. The first section predominantly discusses transit-time broadening within the HC-PCF. Also discussed are power, magnetic and collisional broadening, along with light shifts and Rabi splitting. The second section extends these discussions to model experimental work in an effort to understand broadening mechanisms within the HC-PCF.

5.1 Spectral Broadening Mechanisms

Broadening mechanisms affecting the transition spectra of an effusive vapour loaded within a HC-PCF are many and varied. These are of great consequence if such a system is to be used for an atomic clock or frequency standard, where narrow absorption features are desired. This section aims to address each of the broadening mechanisms present within the HC-PCF analytically or numerically so that they can be compared to experimental observations and possibly avoided.

The most dominant broadening mechanism is Doppler broadening, induced by atomic motion within the vapour as discussed in sections 3.1.1 and 3.1.2. However, this can be avoided using spectroscopic techniques such as saturated absorption spectroscopy or two-photon Doppler-free spectroscopy, discussed in sections 3.1.1 and 3.1.2 respectively. The natural lifetime of the atomic state \( \tau \), generates the unperturbed and unbroadened transition linewidth of \( \gamma = 1/\tau \), as discussed in section 4.3.1. A finite light-atom interaction time can generate substantial transit-time broadening which is discussed in detail in sections 5.1.1 and 5.1.2. Power broadening, discussed in section 4.3.1, can be controlled, as can AC Stark shifts (also known as light shifts) and Rabi splitting, discussed in section 5.1.3. External
CHAPTER 5. EXPERIMENTAL MODELLING

<table>
<thead>
<tr>
<th>Effect</th>
<th>Transition broadening (MHz)</th>
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<td></td>
<td>$5S_{1/2} \rightarrow 5P_{3/2}$</td>
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<td>Doppler broadening</td>
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</tr>
<tr>
<td>Natural broadening ($\gamma$)</td>
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</tr>
<tr>
<td>Transit-time broadening:</td>
<td>$\tau \ll \gamma$</td>
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<tr>
<td></td>
<td>$4\sqrt{2 \ln(2)/\tau}$</td>
</tr>
<tr>
<td>Zeeman Splitting</td>
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<tr>
<td>Power broadening:</td>
<td>$I \ll I_{\text{Sat}}$</td>
</tr>
<tr>
<td></td>
<td>$\gamma \sqrt{T/I_{\text{Sat}}}$</td>
</tr>
<tr>
<td>A.C Stark shift</td>
<td>$\approx \Omega_{p}^{2}/(4\Delta_{p})$</td>
</tr>
<tr>
<td>Background gas collisions</td>
<td>$\approx 18.5$ MHz/Torr</td>
</tr>
<tr>
<td>Rb-Rb collisions</td>
<td>$\approx 4$ GHz/Torr [151]</td>
</tr>
<tr>
<td>Second-order Doppler effect</td>
<td>$&lt; 250$ Hz</td>
</tr>
</tbody>
</table>

Table 5.1: Broadening mechanisms and their contribution to linewidths in descending order.

magnetic fields also affect the spectra within the HC-PCF, discussed in section 5.1.4. Finally, atomic collisions cause dephasing of the atomic dipole which can generate spectral broadening and shifts which are discussed in section 5.1.5. The contribution of each of these mechanisms upon the $D_2$ and two-photon spectra are quantified in table 5.1 and discussed in the following sections.

5.1.1 Transit Time Modelling of Single-Photon Transitions

The most dominant broadening mechanism present within the HC-PCFs is transit-time broadening. Transit-time broadening is caused by a finite light-atom interaction time and it is a major source of broadening for many experiments including atomic clocks and precision spectroscopy [153], and hence has been investigated theoretically in much depth. A variety of papers and text books cover transit-time broadening to varying degrees of complexity [134, 154, 155]. In the following sections, the transit-time broadened spectra is calculated using typical experimental conditions, being a Rb vapour at 100°C generating a most probable transverse velocity of $240 \text{ ms}^{-1}$ through a beam of diameter 36 $\mu$m. This results in a most probable transit time of 150 ns. These experimental conditions are discussed in detail in chapter 6.3.2.
5.1. SPECTRAL BROADENING MECHANISMS

Heisenberg Uncertainty Principle

The simplest approach to estimating the effect transit-time broadening has on atomic spectra is to consider the Heisenberg Uncertainty Principle \[145].

\[
\Delta E \Delta \tau > \frac{\hbar}{2}
\]  \hspace{1cm} (5.1)

An atom passing through a laser beam experiences a pulse of light of duration \(\Delta \tau\). The Heisenberg Uncertainty Principle states that this results in an associated energy uncertainty \(\Delta E = \frac{\hbar}{2(2\Delta \tau)}\).

For a typical transit time of 150 ns this approach yields an energy uncertainty of 597 kHz. This is a crude estimate of the magnitude of transit-time broadening as this simplistic approach neglects the internal dynamics of the atom, the light mode profile and the velocity distribution of atoms in a thermal vapour.

Harmonic Oscillator Model

The next step in complexity is to consider the resulting spectral profile induced by transit-time broadening. First, the assumption is made that the atom’s excited state lifetime is much longer than the transit time through the laser beam. Hence, the atom can be assumed to be an undamped oscillator over a transit time \(\tau = d/|v|\), across a laser beam of diameter \(d\) with a thermal velocity \(v\) [134]:

\[
x(t) = x_0 \cos(\omega_0 t)
\]  \hspace{1cm} (5.2)

where \(\omega_0\) is the oscillation (or transition) frequency and \(x_0\) is the peak amplitude of the oscillation.

The frequency spectrum of this oscillator interacting with a driving laser \(e^{-i\omega t}\) of frequency \(\omega\), over an interaction time \(\tau\) is obtained through a Fourier transform:

\[
A(\omega) = \frac{1}{\sqrt{2\pi}} \int_{0}^{\tau} x(t) e^{-i\omega t} dt
\]  \hspace{1cm} (5.3a)

\[
= \frac{1}{\sqrt{2\pi}} \int_{0}^{\tau} x_0 \cos(\omega_0 t) e^{-i\omega t} dt
\]  \hspace{1cm} (5.3b)

\[
= \frac{x_0}{\sqrt{2\pi}} \frac{-i\omega + i e^{-i\omega_0 \tau} \omega \cos(\tau \omega_0) - e^{-i\omega_0 \tau} \omega_0 \sin(\tau \omega_0)}{\omega^2 - \omega_0^2}
\]  \hspace{1cm} (5.3c)

The observed intensity profile \(I(\omega) = A^* A\) is derived using trigonometric identities and simplified assuming that the spectral profile is interrogated near resonance \(\omega \approx \omega_0\). This eliminates any terms with the quantity \((\omega + \omega_0)\) as their denominator.
resulting in a spectral profile of the form [134]:

\[
I(\omega) = \frac{x_0}{\sqrt{2\pi}} \frac{\sin^2 \left( (\omega - \omega_0)\frac{\tau}{2} \right)}{(\omega - \omega_0)^2} \tag{5.4a}
\]

\[
= \frac{x_0}{\sqrt{2\pi}} \frac{\sin^2 \left( \frac{\Delta \tau}{2} \right)}{\Delta^2} \tag{5.4b}
\]

\[
= \frac{x_0}{\sqrt{2\pi}} \left( \frac{\tau}{2} \right)^2 \text{sinc} \left( \frac{\Delta \tau}{2} \right)^2 \tag{5.4c}
\]

where \( \Delta = (\omega - \omega_0) \) is the laser’s detuning from resonance. Normalising this to the laser’s intensity profile, being \( \tau/\sqrt{2\pi} \) results in a normalised spectral profile of:

\[
I(\omega)_{\text{Norm}} = \frac{\tau x_0}{4} \text{sinc} \left( \frac{\Delta \tau}{2} \right)^2 \tag{5.5}
\]

This results in a FWHM spectral width of [134]:

\[
\Delta \omega_{\text{FWHM}} = 5.566/\tau \tag{5.6}
\]

For typical experimental conditions, as described at the beginning of this section, a transit time of 150 ns leads to a broadening of \( \Delta \omega_{\text{FWHM}} = 37.1 \times 10^6 \text{ rad s}^{-1} \), or in linear frequency units, \( \Delta f_{\text{FWHM}} = 5.91 \text{ MHz} \).

Further detail can be added by considering a temporally varying electric field profile experienced by the atom. For simplicity the optical mode is assumed to be the TE\textsubscript{00} Gaussian mode, hence the electric field of the laser beam is expressed as:

\[
E(r,t) = E_0 e^{-r^2/\sigma^2} \cos(\omega t) \tag{5.7}
\]

in which \( 2\sigma \) is the diameter of the laser beam at the 1/e electric field points. An atom travelling at a transverse velocity \( v_t \), through this laser would experience an electric field of \( E_{\text{atom}}(t) = E(v_t, t) \). Using the simple harmonic oscillator described in equation 5.2, the resulting Fourier transform is:

\[
A(\omega) = \int_{-\infty}^{\infty} E_{\text{atom}}(t) x(t) \, dt \tag{5.8a}
\]

\[
= \int_{-\infty}^{\infty} E_0 x_0 e^{-\frac{(tv)^2}{\sigma^2}} \cos(t\omega) \cos(t\omega_0) \, dt \tag{5.8b}
\]

\[
= \sqrt{\pi} E_0 x_0 \sigma \left( e^{-\frac{\sigma^2(\omega-\omega_0)^2}{4v^2}} + e^{-\frac{\sigma^2(\omega+\omega_0)^2}{4v^2}} \right) \tag{5.8c}
\]

Again, the observed intensity profile is calculated using \( I(\omega) = A^* A \). By dropping oscillating terms with frequencies much larger than the optical detuning \( \Delta \), the
resulting spectral profile is:

\[ I(\omega) = I_0 \exp \left( -\frac{\Delta^2 \sigma^2}{2 \omega^2} \right) \]  \hspace{1cm} (5.9)

This is a Gaussian profile with a spectral FWHM of:

\[ \Delta \omega_{\text{FWHM}} = \frac{4\sqrt{2 \ln(2)}}{\tau_\sigma} \]  \hspace{1cm} (5.10)

\[ \approx \frac{4.71}{\tau_\sigma} \]  \hspace{1cm} (5.11)

where the transit time \( \tau_\sigma \) to cross between the \( 1/e \) electric field points is given by \( \tau_\sigma = 2\sigma/v_t \).

The resulting broadening is \( \Delta \omega_{\text{FWHM}} = 31.4 \times 10^6 \text{ rad s}^{-1} \) or, in linear frequency units, \( \Delta f_{\text{FWHM}} = 4.93 \text{ MHz} \) under typical experimental conditions, as described at the beginning of this section. As can be seen, the inclusion of the electric field profile reduces the calculated broadening by almost 20%. These approaches, however, still do not model the behaviour of an atom for the case where the transit time and decay rate are of similar magnitude. Additionally, this model does not incorporate velocity averaging or different trajectories through the optical beam.

**Inclusion of Internal Atom Dynamics**

Further detail is added when the internal dynamics of the atom is included. This is accomplished through using a simple two-level density matrix model as outlined in chapter 4.3.1. The RWA is used to give the Hamiltonian, as derived in equation 4.33, to be:

\[ \hat{H} = \begin{pmatrix} 0 & \hbar \Omega/2 \\ \hbar \Omega/2 & \hbar \Delta \end{pmatrix} \]  \hspace{1cm} (5.12)

where \( \Omega \) is the Rabi frequency of the interrogating laser. Assuming again that the natural decay time of the atom is much longer than the transit time, equation 4.4 generates the following differential equations of motion:

\[
\begin{pmatrix}
\dot{\rho}_{g,g}(t) \\
\dot{\rho}_{g,e}(t) \\
\dot{\rho}_{e,g}(t) \\
\dot{\rho}_{e,e}(t)
\end{pmatrix} = \frac{i}{2} \begin{pmatrix}
0 & \Omega_0 & -\Omega_0 & 0 \\
\Omega_0 & 2\Delta & 0 & -\Omega_0 \\
-\Omega_0 & 0 & -2\Delta & \Omega_0 \\
0 & -\Omega_0 & \Omega_0 & 0
\end{pmatrix} \begin{pmatrix}
\rho_{g,g}(t) \\
\rho_{g,e}(t) \\
\rho_{e,g}(t) \\
\rho_{e,e}(t)
\end{pmatrix}
\]  \hspace{1cm} (5.13)

where \( g \) and \( e \) label the ground \( |g\rangle \) and excited \( |e\rangle \) states respectively. To examine the effect of a finite duration light pulse, these differential equations are solved using
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a uniform light exposure lasting from $t = 0$ to $t = \tau$. Using standard trigonometric identities the resulting solutions are:

\[
\tilde{\rho}_{gg}(t) = 1 - \frac{t^2 \Omega^2}{4} \text{sinc} \left( \frac{t}{2} \sqrt{\Delta^2 + \Omega^2} \right)^2 \\
\tilde{\rho}_{ee}(t) = \frac{t^2 \Omega^2}{4} \text{sinc} \left( \frac{t}{2} \sqrt{\Delta^2 + \Omega^2} \right)^2 \\
\tilde{\rho}_{ge}(t) = -\frac{t \Omega}{4} \text{sinc} \left( \frac{t}{2} \sqrt{\Delta^2 + \Omega^2} \right)^2 - 2i \text{sinc} \left( t \sqrt{\Delta^2 + \Omega^2} \right) \\
\]

From this, the probing laser’s absorption coefficient can be extracted, as outlined in chapter 4.2. This requires the absorption to be integrated over the atoms transit across the beam. As seen in chapter 4.2.2, the absorption coefficient can be given by $\alpha(t, \Delta) \propto \mathcal{N}_{\text{Scat}} / \mathcal{N}_{\text{Tot}}$. To integrate this properly over the atoms transit across the beam, the number of scattered photons $\mathcal{N}_{\text{Scat}}$, and the number of incident laser photons $\mathcal{N}_{\text{Tot}}$, are integrated separately to give:

\[
\langle \alpha(\Delta) \rangle = \int_0^\tau \alpha(t, \Delta)dt \\
= N \omega \hbar \int_0^\tau \Omega \Im \left[ \tilde{\rho}_{ge}(t) \right] dt \\
= \frac{\tau \Omega^2}{4I} \text{sinc} \left( \frac{\tau}{2} \sqrt{\Delta^2 + \Omega^2} \right)^2
\]

This solution has a similar functional form to that of equation 5.5 whereby $x_0 \rightarrow \Omega^2 / I$. In the limit $\Omega \rightarrow 0$ the spectral width of the solution collapses to:

\[
\Delta \omega_{\text{FWHM}} = 5.566 / \tau
\]

which agrees with the solution for a simple harmonic oscillator in a uniform pulse, equation 5.6.

There is no closed-form expression for the FWHM of equation 5.15c at arbitrary $\Omega$, as the resulting spectra’s side-lobes grow at high Rabi frequencies as seen in figure 5.1. This is caused by the Rabi oscillations of the atom leaving the atom in a superposition of the $|g\rangle$ and $|e\rangle$ state on exiting the beam, along with the rapid intensity changes at the edges of the uniform beam generating additional spectral frequency components. A general solution to the FWHM is not sought here as it does not add value to the discussion, instead a Gaussian excitation pulse is considered.

This two-level density matrix model becomes much more difficult once a non-uniform electric field distribution is taken into account. Again the atom is assumed
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Figure 5.1: Transit-time broadened density matrix solutions (equation 5.15c) for an atom passing through a uniform beam for a fixed transit time of $\tau = 1$ (top) at various values of $\Omega$. Atoms passing through a uniform beam with $\Omega = 0.1$ at various transit times (bottom). The natural linewidth of the transition is assumed to be negligibly small.

to pass through a Gaussian pulse of light, mathematically achieved by making the replacement $\Omega \rightarrow \Omega(t) = \Omega_0 e^{-\left(t/\tau_\sigma\right)^2}$ in the Hamiltonian shown in equation 5.12. Physically this is a laser beam with a maximum Rabi frequency of $\Omega_0$ centred at time $t = 0$, with a $1/e$ electric field of diameter $2\tau_\sigma$.

The resulting system of homogeneous first-order differential equations can not be solved analytically without making some assumptions. The key assumption to make is that the atomic populations are not perturbed greatly throughout the light-atom interaction. That is to say, at all times $\tilde{\rho}_{g,g}(t) \approx 1$ and $\tilde{\rho}_{e,e}(t) \approx 0$ which naturally leads to $\frac{\partial}{\partial t}\tilde{\rho}_{g,g}(t) \approx 0$ and $\frac{\partial}{\partial t}\tilde{\rho}_{e,e}(t) \approx 0$. These assumptions are valid when $\Omega$ is sufficiently small that little population is perturbed throughout the interaction time $2\tau_\sigma$. This is not an unreasonable assumption to make as large optical powers, and hence atomic perturbations induce power broadening and optical saturation effects that mask transit-time broadening. Although, to produce the correct solution, these assumptions are implemented in the final stages of the calculation as shown below.
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To solve equation 5.13 with \( \Omega(t) = \Omega_0 e^{-(t/\tau)^2} \), a transformation is made to the coherence equations of the form [145]:

\[
\tilde{\rho}_{g,e}(t) \rightarrow \hat{\tilde{\rho}}_{g,e}(t) e^{i\Delta t}
\]  

resulting in the following differential equation for the coherence term:

\[
\hat{\tilde{\rho}}_{g,e}(t) = \frac{i\Omega_0}{2} e^{-it\Delta -(t/\tau)^2} (\tilde{\rho}_{g,g}(t) - \tilde{\rho}_{e,e}(t))
\]  

Using integration by parts the solution is found in the following manner:

\[
\hat{\tilde{\rho}}_{g,e}(t) = \frac{i\Omega_0}{2} \int_{-\infty}^{t'} e^{-it\Delta -(t/\tau)^2} (\tilde{\rho}_{g,g}(t) - \tilde{\rho}_{e,e}(t)) dt
\]  

(5.19a)

\[
= \frac{i\Omega_0}{2} \left[ \frac{\sqrt{\pi} \tau}{2} e^{-\Delta^2/4} \operatorname{Erf} \left( \frac{t + i\Delta \tau}{2} \right) (\tilde{\rho}_{g,g}(t) - \tilde{\rho}_{e,e}(t)) \right]_{-\infty}^{t'}
\]

(5.19b)

\[
- \frac{i\Omega_0}{2} \int_{-\infty}^{t'} e^{-it\Delta -(t/\tau)^2} \frac{\partial}{\partial t} (\tilde{\rho}_{g,g}(t) - \tilde{\rho}_{e,e}(t)) dt
\]

+ \frac{i\sqrt{\pi} \Omega_0 \tau}{4} e^{-\Delta^2/4} \left( \operatorname{Erf} \left( \frac{t + i\Delta \tau}{2} \right) (\tilde{\rho}_{g,g}(t) - \tilde{\rho}_{e,e}(t)) + 1 \right)

(5.19c)

where the assumption that \( \frac{\partial}{\partial t} \tilde{\rho}_{g,g}(t) = \frac{\partial}{\partial t} \tilde{\rho}_{e,e}(t) \approx 0 \) was used. Using the assumption that \( \tilde{\rho}_{g,g}(t) \approx 1 \) and \( \tilde{\rho}_{e,e}(t) \approx 0 \), the solution to the atomic coherence between the ground and excited state is:

\[
\tilde{\rho}_{g,e}(t) \approx \frac{i\sqrt{\pi} \Omega_0 \tau}{4} e^{it\Delta - \Delta^2/4} \left( \operatorname{Erf} \left( \frac{t + i\Delta \tau}{2} \right) + 1 \right)
\]  

(5.20)

From here, small changes in populations can be calculated, however, to determine the probing laser’s absorption only the coherence term is required, as outlined above, and in chapter 4.2. The absorption coefficient is integrated over the atom’s transit across the laser field, as demonstrated in equation 5.15b, giving:

\[
\langle \alpha(\Delta) \rangle = \int_{0}^{\tau} \alpha(t, \Delta) dt
\]

(5.21a)

\[= N \omega \hbar \int_{0}^{\tau} \Omega(t) \Im \left[ \tilde{\rho}_{e,g}(t) \right] dt \]

(5.21b)

\[= N \omega \hbar \Im \left[ \pi \tau^2 \Omega_0^2 e^{-\Delta^2/2} \left( -i + \operatorname{Erfi} \left( \frac{\Delta \tau}{\sqrt{2}} \right) \frac{1}{\sqrt{2 \pi}} \right) \right] \]

(5.21c)

\[= N \omega \hbar \frac{\sqrt{\pi} \tau \Omega_0^2}{2 \sqrt{2 I_0}} e^{-\Delta^2/2}
\]

(5.21d)
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Figure 5.2: Absorption coefficient of a two-level atom passing through a Gaussian beam at small optical intensities for various transit times. The transition’s natural linewidth is assumed to be negligibly small.

where ErFi(\(x\)) is the imaginary error function. Evaluation of the integral above was accomplished through parametric integration [156]. Example absorption spectra are shown in figure 5.2 for a variety of transit times. In contrast to figure 5.1, there are no side-lobes present in these spectra since there are no rapid intensity changes, which are present in the uniform pulse model.

The resulting spectra has a Gaussian absorption profile of widths:

\[
\Delta \omega_{\text{FWHM}} = 4 \sqrt{2 \log(2)}/\tau_{2\sigma} \\
\approx 4.71/\tau_{2\sigma} \\
\Delta \omega_{1/e} = 4\sqrt{2}/\tau_{2\sigma} \\
\approx 5.66/\tau_{2\sigma}
\]

(5.22) (5.23)

where \(\tau_{2\sigma} = 2\tau_{\sigma}\) is the time taken for an atom to transit the \(1/e\) electric field diameter of the beam. Spectral absorptions for various transit times are shown in figure 5.2.

These derivations now take into account the internal dynamics of the atoms, however the derivation still assumes that the decay rate of the atom is much longer than the transit time. This is not valid when considering Rb atoms within a HC-PCF with an optical mode diameter of 36 \(\mu\)m where the mean transit time is 150 ns compared to a decay rate of 26.6 ns and 238.5 ns for the Rb \(5P_{3/2}\) and \(5D_{5/2}\) energy levels respectively.
Including Atomic Decay

To account for atomic decay, equation 5.13 is amended, as per equation 4.4 to include decaying terms. The resulting equations of motion are described by:

\[
\begin{pmatrix}
\dot{\rho}_{g,g}(t) \\
\dot{\rho}_{g,e}(t) \\
\dot{\rho}_{e,g}(t) \\
\dot{\rho}_{e,e}(t)
\end{pmatrix} =
\begin{pmatrix}
0 & i\Omega(t)/2 & -i\Omega(t)/2 & \gamma_{e,g} \\
i\Omega(t)/2 & i\Delta - \gamma_{e,g}/2 & 0 & -i\Omega(t)/2 \\
-i\Omega(t)/2 & 0 & -i\Delta - \gamma_{e,g}/2 & i\Omega(t)/2 \\
0 & -i\Omega(t)/2 & i\Omega(t)/2 & -\gamma_{e,g}
\end{pmatrix}
\begin{pmatrix}
\dot{\rho}_{g,g}(t) \\
\dot{\rho}_{g,e}(t) \\
\dot{\rho}_{e,g}(t) \\
\dot{\rho}_{e,e}(t)
\end{pmatrix}
\]

It can be seen there is decay at a rate of \(\gamma_{e,g}\) between \(\ket{e}\) and \(\ket{g}\). This decay is also present in the coherence equations. The model for a uniform pulse of light is not considered here as it is a backward step, however for completeness it is shown in appendix D.

The final detail of this theory is to incorporate both atomic decay with a Gaussian laser beam distribution. This is achieved by making the substitution \(\Omega(t) = \Omega_0 e^{-(t/\tau \sigma)^2}\) in equation 5.24. Using the procedures described by equations 5.17 - 5.20, and using the same assumption that atomic populations are not greatly perturbed (\(\dot{\rho}_{g,g}(t) \approx 1, \dot{\rho}_{e,e}(t) \approx 0,\) and \(\frac{\partial}{\partial t} \dot{\rho}_{g,g}(t) = \frac{\partial}{\partial t} \dot{\rho}_{e,e}(t) \approx 0\)), the coherence between the ground and excited state can be calculated to be:

\[
\dot{\rho}_{g,e}(t) = -\frac{1}{2}(\gamma_{e,g} - 2i\Delta)\dot{\rho}_{g,e}(t) - \frac{i\Omega_0}{2} e^{-(t/\tau \sigma)^2} (\dot{\rho}_{g,e}(t) - \dot{\rho}_{g,g}(t))
\]

\[
\Rightarrow \dot{\rho}_{g,e}(t) \approx \frac{i\sqrt{\pi \tau \sigma} \Omega_0}{4} e^{\left(\frac{\gamma_{e,g} - 2i\Delta}{4t/\tau \sigma}\right)^2 + i(\Delta - \gamma_{e,g})/2} \left(1 + \text{Erf}\left(\frac{t}{\tau \sigma} - \frac{(\gamma_{e,g} - 2i\Delta)/\tau \sigma}{4}\right)\right)
\]

The absorption coefficient of the probe laser is again determined through integration, as shown in equation 5.15b. Parametric integration [156] is again used to evaluate this integral, which results in:

\[
\langle \alpha(\Delta) \rangle = \int_0^\tau \alpha(t, \Delta)dt
\]

\[
= N \omega \hbar \int_0^\tau \frac{\Omega(t) \Im \dot{\rho}_{e,g}(t)}{I(t)dt} dt
\]

\[
= N \omega \hbar \sqrt{\pi \tau \sigma} \Omega_0^2 \text{Re} \left( e^{\frac{1}{2}(\gamma_{e,g} - 2i\Delta)^2/2} \text{Erfc} \left( \frac{(\gamma_{e,g} - 2i\Delta)/\tau \sigma}{2\sqrt{2}} \right) \right)
\]

\[
= N \omega \hbar \Omega_0^2 V(\Delta; \gamma_{e,g}, \sqrt{2}/\tau \sigma)
\]

where \(V(\Delta; \gamma_{e,g}, \sqrt{2}/\tau \sigma)\) is a Voigt function, a convolution of a Gaussian and Lorentzian
Function with a 1/e half width of $\sqrt{2}/\tau_\sigma$ and FWHM of $\gamma_{e,g}$ respectively. More discussion regarding the Voigt function can be found in appendix E. This Voigt spectrum can be seen in figure 5.3, which shows the transit-time broadened spectra for various transit times.

The spectral width of this absorption profile can not be determined analytically, however, there are a number of empirical formula that give a highly accurate FWHM measure of a Voigt linewidth [157].

\[
\Delta \omega_{\text{FWHM}} = 0.54\gamma_{e,g} + \sqrt{0.22\gamma_{e,g}^2 + \left(\frac{4.71}{\tau_{2\sigma}}\right)^2}
\]

(5.27)

where $\tau_{2\sigma} = 2\tau_\sigma$ is the time taken for an atom to transit the 1/e electric field diameter of the beam. The Gaussian contribution to the atoms FWHM is identical to that from the situation where an atom with no decay passes through a Gaussian beam, solved in equation 5.23.

In the limit where transit time becomes much longer than the natural decay

<table>
<thead>
<tr>
<th>Model</th>
<th>Spectral Width FWHM</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>No Decay</td>
</tr>
<tr>
<td>Uniform Pulse</td>
<td>$5.57/\tau$</td>
</tr>
<tr>
<td></td>
<td>$\left(\gamma_{e,g}^{3/2} + \left(\frac{5.56}{\tau}\right)^{3/2}\right)^{2/3}$</td>
</tr>
<tr>
<td>Gaussian Pulse</td>
<td>$4.71/\tau_{2\sigma}$</td>
</tr>
<tr>
<td></td>
<td>$0.54\gamma_{e,g} + \sqrt{0.22\gamma_{e,g}^2 + \left(\frac{4.71}{\tau_{2\sigma}}\right)^2}$</td>
</tr>
</tbody>
</table>

Table 5.2: Summary of spectral FWHM of transit-time broadened atoms for the derived light-atom models.
of the atom $\tau_{2\sigma} \to \infty$, the spectral profile collapses to that of a two-level atom $\langle \alpha(\Delta) \rangle \to \gamma_{e,g}^2 / (\gamma_{e,g}^2 + 4\Delta^2)$, with a spectral width of $\Delta\omega_{\text{FWHM}} \to \gamma$, as described in chapter 4.3.1. On the other hand, if the transit time is much smaller than the natural decay rate, $\tau_{2\sigma}^{-1} \ll \gamma_{e,g}$, the spectra is predominantly Gaussian with a $1/e$ half width of $\sqrt{2}/\tau_{\sigma}$. Here, the spectral shape of the broadened atom tends towards that previously derived for an atom with no decay paths from the excited level which passes through a Gaussian pulse as described by equation 5.21d.

So far, four different light-atom interaction models have been presented and absorption profiles calculated in each case. The spectral width dependence on transit time for these models is summarised in table 5.2 and also displayed in figure 5.4. This clearly shows the similarities and differences between these models. It is clear that the linewidth of both models that include atomic decay asymptote towards the transition’s natural linewidth at long transit times, whereas the two models that do not include decay are infinitely narrow in the same limit. At short interaction times, all models are inversely dependent on transit time with slightly varying coefficients.

Rubidium Implementation

To model the $D_2$ transition of Rb more accurately, the above derived two-level atomic model is extended to a three-level model as described in chapters 4.3.2 and 4.4.1. Here, there are two ground states $|g_1\rangle$ and $|g_2\rangle$, with one excited state $|e\rangle$, and one laser coupling the states $|g_1\rangle$ and $|e\rangle$. Decay from $|e\rangle$ is allowed to both ground states with rates $\gamma_{e,g_1}$ and $\gamma_{e,g_2}$. The Hamiltonian and governing differential equations for
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This system are shown in equations 4.43 and 4.44. This is a trivial extension to the two-level model as comparison of the coherences in these two density matrices show very similar results.

\[
\tilde{\rho}_{g,e}(t) = \frac{1}{2} \left( -i\gamma - 2i\Delta \tilde{\rho}_{g,e}(t) - i\Omega(t) \left( \tilde{\rho}_{e,e}(t) - \tilde{\rho}_{g,g}(t) \right) \right) 
\] (5.28a)

\[
\tilde{\rho}_{g_{1,1},e}(t) = \frac{1}{2} \left( -i\gamma_{\text{Tot}} - 2i\Delta \tilde{\rho}_{g_{1,1},e}(t) - i\Omega(t) \left( \tilde{\rho}_{e,e}(t) - \tilde{\rho}_{g_{1},g_{1}}(t) \right) \right) 
\] (5.28b)

where \( \gamma_{\text{Tot}} = \gamma_{e,g_{1}} + \gamma_{e,g_{2}} \). It can be seen that the governing coherence equations are the same except for a change in decay rate. Hence, a three-level atom passing through a Gaussian pulse with temporal duration of \( 2\tau_{\sigma} \) from 1/e intensity points, has a Voigt spectral profile of:

\[
\alpha = N\omega\hbar \frac{\pi\Omega^2}{2I} V(\Delta; \gamma, \sqrt{2}/\tau_{\sigma}) 
\] (5.29)

\[
\Delta\omega_{\text{FWHM}} = 0.54\gamma_{\text{Tot}} + \sqrt{0.22\gamma_{\text{Tot}}^2 + \left( \frac{4.71}{\tau_{\sigma}} \right)^2} 
\] (5.30)

It should be noted that this spectral width is only valid for low Rabi frequencies, since the assumption is made that the atomic populations are minimally perturbed.

Using the above formula, the transit-time broadening of an atom traversing the optical fibre can be determined. The natural decay rate of the 5\(^{P}_{3/2}\) energy level in Rb is \( \gamma_{\text{Tot}} = 2\pi 6.07 \text{ MHz} \) and the average transit time is \( 2\tau_{\sigma} = 150 \text{ ns} \), under typical experimental conditions as described at the beginning of this section. Using equation 5.30, the spectra FWHM in this situation is \( \Delta f_{\text{FWHM}} = 8.98 \text{ MHz} \). The natural linewidth of the atom is 6.07 MHz, hence there is a 2.92 MHz broadening induced by transit-time broadening.

**Velocity Averaging**

So far the derived transit-time broadening models lack a very important element: the random distribution of velocities and trajectories through the Gaussian beam. This concept was discussed in chapters 4.5 and 4.6, specifically how numerical integration can be used to generate absorption profiles which include this added layer of complexity.

The problem of random atomic trajectories and velocities traversing the optical mode was discussed in detail in a paper published by Bordé et al. [154], in which a saturated absorption spectrum was derived. The derivation conducted in this paper is very detailed and includes: atomic energy level lifetimes, atomic collisions, full
atomic motion dynamics, and the flight time of atoms across the optical mode. Furthermore, they extend the optical mode complexity by modelling a TEM\textsubscript{00} Gaussian mode taking into account the radius of curvature and phase changes of the beam along the propagation axis.

Bordé \textit{et al.} [154] perform a number of different calculations to derive the absorption profile. An intermediate result is the transit-time broadening of a particular velocity class crossing the optical mode, which they reported to be:

\[ \langle \alpha(\Delta) \rangle = N \omega \hbar \frac{\pi \Omega^2}{I} V(\Delta; \gamma, \sqrt{2}/\tau_\sigma) \]  

(5.31)

This is in good agreement with the derived broadening for an atom passing through a Gaussian light pulse as derived above resulting in equation 5.26c.

Extending this result, the paper integrates over trajectories and velocities to derive transit-time broadened linewidths observed in a saturation spectroscopy experiment to be:

\[
\Delta \omega_{\text{FWHM}} = \begin{cases} 
1.511/\sqrt{\gamma \tau_{2\sigma}} & \tau_{2\sigma} < 1/(2\gamma) \\
0.94 + 0.66/(\gamma \tau_{2\sigma}) & \tau_{2\sigma} < 5/\gamma \\
1 + 2.5/(\gamma^2 \tau_{2\sigma}^2) & \tau_{2\sigma} > 5/\gamma 
\end{cases} 
\]  

(5.32)

In reference [154] these equations are presented in terms of the variable \( \eta \), however, the equations have been converted into the notation used here for consistency, where \( \tau_{2\sigma} \) is the transit time across the \( 1/e \) diameter of a TEM\textsubscript{00} optical mode. A comparison is made to the formalisms derived here in figure 5.4. The observed difference arises from averaging over velocities and trajectories. Through this, reference [154] reported a surprising result in that slow moving atoms contribute to the lineshape more than faster atoms, hence producing a narrower linewidth than expected. This is explained by slow moving atoms interacting with the probe beam for longer, hence scattering more photons, whilst simultaneous having a narrower linewidth due to their longer transit time.

Later Bordé published a paper [155], that derived the transit-time broadening in a low-pressure vapour taking into account molecular recoil and the second-order Doppler effect using a complete quantum approach. The result was that there is no broadening associated with the time taken to cross an optical beam “if the medium is homogeneous and isotropic without nearby walls or potentials susceptible to interfere with the absorption process” [155]. The key point here is that a HC-PCF introduces a wall that interferes with the absorption process by terminating it prematurely. Hence, transit-time broadening is still expected to occur within the fibres.
What is presented in equation 5.32 and shown in figure 5.4 is for a saturated spectroscopy experiment [154]. The experiments that were conducted within the fibre were of a slightly different nature, that is they are two-photon transitions.

5.1.2 Transit Time Modelling of Two-Photon Transitions

Transit-time broadening in the case of the two-photon excitation described in section 3.1.2, can be calculated using the density matrix model for lasers arranged in a counter-propagating configuration, as presented in section 4.4.2, in equation 4.49. A Gaussian laser mode is assumed, leading to a temporal light profile of \( \Omega_{g,a} \rightarrow \Omega_{g,a}(t) = \Omega_{g,a,0}e^{-(t/\tau_{\sigma})^2} \) and likewise for \( \Omega_{a,b} \). Again the assumption that the atomic populations are minimally disturbed is employed %\( \tilde{\rho}_{g,g}(t) \approx 1 \) and %\( \tilde{\rho}_{a,a}(t) = \tilde{\rho}_{b,b}(t) = \tilde{\rho}_{c,c}(t) \approx 0 \), leading to the coherence equations:

\[
\begin{align*}
\tilde{\rho}_{g,a}(t) &= (i\Delta_a - \frac{\gamma_{a,g}}{2}) \tilde{\rho}_{g,a}(t) + \frac{i\Omega_{a,b,0}e^{-(t/\tau_{\sigma})^2}}{2} \tilde{\rho}_{g,b}(t) + \frac{i\Omega_{g,a,0}e^{-(t/\tau_{\sigma})^2}}{2} \tilde{\rho}_{a,b}(t) \\
\tilde{\rho}_{g,b}(t) &= (i\Delta_b - \frac{\gamma_{b,tot}}{2}) \tilde{\rho}_{g,b}(t) + \frac{i\Omega_{a,b,0}e^{-(t/\tau_{\sigma})^2}}{2} \tilde{\rho}_{g,a}(t) - \frac{i\Omega_{g,a,0}e^{-(t/\tau_{\sigma})^2}}{2} \tilde{\rho}_{a,b}(t) \\
\tilde{\rho}_{a,b}(t) &= (i(\Delta_b - \Delta_a) - \frac{\gamma_{a,g} + \gamma_{b,tot}}{2}) \tilde{\rho}_{a,b}(t) - \frac{i\Omega_{g,a,0}e^{-(t/\tau_{\sigma})^2}}{2} \tilde{\rho}_{g,b}(t) + \frac{i\Omega_{a,b,0}e^{-(t/\tau_{\sigma})^2}}{2} \tilde{\rho}_{g,a}(t) 
\end{align*}
\] (5.33)

As they stand above, the coherence equations can not be solved analytically, hence numerical methods were employed. To simplify the numerical solutions, the random velocity and trajectories of atoms traversing the optical mode were ignored as they can be averaged over at a later stage as discussed in section 4.6.

The numerical method used to model the transit-time broadening for arbitrary light pulse durations is similar to that discussed in section 5.2, where equation 4.57 was solved over the parameter set \( (x, y, v_t, \Delta) \). Here, the Gaussian mode-shape of the beam is ignored momentarily (removing the \( x, y \) and \( v_t \) parameter space), and replaced with a solution set over transit time, detunings, and Rabi frequency, i.e \( (\tau, \Delta_a, \Delta_b, \Omega_{g,a}, \Omega_{a,b}) \). The model was initiated at \( \tau = -3\tau_{\sigma} \) from the light pulse centre, where the light field is negligible (i.e. \( 10^{-4} \) of the peak field strength) and the coherences in equation 5.33 are initially set to zero. The atom is propagated through the light field and numerical methods are used to calculate the coherences at each time step until the model in terminated at \( \tau = 3\tau_{\sigma} \).

The absorption coefficient was calculated by integration as described by equa-
Figure 5.5: Comparison between the two- and single-photon transition’s transit-time broadened linewidth. The data points represent results from the numerical model for both single- and two-photon transitions. A fit to the two-photon transition linewidth is presented, along with the single-photon transition analytic result derived in equation 5.27. Both the spectral widths and transit times are normalised to the atoms natural decay rate $\gamma_{e,g}$.

Equation 5.15b to give:

$$\alpha_{a,b} = N\hbar \omega_{a,b} \frac{\int_{-3\tau_{\sigma}}^{3\tau_{\sigma}} \Omega_{a,b}(t) \Delta \rho_{a,b}(t, \Delta) d\tau}{\int_{-3\tau_{\sigma}}^{3\tau_{\sigma}} I_{a,b}(t) d\tau} = N\hbar \omega_{a,b} \frac{\Omega^2_{g,a,0} \Omega^2_{a,b,0}}{I_{a,b,0}(4\Delta^2_a + \gamma^2_{a,g})} V\left(\Delta_b; \gamma_{b,tot}, \frac{2}{\tau_{\sigma}}\right)$$

(5.34)

where $\Omega_{g,a,0}$ and $\Omega_{a,b,0}$ are the peak Rabi frequencies for the lasers coupling the $|g\rangle \rightarrow |a\rangle$ and $|a\rangle \rightarrow |b\rangle$ transitions respectively, and $V\left(\Delta_b; \gamma_{b,tot}, \frac{2}{\tau_{\sigma}}\right)$ is the Voigt profile described in chapter E. In the limit of $\tau_{\sigma} \rightarrow \infty$, equation 5.34 simplifies to the product of the first two Lorentzian factors in equation 4.55, being the steady state solution. The single-photon absorption described in equation 4.55 is present in the numerical solutions, however, is not presented in equation 5.34 as it is too far detuned to be observed experimentally, hence not modelled.

The pre-factor to the Voigt function matches that of the steady-state solution described in section 4.4.2, equation 4.55. The Voigt profile $V\left(\Delta_b; \gamma_{b,tot}, \frac{2}{\tau_{\sigma}}\right)$ has a FWHM which is described by:

$$\Delta\omega_{\text{FWHM}} = 0.54\gamma_{b,tot} + \sqrt{0.22\gamma^2_{b,tot} + \left(\frac{8\sqrt{\log(2)}}{\tau_{2\sigma}}\right)^2}$$

(5.35)
5.1. SPECTRAL BROADENING MECHANISMS

where the linewidth is composed of a Lorentzian with FWHM of $\gamma_{b,\text{tot}}$ and Gaussian FWHM $8 \sqrt{\log(2)} / \tau_{2\sigma} \approx 6.66 / \tau_{2\sigma}$, where $\tau_{2\sigma}$ is the 1/e electric field beam diameter. It can be seen that in the limit that $\tau_{2\sigma} \to \infty$ the numerical modeling suggests that $\Delta \omega_{\text{FWHM}} \to \gamma_{b,\text{tot}}$ which is in agreement with the steady-state solution. The numerical methods used here were double checked by modelling transit-time broadening for a two-level system, which was derived analytically and is shown in figure 5.5. The results matched the analytic expressions exactly giving confidence to the methods used.

The results of this modelling show a different spectral width to that of a single-photon transition as seen in figure 5.5. The transit-time broadened spectral width of the two-photon transition is a factor of $\sqrt{2}$ larger than that of the single-photon transition. This is due to the intensity dependence of the two-photon transition effectively narrowing the optical mode as seen in figure 5.6.

Monte-Carlo simulations and analytic methods were used to include averaging over the Gaussian laser beam, described in chapters 4.6.2 and 4.6.1 respectively. For both of these methods, the two-photon lineshape was given by equation 5.34. The Monte-Carlo simulation accumulated the transit-time broadened two-photon absorption lineshape of an ensemble of random atomic velocities and trajectories crossing the fibres optical mode. This was compared to numerical integration methods by replacing $O \to \alpha_{a,b}$ in equation 4.65 by the two-photon lineshape. The results are shown in figure 5.6 to be in good agreement.
5.1.3 Power Broadening and Light Shifts

Solutions to the two-level density matrix equations, as derived in section 4.3.1, show that the spectral width of an atomic transition can be broadened by the interrogating laser beam power. Equation 4.40 derived that the absorption coefficient for a steady-state light-atom interaction takes a Lorentzian profile of:

\[ \alpha(I, \Delta) = N \omega \hbar \frac{\Omega^2}{I} \frac{\gamma^2}{\gamma^2 + 2\Omega^2 + 4\Delta^2} \]  

(5.36)

with a spectral FWHM of:

\[ \Delta \omega_{\text{FWHM}} = \gamma \sqrt{1 + \frac{I}{I_{\text{sat}}}} \]  

(5.37)

where \( \gamma \) is the excited state’s decay rate, \( \omega \) is the transition frequency, \( \Omega \) is the Rabi frequency and \( I \) is the intensity of the probing light.

The physical origin of this broadening mechanism is the saturation of the atomic transition [144]. As the optical intensity of the driving laser increases, the atomic populations are driven towards \( \tilde{\rho}_{g,g} = \tilde{\rho}_{e,e} = 1/2 \) at which point they are saturated. As the transition saturates, the spectral width of the transition broadens in conjunction with a reduction in absorption [144].

The presence of a strong optical pump also causes Rabi splitting of the atomic energy levels, whereby the pumping laser’s optical field splits the energy of the atomic levels being pumped [144]. Such splitting can not be observed in the spectrum of the pumping laser, as the induced energy splitting shifts with respect to pump laser detuning as shown in figure 5.7. A weak probe laser coupled to an independent excited state is required to observe the splitting, as shown in figure 5.7. Solving the steady-state density matrix equations for this energy level configuration produces a map revealing how the energy levels shift with respect to pump laser detuning, as shown in figure 5.7. In this example, the decay rates are set to be equal \( \gamma_{g,a} = \gamma_{g,b} = \gamma \), the pumping Rabi frequency was set to \( \Omega_p = 10\gamma \), while the probe/signal Rabi frequency was \( \Omega_s = 0.01\gamma \).

It can be seen in figure 5.7 that for a pump detuning of \( \Delta_p = 0 \) the atomic transition is split into two absorption features of equal strength, located at a frequency detuning of \( \Delta_p = \pm \Omega_p/2 \). A splitting in the ground state \( |g\rangle \) energy level is the cause of the observed splitting on the \( |g\rangle \rightarrow |b\rangle \) transition. As a function of pump
5.1. SPECTRAL BROADENING MECHANISMS

Figure 5.7: An energy level diagram (left) for a modelled atom that exhibits light shifts and Rabi splitting. A resulting spectral map showing energy level splitting (right). In this example the pump Rabi frequency was $\Omega_p = 10\gamma$. The detuning axes are normalised to the decay rates $\gamma_{g,b} = \gamma_{a,g} = \gamma$.

As the pump is detuned away from resonance, there is a residual shift in the ground state energy level, seen in figure 5.7 at large detunings of $\Delta_p$. This effect is known as a light shift or AC Stark shift. It is this shift, induced by the pump beam, which gives rise to dipole trapping and other such effects.

Both the splitting and shifting of the atomic energy levels are masked within an effusive atomic vapour due to the random velocities and trajectories of atoms through the pump and probe beams. Atoms of different trajectories passing through the beam sampling different intensity profiles, hence experience different amounts of splitting and shifting. To fully model this in an effusive vapour, numerical techniques such as those discussed in sections 4.5 and 4.6 are used, which is discussed in section 5.2.
5.1.4 Magnetic Field Broadening

Magnetic field broadening arises from Zeeman splitting that is not spectrally resolved within a magnetic field where the induced splitting is under the natural linewidth. As discussed in section 3.1, the hyperfine energy levels of Rb, labelled by the integer \( F \), consist of \( 2F + 1 \) magnetic sub-levels. These levels are labelled with the integer value \( m_F \) which ranges from \( -F \leq m_F \leq F \). In the absence of a magnetic field these sub-levels are degenerate, however, when a magnetic field is applied, the degeneracy is lifted [147, 152]. This is caused by the electronic angular momentum \( J \) and nuclear spin \( I \) vector precessing around the total angular momentum vector \( F \), which is itself precessing the applied magnetic field [147, 152]. In the weak magnetic field limit, the energy shift of the magnetic sub-level \( m_F \) is [147, 152]:

\[
\mathcal{E} = g_F \mu_B B m_F \tag{5.40}
\]

where \( \mu_B \) is the Bohr Magneton, \( B \) is the applied magnetic field, \( m_F \) is the magnetic sub-level number. The hyperfine Landé factor \( g_F \) is given by [130]:

\[
g_F \simeq g_J \frac{F(F + 1)I(I + 1) + J(J + 1)}{2F(F + 1)} \tag{5.41}
\]

and:

\[
g_J \simeq 1 + \frac{J(J + 1) + S(S + 1)L(L + 1)}{2J(J + 1)} \tag{5.42}
\]

where the approximations \( g_S \approx 2 \) and \( g_L \approx 1 \) were used along with ignoring nuclear terms being as they are much smaller. These approximations are correct to \( \approx 0.1\% \) level [130].

Splitting of the magnetic sub-levels by a constant magnetic field is not strictly a broadening mechanism. However, if the magnitude of the splitting is less than the linewidth of the sub-levels, the levels will blur together to form a broader spectral profile compared to the case where there is no magnetic field present. Furthermore, if a fluctuating magnetic field is present (for example from electrical components) the energy shift of the magnetic sub-levels will fluctuate, broadening their spectral profile.

The overall shape of the broadened lineshape is affected by the polarisation of the probing laser [147]. Optical pumping between magnetic sub-levels is generated by the interaction of a magnetic field and optical fields. The coupled sub-levels depends on the orientation of the magnetic quantisation axis relative to the optical polarisation axis. Optical pumping of this nature has the ability to change the
absorption spectrum of the transition.

Incorporation of the aforementioned effects into the density matrix equations is not necessary as each sub-level transition can be treated as independent. Hence, this effect can be included after other broadening mechanisms are modelled.

5.1.5 Pressure Broadening and Shifts

Atomic collisions between optically excited atoms and background gases are another mechanism that generates spectral broadening. Such collisions can either be elastic or inelastic, both of which effect the spectral lineshape of the resulting absorption profile. Elastic collisions do not cause a state change as this would require energy to leave the optically excited atom. However, phase changes of the atomic dipole can occur which result in broadening and shifting of atomic absorption features. On the other hand, inelastic collisions have the ability to cause a change of atomic state, prematurely shortening the atomic state’s lifetime, and hence effecting the spectral absorption profile.

Much research has been conducted in this area, owing to the substantial effect collisions with background gases have on absorption spectra. In the case of Rb collisions, background gases that have been investigated include: the noble gases [158–161], Nitrogen [160], Hydrogen [162, 163] as well as organic molecules like: Methane [163], Ethane and Butane [164]. Pressure broadening has been measured for both the Rb $D_1$ and $D_2$ transitions [160, 163, 164]. Highly excited Rb states (Rydberg states) have also attracted much research [158, 162, 165, 166], as the collisional broadening has some dependence on the excited state of the colliding atom. A summary of the results from these publications are shown in figure 5.8. Interestingly, no previous work has reported the pressure broadening and shift of the $5S_{1/2} \rightarrow 5D_{5/2}$ transition, although measurements have been made of the $5S_{1/2} \rightarrow 6P_{3/2}$ transition [159] which is a decay route from the $5D_{5/2}$ energy level, discussed in chapter 3.

Background Pressure Broadening

From the data summarised in figure 5.8, a rough estimate of the broadening expected by the $5D_{5/2}$ energy level can be predicted. For moderate to high principal quantum numbers $n$, discussed in chapter 3.1, figure 5.8 shows a decreasing rate of pressure broadening. This is because the size of an excited atom increases proportional to $n^2$ (according to the Bohr model of an atom [144]), hence the atoms core and valence electron become increasingly separated. Consequently the probability of finding the
Figure 5.8: A summary of published data for pressure broadening (top) and shifts (bottom) due to an Argon buffer gas on a range of excited atomic states. The $D_2$ averaged data is taken from [163, 167–169], $6P_{3/2}$ data is taken from [159], Bruce et al. is from [166] and Weber et al. is from [158].

For the low to intermediate principal quantum numbers, a van der Waals interaction is a good first-order approximation for the pressure broadening mechanism [158, 164]. This approximation is based on the long-range second-order dipole-dipole interaction and assumes that the excited atom and its collisional partner are not identical. Furthermore, the collisional partner’s energy level separations are assumed to be much greater than those of the excited atoms [144]. The van der Waals approximation is given by [144, 158, 164]:

$$
\gamma/N = \frac{8.16}{2\pi} \bar{v}^{3/5} C_6^{2/5}
$$

(5.43)

where $N$ is the number density of the vapour, $\bar{v}$ is the mean relative speed of the collision pair and $C_6$ is the van der Waals constant or Lennard-Jones constant. The left hand side of equation 5.43 is in units of broadening rate per unit of pressure.
The van der Waals constant, $C_6$, is given by [144, 158, 164]:

$$C_6 = \frac{e^2 \alpha}{\hbar} \left( \langle \tilde{R}_e \rangle^2 - \langle \tilde{R}_g \rangle^2 \right)$$  \hspace{1cm} (5.44)

where $e$ is the charge of an electron, $\alpha$ is the ground state dipole polarisability of the collisional partner, and $\langle \tilde{R}_e \rangle^2 - \langle \tilde{R}_g \rangle^2$ is the difference in the square radii of the orbits of the ground and excited states of the optically excited atom. This square radius of an orbit is given by [144, 158, 164]:

$$\langle \tilde{R}_i \rangle^2 = \frac{(n_\ast^i)^2 a_0^2}{2} \left( 5(n_\ast^i)^2 + 1 - 3l_i (l_i + 1) \right)$$  \hspace{1cm} (5.45)

where $n_\ast^i$ is the effective principal quantum number and $l_i$ is the orbital angular momentum quantum number for the ground and excited states $i = g, e$ and $a_0$ is the Bohr radius. It should be noted that when calculating equations 5.43 and 5.44, cgs units were used as the polarisability is typically quoted in these units [170].

Predictions of broadening rates from equations 5.43 and 5.45 match literature reported values, as shown in figure 5.8. It can be seen that the theory agrees with the broadening value for the $5P_{3/2}$ and $6P_{3/2}$ levels. Extending this to the $nD$ levels shows that the theoretical broadening agrees with experimental results for $n = 7, 8$ where, from this point on, the broadening starts to decrease with increasing $n$ which is not captured by this theory. For the $5D$ excited state, the calculated pressure broadening due to an Argon buffer gas, is $\approx 37$ MHz/Torr. Pressure broadening of the $5D_{5/2}$ state with a Ne buffer gas has been indirectly measured through an EIT experiment [161]. They reported a broadening of $6 \pm 1$ MHz/Torr which is in broad agreement (within a factor of 5) with that predicted from equations 5.43 and 5.45 which is 27 MHz/Torr. A similar broadening rate is calculated for collisions between nitrogen molecule ($N_2$), oxygen molecule ($O_2$), and carbon dioxide ($CO_2$), as their average electric dipole polarisabilities ($N_2$: 1.740310$^{-24}$ cm$^3$; $O_2$: 1.568910$^{-24}$ cm$^3$; $CO_2$: 2.91110$^{-24}$ cm$^3$ [170]) are of the same order of magnitude to that of Argon 1.641110$^{-24}$ cm$^3$.

**Background Pressure Shifts**

Accompanied with pressure broadening are shifts of the atomic spectra caused by the interaction of the excited atom with the background gas. The shift rate per unit pressure can be estimated using a van der Waals interaction approximation in the same way the broadening rate was calculated in equation 5.43. The pressure shift
Figure 5.9: A summary of published data for Rb-Rb self-pressure broadening effects (top) and lineshifts (bottom) on a range of highly excited atomic states (Rydberg states). Weber et al. is taken from [171] and Stoicheff et al. is taken from [172]. The theory curve is $\propto n^{2.4}$ [172].

is given by [164]:

$$\delta f/N = \frac{-2.96}{2\pi} \bar{v}^{3/5} \epsilon_6^{2/5}$$  (5.46)

The only difference between the pressure shift and broadening calculations (equations 5.43 and 5.46) is the scaling pre-factor, where the shift is approximately a factor of 2.76 smaller. Figure 5.8 shows that this approximation is in agreement with measurements well between the principal quantum numbers 7 to 12 [158]. Beyond this region, the large separation between the nucleus and valence electron starts to play a role. This can also be seen to occur in the pressure broadening and described previously. For principal quantum numbers below 8, equation 5.46 gives a small overestimate of the shifts expected.

Self Broadening and Shifts

In addition to broadening and shifts caused by alien gases, collisions between Rb atoms also induce broadening and shifts to the absorption spectra. The polarisability
of Rb atoms in their ground state are an order of magnitude larger than that of the buffer gases mentioned above \((47.310^{-24}\ \text{cm}^3\ [170])\). Hence, the broadening and shifts due to Rb-Rb collisions are an order of magnitude more severe, as seen in figure 5.9. Furthermore, measurements of the broadening rate of the \(D_1\) and \(D_2\) transitions yielded \(\approx 2\ \text{GHz}/\text{Torr}\) and \(\approx 4\ \text{GHz}/\text{Torr}\) respectively \([151, 173]\).

To predict the pressure broadening and shifts expected for the \(nD\) states, the equations used above (equations 5.43 and 5.46) unfortunately cannot be used. This is because they assume that the collision partners are not identical and their energy levels are very different \([144]\), both of which are violated in the case of Rb-Rb collisions.

The effects of Rb-Rb self broadening and shifting was investigated by Weber et. al. \([171]\) and Stoicheff et. al. \([172]\) and the data presented in these papers is summarised in figure 5.9. An extrapolation of this data to the \(5D\) excited state can be obtained using a shift and broadening rate proportional to \(n^{2.4}\) \([172]\). This results in an estimation of the broadening and shift rates for the \(5S \rightarrow 5D\) transition of \(\approx 50\ \text{MHz}/\text{Torr}\) and \(\approx -16\ \text{MHz}/\text{Torr}\) respectively.

5.2 Population Pumping

One of the main focuses of the work presented within this thesis was to determine the broadening mechanisms present within the HC-PCF. As discussed in the previous chapter, there are many various types of broadening mechanisms which have been modelled to varying degrees. To experimentally investigate what processes effect the spectral lineshape within the fibre, a hole-burning experiment was conducted. This is discussed in chapter 7, using the \(D_1\) and \(D_2\) transitions, which were discussed in section 3.1.1. In such an experiment, a strong laser field at a fixed frequency, pumps a specific velocity class of atoms from one ground state to the other through an excited state, as shown in figure 5.10. The resulting population profile is then probed with a weak laser field, coupled to a different excited state, revealing a lack of absorption on the pumped ground state, known as a “hole”, and increased absorption on the collecting ground state.

A density matrix model describing this system was presented in section 4.4.2 and will be briefly revisited here. A diagram showing the energy level configuration, coupling lasers and possible decay routes is shown in figure 5.10. The ground state \(|g_2\rangle\) is pumped with a strong laser of Rabi frequency \(\Omega_{g_2,a}\), to the excited state \(|a\rangle\), at a fixed detuning of \(\Delta_a\). Once in this state, the atom can decay back either ground state \(|g_2\rangle\) or \(|g_1\rangle\). It is the \(|a\rangle \rightarrow |g_1\rangle\) decay channel which creates the population
hole in the $|g_2\rangle$ ground state, and hence abundance of population in the $|g_1\rangle$ ground state. A weak laser of Rabi frequency $\Omega_{g_2,b}$, probes the $|g_2\rangle \to |b\rangle$ transition, and by scanning the detuning $\Delta_b$, the ground state population changes, resulting from the pump laser, can be observed. In the experiments conducted in chapter 7, the excited states $|a\rangle$ and $|b\rangle$ correspond to $5P_{1/2}$ and $5P_{3/2}$ respectively.

There are a number of main broadening mechanisms present within the HC-PCF for this experiment, namely transit-time and power broadening. An average transit time of 150ns across the 36 $\mu$m optical mode will generate transit-time broadening. Optical intensities generated within the HC-PCF can be very high, even for modest input powers, due to its small cross-section. For example, $\approx 1 \mu W$ of optical power within the fibre generates intensities of $\approx 1 kW/m^2$. As a result, the strong $D_1$ and $D_2$ transitions saturate at low input power, generating considerable power broadening at typical operating powers. Furthermore, at these intensities light shifts of the atomic levels will occur within the vapour.

To disentangle transit-time broadening from power broadening, an extrapolation to low optical power is required for the experimental data collected. However, this requires a functional form which can be fitted to the data and used for extrapolation. As discussed in section 5.1.1, transit-time broadening can be calculated under the assumption that optical powers are small enough, such that negligible populations perturbations occur. Hence, this is not conducive to deriving a functional form that includes power broadening and light shifts. Furthermore, chapter 5.1.3 derived expressions describing power broadening and light shifts under steady-state conditions, which is clearly violated in the fibre due to the short interaction time. As a result of these problems, the only avenue is to numerically model the whole problem so that transit-time broadening, power broadening and light-shift are incorporated.

This was achieved by numerically solving the equations of motion for this light-atom configuration as described in section 4.4.1 by equations 4.45 and 4.46. The numerical methods used to solve the equations of motion are those described in section 4.5. An absorption coefficient, and hence spectral absorption profile, were then calculated as described in section 4.6.1 by evaluating equation 4.58 and making the substitution described in equation 4.62. An example spectra returned from these numerical methods is shown in figure 5.10. In this example the following parameter were used: a pump Rabi frequency of $\Omega_{g_2,a} = 5$, probe/signal Rabi frequency of $\Omega_{g_2,b} \ll 1$, pump detuning of $\Delta_a = 0$, a beam diameter of 36 $\mu$m, and Rb temperature of 60 °C. In figure 5.10 the reduction in absorption is clearly evident in the Doppler broadened profile.

The burnt hole takes the form of a Voigt profile, as described in appendix E,
5.2. POPULATION PUMPING

Figure 5.10: Energy level diagram of the hole-burning experiment (left) presented in chapter 7. An example hole-burn spectra returned from numerical modelling (right, dots) for the parameters $\Omega_{g_2,a} = 5$, $\Omega_{g_2,b} \ll 1$, $\Delta_a = 0$, a beam diameter of 36 $\mu$m, $T = 60^\circ C$ and vapour cell length of 0.1 cm. A fit to the returned spectra using equation 5.47 is also shown (line).

which is fitted to the simulated data using the function:

$$T(\Delta_b) = \exp[\alpha (1 - \beta V(\Delta_h; \gamma_H, \sigma_H)) V(\Delta_b; \gamma_D, \sigma_D)]$$  \hspace{1cm} (5.47)

where $\alpha$ and $\beta$ are the depth of the Doppler absorption and hole respectively, $\gamma$ and $\sigma$ represent the Lorentzian FWHM and Gaussian $1/e$ half-width respectively for the Doppler absorption and hole, labelled by the subscripts $D$ and $H$.

Running these numerical methods for a variety of pump Rabi frequencies $\Omega_{g_2,a}$ provides information as to how the hole broadens with input power. The Voigt FWHM was extracted from fitting the resulting spectra from a variety of pump Rabi frequencies which are presented in figure 5.11. Both the Lorentzian and Gaussian components broaden with increasing intensity due to power broadening, Rabi splitting and light shifts being averaged over all possible velocities and trajectories. A fit to the resulting Voigt FWHM takes the form:

$$\gamma_H(I) = \gamma_{a,tot} \sqrt{1 + \frac{2\eta I_{g_2,a}}{I_{Sat}}} + \gamma_{b,tot} + \gamma_T$$ \hspace{1cm} (5.48a)

$$= \gamma_{a,tot} \sqrt{1 + \frac{\eta I_{g_2,a}}{I_{Sat}}} + \gamma_{b,tot} + \gamma_T$$ \hspace{1cm} (5.48b)

where $\gamma_{a,tot} = \gamma_{a,g_1} + \gamma_{a,g_2}$ and similarly for $\gamma_{b,tot}$, $\eta$ is a fitting parameter, $\gamma_T$ is the transit-time broadening and $I_{Sat}$ is the saturation intensity of the $|g_2\rangle \rightarrow |a\rangle$ transition.

Neglecting transit-time broadening, the spectral hole width described by equation 5.48 contains two components: the power broadened hole induced by the strong
pump, and a contribution of $\gamma_{b,tot}$. The power broadened component is induced by the population which is removed from the ground state $|g_2\rangle$. This is the feature which gives rise to the hole. The additional contribution of $\gamma_{b,tot}$ is caused by the probe laser scanning through the $|g_2\rangle \rightarrow |b\rangle$ transition. This causes a convolution between the population excited by the probe, of spectral width $\gamma_{b,tot}$, and the power broadened hole. Hence, the summation of these two linewidths.

A fit to the data presented in figure 5.11 yielded $\eta = 0.41 \pm 0.01$ and $\gamma_\tau = 0.44\gamma_{b,tot} \pm 0.04\gamma_{b,tot}$. The expected power broadening for a steady-state stationary two-level atom, as discussed in section 5.1.3, results in $\eta = 1$, hence there is a scaling applied to the saturation intensity $I_{sat}$ to yield the numerically generated results. This is not surprising as optical pumping and atomic saturation greatly perturb the observed saturation intensity [153]. The extracted transit-time broadening from figure 5.11 failed to converge to a stable resultant line width with the available computational power and time. A Monte-Carlo numerical integration method, as described in chapter 4.6, was used to evaluate the zero-power transit-time broadening which returned 0.44 MHz. This value is smaller than the broadening of 2.9 MHz calculated in section 5.1.1 as it averages over all possible atomic trajectories and velocities through the laser beam. Specifically, it indicates that atoms that spend a long time in the beam (slower moving atoms passing through the centre of the beam) contribute substantially to the line shape when comparison to the faster moving atoms, as they are able to scatter more photons.

From this modelling, it can be seen that equation 5.48 is a suitable theoretical
5.2. POPULATION PUMPING

estimate for the expected power and transit-time broadening within the fibre. This line broadening model is used in section 7 to extrapolate the measured spectral hole-burn linewidths to low powers to extract the transit-time broadening contribution within the HC-PCF.
Part III

Experiment and Results
This chapter provides an overview of the components used to conduct experiments discussed in the final few chapters. The optical set-up used for these experiments is discussed in section 6.1. Section 6.2 describes the vacuum system used to load the HC-PCF, while the loading performance of the fibres are discussed in section 6.3. Finally, section 6.4 describes a laser which was custom designed for use in the experiments reported here.

6.1 Optical Experimental Overview

This section provides a broad overview of the optical experimental components used to investigate broadening mechanisms within the HC-PCF, build a frequency standard and explore cross-phase modulation, discussed in chapters 7 - 10. The first section gives a broad overview of the base optical layout, with the following sections discussing the lasers used, frequency measurement techniques, a reference Rb system, and optical detection methods.

6.1.1 Optical Setup

The major optical components used for the experiments discussed in future chapters are shown in figure 6.1. From experiment to experiment, certain parts of the optical system were altered or added to, however the permanent base components are displayed in figure 6.1. There are three major sections to the experiment: the 780 nm laser and its frequency calibration setup, highlighted in red; the HC-PCF setup, highlighted in green; and Rb reference cell for comparison, highlighted in blue.

Each section was linked with commercial single mode fibre (SMF) and fibre based beam splitters to make the whole experiment as reconfigurable as possible. This was essential as the hole-burning experiment, described in chapter 7, and the two-photon
Figure 6.1: Overview of the optical experimental set-up. Four main set-ups are highlighted: Red - 780 nm ECDL and frequency measurement; Grey - Ti:S frequency modulation; Blue - Rb reference cell spectroscopy; Green - HC-PCF spectroscopy.

experiments, described in chapters 8 - 10, required excitation with lasers from different directions. For the hole-burning experiment, two lasers were required at 780 nm and 795 nm which were delivered into the fibre in a co-propagating configuration. In the two-photon experiments, lasers at 780 nm and 775 nm were coupled into the fibre in a counter-propagating configuration to produce Doppler-free excitation as described in section 3.1.2.

6.1.2 Lasers

Two main lasers were used in the majority of the experiments. The first was an extended cavity diode laser (ECDL), tuned to $\approx 780 \text{ nm}$ for spectroscopy of the Rb $D_2$ transitions. This ECDL was designed and built specifically for high-resolution spec-
6.1. OPTICAL EXPERIMENTAL OVERVIEW

troscopy experiments. It was designed to have a narrow spectral linewidth, achieved through minimising the laser frequency’s sensitivity to environmental acoustic noise (this will be discussed in full in section 6.4). As a result, its linewidth was \( \approx 880 \text{kHz} \) for 1 s integration time. The laser was also capable of fast frequency scan speeds of more than 2000 GHz/s, providing fast experimental feedback.

The second laser was a continuous wave titanium:sapphire (Ti:S) laser, capable of tuning from \( \approx 750 \text{nm} \) to \( \approx 800 \text{nm} \). This allowed access to either: the Rb \( D_1 \) transition at 795 nm, the single-colour two-photon transition at 778 nm, or stepwise two-photon transition at 775 nm. The Ti:S system was a monolithic block resonator (MBR) which was pumped with a Verdi (a diode-pumped solid-state laser operating at 532 nm) both of which were purchased from Coherent Scientific. The linewidth of the Ti:S was \( \approx 210 \text{kHz} \) for 1 s integration time, however its frequency scan speed and feedback was much slower than the ECDL.

6.1.3 Frequency Calibration and Measurement

To measure the frequency of the ECDL and Ti:S, three complementary systems were used. The first method comprised of a Doppler-free atomic spectrometer which provided relatively course absolute frequency information, while simultaneously a optical cavity generated finer relative measurements. The absolute frequency reference used saturated absorption features of \(^{87}\text{Rb}\) to provide absolute frequency markers. The hyperfine transitions used were the \( F = 2 \rightarrow F' = 3 \) and \( F = 1 \rightarrow F' = 2 \), as marked in figure 6.2. The absolute frequency of these transitions are 384.228, 115, 202 THz and 384.234, 683, 234 THz respectively, hence provided a course absolute scale as they are separated by \( 6.568031 \text{GHz} \).

Resonances of a Fabry Pérot optical cavity provided relative frequency measurements as shown in figure 6.2. The length of the optical cavity was \( \approx 0.5 \text{ m} \) producing a free spectral range (FSR) of 297 MHz, calibrated using the aforementioned \(^{87}\text{Rb}\) hyperfine transition’s frequency separation of 6.568031 GHz. The cavity mirrors had a radius of curvature of 500 mm and reflectivity > 95%, which provided a finesse of \( \approx 75 \) and cavity bandwidth of \( \approx 4 \text{ MHz} \). A contrast of 50% was obtained producing excellent signal-to-noise frequency markers every 297 MHz.

The wavelength of both lasers could also be measured using a wave-meter which provided wavelength measurements with a resolution of 0.01 pm, corresponding to \( \approx 5 \text{ MHz} \) at 780 nm. The absolute accuracy of the measurements was limited to \( \approx 200 \text{ MHz} \) due to temperature drifts and in-coupling alignment into the optical cavities within the wave-meter.
Figure 6.2: A typical frequency calibration spectra. The resonances of an optical cavity are shown along with a saturated spectra of the Rb D2 transition.

The most accurate measurement of the laser’s frequency was acquired by comparison to an optical frequency comb. A Hydrogen maser provided stable timing signals for the comb, resulting in frequency measurements with a potential precision and accuracy of 100 Hz. Measurements were made by producing an optical beat note between the optical modes of the comb and both the ECDL and Ti:S, which were detected using fast photo-diodes and measured using counters. This frequency measurement scheme was used when the highest precision and accuracy measurements were required, such as when measuring the stability of optical frequency standards. It was impractical to use the frequency comb for all frequency measurements due to “dead zones”, where frequency measurements became ambiguous. These zones are described in detail in appendix B, which contains a journal paper resulting from experimental work which circumvented these “dead zones”.

6.1.4 Comparison Rb Cell Setup

A reference Rb bulk cell was used in order to compare the spectra retrieved from the Rb loaded HC-PCF to that of an unperturbed system. The experimental set-up for this reference is shown by the blue section in figure 6.1. Using this set-up, both the hole-burning and two-photon experiments conducted within the HC-PCF were able to be compared to an unperturbed system. The reference-cell had a diameter of 25 mm and length 70 mm containing a natural mixture of Rb isotopes with no buffer gas, hence there was no spectral broadening or shifts from collisions with background gas or with the cell wall, both of which are discussed in chapter 5.
6.1. OPTICAL EXPERIMENTAL OVERVIEW

Figure 6.3: Photo of the Rb reference-cell holder. Key elements are indicated.

The reference-cell was held within an aluminium block, as shown in figure 6.3, to minimise temperature gradients. A photomultiplier tube (PMT) was mounted on-top of the cell to detect decaying 420 nm fluorescence from the two-photon transition, as described in section 3.1. At either end of the holder were two solenoids, used to apply an artificial magnetic field to investigate Zeeman splitting. These were arranged in a Helmholtz configuration in an effort to produce a uniform magnetic field throughout the cell, within the constraints of the holder’s geometry.

A 100W thick-film power-resistor was able to heat the aluminium holder up to 150°C as necessary. A thick-film power-resistor was used as it produces substantially lower magnetic fields when compared to a typical wire-wound power-resistor. To ensure efficient heat transfer between the holder and Rb cell, and minimise temperature gradients within the cell, the holder was designed and manufactured with tolerances of < 1 mm between these two components. A thermocouple was mounted within the holder to give an accurate measure of the block’s temperature, and hence the temperature of the Rb cell.

The reference-cell, its holder and peripherals were placed within a mu-metal shield to reduce ambient magnetic fields, which would perturb the energy level structure of Rb through Zeeman splitting, discussed in section 5.1.4. An attenuation ratio of > 100 was measured, limited by the magnetic-field sensor, which reduced the ambient magnetic field from ≈ 500 mG to < 5 mG.

As a result of its design, the spectral absorption from reference-cell had negligible unwanted broadening effects such as: transit-time, pressure and magnetic field broadening (all of which are discussed in chapter 5). Hence this provided a good source of reference spectra for comparison. Spectra for both the HC-PCF and
CHAPTER 6. EXPERIMENTAL APPARATUS

reference-cell were taken simultaneously to ensure systematic noise, sources such as laser frequency noise, are common to both measurements. These spectra were compared to isolate spectral broadening mechanisms and shifts occurring within the HC-PCF.

Either absorption or fluorescence measurements were used to detect Rb transition spectra within the reference-cell. For detection of the two-photon transition, 420 nm fluorescence measurements were made using the above mentioned PMT, as absorption of the pump lasers was minimal. The PMT required shielding from external light sources as the 420 nm fluorescence was detected at power levels varying between 1 pW - 10 nW, depending on experimental conditions. To provide this shielding, the reference-cell and PMT were placed within two layers of wooden boxes.

6.1.5 Detectors and Detection Methods

A number of different detection methods were used throughout the experimental work presented here. Typically, detection of low-power probe-beams or fluorescence was necessary, requiring photomultiplier tubes or avalanche photo-diodes. For both of the major experiments, either hole-burning or the two-photon transition, two beams were used in counter- or co-propagating configurations. This required efficient methods for separating the beams after passing through either the cell or HC-PCF. Both the detectors and methods for efficiently separating the bi-chromatic beams are discussed below.

Detectors

Typically, low-optical powers on the order of 1 nW to 1 μW, between 776 – 795 nm were used to probe Rb transitions, while avoiding atomic transition saturation effects within the HC-PCF. Furthermore decaying 420 nm fluorescence from the two-photon excitation within the reference-cell was of the order of 1 pW - 10 nW. These low powers and varying wavelengths provided challenges for efficient and low-noise detection. There were two options for detectors, either a photomultiplier tube (PMT) or an avalanche photo-diode (APD).

A variety of PMTs were used for situations where low-optical-powers were involved. Photomultiplier tubes consist of a photo-cathode which is coated in a material with a low work-function. Incoming photons liberate electrons from the photo-cathode material through the photoelectric effect. A high driving voltage accelerates the electrons through an array of dynodes, which amplify the number of photoelectrons initially produced. The final dynode collects these amplified elec-
6.1. OPTICAL EXPERIMENTAL OVERVIEW

...trons which form the PMT’s output. As a result of being based on the photoelectric effect, PMTs are most efficient at short wavelengths. This is ideal for detection of the 420 nm decaying fluorescence from the two-photon excitation. However, when detecting infra-red wavelengths, such as those typically used here: 776 nm, 780 nm, or 795 nm, the efficiency of photo-electron production is reduced. For this task, specially designed PMTs were used which had photo-cathode materials with lower work-functions. The Hamamatsu PMTs used had quantum efficiencies of \( \approx 30\% \) at 420 nm, dropping to \( \approx 3\% \) at 780 nm. Although there is an order of magnitude drop in quantum efficiency towards the long wavelengths, this level of efficiency is still orders of magnitude better than PMTs not designed for this task.

When in dark conditions, the major component of noise within the PMT is thermionic emission [174], for typical accelerating voltages used here. This effect is caused by the low work-function of the photo-cathode material, causing thermionic electrons to be emitted from the photo-cathode. The flux of these electrons \( i_s \), is temperature dependent, taking the form [174]:

\[
i_s \propto T^{5/4} e^{-e\phi/(k_B T)}
\]  

where \( e \) is the electron charge, \( \phi \) is the photo-cathode material’s work function, \( k_B \) is the Boltzmann constant and \( T \) the absolute temperature of the photo-cathode. This temperature dependence became an issue when heating either the reference-cell or HC-PCF to increase the Rb density, and hence detectable signal. The proximity of the PMT to the heated HC-PCF or reference-cell, led to an elevated operating temperature where thermionic emissions within the PMTs generated substantial noise. As a result, the experiments were run at temperatures which compromised between thermionic emission noise and photon shot-noise.

When exposed to incident light \( > 1 \) pW, the PMT’s output noise is dominated by shot-noise, originating from the random arrival times of photons. In the PMT, this manifests as a fluctuating rate of photon-electrons from the photo-cathode [44]. The magnitude of the shot-noise depends on the number of detected photons, and hence the PMT’s quantum efficiency, \( \eta \) [174]. The shot-noise induced standard deviation of a detected signal takes the form [44]:

\[
\sigma = \sqrt{2ei} \Delta f
\]

where \( e \) is the charge of an electron, \( i \) is the average photo-current and \( \Delta f \) is the measurement bandwidth. The average photo-current can either be measured or...
calculated from the incoming photon flux $\Phi$, to be $[44]$:

$$i = e\eta \Phi \quad (6.3)$$

Thus the signal-to-noise ratio scales as:

$$(S/N) = \sqrt{i}/\sqrt{2e\Delta f} \quad (6.4a)$$

$$= \sqrt{\eta \Phi}/\sqrt{2\Delta f} \quad (6.4b)$$

It is evident that the more efficient the detector, the greater the signal-to-noise ratio of the detected signal.

For certain applications APD’s were used as their quantum efficiency at longer wavelengths is superior to that available using PMTs. An APD is a highly reversed-biased photodiode (PD) in which photoelectrons are capable of exciting more electrons (into an avalanche) before being detected $[44]$. As a result, APDs are also efficient at low-power level. Unfortunately the available APDs were filtered such that only frequencies between $5 - 1000$ MHz were present at their outputs. Hence they were used for low-power self-heterodyne measurements where an optical beat note is generated in the range of the available APDs’ output, as discussed in chapter 9.

For measurements of optical powers above $1 \mu W$, standard photodiodes were used. These consisted of large- and small-area photo-diodes depending on the bandwidths noise requirements.

Detection Methods

A range of methods were used in order to separate bi-chromatic beams which were typically used in the experiments discussed here. The simplest method was to produce a bi-chromatic beam consisting of two lasers with opposite linear polarisations. Hence, after the Rb cell or HC-PCF, the two beams could be separated using a polarising beam splitter (PBS) to the 1% level. However, often a beam could not be created with adequate control over the polarisation for this to be used. These situations arose when: two lasers are mixed in a fibre-based beam splitter, or when retro-reflections with undefined polarisation arises in a bi-chromatic beam being formed from originally counter-propagating beams. In both of these situations polarisation selection could not be employed.

An alternative method to separate two beams was to use frequency discrimination. Assuming the two optical beams were separated by more than $1$ nm in
6.1. OPTICAL EXPERIMENTAL OVERVIEW

Figure 6.4: Diffraction grating with groove spacing $d$ and light incident at an angle $\alpha$ is diffracted at an angle $\beta$ related by equation 6.5.

wavelength (for wavelengths around 780 nm) a diffraction grating was used to separate the beams. The angle from the normal at which a beam is diffracted off a diffraction grating is given by [175]:

$$\beta(\lambda, \alpha) = \sin\left(\frac{m\lambda}{d} - \sin(\alpha)\right)$$

where $\lambda$ is the wavelength of light, $m$ is the diffraction order, $d$ is the grating’s groove spacing and $\alpha$ is the angle of incidents, from the normal, of the incoming beam as illustrated in figure 6.4. The minimum wavelength separation between two beams was typically $\approx 5$ nm, between 775 nm and 780 nm, which, when using a grating with 1800 grooves/mm, resulted in a angular separation of $\approx 1^\circ$ for an input angle of $\approx 30^\circ$. Using this method, beams of random polarisations, but separated in wavelength, could be easily separated.

For the 420 nm decay fluorescence measurements, spectral filtering was required to eliminate stray reflections from the 780 nm and 775 nm pumps, since their powers were orders of magnitude larger than the power of the fluorescence. To achieve this, dichroic filters were used which displayed intensity attenuation of $> 20$ dB around 780 nm while only 1 dB around 420 nm.

Detection Scheme

Within the reference-cell, the optical intensities used to excite the two-photon transition were insufficient to generate substantial absorption within the exciting beams. As a result, fluorescence measurements were used to monitor the rate of two-photon excitation. Fluorescence is generated during a cascading decay from the excited $5D_{5/2}$ state, through the $6P_{3/2} \rightarrow 5S_{1/2}$ transition which generated 420 nm fluorescence. To detect this fluorescence, a PMT was placed as close as possible to the Rb cell to increase the solid angle of the fluorescence projected onto the PMT. This
seen in figure 6.3, where the PMT is mounted directly above the Rb cell.

To detect fluorescence within the fibre a PMT was placed on-top of the fibre as indicated in figure 6.1. This produced an excellent capture efficiency, however, the short transit time across the fibre quenched a large fraction of the atoms before they were able to decay and emit 420 nm fluorescence.

A substantially larger two-photon absorption coefficient within the HC-PCF, enabled absorption measurements to be conducted that were not possible for the two-photon transition within the reference-cell. Absorption measurements were conducted for the hole-burning experiments after the HC-PCF and reference-cell. For these measurements, the laser selection methods described above were employed and detected with the aforementioned detectors. The detected fibre transmission was a direct measure of the absorption coefficient $\alpha(\omega)$, derived in chapters 4 and 5 through the Beer-Lambert Law:

$$I(\omega, z) = I(\omega, 0)e^{-\alpha(\omega)z} \quad (6.6)$$

where $z$ is the path length through the Rb vapour. Absorption measurements were only conducted within the cell when conducting the hole-burning experiments.

The discussion thus far has concerned the optics and reference-cell. The next two sections describe the vacuum system used to load the HC-PCF with Rb and loading performance of both HC-PCF’s used.

### 6.2 Vacuum System

A vacuum system was employed to load the HC-PCF with Rb for a number of reasons: firstly, Rb has a low vapour pressure, as discussed in section 3.2.1; second, Rb is highly reactive, as discussed in section 3.2.2, and would readily react with atmosphere providing no useful vapour within the HC-PCF; finally, the absence of background gas avoids pressure broadening and shifts of the Rb spectra, as discussed in section 5.1.5. As a result a vacuum system was required, capable of reaching pressures of at least $10^{-7}$ Torr in order to provide a clean system in which to load the HC-PCF with Rb.

To generate long interaction lengths between the confined atoms within the HC-PCF and optical field, a long length of fibre was needed to be filled with Rb. As a result, it was practical to build the vacuum system such that both ends of the HC-PCF were within the vacuum while the mid-section was outside and able to be coiled up as necessary. This led to a design as shown in figure 6.5 and discussed
6.2. VACUUM SYSTEM

6.2.1 Chamber Design

The two vacuum chambers were constructed to hold each end of the HC-PCF. These were built from six-way crosses so that there was sufficient optical and vacuum access into each chamber. The top most chamber flange was connected to the rest of the vacuum system so that each chamber could be pumped on. At this connection, both chambers had vacuum valves, as shown in figure 6.5, so they could be sealed off from the supporting vacuum system in the event that it needed to be brought up to atmosphere.

A pair of flanges on opposite sides of the vacuum system were fitted with windows for optical access, to allow monitoring of Rb pressures within the vacuum chambers. A further optical access port was present for coupling into and out of the HC-PCF. Opposite this coupling window, a fitting was made to hold the fibre and create a vacuum seal around it. Further details regarding the mounting of the fibre within the vacuum system are discussed in section 6.2.3.

Only one of the chambers was loaded with a Rb vapour, in this chamber the bottom flange was used to connect the Rb source which is described in more detail in the next section. Again, at this connection there was another valve to form a seal between the chamber and Rb source. This was used in the event that the Rb source required replacement as it minimised exposure of the chamber and HC-PCF to atmosphere. On the other hand, it also isolated the Rb from exposure to atmosphere when the chamber required opening.

Figure 6.5: Schematic (left) and photo (right) of vacuum system used to load the HC-PCF. Key elements are indicated.
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6.2.2 Rubidium Source

To load the HC-PCF with Rb, a reliable Rb source was required which ideally did not require continuous replacing, as this would result in exposing the vacuum system and fibre to atmosphere to some degree. There were two Rb source options available: the first was a Rb getter, a Rb impregnated resistive strip of metal that releases Rb as it is heated; the second was a glass ampoule filled with 1 g of Rb. Both of these sources have their downsides: the getter only contains micrograms of Rb, while the ampoule needs to be broken within the vacuum adding complexity to the vacuum system. The large quantities of Rb available in the ampoule was an overriding factor, and hence an ampoule was used.

To break the glass ampoule within the vacuum required a custom vacuum fitting to be manufactured. A tube of thin-walled vacuum-quality stainless steel was used to house the Rb ampoule, which was designed to have tolerances between the ampoule and tube walls of < 5 mm. The thin wall thickness was ≈ 0.5 mm, and hence was easily malleable. To crack the Rb ampoule under vacuum, a pair of pliers was used to apply force through the thin-walled stainless steel onto the ampoule. This resulted in shattering the glass ampoule within the vacuum. This technique proved to be reliable and repeatable, as no leaks were formed within the thin-walled stainless steel over a number of trials.

6.2.3 Fibre Holder

To ensure the integrity of the vacuum system, the vacuum seal onto the fibre had to be sufficient to maintain a pressure of $10^{-7}$ Torr within the vacuum chamber. This was achieved by manufacturing the fibre holder and vacuum seal shown in figure 6.6. A commercial vacuum flange blank was fitted with a conical housing, with a threaded exterior, through which the fibre passed. A cap screwed onto the conical housing, compressing a Viton gasket within it, and hence onto the HC-PCF. This formed the vacuum seal between the vacuum system and fibre. This seal proved to be reliable and durable, able to withstand temperatures of up to 150 °C while maintaining vacuum pressures of ≈ $10^{-7}$ Torr within the loading chamber.

A stainless steel support fitted with a hypodermic needle supported the fibre within the vacuum chamber, as seen in figure 6.6. This was necessary as the focussing optics required to couple efficiently into the HC-PCF, placed constraints on the distance between the coupling lens and HC-PCF. Furthermore, vibrations of the end of the fibre needed to be minimised to ensure efficient in-coupling into the HC-PCF. Typically the fibre was within 15 mm of the coupling window, hence ≈ 50 mm
of fibre was supported in this fashion.

### 6.2.4 Temperature Control

To load the HC-PCF efficiently a large flux of Rb atoms needs to be present at the entry to the fibre. This was achieved by heating the Rb ampoule and vacuum chambers to increase the vapour pressure, hence increasing the Rb flux into the HC-PCF. Difficulty was experienced because of the strong temperature dependence of Rb vapour pressure, which caused Rb to condense onto the coolest point of the vacuum system. The following subsections describe the methods involved to increase the vapour pressure while avoiding Rb condensing on critical elements within the vacuum system.

**Ampoule Heater**

In order to increase the Rb vapour pressure within the loading chamber, the ampoule was heated to a range of 100 – 150°C. This resulted in Rb atoms evaporating off the 1 g source and moving towards the loading vacuum chamber. The temperature of the ampoule was set to below that of the loading chamber by at least 10°C to avoid all of the Rb being transported to a cooler part of the vacuum system. A resistive wire, driven by a current controlled power supply, was used to heat the Rb ampoule and the vacuum section connecting it to the loading chamber.

**Chamber Heater**

The vacuum chambers themselves were wrapped with resistive wire and heated to a range of 100 – 150°C to maintain the increased vapour pressure produced at the ampoule. To reduce the number of cold points, the chamber and heaters were wrapped in aluminium foil to ensure even heating. A thermocouple was used to monitor the chambers temperature.
Window Heaters

The coldest point of the vacuum chambers were their windows which allowed coupling into the HC-PCF and probe beams to monitor the Rb pressure. Initially these windows were not heated and Rb condensed on them readily, degrading optical throughput. Heaters were designed and installed onto the windows to provide even heating while still allowing optical access. The temperature of the windows was held at a higher temperature than the rest of the vacuum chamber so that Rb did not condense upon them. This technique proved successful as it eliminated the Rb condensation onto the windows.

Fibre Heater

The section of HC-PCF which was outside the vacuum chambers required heating to promote transportation of Rb along the fibre’s length. As this was the desired location for the Rb, the fibre was typically held just below the temperature of the chamber. To achieve this, the fibre was rested on-top of a 10 mm thick aluminium plate. On the underside of this plate a 100 W thick-film power-resistor was mounted which enabled heating of the plate up to 150°C. A thermocouple monitored the aluminium plates temperature, and hence the HC-PCF. A post made of fibre glass supported the heating plate and also isolated it from the environment.

Cold Points

Certain elements of the vacuum were cooled down to \(< 10 \, ^\circ\text{C}\) to constrain Rb from flooding the entire vacuum system. A water-cooled thermoelectric cooler (TEC) was used to reduce the temperature to this level. Thermal conductivity between the TEC and vacuum system was improved through manufacturing close fitting aluminium blocks to encase the vacuum sections to be cooled. Thermally conductive paste was also used to ensure maximum heat transfer. As a result, these blocks achieved a Rb pressure drop from \(10^{-5}\) Torr to \(10^{-7}\) Torr.

6.2.5 Buffer Gas

To facilitate cleaning the HC-PCF of containments, the vacuum system was designed so that a high purity gas could be flushed through the HC-PCF. A vacuum port was capable of being connected to standard Swagelok fittings, which enabled gases to be vented into the system in a controlled manner. The point at which the gas was introduced, as shown in figure 6.5, enabled only the Rb loading chamber to be filled,
creating a large pressure differential between the two ends of the fibre, and hence a continuous stream of gas through the fibre was able to be produced. Methods for cleaning and loading the fibre with Rb are discussed in further detail in the next section.

6.2.6 Pumps

Two types of vacuum pump were used extensively with this vacuum system, being a: turbo pump, and ion pump. Initial roughing of the vacuum system from atmosphere to pressures in the range of $10^{-5} - 10^{-6}$ Torr were achieved with the turbo pump. Furthermore, this pump was used when the vacuum system was baked out to remove contaminants on the vacuum walls.

Ion pumps are designed to pump on low pressure systems, hence were only used after the system was baked out using the turbo pump. This pump has the advantage that there are no mechanical parts, hence does not introduce acoustic vibrations to the system which would significantly affect coupling into the HC-PCF. A Varian ion pump was used, which provided a pressure read-out at the pump itself. Prior to installation of the cold points onto the vacuum system, an ion pump would last less than a year pumping on the system. After inclusion of the cold points, the ion pump lifetime was extended to at least two years.

6.3 Loading the HC-PCF

The vacuum system and components described in the last section were used in attempts to load two separate fibres. Initial loading attempts made use of a 7-cell defect HC-PBF, which is discussed in sections 2.2 and 2.4, and a SEM cross-section image and transmission plot of this fibre shown in figure 2.3 (a). Loading this fibre proved to be unsuccessful due to poor vacuum conductance as discussed in the next two sections. The second loading attempt proved to be successful, in which a single-cell defect kagomé HC-PCF was used, discussed in sections 2.2 and 2.4, with a SEM cross-section image and transmission plot of this fibre shown in figure 2.3 (b). This section gives a brief description regarding vacuum conductance within a HC-PCF, the consequential Rb loading performance and effects of the Rb upon the HC-PCFs used.
CHAPTER 6. EXPERIMENTAL APPARATUS

6.3.1 Vacuum Conductance

The conductance of a vacuum system is a measure of how easily atoms flow between two parts of the system. When considering Rb loaded within a HC-PCF it is valid to classify the system as being in the ballistic flow, or molecular flow regime, as the atoms are unlikely to collide with each other. For a Rb vapour heated to $100^\circ C$, the mean free path of a Rb atom before collision with another Rb atom is $\approx 0.5 \text{ m}$, much longer than the geometry of the HC-PCF or loading vacuum chamber. In this regime, the conductance of a long tube is given by:

$$C \propto v_0 \frac{d^3}{L}$$  \hspace{1cm} (6.7)

where $d$ and $L$ are the diameter and length of the tube respectively, and $v_0$ is defined as in equation 4.61. It is evident that the core size of the fibre will be a critical factor in determining the rate of fibre filling.

6.3.2 Loading Performance

As discussed previously, attempts were made to load two different fibres: a 7-cell defect HC-PBF, with core diameter of $5 \mu\text{m}$; and a single-cell defect kagomé HC-PCF with core dimensions of $45 - 51 \mu\text{m}$ for the short and long axis. These fibres were discussed in more detail in section 2.4. A number of techniques and loading procedures were trialled and tested to load both fibres, which are discussed in the next two sub-sections.

Absorption measurements of Rb through the vacuum chamber and fibres were made in order to measure the quantity of Rb loaded into the fibres. Fitting measured absorption spectra using the Beer-Lambert law, as described by equation 4.25c, produced an absorption depth $A$ for both the fibre and cell, which is related to the absorption coefficient by $A = \alpha L$. Using the known length of the vacuum chamber through which a monitor laser passes $L_{\text{Chamber}}$, an absorption coefficient is obtained. Assuming that the fibre is at the same temperature as the chamber implies that the absorption coefficient in the HC-PCF and chamber are approximately equal $\alpha_{\text{Fibre}} \approx \alpha_{\text{Chamber}}$. This is a valid assumption for the first 50 mm of fibre which is within the chamber. A corresponding vapour length within the fibre $L_{\text{Fibre}}$ can be obtained:

$$L_{\text{Fibre}} = \frac{A_{\text{Fibre}}}{A_{\text{Chamber}}/L_{\text{Chamber}}} \hspace{1cm} (6.8)$$

This gave a direct measure of how successful the loading attempt and technique was.
Initial attempts were made at loading the 5 \( \mu \text{m} \) HC-PBF, which were followed with attempts to load the 45 \( \mu \text{m} \) kagomé HC-PCF.

**HC-PBF**

Initial attempts to load a HC-PCF started with the 5 \( \mu \text{m} \) HC-PBF. This fibre was chosen as its small core diameter produces high intensities, ideal for excitation of the two-photon \( 5S_{1/2} \rightarrow 5D_{5/2} \) transition in Rb. Four different attempts to load this fibre were carried out with lengths of fibre ranging from 0.5 m to 1 m. To load the HC-PBF the temperature of the chamber and ampoule were heated to \( \approx 100 \degree \text{C} \) as discussed previously, and hence produced a dense Rb vapour. Under these conditions, the atom flux into the HC-PBF is orders of magnitude larger than at room temperature.

Data from each of these loading attempts showed that the length of loaded HC-PBF with Rb vapour was minimal and typically could not be resolved with the above mentioned technique. Loading data from one of these attempts is displayed in figure 6.7, where the optical depths within the chamber and fibre were monitored for almost a month after exposure to Rb, revealing no significant increase in absorption length within the fibre.

A possible reason for the lack of Rb within the HC-PBF was contamination of the fibre’s core with water, oxygen or any other element or molecule that reacts with
Figure 6.8: Optical depth measurements of the kagomé HC-PCF and chamber over the length of a month. Using these measurements and equation 6.8, the length of the kagomé HC-PCF is presented.

Rb. This seriously degrades the loading performance as described in section 3.2.2. To remove contaminants, high purity Helium was flushed through the fibre to clean the HC-PBF. This was achieved by loading one chamber with 2 atm, 1520 Torr, while maintaining a vacuum in the other chamber using a turbo pump. No substantial change in loading performance was observed from implementing this technique.

It was concluded that loading Rb into such a small core with long fibres needs a different technique due to the low vacuum conductance within the fibre. Such a small core increases the resistance that Rb atoms experience within the fibre, making any motion down the fibres length extremely slow. Furthermore, the small core size restricts the rate of atoms entering the fibre though their ballistic motion.

**Kagomé HC-PCF**

A switch to the larger 45 μm kagomé HC-PCF was made to alleviate, to some extent, the low vacuum conductance within the fibre as well as the small flux of atoms entering the fibre. Increasing the fibre core size from 5 μm to 45 μm increased the vacuum conductance, and hence loading efficiency, by a factor of > 700. A length of kagomé HC-PCF used in these loading attempts was ≈ 0.7 m.

Using the same techniques as that for the HC-PBF, as described above, the length of HC-PCF filled with Rb was observed to increase within a month of being exposed, as shown in figure 6.8. After being exposed to Rb for four months, strong
fluorescence from the Rb $D_2$ transitions was detected in the fibre outside the vacuum chamber, hence more than 10 cm of fibre was eventually loaded with Rb.

Although the initial loading time is long, it was observed that refilling the same fibre without exposing it to air progressed much faster, as shown in figure 6.9. Refilling the same fibre was required at times when the vacuum system required opening for reasons such as replacing the Rb source or optical windows. When this was required, care was taken to ensure the HC-PCF was minimally contaminated by atmosphere. The fibre refilled much faster as contaminants present in the initially loading period, were permanently removed through Rb reactions; this was the cause of the long initial loading time. However, when subsequently reloaded, these contaminants were not present, hence loading occurred unhindered by this process.

It was this 45 $\mu$m kagomé HC-PCF loaded with Rb which produced the results presented in the following chapters. Further efforts to load the 5 $\mu$m HC-PBF were not attempted as the loading time would be too long to prove practical. This particular loading technique, using a dense thermal cloud of Rb atoms, is probably not the most efficient method for loading HC-PCF of any description. Other possible methods are described in chapter 12.
6.3.3 Throughput Reduction

Although the kagomé HC-PCF was successfully loaded with Rb, there was an unexpected side effect, a reduction in off resonance laser transmission through the fibre. This effect was most noticed in the 5 μm HC-PBF as shown in figure 6.10, for one of its loading attempts. The 45 μm kagomé HC-PCF also showed a reduction in transmission, however not to the extent seen in the HC-PBF. A transmission reduction of ≈ 60%, resulting in a ≈ 10% throughput, through the 40 cm of loaded kagomé HC-PCF was observed after loading for 8 months.

There are two possible explanations for this effect occurring: firstly, Rb could be forming large lumps within the fibre which scatters large amounts of light out of the core. This seems the most likely as cutting back the end of a loaded fibre by ≈ 5 mm typically increase the fibres transmission back to its original value. This points towards solid Rb forming at the entrance to the fibre and scattering in-coupled light. A second explanation could be because Rb is coating the core walls of the fibre, producing a layer of material with a refractive index very different to the silica walls. As the fibre is not designed for this, it is possible that the optical guidance properties of the fibre is reduced. This second explanation has not been able to be tested.

Figure 6.10: Reduction in laser throughput of HC-PBF during Rb load.
6.4 Acoustically Immune Extended Cavity Diode Laser

To conduct the spectroscopy experiments discussed in the next few chapters, an acoustically immune extended cavity diode laser was designed. A number of factors drove the motivation behind this design. Firstly to make precise measurements of the Rb absorption features, a single mode, narrow linewidth laser was required, especially as the two-photon absorption features are on the order of a MHz in width. This makes interpretation of the observed atomic spectra much simpler as there is negligible width added into the spectra from the laser. Hence, determining the spectral broadening mechanisms within the HC-PCF would be greatly simplified.

Secondly, a narrow linewidth laser also simplifies the equipment and procedures required to stabilise the laser to these absorption features in order to create a frequency standard. Small deviations from the atomic transition lessen the requirement on the electronic bandwidth necessary to maintain the laser’s frequency in resonance with the atomic transition. Furthermore, long-term laser frequency stability is desired, as the electronic feedback would not require large ranges which are often difficult to engineer reliably. Additionally, long-term stability is preferable for experiments conducted over extended periods of time, so that active frequency control is not required.

The dominant factor that affects the linewidth of an extended cavity diode laser ECDL laser are acoustic vibrations within the laser cavity. In a tunable laser, springs or flexures are used to add frequency tunability, however they also vibrate and generate frequency side-bands on the laser’s carrier frequency. These vibrations are typically driven by ambient acoustic noise in the environment, picked-up by the laser’s structures. As an example, a standard designed ECDL which was previously used within the laboratory, was sensitive to acoustic noises and displayed a laser linewidth of 1.9 MHz at an integration time of 1 s. This particular laser was su-

![Figure 6.11: Littrow configuration for an ECDL. The extended cavity is formed between the laser diode and diffraction grating.](image-url)
perseded by the new design as its linewidth was simply too large for the precision spectroscopy needed for this experimental work.

The following sections describe certain aspects of the laser’s design. Specifically the design of the flexures to avoid acoustic sensitivity is presented, along with the cavity design to provide optimal linewidth, tuning range and long-term stability. Finally the performance of the laser is characterised.

### 6.4.1 Laser Design

The laser was designed in a *Littrow* configuration, whereby the first order reflection off a diffraction grating is reflected back into the laser diode to form an external cavity [176], as seen in figure 6.11. In this configuration, the laser oscillates at the frequency which maximises the overall laser gain [176]:

\[
G_{\text{total}} = G_D D T_D T_{\text{inner}} T_{\text{outer}}
\]

where \(G_D\) is the laser diode gain profile, \(D\) is the dispersion of the diffraction grating, \(T_D\), \(T_{\text{inner}}\) and \(T_{\text{outer}}\) are the transmissions of the three optical cavities formed between
6.4. ACOUSTICALLY IMMUNE EXTENDED CAVITY DIODE LASER

Figure 6.13: Complete design of the acoustically immune ECDL with its lid removed. The flexure is visible inside the laser’s walls.

front and rear facets of the diode, front facet and diffraction grating, and rear facet and the diffraction grating respectively. Details regarding the functional form of each of these transmission, dispersion and gain terms can be found in reference [176]. Using the design parameters for this laser, a gain profile is shown in figure 6.12.

The transmission of the optical cavities formed between the laser diode and the diffraction grating depends on the spacing between the diode and grating. By making the length of the external cavity longer, the extended cavity’s FSR decreases, increasing its frequency selectivity, thus producing a narrower linewidth laser. The drawback to this is that the finely spaced modes of the cavity tend to produce a limited frequency scan range as the laser mode-hops from one cavity FSR to the next. This is evident when computing the cavity transmission spectra $T_D$, $T_{\text{inner}}$, and $T_{\text{outer}}$ as given in reference [176]. As a result, a compromise must be made between intrinsic laser linewidth and frequency tunability without mode-hops.

The dispersion of the diffraction grating can be shifted dependent on the angle of the diffraction grating relative to the laser diode, and hence can be used to tune the lasing frequency of the diode. By tuning the grating angle far enough, the maximum gain point can change from one cavity mode to another, inducing a mode-hop. By simultaneously tuning the length of the cavity and the angle of the grating, these mode-hops can be reduced by maintaining the maximum gain on a single cavity resonance.

It is evident that for long-term stability the length of the extended cavity needs to be held constant. As the temperature of the environment changes, the structure that determines the length of the external cavity shrinks and expands, inducing mode-
hops. Therefore, the temperature of the laser requires stabilisation and control, this can be achieved through both passive design and active control.

The requirement for a narrow linewidth with large tuning range, and good long-term stability lead to the design shown in figure 6.13. An extended cavity length could be set between 15 – 25\,\text{mm} with a grating angle of 44.95° which together provided a laser capable of scanning over the entire Rb $D_2$ transition at 780.24\,\text{nm}. Passive temperature stability was engineered by encasing the extended cavity in aluminium box with wall thickness’s of at least 10\,\text{mm}. Active temperature controlled ensured that the temperature of the laser did not drift with its environment.

The walls, lid and base of the laser design have another role other than just temperature control. Acoustic vibrations within the laser structure changes the cavity length, causing the laser linewidth to broaden as laser power is shifted into acoustic side-bands of the carrier frequency. To minimise the effect of this, the walls, lid and base, shield the cavity from ambient noise sources, but more importantly provide a rigid, stiff structural support for the extended cavity. By clamping the flexure that hold the grating between the lid and base, the acoustic vibrational modes were greatly reduced as it removed a degree of freedom.

6.4.2 Flexure Design

The key element of the laser design is the flexure that holds the diffraction grating that tunes the laser’s frequency. The design philosophy was to make this flexure monolithic to increase its stiffness, hence reducing the motion of vibrational modes thus reducing the laser’s linewidth. As can be seen in figure 6.14, the flexure was manufactured from a solid block of aluminium.

This flexure needs to be flexible so that the frequency of the laser can be tuned.
6.4. ACOUSTICALLY IMMUNE EXTENDED CAVITY DIODE LASER

Figure 6.15: Finite element modelling of the ECDL flexures. The fine adjustment (left) course adjustment (middle) and vertical adjustment (right) flexures are shown.

However, it also needs to be stiff to minimise acoustic resonances which modulate the extended cavity length, hence lasing frequency. These two criteria require a degree of compromise and hence this issue had to be designed accordingly.

To avoid large acoustic resonances, two levels of flexures were introduced: one very weak flexure for frequency tuning; and one stiff for finer tuning. These are both visible in figure 6.14, where the broad tuning flexure is at the base of the block and the fine tuning flexure is at the front. The weak flexure was designed to only be active during the initial course tuning stage. The bar of this flexure was held rigid in the laser’s base plate, however, the flexure support was not restricted by the base. A screw was used to pivot the support, hence grating around the weak flexure, providing course tuning. Once coarsely tuned, this flexure was disabled by clamping the support on the top and bottom surfaces using the lid and base of the laser enclosure. This isolated the weak flexure from the laser cavity. As a result, only a stiff flexure remains in the laser cavity for fine tuning of the laser’s frequency.

The design of both of these flexures were constrained by the physical properties of aluminium. Ideally each flexure should have a large angular range over which they cover, however aluminium can only deform so far until its permanently deformed. Hence the material stresses around the flexure were modelled, as shown in figure 6.15, to find a design that allowed a large angular range without permanently deforming. The final design, shown in figure 6.14, consists of broad curved flexures with minimal tight corners. Stress and strain is distributed along the length of these flexures reducing the stress at each point and giving a larger angular range before permanent deformation. In total there were three flexures designed and implemented. These were the course and fine frequency adjustment flexures discussed above, and a third for adjusting the vertical alignment of the grating to align the extended cavity.

To modulate the grating angle and cavity length a piezoelectric transducer was placed behind the stiff, fine-tuning flexure. The length of the fine tuning flexure was designed such that the angular change of the grating, and extended cavity length
Figure 6.16: Power spectral density plot of the original, acoustically immune and Vortex II ECDLs frequency noise in ambient noise conditions. An acoustically driven frequency noise PSD is shown for the original ECDL. This is not shown for the acoustically immune and Vortex II ECDLs as there was negligible deviation from their ambient PSD measurements.

changed at compensating rates to increase the mode-hope frequency scanning range. By designing the stiff flexure carefully, its resonant frequency was increased, enabling fast control of the laser frequency through the piezoelectric actuator.

### 6.4.3 Performance

The performance of features designed into the ECDL were extensively tested to ensure the quality of the laser. The crucial performance indicators are the frequency linewidth of the laser’s output, piezo modulation bandwidth, and the long-term stability. All of these can be determined from measurements of the frequency fluctuations of the ECDL which were made using a variety of frequency discriminators including: saturated absorption features within a Rb vapour, an unbalanced Mach-Zehnder interferometer, and comparison to an ultra-stable laser. For short-term frequency fluctuation measurements, the Mach-Zehnder interferometer was used due to its frequency sensitivity, however, it was not suitable for long-term measurements due to arm-length drifts. For long-term frequency measurements, saturated absorption features within a Rb vapour or an ultra-stable laser were used.

To measure the ambient frequency noise of the ECDL, an unbalanced Mach-Zehnder interferometer was used, created using lengths of optical fibre and fibre
beam splitters. The contrast of the Mach-Zehnder was $> 90\%$ and the fringe spacing was $\approx 15 \text{ MHz}$ creating a very sensitive frequency discriminator. The frequency fluctuations were recorded using a spectrum analyser, giving the power spectral density (PSD) of the laser’s frequency fluctuations. A power spectral density plot is a common method of presenting how signal power is spread over Fourier frequency space, i.e. it displays the power per unit frequency [44].

Figure 6.16 shows PSD of the acoustically immune ECDL frequency noise under ambient excitation conditions. Spectral peaks at harmonics of 50Hz are present, which correspond to the power line frequency. These features are associated with noise in the current driver, which was confirmed by direct current noise measurements of the driver’s output. At higher Fourier frequencies, between 2kHz to 20kHz, there are a number of mechanical resonances associated with the flexures holding the diffraction grating. For comparison, figure 6.16 also presents the frequency noise PSD for the superseded Littrow design, and a commercial high-quality Newport Vortex II ECDL tuned to the caesium $D_1$ transitions at 894nm. It can be seen that the acoustically immune ECDL and its predecessor have similar noise floors between 10Hz - 10kHz, while the Vortex II has a noise floor an order of magnitude lower.

Mathematical integration techniques can be used to convert a measured PSD to laser linewidth over a given time-scale [177]. The linewidths of these three lasers were calculated to be $\approx 1.9 \text{ MHz}$, $\approx 880 \text{ kHz}$, and $\approx 830 \text{ kHz}$ for the original ECDL, acoustically immune ECDL and the Vortex II respectively, integrated over 1s. The main contribution to all of these laser linewidths is from the power line resonances at harmonics of 50Hz.
Tests of the laser’s acoustic immunity was made by projecting audio tones from a high-powered audio amplifier and speaker at the ECDLs. Over the whole audio range ($\approx 10$ Hz - 20 kHz) there was little elevation from the ambient PSD measurements for the acoustically immune ECDL and Newport Vortex II ECDL. On the other hand, the previous ECDL design was highly susceptible to acoustic excitation, evident by the large increase in frequency noise shown in figure 6.16.

Measurements were also made of the modulation bandwidth available through manipulations of the grating by its piezoelectric actuator. A transfer function was measured by modulating the piezo actuator at a given frequency and measuring the magnitude of the resulting laser frequency modulation. This directly excites the mechanical resonances of the flexure design as seen in figure 6.17. A small resonance is observed at 2.3 kHz, which is dwarfed by the next resonance at 4.2 kHz. Beyond 10 kHz limited response is observed due to mechanical loading of the piezo actuator. As a result of the flexure design, a modulation bandwidth up to 2.3 kHz is possible using only the piezo control of the grating angle and cavity length. A similar test was performed on both the previous ECDL and Vortex II. The original ECDL showed large acoustic resonances, starting at 500 Hz and extending to $> 10$ kHz. These resonances are the source of the laser’s acoustic sensitivity and are caused by the commercial optics mounts used as part of the ECDL design. On the other hand, the Vortex II displays only one resonance at 6 kHz, although this has been filtered somewhat by a 2 kHz low-pass filter on the input-port of the piezo control.

Long-term measurements of the ECDL’s frequency were made in the process of measurement shown in chapter 9. These measurements were made using a commer-
cially available Menlo optical frequency comb, which has a long-term stability orders of magnitude more stable than the ECDL. An optical beat note between the ECDL and the nearest comb mode was detected and its frequency counted to provide the measurements in figure 6.18. This shows excellent stability over time scales from 1 – 1000 seconds with a maximum measured frequency drift of ≈ 2 MHz over 650 s. This stability is much better than that of the previous ECDL which demonstrated > 10 MHz over 200 s. Measurements of the Vortex II were not made, and hence are not presented.
7
Broadening Mechanisms of the D Transitions

This chapter introduces and presents the peer-reviewed publication “High Resolution Optical Spectroscopy in Hollow-Core Photonic Crystal Fiber”, published in the journal Physical Review A.

7.1 Overview

A hole-burning experiment was conducted within a kagomé HC-PCF to investigate spectral line broadening mechanisms associated with the atoms being confined within the fibre’s core. A detailed understanding of these mechanisms is required when using this system in applications such as optical atomic frequency standards or when inducing photon-photon interactions for quantum logic operations.

For the experiment presented in this paper, a kagomé HC-PCF was used (as discussed in chapter 2) which demonstrated broad wavelengths guidance, as shown in figure 2.3. A scanning electron microscope image of the fibre’s cross-section is shown in figure 2.3 which shows the fibre’s core dimensions were 45 μm and 51 μm for the short- and long-axis respectively.

To measure the broadening mechanisms within this fibre, a spectral hole-burning experiment was conducted. In this approach, a single atomic velocity class’s population, hence absorption, was perturbed through optical pumping as discussed in sections 4.4.1 and 5.2. This revealed the spectral absorption profile of the pumped velocity class, allowing detailed analysis of the atom’s absorption spectra as discussed within this paper. Saturation absorption spectroscopy experiment were not conducted as the optical depth of the Rb filled HC-PCF was large to perform such measurements.
The $D_1$ and $D_2$ transitions, described in section 3.1.1, were used to pump and probe the atomic populations respectively. These transitions were used as they are well studied and understood, making interpretation of the resulting spectra simpler.

The atomic populations were pumped with the Ti:S which was tuned to the $D_1$ transition at 795 nm, while the 780 nm ECDL was tuned to the $D_2$ transition to monitor the resulting atomic populations. The experimental set-up, as described in chapter 6, was configured so that these lasers were co-propagating through both the HC-PCF and reference-cell. A diffraction grating was used to separate the two beams after the Rb vapour.

Extensive numerical modelling of the population hole-burning, for both the HC-PCF and reference-cell, was conducted using the methods described in chapters 4 and 5. Specifically, the results from the numerical model presented in section 5.2 was used to extrapolate the linewidth of the spectral features to low-power, to avoid optical saturation effects and reveal the inherent broadening mechanisms.

It was determined that the additional broadening caused by transit-time broadening was $1.0 \pm 0.7$ MHz through these techniques.
High-resolution optical spectroscopy in a hollow-core photonic crystal fiber

C. Perrella, 1 P. S. Light, 1 T. M. Stace, 2 F. Benabid, 3 and A. N. Luiten 1

1Department of Physics, University of Western Australia, Perth, Western Australia 6009, Australia
2ARC Centre of Excellence in Engineered Quantum Systems, University of Queensland, Brisbane, Queensland 4072, Australia
3Centre for Photonics and Photonic Materials, University of Bath, Bath BA2 7AY, United Kingdom

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In this paper, we present detailed high-resolution spectroscopy of rubidium (Rb) vapor confined within a hollow-core photonic crystal fiber (HC-PCF). We find a very low level of additional frequency broadening associated with this confinement, with spectral features being only 1 MHz broader than the natural linewidth of the excited state. We show that this additional broadening is consistent solely with the atoms’ transit across the fiber’s optical mode. This low level of decoherence opens the door to a wide variety of applications including compact frequency standards and new types of quantum optical devices based on alkali-metal-loaded HC-PCFs. We highlight the low level of decoherence through observation of electromagnetically induced transparency in the confined vapor.

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I. INTRODUCTION

A recent innovation in photonics is the development of hollow-core optical fibers that simultaneously confine vapors and guide light within their cores over large distances [1]. This capability has attracted great interest for applications in frequency stabilization [2,3], electromagnetically induced transparency (EIT) [4–6], slow light [7], few-photon optical switches [8], and deterministic quantum logic gates [9]. However, the full potential of this strong light-atom interaction can only be realized if the deleterious effects of fiber-atom interaction can be minimized.

Previous work utilizing vapors [10,11] in hollow-core photonic crystal fiber (HC-PCF) has shown large dephasing effects caused by transit time [3,10,12] or pressure broadening [2,6,7]. Such effects limit this technology in applications where optical coherences play a key role.

Here we report a very low dephasing rate (∼1 MHz) within HC-PCF, which opens the door to a wide range of applications. This was achieved in untreated silica fiber loaded with rubidium (Rb) vapor. We show that small residual broadening effects are consistent with finite light-atom interaction time. We also explain our observation that the Rb vapor exhibits greatly increased saturation intensity in this geometry compared to that in a conventional cell.

Using the optical setup shown in Fig. 1, we investigated dephasing and decay processes of Rb vapor confined to the fiber core and in a reference bulk cell. Via direct fluorescent decay measurements and the saturation behavior of spectral features, we measured the influence of finite interaction time on the atomic population dynamics. Finally, we demonstrate how the low level of decoherence enables creation of highly efficient EIT. These observations are consistent with a four-level atomic model, which we discuss.

II. SETUP

We used kagome-lattice [13] HC-PCF with a single-cell core defect of diameter 45 µm (see Fig. 1). This fiber exhibits low-loss guidance from 600 to 1600 nm. Each end of the 40-cm fiber was held in separate vacuum chambers while the midsection was outside the vacuum system. Each chamber had windows to couple light into the fiber and to allow monitoring of Rb vapor densities within the vacuum chambers.

The whole vacuum system, including fiber and chambers, was outgassed at 100 °C, achieving pressures below 10−7 Torr measured at the ion pump. Rubidium was released into one chamber from a Rb ampoule at these elevated temperatures producing high Rb vapor densities and promoting diffusion into the fiber from the vacuum chamber’s high-pressure environment. Observation of Rb fluorescence scattered out the side of the fiber indicates that over half its length was filled after four months of loading. On-resonance transmission of the 85Rb D2 transition through the fiber showed an absorption coefficient αL ∼ 10. We believe this slow filling rate is associated with Rb atoms coating and pacifying the fiber core surface. After the initial load, we were able to recover the original optical depth in ∼2 days by returning the vacuum system to the loading conditions specified above. Rb vapor was observed within the fiber from 20 °C to 120 °C with the density strongly dependent on the fiber temperature. For the experiments discussed below, the temperature of the vacuum system was lowered to 60 °C to avoid extinction of the laser beam on resonance.

Two lasers were used to conduct hole-burning spectroscopy within the fiber and bulk cell: the pump was an intense Ti:sapphire laser tuned to the 795-nm D1 transition, while the probe was a tunable extended-cavity diode laser (ECDL), which scanned through the 780-nm D2 transition. In order to measure frequencies precisely, part of the probe laser was directed to an optical cavity and a saturated absorption spectrometer. The resonances of the optical cavity provided equally spaced frequency markers with a free spectral range (FSR) of ∼300 MHz. The absolute frequency of the probe and an accurate estimation of the optical cavity’s FSR were obtained using the known spacings between several of the saturated absorption features. These elements provided a frequency axis with an accuracy of 2%.

A chopper wheel or an acousto-optic modulator (AOM) enabled independent amplitude modulation of the lasers, which were combined on a polarization beam splitter as shown in Fig. 1. The combined beam was focused into the fiber core and also sent to a Rb cell. The 1/e2 intensity beam diameters
FIG. 1. (Color online) Schematic of the optical experimental setup. HWP (QWP), half (quarter) wave plate; NPBS (PBS), nonpolarizing (polarizing) beam splitter; CH, chopper wheel; AOM, acousto-optic modulator; OC, optical cavity; VNDF, variable neutral density filter; RA, rubidium ampoule; FM, flip mirror; DG, diffraction grating; PD, photodiode; and PMT, photomultiplier tube. Inset, top right: scanning electron microscope image of the kagome HC-PCF cross section.

The frequency axis is detuned from the $87\text{Rb}$ fiber on the $87\text{Rb}$ transition and probe tuned to the $D_1$ transition.

for the fiber and cell were 36 $\mu$m and 2 mm, respectively. After the fiber or cell, the pump and probe beams were separated using a diffraction grating, detected by either a photomultiplier tube (PMT) or photodiode, and then demodulated using a lock-in amplifier. Typical spectra obtained from the fiber are shown in Fig. 2(a).

III. BROADENING PROCESSES

For the remainder of this paper, we primarily consider the $^{87}\text{Rb}$ isotope and its $D_1$ and $D_2$ transitions. We denote the two $5S_{1/2}$ ground states as $|g_F\rangle$, where $F = 1, 2$ labels the total atomic angular momentum quantum number. Similarly, we denote the $5P_{3/2}$ excited state manifold as $|a_F\rangle$ and the $5P_{3/2}$ manifold as $|b_F\rangle$, where the subscripts label the hyperfine states of each manifold. This labeling convention is shown in Fig. 2(b).

To measure the broadening processes within the fiber, we used hole-burning spectroscopy, which avoids Doppler broadening effects. The Ti:sapphire laser was tuned to the $D_1$ transition to perturb the thermal equilibrium characteristics of the ground states. The resulting disturbance was measured with the ECDL, as it scanned across the $D_2$ transitions. With the Ti:sapphire laser resonant with the $|g_1\rangle \rightarrow |a_2\rangle$ transition, we observed spectral holes in the $|g_1\rangle \rightarrow |b_2\rangle$ set of transitions while also observing excess population features in the $|g_2\rangle \rightarrow |b_1\rangle$ transition [see Fig. 2(a)]. We observed three holes and three corresponding peaks due to the three excited-state hyperfine transitions in the $D_2$ Doppler-broadened resonances. We used two-color hole-burning spectroscopy rather than conventional saturation spectroscopy to avoid complications arising from optical interference and Zeeman pumping effects, which make interpretation of saturation spectroscopy features very complex.

To extract the linewidth of the resulting features, we fit the probe absorption spectrum with the form [14]

$$\exp\left\{-\frac{\alpha_{ij}}{\mu_{ij}} \left( 1 + \frac{h_{ij}\Gamma_{ij}^2}{h_{ij}^2 + 4\Delta_{ij}^2} \right) \exp\left[ -\frac{1}{2} \left( \frac{\Delta_{ij}}{\Delta_{Dop}} \right)^2 \right] \right\},$$

where $\alpha_{ij}$ and $\mu_{ij}$ are the $|g_i\rangle \rightarrow |b_j\rangle$ transition absorption coefficient and spectroscopic strength factor [15], respectively, $\Gamma_{ij}$ and $h_{ij}$ are the width and fractional height of the burnt hole (or excess population feature) respectively, $\Delta_{ij} = \omega_{ij} - \omega_{ij}$ is the laser detuning, and $\Delta_{Dop}$ is the Doppler absorption width. Varying the pump power yields the power dependence of the linewidth, $\Gamma_{ij}$, and the result is shown in Fig. 3(a).

The driven system was modelled using a simple four-level atomic model, consisting of the two excited states, $|a_j\rangle$ and $|b_j\rangle$, and the two ground states, $|g_1\rangle$ and $|g_2\rangle$. States $|g_1\rangle$ and $|a_j\rangle$ were coupled by a pump laser with Rabi frequency, $\Omega_p(t)$, and $|a_j\rangle$ and $|b_j\rangle$ were coupled by a probe laser with Rabi frequency, $\Omega_p(t)$, and $|b_j\rangle$ and $|g_2\rangle$ were coupled by a probe laser with Rabi frequency, $\Omega_p(t)$.

FIG. 2. (Color online) (a) Transmission spectra of the Rb-filled fiber on the $^{87}\text{Rb} D_2$ transitions with (blue) and without (black) the pump laser. The frequency axis is detuned from the $^{87}\text{Rb} g_1 \rightarrow b_1$ transition. Arrows indicate the position of the pumped features. (b) An energy-level diagram showing the pump tuned to the $D_1$ transition and probe tuned to the $D_2$ transition.

FIG. 3. (Color online) (a) The intensity dependence of the linewidth of the hole-burn features for the fiber and a bulk cell. The solid lines are theoretical fits to the data. (b) Fluorescence decay measurements of both the fiber and cell.
while a probe laser of strength $\Omega_p(t)$ was scanned across the resonances $|g_1\rangle \leftrightarrow |b_j\rangle$ and $|g_2\rangle \leftrightarrow |b_j\rangle$. The excited states decayed into the ground-state manifold with known branching ratios [16]. The evolution of a particular atom crossing the beam is governed by the master equation

$$\dot{\rho} = -i[H, \rho] + \sum_k \Gamma_{g_k}^{\mu} D(|a_k\rangle\langle b_j|\rho + \sum_k \Gamma_{g_k}^{\mu} D(|b_j\rangle\langle g_k|\rho,$$

$$H = (\omega_{g_k} - \omega_{p}) |b_j\rangle\langle b_j| + \omega_{a_k} |a_k\rangle\langle a_k| + \omega_{g_2} |g_2\rangle\langle g_2| + \Omega_p(t)|b_j\rangle\langle g_k|/2 + \Omega_p(t) |a_k\rangle\langle g_1|/2 + \text{h.c.}),$$

(2)

where $|g\rangle = |g_1\rangle + |g_2\rangle$ [17]. Absorption of the probe is then proportional to $\text{Im}[\Omega_p(g|b_j\rangle]$. Atoms with different speeds and trajectories will see different time-dependent pump and probe fields. To account for this, we averaged the absorption over the Maxwell-Boltzmann distribution similar to that presented in Ref. [18]. In a pump-probe experiment of this type, the absorption spectrum of the probe laser will be affected by both the pump-induced Rabi splitting [14] and transit-time-broadened Voigt profile [15]. Our calculations show that both of these effects are obscured by the distribution of atomic trajectories, resulting in a line shape that is close to Lorentzian.

The model was solved numerically, showing that $\Gamma_{ij}$ varies with intensity with a form given by

$$\Gamma(I) = \Gamma_{D_1} \sqrt{1 + I/(2I_{\text{Sat}})} + \Gamma_{D_2} + \Gamma_{\text{trans}},$$

(3)

where $\Gamma_{D_1}$ and $\Gamma_{D_2}$ are the “zero-power” linewidths of the pump and probe transitions, $\Gamma_{\text{trans}}$ is the transit time broadening, and $I_{\text{Sat}}$ is a phenomenological saturation parameter. Fitting Eq. (3) to experimental data and extrapolating to zero intensity gives FWHM linewidths of the features within the fiber of $16.8 \pm 0.4$ MHz and $15.9 \pm 0.6$ MHz for the cell.

The factors contributing to the “zero-power” widths are summarized in Table I. The linewidths of the lasers were estimated from the frequency noise inferred from intensity noise on the side of a Rb $D_2$ resonance. The ECDL Voigt linewidth was 2.55 MHz, consisting of Lorentzian and Gaussian components of 966 kHz and 1.74 MHz respectively. The Ti:sapphire Lorentzian linewidth was 62 kHz. These linewidths were convolved with the atomic linewidths to give the broadening in Table I. Ambient magnetic fields contributed a further $2.0 \pm 0.5$ MHz broadening to both the fiber and cell spectra, as estimated by comparison of saturated spectra in the cell with and without magnetic shields. In total, the fiber exhibits a “zero-power” linewidth of $1.0 \pm 0.7$ MHz broader than that of the cell. This is consistent with a predicted transit-time broadening of 0.44 MHz calculated using our four-level atomic model.

To confirm these conclusions, we also measured fluorescence decay rates. The Ti:sapphire laser was tuned to the $^{85}$Rb $g_{2} \rightarrow d_{2.3}$ transitions and an AOM was used to extinguish the light in 7 ns. A PMT detected scattered light from the Rb vapor in the fiber and the cell. Figure 3(b) shows the average of 4000 of these decay measurements. The fiber lifetime was $21.7 \pm 1.7$ ns, and that in the cell was $27.8 \pm 0.7$ ns. The cell measurement is consistent with previous measurements of the $D_2$ lifetime [16], while the reduced lifetime in the fiber is associated with quenching of excited state through collisions with the fiber wall. This is in good agreement with the prediction of average atomic lifetime within the fiber of $22.9 \pm 0.2$ ns.

### IV. Optical Saturation

There is a large difference in the effective saturation intensity between the cell and fiber measurements in Fig. 3(a). To understand this in more detail, we measured the optical saturation behavior of the $D_1, |g_1\rangle \rightarrow |a_2\rangle$ Doppler-broadened transition for which the hyperfine energy levels of this excited state are fully resolved. This is shown in Fig. 4 for both the fiber and cell, together with a theoretical fit based on the expected saturation form of the absorption [14]:

$$\alpha(I) = \alpha_0 / \sqrt{1 + I/I_{\text{Sat}}},$$

(4)

where $\alpha_0$ is the “zero-power” optical depth, $I$ is the probe laser’s intensity, and $I_{\text{Sat}}$ is the fitted saturation intensity. We find $I_{\text{Sat}}^{\text{(cell)}} = 1.2 \pm 0.1$ W/m$^2$, while $I_{\text{Sat}}^{\text{(fiber)}} = 102 \pm 7$ W/m$^2$, which is a factor of ~85 times larger than $I_{\text{Sat}}^{\text{(cell)}}$ and a factor of 2 above the two-level saturation intensity of 53.9 W/m$^2$ for this transition [16].

Ordinarily, optical saturation originates from optical pumping into uncoupled ground states. In the fiber case, the short transit time means that pumping is substantially reduced, leading to a much higher saturation intensity. A model for the saturation behavior of an open effusive atomic system crossing a Gaussian laser beam in time $t$ has been previously presented in Ref. [19]. The degree of optical pumping is

**TABLE I. Zero-power hole broadening budget.**

<table>
<thead>
<tr>
<th>Effect</th>
<th>Contribution (MHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D_1$ natural linewidth [16]</td>
<td>5.75</td>
</tr>
<tr>
<td>$D_2$ natural linewidth [16]</td>
<td>6.07</td>
</tr>
<tr>
<td>Laser linewidth (Lorentzian component)</td>
<td>1.03 ± 0.07</td>
</tr>
<tr>
<td>Laser linewidth (Gaussian component)</td>
<td>1.74 ± 0.07</td>
</tr>
<tr>
<td>Zeeman splitting</td>
<td>2.0 ± 0.5</td>
</tr>
<tr>
<td>Resultant</td>
<td>15.3 ± 0.5</td>
</tr>
<tr>
<td>Cell experimental value</td>
<td>15.9 ± 0.6</td>
</tr>
<tr>
<td>Fiber experimental value</td>
<td>16.8 ± 0.4</td>
</tr>
<tr>
<td>Additional fiber broadening</td>
<td>1.0 ± 0.7</td>
</tr>
<tr>
<td>Transit-time broadening (theory)</td>
<td>0.44</td>
</tr>
</tbody>
</table>
characterized by a parameter $\mathcal{F}$, which is an estimate of the pump-induced population transfer. $\mathcal{F} = 0$ represents no perturbation to the thermal equilibrium situation, while $\mathcal{F} = 1$ represents complete complete population transfer to the uncoupled ground state (and thus complete transparency). The condition $\mathcal{F} \sim 0.5$ corresponds to $I = I_{\text{sat}}$. For an atomic transit time $t$, and when the population transfer is small, $\mathcal{F}$ takes the form [19]

$$\mathcal{F} = \begin{cases} \frac{\{\Omega_p t/2\}^2}{\Omega_p^2 t/(2\Gamma)} & \Gamma t \ll 2 \\ \frac{\Omega_p t/(2\Gamma)}{\Gamma} & \Gamma t \gg 2, \end{cases} \quad (5)$$

Both fiber and the cell are in the regime $\Gamma t > 2$, where the perturbation to the ground state is proportional to both the pump power and the transit time. Therefore, the ratio of the saturation powers should be approximately the inverse of the ratio of the transit times for the two cases. In our case, $t_{\text{cell}}/t_{\text{fiber}} \approx 60$, while $I_{\text{sat}}^{\text{fiber}}/I_{\text{sat}}^{\text{cell}} = 85$, which are in reasonable agreement given that Eq. (5) is only valid for small $\mathcal{F}$ and ignores the distribution of transit times.

V. ELECTROMAGNETICALLY INDUCED TRANSPARENCY

A final deep insight into the effect of the pump on the probe transmission can be gained by comparing the magnitude of the spectral hole features seen in the $|g_i\rangle \rightarrow |b_j\rangle$ manifold to that of the peak features seen in the $|g_2\rangle \rightarrow |b_1\rangle$ manifold (see Fig. 2).

The ratio of hole-to-peak heights, $R = -h_{g_2}/h_{b_1}$, in Eq. (1) yields an absorption ratio, which will be 1 if the entire population from one ground state had been transferred to the other ground state. A ratio of less than unity indicates that the optical saturation behavior cannot simply be explained by population transfer between ground states.

Figure 5 shows $R$ for both the fiber and cell. We find $R_{\text{cell}} = 1.02 \pm 0.08$ and $R_{\text{fiber}} = 0.53 \pm 0.06$, independent of both intensity and $i$. Also shown are solid lines calculated from the four-level atomic model, which are in close agreement with the experimental data.

In this cell, the transit time is long enough that all excited-state population decays to the other ground state, and hence the ratio is essentially unity. In the fiber, the ratio is approximately 0.5, so population transfer alone is not sufficient to explain the apparent decrease in absorption from the pump-coupled ground state. The physical origin for the lower ratio in the fiber lies in the much higher intensities in the fiber together with a transit time comparable to the lifetime of the excited state. The short transit time and strong coherent driving of the pump laser lead to a substantial Rabi splitting of the coupled ground state. The increased transparency of the vapor is therefore explained by EIT. The probe and pump together form a V configuration for EIT, which is absent when the probe is tuned to the $|g_2\rangle \rightarrow |b_j\rangle$ [20]. Since $t^{-1} \sim \Gamma \ll \Omega_p$, implies that neither the dephasing effects of decay nor the masking effects of optical pumping can hide EIT, as they usually do in the V configuration, the effect of EIT in the fiber is substantial. Our model indicates that EIT accounts for half the hole depth seen in the $|g_i\rangle \rightarrow |b_j\rangle$ transitions. It is the action of this EIT peak that results in $R$ approaching 0.5 at the highest intensities.

VI. CONCLUSIONS

We have demonstrated that a Rb-filled HC-PCF can show narrow spectral features that are only $\sim 1$ MHz broader than the natural atomic linewidth. This additional broadening arises solely from coherent transit-time effects and can thus be further reduced with larger core fibers. The absence of any additional dephasing supports the potential for use of this technology in miniature, robust clocks and quantum optics experiments. Measurements of the fluorescent decay time of fiber-confined atoms directly demonstrates the influence of the walls on the excited-state lifetime of the atoms. Finally, in contrast to the usual observation in cells, we observed a saturation intensity of the atoms in the fiber that is of the same order as the value expected from a simple two-level model [16]. The small influence of optical pumping and the low level of decoherence are confirmed by the demonstration of EIT in the fiber confined atoms.

ACKNOWLEDGMENTS

The authors acknowledge financial support from the Australian Research Council. We also acknowledge Francois Knabe for fabricating the HC-PCF and the Australian Microscopy and Microanalysis Research Facility at the University of Western Australia, a facility funded by the university and state and commonwealth governments.

Characterisation of the Two-Photon Transition

This chapter introduces and presents the peer-reviewed publication “High Resolution Two-Photon Spectroscopy of Rubidium within a Confined Geometry”, published in the journal *Physical Review A*.

### 8.1 Overview

Characterisation of the Rb two-photon \(5S_{1/2} \rightarrow 5D_{5/2}\) transition is presented for a Rb vapour confined within a HC-PCF. Understanding the broadening mechanisms of this transition, and its absorption strength’s dependence on power, is required when using this system for applications such as optical atomic frequency standards or inducing photon-photon interactions for quantum logic operations.

A kagomé HC-PCF, as discussed in chapter 2, was used for the experiments presented in this paper. Both the fibre’s transmission, and a scanning electron microscope image of the fibre’s cross section are shown in figure 2.3. The fibre’s core dimensions were 45 \(\mu\)m and 51 \(\mu\)m for the short- and long-axis respectively.

The two-photon transition, as discussed in section 3.1.2, was excited using two lasers of different frequencies. The ECDL was tuned near the \(5S_{1/2} \rightarrow 5P_{3/2}\) transition at 780 nm, while the Ti:S was tuned to 776 nm to complete the two-photon transition to the \(5D_{5/2}\) state. A small detuning of typically < 10 GHz from the intermediate \(5P_{3/2}\) state was used to enhance the two-photon transition rate, and hence absorption strength as described in section 4.4.2.

The experimental set-up discussed in chapter 6 was arranged with the two lasers in a counter-propagating configuration to reduce Doppler broadening of the two-photon transition, as discussed in section 3.1.2. To monitor the two-photon transi-
CHAPTER 8. CHARACTERISATION OF THE TWO-PHOTON TRANSITION

tion strength, the absorption of the 780 nm ECDL was observed as this displayed stronger absorption than the 776 nm Ti:S due to a larger atomic dipole moment for this transition. The two-photon absorption strength and linewidth were explored for: varying detunings from the intermediate level, and a variety of Ti:S powers. These results were seen to be in good agreement to the theory presented in sections 4.4.2 and 5.1.2.

Spectral broadening mechanisms of the two-photon transitions were able to be more carefully examined, in comparison to those presented in chapter 7 as the analysis was simplified in this case. The counter-propagating Doppler-free configuration used intrinsically generates narrow two-photon absorption features, with no Doppler background. Tight constraints were placed on pressure broadening within the fibre by comparing spectral shifts between the reference-cell and HC-PCF. As discussed in section 5.1.5, atomic collisions cause spectral broadening and shifts, hence accurately measuring spectral shifts places constraints on the expected pressure broadening. A shift of 160 ± 130 kHz was measured, placing an upper bound on the pressure broadening within the HC-PCF of 450 ± 360 kHz. This value is both small and statistically insignificant in comparison to other broadening mechanisms. A two-photon linewidth of 10.4 ± 0.5 MHz was measured with the main contributions to the spectral width being: transit-time broadening, discussed in section 5.1.1; residual Doppler broadening, discussed in section 3.1.2; and magnetic field broadening, discussed in section 5.1.4. The transit-time contribution to the linewidth for the two-photon transition reported here was 5.61 ± 0.05 MHz compared to 1.0 ± 0.7 MHz measured using the $D_1$ and $D_2$ transitions in chapter 7. The difference is due to the small mode overlap of 13 ± 1 μm between the two driving lasers in the two-photon experiment. In contrast, for the single-photon transitions the atoms are free to interact with the light field over the full optical mode diameter of 36 μm.

Using the results from this paper, efficient design of an optical atomic frequency standard was achieved, as discussed in chapter 9, and a strong photon-photon interaction could be created, as discussed in chapter 10.
High-resolution two-photon spectroscopy of rubidium within a confined geometry

C. Perrella,1 P. S. Light,1 J. D. Anstie,1 T. M. Stace,2 F. Benabid,3 and A. N. Luiten1,4
1School of Physics, University of Western Australia, Perth, Western Australia 6009, Australia
2Department of Physics, University of Queensland, Brisbane, Queensland 4072, Australia
3Xlim Research Institute CNRS, University of Limoges, Limoges, France
4Institute of Photonics and Advanced Sensing (IPAS) and the School of Chemistry and Physics,
The University of Adelaide, Adelaide SA 5005, Australia
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We present two-photon spectroscopy of a thermal rubidium vapor confined to the hollow core of a photonic-crystal fiber. Linewidths as narrow as 10 MHz were observed on the 5S1/2 → 5D3/2 transition enabling the hyperfine splitting of the excited state to be resolved. Very strong nonlinear absorption (>90%) was observed, with substantial absorption maintained over large detunings (9 GHz) from an intermediate state. These attributes make this system ideal for many frequency metrology and quantum optics applications.

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I. INTRODUCTION

Atomic transitions involving the absorption of two photons [1] offer a platform for all-optical switches [2–4], single-photon generation [5], photon coherence measurements, and quantum logic gates [6,7], due to their ability to modulate, or sense, one light field with another. Such transitions also offer narrow linewidths beneficial for frequency metrology applications such as atomic clocks and frequency stabilization [8,9]. For these applications, nonlinear transitions are most useful when they deliver strong absorption at low optical powers. This characteristic has previously been demonstrated using resonant enhancement through an intermediate energy level together with strongly focusing optics [1,10]. Here, we extend this concept by utilizing the technology of hollow-core photonic-crystal fibers (HC-PCF) [11,12], which can deliver high optical intensities for low input powers. Furthermore, the fiber maintains tight overlap of the highly confined optical mode and atomic vapor over arbitrary lengths.

Three key properties determine the performance of two-photon transitions in the aforementioned applications: the ultimate absorption strength, the absorption linewidth, and the range of frequency detunings, from the intermediate state, over which it is possible to maintain these properties. In this paper we explore these performance criteria for the 5S1/2 → 5D3/2 two-photon transition of rubidium (Rb) [13,14] loaded into a HC-PCF. This system was chosen for its potentially narrow linewidth and strong absorption, which was further enhanced by driving the transition in near resonance with the 5S1/2 → 5P3/2 single-photon transition.

II. SETUP

Two lasers, of wavelengths 780 and 776 nm, drove the 5S1/2 → 5D3/2 two-photon transition as shown in Fig. 1(a). Specifically, we focus on the 85Rb 5S1/2 (F = 2) → 5D3/2 (F′ = 1–4) set of transitions for the majority of this paper. From the excited state, Rb can decay to the ground state through either the original intermediate state, or via the 6P3/2 energy level, which emits 420 nm fluorescence on decay back to the ground state [see Fig. 1(a)]. For the rest of this paper, the ground (5S1/2) state will be labeled |g⟩, while the intermediate (5P3/2) and excited (5D3/2) energy levels will be labeled |i⟩ and |e⟩, respectively, with natural linewidths γi and γe. Detunings from these states will be labeled Δi = ωe − ω580 and Δe = ωg − ω580 = (ω780 + ω776), where ωjk denotes the |j⟩ → |k⟩ transition frequency. The optical power coupling energy levels |j⟩ and |k⟩ will be denoted as Pjk.

The fiber used in this work was kagome-structured HC-PCF [15], with core diameter 45 μm providing low-loss guidance from 1600 to 600 nm [the fiber’s cross section is shown in Fig. 1(b)]. Each end of the fiber was held within separate vacuum chambers, only one of which was loaded with Rb, while the midsection was outside the vacuum. The fiber was curved between the vacuum chambers, which, combined with its large core diameter, resulted in coupling to higher-order transverse optical modes. The Rb vapor density within the fiber was controlled by adjusting the temperature

FIG. 1. (Color online) (a) Two-photon transition energy level diagram. Solid arrows are driving lasers; dashed arrows show decay routes. (b) Scanning electron microscope image of the kagome HC-PCF used. (c) Schematic of the optical experimental setup. AOM, acousto optic modulator; SMF, single mode fiber; HWP, half wave plate; PBS, polarizing beam splitter; DG, diffraction grating; PMT, photomultiplier tube; PD, photodiode; and RA, rubidium ampoule.
of the fiber and vacuum chamber, nominally kept at 90 °C for the reported experiments. Fluorescence measurements probed the Rb density within the fiber showing over half the 40-cm fiber was loaded. Furthermore, the Rb density was observed to decrease along the fiber’s length away from the loading chamber. Nonetheless, pressure broadening and line shifts (∼12 kHz [8,16,17]) for the largest vapor densities (∼2 × 10^{18} m^{-3} [18]) are below the resolution of this experiment.

The optical setup used to excite the two-photon transition is shown in Fig. 1(c). This consisted of two tunable lasers: an extended cavity diode laser (ECDL) at ∼780 nm and a Ti:sapphire laser at ∼776 nm. For high-resolution measurements, laser noise was minimized by stabilizing the ECDL to a reference optical cavity yielding a beat note between the Ti:sapphire and ECDL with a full width at half maximum (FWHM) of < 300 kHz. The 776-nm laser passed through an acousto optic modulator to provide power control. Both lasers passed through single mode fibers, before their polarizations were set to be orthogonal. Two 60-nm lenses then coupled each laser into the HC-PCF from either end to allow Doppler-free spectroscopy of the two-photon transition. Optical powers \( P_{g} \) and \( P_{e} \) were measured after the fiber so that optical in-coupling losses into the fiber were included. We simultaneously observed 420 nm fluorescence and absorption of both excitation beams using photomultiplier tubes and a photodiode with suitable dichroic filters.

The two-photon transition was also excited within a room temperature bulk Rb cell, in order to compare the characteristics of the fiber and bulk cell systems. Both lasers had a 1/e^2 waist diameter of ∼1.3 mm with typical laser powers of \( P_{g} \approx 1 \) mW and \( P_{e} \approx 20 \) mW. These powers were sufficiently low to avoid saturation effects. A mu-metal shield reduced magnetic fields inside the cell below the resolution of the experiment. The temperature of the cell was 22 °C, which corresponded to a vapor density of ∼7 × 10^{17} m^{-3} [18].

The frequency of the Ti:sapphire laser was measured using a wave meter with a relative accuracy of 5 MHz. The ECDL’s frequency was measured using a combination of an optical cavity, providing equispaced frequency markers (300 MHz), and the saturated absorption features from a Rb reference cell, providing an absolute frequency reference. This procedure provided a frequency scale with an estimated accuracy of 2 MHz.

### III. TWO-DIMENSIONAL MAPS

The two-dimensional maps in Fig. 2 show the dependence of the two-photon transition strength on detunings from both the intermediate and excited state transitions. This was achieved by scanning both lasers to completely explore a 23 × 23 GHz two-dimensional frequency detuning space centered on the single- and two-photon transitions while detecting both the 420 nm fluorescence [Fig. 2(a)] and 776-nm absorption [Fig. 2(b)]. The 780-nm absorption map is complicated by single-photon absorption features, and hence, is not shown. However, when well detuned from single-photon resonance (\(|\Delta_{1}\| \gg 900 \) MHz), and in two-photon resonance, the 780-nm beam typically displayed stronger absorption than the 776-nm beam due to its larger dipole moment. Hence, the rest of this paper presents absorption of the 780-nm beam while in two-photon resonance. We will term this 780-nm absorption as the two-photon absorption for the remainder of this paper.

The four strong diagonal lines apparent in both panels of Fig. 2 represent the two-photon Doppler-free transition between the ground and excited states. Each isotope of Rb contributes two lines due to ground-state hyperfine splitting, with the two central lines belonging to \(^{85}\)Rb, and the outer two to \(^{87}\)Rb. These lines have a measured gradient of ∼1,000(1), and hence map out a diagonal line of constant sum frequency corresponding to \(\Delta_{1} = 0\).

Each of the four main absorption features in Figs. 2(a) and 2(b) consist of multiple absorption lines originating from excited state hyperfine splitting. These lines are not visible in Fig. 2 due to their narrow frequency spacing, but are readily resolved in our setup. A spectra taken through one of the main absorption lines [depicted by the horizontal dashed line in Fig. 2(b)] reveals the excited state hyperfine splitting, shown in Fig. 3 for both the HC-PCF and reference cell. Theoretical analysis of the two-photon transition [1,10] shows the spectral

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**FIG. 2.** (Color online) Frequency map of the \(|g\rangle \rightarrow |e\rangle\) transition. Map (a) shows 420 nm fluorescence. Hyperfine ground-state splittings are highlighted. Map (b) shows 776 nm absorption. The dashed line and arrow depict how the data presented in Figs. 3 and 4, respectively, are highlighted. Map (a) shows 420 nm fluorescence. The dashed line and arrow depict how the data presented in Figs. 3 and 4, respectively, are highlighted. Map (b) shows 776 nm absorption. The dashed line and arrow depict how the data presented in Figs. 3 and 4, respectively, are highlighted.
lineshape takes the form of a Voigt profile:

$$\alpha(\Delta_e) \propto \int_{-\infty}^{\infty} \frac{e^{-(\Delta_e/\gamma_e)^2}}{4(\Delta_e - \Delta_i)^2 + \gamma_i^2} d\Delta_i.$$  

(1)

The Gaussian component of the Voigt profile arises from residual Doppler broadening as the two counterpropagating laser frequencies are not identical [1]. The fiber and cell give residual Doppler broadenings of $\gamma_{DF} = 1.9$ and $1.7$ MHz, respectively, with the variation due to different operating temperatures (90 °C and 22 °C, respectively). The Lorentzian component arises from the $|g\rangle \rightarrow |e\rangle$ transition line shape with a natural FWHM of $667$ kHz (lifetime of 238 ns [19]) which can be power broadened. The optical depth observed in the $|g\rangle \rightarrow |i\rangle$, 780-nm coupling laser, $\alpha(\Delta_e)$, is related to observed absorption via $\alpha(\Delta_e) = 1 - e^{-\alpha(\Delta_e)}$ and the two-photon transition rate $w(\Delta_e)$ by $\alpha(\Delta_e) \propto w(\Delta_e)/P_{gi}$. Equation (1) was fitted to the retrieved spectra, as shown in Fig. 3, with no fixed fitting parameters. The individual Lorentzian and Gaussian widths could not be extracted as the signal-to-noise ratio of the data was not sufficient to retrieve both values reliably. However, the FWHM of the Voigt consistently returned reliable values, which are presented here. These fits allowed the amplitude, linewidth, and excitation bandwidth of this transition to be investigated along with their dependence on experimental parameters such as laser powers and detuning from the intermediate level.

IV. TWO-PHOTON CHARACTERISTICS

The two-photon absorption as a function of detuning from the intermediate state is shown in Fig. 4(a). This data corresponds to tracking along the absorption line, depicted by the dashed diagonal arrow on the 2D map in Fig. 2(b) (i.e., $\Delta_i = 0$), by postprocessing 780-nm absorption spectra similar to Fig. 3 for varying $\Delta_i$. For large detunings, of up to 9 GHz, significant absorption (>10%) is still observed. Stepwise excitation through $|i\rangle$ is negligible in Fig. 4 as $|\Delta_i| \gg \gamma_{Dop}/2$, where $\gamma_{Dop} \approx 1.8$ GHz is the Doppler-broadened FWHM of the intermediate state. In this limit, the resonant two-photon transition rate $w(0)$ for a stationary atom in a low-power laser field (where the absorption is power independent [20]) depends upon single-photon detuning $\Delta_i$, as [1,10]

$$w(0) \propto \frac{P_{gi} P_{ie}}{\gamma_e (4\Delta_i^2 + \Gamma_i^2)}.$$  

(2)

This is a Lorentzian line shape centered on the $|g\rangle \rightarrow |i\rangle$ transition frequency [1] with a width $\Gamma = \gamma_i$ being the linewidth of the intermediate state. However, when considering a thermal vapor, the Maxwell-Boltzmann velocity distribution naturally broadens this bandwidth. Furthermore, power dependent broadening of $\Gamma$ is expected [21] and was observed in the experiment. For optical powers of $P_{gi} \approx 60$ nW and $P_{ie} \approx 640$ $\mu$W, a Lorentzian fit to the data presented in Fig. 4(a) returned a two-photon excitation bandwidth FWHM of $\Gamma = 2.50 \pm 0.05$ GHz.

The second key characteristic for many applications is the dependence of two-photon absorption strength on input powers. A summary of this is shown in Fig. 5(a) for a detuning of $\Delta_i \approx 2$ GHz. The two-photon optical depth, defined using Eq. (2) as $\alpha(0) \propto w(0)/P_{gi}$, describes the absorption strength for sufficiently low optical powers where atomic saturation and optical pumping are negligible [10,20]. Atomic saturation was observed for $P_{gi} \approx 850$ nW and $P_{ie} > 500$ $\mu$W as seen in Fig. 5 by the combined reduction in absorption strength and increase in the spectral linewidth. A maximum two-photon optical depth of $\alpha(0) > 2.4$ [absorption $\alpha(0) > 90\%$] was observed for input powers of $P_{gi} \approx 870$ $\mu$W and $P_{ie} < 100$ nW.

Finally, for many applications the most important characteristic is minimizing the linewidth and decoherence of the two-photon transition. The behavior of the two-photon transition’s linewidth $\gamma_{FWHM}$ can be broken into two regions [1,10,14]:

$$\gamma_{FWHM} = \begin{cases} \gamma_e + s_1 \sqrt{1 + P_{gi}/P_{sat}} & |\Delta_i| < \gamma_{Dop} \\ \gamma_e & |\Delta_i| \gg \gamma_{Dop}. \end{cases}$$  

(3)
where \( s = (\omega_{gi} - \omega_{ei})/\omega_{gi} \), for this configuration \( s \approx 0.005 \), and \( P_{\text{sat}} \) is the transition’s effective saturation power. It can be seen from Eq. (3) that when the lasers are resonant with the intermediate transition, the power broadened linewidth associated with the intermediate level contributes to the two-photon linewidth. This is confirmed experimentally in Fig. 4(b) where the linewidth expands for intermediate state detunings approaching \( \gamma_{\text{Dop}} \). In the limit that \( |\Delta_{i}| \gg \gamma_{\text{Dop}} \) the linewidth is seen to be constant as predicted from Eq. (3).

The linewidth was also seen to vary as a function of input power for a fixed frequency detuning, as shown in Fig. 5(b). In contrast to Eq. (3) and Fig. 4(b), the increase in two-photon spectral linewidth observed here is not induced by the intermediate state. Rather, it is the effect of inhomogeneous ac Stark shifts experienced by the atoms in different regions of the fiber’s optical mode [9].

V. ULTIMATE LINEWIDTH

The ultimate linewidth achieved was 10.4 ± 0.5 MHz, substantially broader than expected from the natural linewidth alone being 667 kHz [19]. Technical broadening mechanisms include intermediate level effects, light shifts, and laser noise. All were minimized below the resolution limit of the experiment. However, the effects of magnetic fields, residual Doppler, and transit-time broadening, could not be removed. Each of these effects are discussed below and their contribution summarized in Table I.

The effect of the intermediate level [as discussed in Eq. (3)] was minimized by driving the transition with a detuning of \( \Delta_i \approx 3 \) GHz. Optical powers were also minimized (\( P_{gi} < 100 \) nW and \( P_{ei} < 20 \) \( \mu \)W), to ensure inhomogeneous light shifts did not broaden the spectral linewidth as shown in Fig. 5(b). Laser noise was minimized by stabilizing the ECDL to the reference optical cavity reducing frequency noise below the resolution limit of the experiment.

It was not feasible to shield the fiber from ambient magnetic fields due to the complexity of the vacuum system. An estimate of this broadening was determined by observing the two-photon linewidth while increasing the local magnetic field. Extrapolation of this data to the ambient field level provides an estimated ambient magnetic field broadening of \( \approx 2.6 \pm 1.0 \) MHz. Residual Doppler broadening is also present resulting in a contribution of 1.9 MHz \( 1/e \) half-width Gaussian broadening within the heated fiber. This was verified with identical measurements on a conventional macroscopic cell at room temperature.

The fiber core’s confined geometry leads to transit-time broadening caused by short light-atom interaction times [22]. This effect was modeled using a four-level density matrix model which included a Maxwell-Boltzmann thermal velocity distribution which accounts for residual Doppler broadening; random atomic trajectories and the laser beam geometry [20] resulting in transit-time broadening; and splitting of the Zeeman sublevels which accounted for the observed magnetic field broadening. These effects were all convolved together to produce an expected spectral line shape. An estimate of the overlap of the counterpropagating modes within the fiber was measured by imaging the transmitted modes at both ends of the fiber. This showed an overlap region with diameter 13 ± 1 \( \mu \)m. Using this as the effective optical mode diameter, a transition spectra was generated from the model, which is shown in Fig. 3 to be in excellent agreement with the experimental data. As shown in Table I, the width of the modeled spectra is predicted to be 10.1 ± 0.4 MHz, which is consistent with the observed result.

In contrast, the reference cell was seen to have a linewidth of 4.7 ± 0.2 MHz as seen in Fig. 3. This is because the cell was magnetically shielded and the optical beam had a \( 1/e^2 \) diameter of 1.3 mm causing negligible transit-time broadening. Minor misalignment between the 780- and 776-nm beams within the cell (< 0.1\°) caused additional Doppler broadening as indicated in Table I. This was avoided in the fiber due to its guidance properties.

To confirm transit-time broadening as the dominant effect within the fiber, and to rule out additional population relaxation processes, the excited state lifetime was directly observed through fluorescence decay measurements, shown in Fig. 6. If there are no other population relaxation mechanisms, the atomic fluorescence lifetime will be limited by the time of flight of the atoms across the fiber core. Observation of this fluorescence decay was enabled by rapidly switching off the excitation laser (\( 1/e \) falloff time of 55 ns). Analysis of the

<table>
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<th>TABLE I. Two-photon linewidth budget.</th>
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<tr>
<td>Broadening effect</td>
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<tr>
<td>Natural linewidth [19]</td>
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<tr>
<td>Magnetic field</td>
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<td>Residual Doppler</td>
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<td>Beam misalignment</td>
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<td>Convolved resultant</td>
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*Theoretically calculated values based on measured temperature, mode area, and beam alignment.*
decay using methods described in [19] gave an excited state lifetime $\tau_l$ of $81 \pm 2$ and $226 \pm 8$ ns for the fiber and cell, respectively. The decay rate in the cell agrees with previous measurements (238 ns [19]). On the other hand, the fiber’s excited state lifetime is shortened by 157 ns corresponding to a time of flight across the fiber core of $123 \pm 3$ ns using the formalism

$$\frac{1}{\tau_f} = \frac{1}{\tau_e} + \frac{1}{\tau_{tt}},$$

(4)

where $\tau_e = 238$ ns is the natural lifetime of the excited state [19] and $\tau_{tt}$ is the transit time of the atoms across the fiber. This is consistent with a theoretical estimate of the expected time of flight across the fiber core of 132 ns, obtained by averaging over all possible velocities and trajectories. This agreement suggests that the dominant population relaxation process is from atom-wall collisions, ruling out other population relaxation mechanisms which could cause the observed broadening. Furthermore, the agreement confirms the earlier modeling underpinning the theoretical line shape (Fig. 3). Thus, this confirms that the limited transit time across the optical mode is the origin of the observed increased linewidth in the fiber.

VI. CONCLUSION

We have demonstrated the ability to drive the $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition within a Rb-filled HC-PCF. Even for very modest optical powers ($< 500 \mu W$) it is possible to obtain two-photon absorption depths close to 100%. Furthermore, it is possible to maintain this substantial absorption over large optical detunings $>9$ GHz from the intermediate level. The ultimate linewidth of the two-photon transition was shown to be $10.4 \pm 0.5$ MHz, which is dominated by transit-time broadening and is consistent with theoretical modeling. Such narrow linewidths enable the two-photon excited state hyperfine splitting to be resolved.

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This chapter introduces and presents two papers. The first is published in the peer-review journal *Optics Letters* and named “Two-Colour Rubidium Fibre Frequency Standard”. The second paper is called “Two-Photon Two-Colour Rubidium Frequency Standard”, a decision has been made not to publish this work at this point in time.

### 9.1 Fibre

A frequency standard was built and characterised based on a Rb filled HC-PCF which is presented in this paper. Potentially such a frequency standard could be a very compact, stable standard rivalling the commercial frequency standards discussed in section 1.2.

The optical transition used was the Rb two-photon $5S_{1/2} \rightarrow 5D_{5/2}$ transition as described in section 3.1.2. This transition was excited using two laser of different wavelengths such that the detuning from the intermediate $5P_{3/2}$ energy could be controlled and was typically $< 10$ GHz as described in chapter 8. Such a small detuning from the intermediate state greatly enhanced the two-photon excitation rate. In depth characterisation of this transition, excited in this manner, was presented in chapter 8 which enabled the driving laser’s optical powers and intermediate state detuning to be tailoring to produce the optimal excitation regime. As a result, it was the sum frequency of these lasers which is the stabilised optical signal.

The Rb vapour being interrogated was loaded within a kagomé HC-PCF, as discussed in chapter 2. Both the fibre’s transmission, and a scanning electron microscope image of the fibre’s cross section are shown in figure 2.3. The fibre’s core dimensions were 45 μm and 51 μm for the short- and long-axis respectively.

The experimental set-up discussed in chapter 6 was arranged with the two lasers
in a counter-propagating configuration within the HC-PCF to reduce Doppler broadening of the two-photon transition, as discussed in section 3.1.2. In order to stabilise the optical signal, a frequency discrimination signal was produced using a variation of modulation transfer spectroscopy [178]. This was achieved through frequency modulating the 776 nm Ti:S laser at 5 MHz (modulation depth ~5 MHz) using an acoustic-optic modulator. Transfer of this frequency modulation into amplitude modulation on the 780 nm laser was produced through the non-linear light-atom interaction. An avalanche photo-diode (APD), as discussed in chapter 6, was used to detect the 5 MHz amplitude modulation on the 780 nm laser.

Using these techniques, a fractional frequency stability of $9.8 \times 10^{-12}$ at an integration time of $\tau = 1$ s was measured by comparison to an optical frequency comb. The limits of this frequency standard are discussed in the paper, with the predominant limitation being alignment fluctuations into the HC-PCF. By experimentally removing the source of these fluctuations, a predicted stability of $\approx 1 \times 10^{-13} \tau^{-1/2}$ could be achieved, rivalling the frequency stability of the compact frequency standards discussed in section 1.2.
Two-color rubidium fiber frequency standard

C. Perrella,1,3⁎ P. S. Light,1,3 J. D. Anstie,1,3 F. N. Baynes,1 F. Benabid,2 and A. N. Luiten1,3

1School of Physics, University of Western Australia, Perth, Western Australia 6009, Australia
2GPPMM Group, Xim Research Institute, CNRS, Universite de Limoges, France
3Institute of Photonics and Advanced Sensing (IPAS) and the School of Chemistry and Physics, The University of Adelaide, Adelaide SA 5005, Australia

⁎Corresponding author: chris.perrella@adelaide.edu.au

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We demonstrate an optical frequency standard based on rubidium vapor loaded within a hollow-core photonic crystal fiber. We use the $^5S_{1/2} \rightarrow ^5D_{5/2}$ two-photon transition, excited with two lasers at 780 and 776 nm. The sum-frequency of these lasers is stabilized to this transition using modulation transfer spectroscopy, demonstrating a fractional frequency stability of $9.8 \times 10^{-12}$ at 1 s. The current performance limitations are presented, along with a path to improving the performance by an order of magnitude. This technique will deliver a compact, robust standard with potential applications in commercial and industrial environments. © 2013 Optical Society of America

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There is a great deal of interest in the development of compact, robust, efficient, and relatively inexpensive atomic frequency standards aimed at commercial and industrial markets, where frequency stabilities in the range of $10^{-10}$ to $10^{-13}$ are desired. These devices, based on either optical [1,2] or microwave [3,4] atomic transitions, are aimed at applications such as telecommunications [4] and navigation systems [5]. One such commercial device has achieved a fractional frequency stability of $2.5 \times 10^{-10}$ for integration times of $1 < \tau < 1000$ s [4]. Here, we demonstrate a device that could be of similar size, that outperforms this commercial product by an order of magnitude over short time scales with another order of magnitude in performance through to be achievable. This is achieved through a novel twin approach: first, a hollow-core photonic crystal fiber (HC-PCF) is used in combination with a two-photon transition in rubidium (Rb) and second, exploitation of the energy level structure of Rb to enhance the two-photon transition rate using a two-color excitation technique.

The most stable laboratory-based optical Rb frequency standards utilize the $^5S_{1/2} \rightarrow ^5D_{5/2}$ two-photon transition and have demonstrated a frequency stability of $3 \times 10^{-13}$ for $1 < \tau < 1000$ s [6,7]. This transition can be excited in a Doppler-free configuration [8], resulting in a linewidth of $334$ kHz, limited by the natural lifetime of the $^5D_{5/2}$ state. These standards use a single 778 nm laser tuned to half the energy difference between the $^5S_{1/2}$ and $^5D_{5/2}$ states. To increase the Rb two-photon transition rate, and thereby the potential frequency stability, an optical cavity is typically used to increase the optical power interrogating the transition [6,7]. This makes these clocks complex and fragile devices.

This Letter presents an alternative approach that avoids the complexity of an optical cavity [6,7] through the use of HC-PCF. The geometry of a HC-PCF allows production of high optical intensities, at low input powers, over arbitrary lengths. Such attributes enhance light-atom interactions, improving the signal-to-noise ratio of frequency stabilization signals. Moreover, the flexibility and small volume of HC-PCF makes it an excellent basis for a compact, robust, and efficient high-performance frequency standard.

In our scheme, further enhancement of the two-photon transition is gained over previous work [6,7] by using a two-color excitation technique. Two lasers at 780 and 776 nm were used to drive the two-photon $^8Rb ^5S_{1/2}(F = 2) \rightarrow ^5D_{5/2}(F' = 4)$ transition, seen in Fig. 1(a), with a small detuning from the intermediate $^5P_{3/2}$ state. A detuning of $\Delta \approx 2$ GHz was used which, when compared to the single laser 778 nm based standards [6,7] with $\Delta \approx 1$ THz, provided a $10^8$ enhancement in transition rate [10]. The two-photon transition displayed a spectral full-width at half-maximum of 10.4 MHz, limited predominantly by transit time broadening in combination with residual Doppler and magnetic field broadening [11]. At typical driving powers used here, the transition

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Fig. 1. (a) Two-photon transition energy level diagram. Solid arrows are driving lasers, dashed arrows show decay routes. (b) Scanning electron microscope image of the kagome HC-PCF used. (c) Schematic of the experimental setup. AOM, acousto-optic modulator; SMF, single-mode fiber; HWP, half-wave plate; PBS, polarizing beam splitter; DG, diffraction grating; PMT, photomultiplier tube; PD, photodiode.
exhibited $\approx 5\%$ absorption of the 780 nm laser within the HC-PCF. In this scheme, neither the 780 nor 776 nm lasers were individually frequency stabilized: it was the sum-frequency of these lasers that was stabilized to the $^5S_{1/2}$ and $^5D_{5/2}$ energy level difference.

Figure 1(c) shows the optical setup used to excite the two-photon transition, along with the electronic feedback systems. The two driving lasers were an extended cavity diode laser at $\sim 780$ nm and a titanium:sapphire laser at $\sim 776$ nm. The lasers’ optical modes were spatially filtered by single mode fibers, and their polarization settings were set to be linearly orthogonal, enabling their separation after the HC-PCF. Doppler-free spectroscopy of the two-photon transition was achieved by coupling the lasers into opposite ends of the HC-PCF using 60 mm lenses.

The HC-PCF used had a kagome-style cladding structure [12], shown in Fig 1(b), with a core diameter of 45 $\mu$m, which together provided low-loss multimode guidance from 600 to 1600 nm. Observation of the Rb vapor’s fluorescence was used to qualitatively measure variations of the Rb density along the length of the fiber. This revealed over half of the 40 cm fiber contained measurable quantities of Rb, although the density decreased along the fiber’s length.

After the HC-PCF, the lasers were separated using polarizing beam splitters. A diffraction grating further isolated the 780 nm beam from any residual 776 nm light before detection with an avalanche photodiode. A frequency discrimination signal was generated using a variant of modulation transfer spectroscopy (MTS) [13]. This was achieved through frequency modulating the 776 nm laser at 5 MHz (modulation depth $\sim 5$ MHz) using an acousto-optic modulator (AOM). Through the nonlinear light-atom interaction, this modulation was transferred into amplitude modulation on the 780 nm laser. This technique avoids background contamination [13] that would otherwise deteriorate the frequency discriminator signal.

Stabilization of the lasers’ sum-frequency to the MTS discriminator signal was achieved by controlling the frequency of the 780 nm laser. The control loop bandwidth was 100 kHz, limited by the laser controller. This stabilization effectively compensated for the frequency fluctuations of the uncontrolled 776 nm laser ($\sim 5$ MHz over 1000 s).

The frequencies of both lasers were compared to two modes of a commercial fiber frequency comb that was locked to a hydrogen maser. Each optical comb mode exhibited a frequency stability of $\approx 5 \times 10^{-13}$ at 1 s. The mixing products between the closest comb modes and the two lasers were separately measured on synchronously triggered counters. The sum-frequency was obtained by post processing the recorded beat notes.

The typical optical powers used to drive the two-photon transition were 3 and 4 $\mu$W for the 780 and 776 nm lasers, respectively. Even for these low powers, intensities within the fiber were large enough to produce substantial light shifts, thus the laser power coupled into the fiber were actively controlled. This was achieved by detecting the transmitted power of both lasers with a photomultiplier tube and photodiode, respectively. These signals were stabilized via attenuation of the corresponding

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<td>776 nm power</td>
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<td>776 nm frequency</td>
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AOM. Once stabilized, the light shift induced by each beam were investigated by varying the set point of each power control loop independently and monitoring the induced shift in the sum-frequency. The values obtained were $540 \pm 20$ kHz/$\mu$W and $29.7 \pm 0.2$ kHz/$\mu$W for the 780 and 776 nm beams, respectively. Using these measured light shift sensitivities, the frequency shifts and fluctuations calculated from the operating optical powers and residual fluctuations are presented in Table 1.

The scale of the light shifts, measured above, depend on the detuning of both the 780 and 776 nm lasers from the intermediate state. Hence, uncontrolled frequency fluctuations of the 776 nm laser, and corresponding 780 nm corrections, cause additional light shifts. These shifts are proportional to $1/(\omega^2_{ij} - \omega^2_l)$, where $\omega_{ij}$ is the transition frequency from level $|i\rangle$ to $|j\rangle$ and $\omega_l$ is the laser frequency. Using the measured drift of the 776 nm laser and the calibrated light shifts discussed above, an estimate of $10^{-14}$ stability is calculated for this noise source.

The measured fractional frequency stability of the stabilized sum-frequency is shown in Fig. 2. Also shown is the instability in the sum-frequency when both lasers are free-running. A stability of $9.8 \times 10^{-12}$ was measured at an integration time of 1.3 s, which averaged down to a minimum of $5.9 \times 10^{-12}$ at an integration time of $\sim 10$ s.

The dominant noise source was associated with alignment-driven coupling fluctuations of the lasers to the HC-PCF. Varying alignment of the input light modified the particular set of high-order optical modes that are excited within the fiber. Consequently, the mode

![Figure 2](image-url)
shape and intensities are altered in the region where the 780 and 776 nm lasers overlap within the HC-PCF. These created varying light shifts that cannot be compensated for by simply controlling the overall power in the counterpropagating modes. An estimate of the frequency noise induced by this effect was made using a quadrant photodiode to measure the alignment fluctuations associated with movement of the HC-PCF within the vacuum system. Calibration of this motion, in terms of frequency shift, was made by displacing the in-coupling lens by a known amount and measuring the associated summed frequency shift, which corresponded to $63 \pm 15 \text{ kHz/\mu m}$. During this calibration the power control loops were engaged, eliminating overall power fluctuations as a cause of the frequency shifts. The resulting frequency noise estimate is within a factor of two of the measured stability, as shown in Fig. 2.

Frequency noise induced by collisional and magnetic field noise are negligible at current performance levels. At the operating system temperature (90 $\pm$ 5°C), Rb–Rb collisions produce frequency fluctuations and shifts [4], which are presented in Table 1. Shifts induced by magnetic fields were estimated by observing the two-photon transition while increasing the local magnetic field, yielding shifts of $\approx -1.8 \pm 1.0 \text{ MHz/G}$. Hence, ambient magnetic field levels of $\approx 0.5 \text{ G}$ with a noise of $<1 \text{ mG}$ caused a frequency shift of $-960 \text{ kHz}$ and noise of $10^{-12}$.

Electronic and optical noise sources were investigated by monitoring the frequency error signal when detuned out of resonance with the two-photon transition. A limit of $\approx 10^{-13}$ fractional frequency stability was obtained originating from the shot-noise of the 780 nm light detected by the APD. These values demonstrate the potential stability achievable at this excitation level if the noise associated with fluctuating transverse mode structure within the HC-PCF could be eliminated.

A pathway for improvement is seen by removing optical mode fluctuations within the fiber. Similar alignment fluctuations have previously been reduced by more than an order of magnitude using active alignment systems [1]. The ultimate approach consists of loading Rb within an HC-PCF supporting only single mode guidance at 780 nm and sealing the fiber by splicing single mode fibers onto both ends. This approach removes both in-coupling alignment fluctuations along with the need of a vacuum system. This is a route for this platform to achieve frequency stabilities at the level of $\approx 10^{-13}$, which would match previously demonstrated Rb two-photon frequency standards [6] and begins to rival the stability of Rb fountain clocks [14] and hydrogen masers.

In conclusion, we demonstrate an optical frequency standard with a fractional frequency stability of $9.8 \times 10^{-12}$ at a 1.3 s integration time. This standard is based on a two-photon two-color Doppler-free transition of a Rb vapor loaded within an HC-PCF, to which the sum-frequency of the two lasers stabilize. Stability was limited by optical mode shape changes within the fiber, driven by in-coupling alignment fluctuations. A route for improved stability is outlined, which should achieve $\approx 10^{-13}$ fractional frequency stability at integration times of 1 s. This architecture could lead to compact, robust, and efficient frequency standards.

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References
CHAPTER 9. TWO-PHOTON FREQUENCY STANDARD

9.2 Cell

A similar frequency standard was built based on the Rb two-photon transition excited inside a the bulk reference-cell. Again, this system could potentially be a very compact and rival the commercial standards discussed in section 1.2.

The two-photon transition was excited in the same way as the fibre standard discussed in the previous section. Again, it was the sum frequency of these lasers which is the stabilised optical output signal.

The experimental set-up discussed in chapter 6 was arranged with the two lasers counter-propagating through the reference cell to reduce Doppler broadening of the two-photon transition, as discussed in section 3.1.2. As discussed in chapter 6, the absorption of the two-photon transition within the reference-cell was too small to be used for frequency stabilisation, hence 420nm fluorescence from the decaying $5D_{5/2}$ excited state was used. The rate of modulation was limited by the excited state life-time, a modulation rate of 333 kHz with a modulation depth of 2 MHz was applied to the 776 nm Ti:S laser. This modulated population within the $5D_{5/2}$ state, and hence the 420 nm fluorescence detected by a PMT.

Using these techniques a fractional frequency stability of $8 \times 10^{-13}$ at $\tau = 1$ s was measured by comparison to an optical frequency comb. A number of limiting factors for this frequency standard are highlighted within the paper. By removing these limitations, a predicted stability of $\approx 1 \times 10^{-14} \tau^{-1/2}$ could be achieved, rivalling the frequency stability of the compact frequency standards discussed in section 1.2.
Two-Photon Two-Colour Rubidium Frequency Standard

C. Perrella, 1 P. S. Light, 1 J. D. Anstie, 1 F. N. Baynes, 1 and A. N. Luiten 1, 2

1) School of Physics, University of Western Australia, Perth, Western Australia 6009, Australia
2) Institute of Photonics and Advanced Sensing (IPAS) and the School of Chemistry and Physics, The University of Adelaide, Adelaide SA 5005, Australia

We demonstrate an optical frequency standard based on the 5S1/2 → 5D5/2 two-photon transition of rubidium. The transition is interrogated, using a Doppler-free arrangement, by two lasers at 780 nm and 776 nm, and it is the sum frequency of the two lasers that is frequency locked to the two-photon transition. We measure the fractional frequency stability of the frequency standard to be 8 × 10−13 at 1 s by comparison to an optical frequency comb. Performance limitations are presented and a clear pathway to an improvement by another order of magnitude is demonstrated. This platform has the potential to deliver a compact, robust standard for commercial and industrial applications.

Frequency standards can be split into two broad domains: the first is that of ultra-high stability frequency standards based on laser-cooled atoms and ions 1–12, which are reaching fractional frequency stabilities of 10−17. These high-precision devices have been used for tests of general relativity 13–17, investigation of the temporal stability of fundamental constants 18–20, and exploration of atomic physics 21–24. The second territory is that of compact atomic frequency standards aimed at commercial and industrial markets, where stabilities in the range of 10−10 to 10−13 are desired. These devices, based on either optical 11,12 or microwave 12–15 atomic transitions, are aimed at applications in the fields of telecommunications 13 or navigation 14. Here we aim to bridge the gap between these two domains by building a standard that demonstrates a frequency stability performance similar to the very best commercial frequency standards (e.g. the hydrogen maser with a fractional frequency stability of ∼10−13 at 1 s integration time) but which exhibits the compactness and robustness of the new portable standards. We propose to achieve this by exploiting the energy level structure of rubidium (Rb) to efficiently drive a narrow linewidth two-photon transition.

Previous work has shown the Rb 5S1/2 → 5D5/2 two-photon transition to be a promising candidate for a high stability frequency standard, with frequency stabilities of 3 × 10−13 τ−1/2 for 1 < τ < 1000 s being reached. 17–20 These standards used a Doppler-free excitation technique that produces a spectral linewidth of 334 kHz, limited solely by the lifetime of the 5D5/2 state. A single 778 nm laser drives the two-photon transition in these standards, with its frequency set to exactly half the energy difference between the 5S1/2 and 5D5/2 states. In this configuration the optical detuning of the laser from a fortuitous intermediate energy level is ∼1.1 THz, producing a rather weak two-photon transition. To overcome this intrinsic weakness, an optical cavity is typically used to increase the effective optical intensity interrogating the Rb two-photon transition, thereby increasing the two-photon scattering rate. This leads to better frequency stability, although at the cost of a more complex and fragile device.

The scheme presented in this letter avoids the need for an optical cavity by more effectively exploiting the intermediate atomic energy level. This is achieved by using a two-colour excitation technique to drive the 87Rb 5S1/2 (F = 2) → 5D5/2 (F = 4) transition as shown in Fig. 1(a). The detuning from the intermediate 5P3/2 state is reduced to 2 GHz, producing a factor of 108 enhancement in two-photon transition rate when compared to the aforementioned Rb two-photon standards 17–20. It is important to note that in this approach neither of the two lasers are individually frequency stabilised, instead, it is the sum frequency of the two lasers that is frequency locked to the two-photon transition and is thus the sum frequency that is the output of the standard.

The two tunable lasers used to excite the two-photon transition are shown in Fig. 1(b): an extended cavity diode laser at ∼780 nm and a Titanium:sapphire laser at ∼776 nm. The lasers’ optical modes were spatially filtered using single mode fibres, and their polarizations set to be circular and of opposite helicity. A counter-propagating beam arrangement was employed to excite the Rb: this allowed Doppler-free spectroscopy of the two-photon transition with minimal Doppler-broadened background 21. The laser beams were focused at the centre of the Rb cell using 250 mm lenses to produce a 44 ± 1 μm waist, thereby increasing the optical intensity.
The frequency of the two lasers, $f_{776}$ and $f_{780}$, were mixed with the respective closest optical modes (numbered $n_1$ and $n_2$) of a frequency-stabilized commercial fibre frequency comb. The fractional frequency stability of the comb modes was $\sim 5 \times 10^{-13}$ at 1 s as it was stabilised to a hydrogen maser. The frequency of the $n^{th}$ mode can be expressed as: $f_n = n \cdot f_{RR} + f_0$ where $f_{RR}$ is the repetition rate of the comb, and $f_0$ is the carrier-envelope offset frequency. The two mixing products were then mixed together to produce a new signal at a frequency: $(f_{776} - f_{n1}) - (f_{780} - f_{n2}) = f_{776} + f_{780} - (n1 - n2)f_{RR}$. This new doubly-mixed signal is equal to the sum frequency of the two lasers, although shifted to the radiofrequency domain ($\sim 150$ MHz). Synchronously triggered counters recorded both this shifted sum frequency signal and the 776 nm laser frequency. From this data the individual frequencies of both lasers could be deduced.

The two-photon fluorescence signal demonstrated a Voigt profile with a full width at half maximum (FWHM) of $5.45 \pm 0.01$ MHz. This was substantially larger than the natural linewidth of $667$ kHz, predominantly due to $3.09 \pm 0.01$ MHz of residual Doppler broadening as the two exciting lasers had different wavelengths. An additional broadening effect originates from limited light-atom interaction time due to the small waist of the exciting laser beams. The time taken for an atom to traverse the optical mode, averaging over velocity and trajectory, is $\sim 380$ ns which is fractionally larger than the decay rate of the excited state of $238.5$ ns. This leads to transit-time broadening of $1.9$ MHz. A final broadening of $0.7 \pm 0.2$ MHz was associated with $\sim 0.06^\circ$ angular misalignment in the counter-propagating beams. All of these contributions convolve together to give an estimated FWHM of $5.4 \pm 0.2$ MHz, which agrees well with the observed value.

The two-photon transition was driven with optical powers of $\sim 30 \mu W$ and $\sim 280 \mu W$ for the 780 nm and 776 nm beams respectively. This corresponds to an intensity of $5 \text{kW/m}^2$ and $45 \text{kW/m}^2$ at the beams waist, sufficient to produce substantial light shifts. To minimise frequency variations arising from power fluctuations, both laser powers were stabilised by detecting their respective powers after the cell and actively controlling the incident intensity using their corresponding AOMs. This control system also enabled a careful investigation of the light shifts induced by each of the beams, by modulating its incident power and monitoring the associated frequency shift in the output of the standard. These measurements yielded a sensitivity of $-5.72 \pm 0.03$ kHz/$\mu W$ and $280 \pm 10$ kHz/$\mu W$ for the 780 and 776 nm beams respectively. Using these measured shifts, frequency noise induced by residual optical power fluctuations was estimated to be below the $10^{-14}$ fractional frequency stability level for both lasers at 1 s integration times (see Table 1). Frequency shifts from the unperturbed atomic resonance frequency induced by the operating optical powers was $177 \pm 1$ kHz and $80 \pm 1$ kHz for the 780 and 776 nm beams respectively.
Light shifts also arise from frequency fluctuations of the uncontrolled 776 nm laser which, along with the 780 nm frequency corrections, change the effective light-atom coupling through changing the detuning from the intermediate state. The magnitude of this effect was determined by scanning the 776 nm laser and observing corresponding shifts in the stabilised sum frequency, which corresponded to 216±5 Hz/MHz. During the measurements presented below, the fluctuations of the 776 nm laser were monitored against the frequency comb, thus unwanted frequency variations arising from this mechanism could be removed from the measurements.

Figure 3 presents the fractional frequency stability of the sum frequency when stabilised to the two-photon transition. Also shown is the measured frequency stability when the both lasers are uncontrolled. A fractional frequency stability of 7.9×10⁻¹³ was measured at an integration time of 1.3 s, which averaged down to 3.9×10⁻¹³ at an integration time of ~10 s. This was achieved by balancing the noise contributions from alignment-driven frequency fluctuations, and that of shot-noise and electronic noise by operating at low optical powers. These noise sources are described below and summarised, along with other relevant noise sources, in Table I. The influence of the three main noise sources on the standards frequency stability is also shown in Fig 3.

Variations of up to 60 kHz were observed in the locking point upon multiple optical realignments, even though no noticeable change in the frequency discriminator amplitude was observed. Thus, alignment fluctuations into the Rb cell are the current key limitation to the performance of the standard. The effect of alignment fluctuations on the standard was quantified by displacing one of the incident beams by a known amount and then measuring the resulting sum frequency shift; this led to a sensitivity of 17±5 kHz/μm of displacement. This value, when combined with the measured beam directional fluctuations (estimated using a quadrant photodiode), provided an estimate of alignment-based frequency fluctuations of 4.5×10⁻¹³ at 1 s (see Fig 3). These frequency shifts originate from the effective intensity changes seen by atoms in the region where the two counter-propagating beams overlap. These effective intensity changes, which cannot be suppressed by power control of the overall beam, cause a frequency shift through the light shift.

The effect of electronic noise was estimated by monitoring the frequency discriminator noise when the lasers were tuned to be out of resonance with the two-photon transition. These fluctuations corresponded to a fractional frequency stability of 2×10⁻¹³ at τ=1 s and are attributed to dark-current of the PMT. The effect of shot noise in the fluorescence was calculated using the detected PMT photocurrent to be ~5×10⁻¹³ at the operating powers.

The effect of other noise sources are negligible at the current level of performance, although for completeness they are summarised in Table I. The Rb cell was heated to 89.7(1) °C to increase the Rb density. At this temperature, Rb-Rb collisions produce a frequency shift of −1.82±0.01 kHz while temperature fluctuations influence the long-term stability below the 10⁻¹⁵ level²⁶,²⁷. A mu-metal shield reduced ambient magnetic fields to the level of 2.0±0.5 mG with a noise of 0.01 mG over 100 s. Hence, the associated frequency shift and noise are at the level of <1.5 kHz and <10⁻¹⁵ for τ<100 s respectively.

Substantial performance improvements of this standard can be gained by eliminating alignment fluctuations and increasing the signal-to-noise ratio of the frequency discriminator. The long optical path length between the Rb cell and the single-mode fibres exacerbates alignment fluctuations substantially — this path length could be reduced by a factor of 100 by launching the laser beams from optics that are mounted directly on the Rb cell. Further, the frequency standard is currently using very low optical power levels to minimise unwanted alignment-

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FIG. 3. Fractional frequency stability of the sum frequency when the lasers stabilised and free-running. Also shown are the major noise sources in the standard.
related frequency fluctuations. If the alignment-related problems can be overcome then the optical powers can be increased by a factor of 10-100, which brings a factor of 10-100 improvement\(^7\) in the signal-to-noise of the two-photon fluorescence signal. This would push the shot-noise and electronic noise floors into the low 10\(^{-14}\) regime and thus allow this compact platform to potentially compete in performance with the much larger and more expensive hydrogen maser.

We have demonstrated an optical frequency standard based on a two-colour excitation of a Doppler-free transition in Rb. The standard demonstrates a fractional frequency stability of 7.9\times10^{-12} at 1.3 s integration time. This architecture can potentially bring Hydrogen-maser performance but in a frequency standard of a few liters in volume. The current performance limits have been identified as associated with alignment fluctuations as well as from photon shot-noise and detector noise. There are well-known techniques to solve these problems and we suspect that by implementing such changes, this platform can offer a route to 10^{-14} fractional frequency stability at integration times of 1 s.

The authors acknowledge financial support from the Australian Research Council under grants DP0877938, DE120102028 and FT0991631.

This chapter introduces and presents the paper “High-Efficiency Cross-Phase Modulation in a Gas-filled Waveguide”, published in the peer-reviewed journal Physical Review A.

10.1 Overview

Photon-photon interactions are keenly sought in the field of optical quantum computing as was discussed in section 1.3. One method for creating such interactions is through the natural coupling between two lasers that are tuned to a two-photon atomic transition. In such an atomic transition, the refractive index, and hence phase-shift, observed by one of the driving lasers is dependent upon the intensity of the other driving laser. The refractive index and phase-shift were derived and discussed in sections 4.4.2 and 4.2.1 respectively. Therefore, by efficiently driving a two-photon transition, an efficient photon-photon interaction can be generated. The approach taken in this paper was to use a Rb filled HC-PCF to produce an efficient two-photon transition.

The efficiency of the photon-photon interaction is linearly dependent on the photon’s intensity as discussed in section 4.4.2. Hence, to increase the intensity a kagomé HC-PCF was used, the same HC-PCF as was used for the previous experiments in chapters 7 - 9. This fibre is discussed in chapter 2, and both the fibre’s transmission, and a scanning electron microscope image of the fibre’s cross section, are shown in figure 2.3. The fibre’s core dimensions were 45 μm and 51 μm for the short- and long-axis respectively.

Similarly to chapters 7 - 9, the optical transition was the Rb two-photon $5S_{1/2} \rightarrow 5D_{5/2}$ transition, as described in section 3.1.2. This transition was excited using two laser of different wavelengths such that the detuning from the intermediate $5P_{3/2}$
energy could be controlled, and was typically $< 10 \text{GHz}$ as described in chapter 8. Such a small detuning from the intermediate state greatly enhanced the two-photon excitation rate, and hence the achievable phase-shift. In depth characterisation of this transition, excited in this manner, was presented in chapter 8 which enabled the intermediate state detuning to be tailoring to produce the maximum phase-shift.

The phase-shift induced by the two-photon transition was measured using a self-heterodyne technique. For this, the 780 nm ECDL was first passed through an acoustic-optic modulator to frequency shift $\approx 50\%$ of its optical power by 80 MHz. Both the un-shifted and shifted beams were combined using SMF fibre beam splitters to ensure mode overlap, as shown in chapter 6. An optical beat note between these two beams was detected before and after the Rb filled kagomé HC-PCF. The relative phase of these two beat notes was compared to determine the phase-shift induced by the Rb two-photon transition.

Using the above mentioned techniques, this paper presents a maximum phase-shift of $0.7 \times 10^{-6}$ rad per photon. This paper also examines photon-loss through two-photon absorption and compares it to the phase-shift produced. It is demonstrated that the ratio between the induced photon phase-shift falls and photon-loss increases as the detuning from two-photon resonance is increased, demonstrating a pathway to non-demolition photon-photon interactions and photon number measurements.
High-efficiency cross-phase modulation in a gas-filled waveguide

C. Perrella,1,2 P. S. Light,1,2 J. D. Anstie,1,2 F. Benabid,3 T. M. Stace,4 A. G. White,5 and A. N. Luiten1,2
1School of Physics, University of Western Australia, Perth, Western Australia 6009, Australia
2Institute for Photonics and Advanced Sensing (IPAS) and the School of Chemistry and Physics, The University of Adelaide, Adelaide, South Australia 5005, Australia
3GPPMM Group, Xlim Research Institute, CNRS, Universite de Limoges, France
4Centre for Engineered Quantum Systems, School of Mathematics and Physics, University of Queensland, Brisbane, Queensland 4072, Australia
5Centre for Quantum Computing and Communication Technology, School of Mathematics and Physics, University of Queensland, Brisbane, Australia

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Strong cross-Kerr nonlinearities have been long sought after for quantum information applications. Recent work has shown that they are intrinsically unreliable in traveling-wave configurations: cavity configurations avoid this, but require knowledge of both the nonlinearity and the loss. Here we present a detailed systematic study of cross-phase modulation and absorption in an Rb vapor confined within a hollow-core photonic crystal fiber. Using a two-photon transition, we observe phase modulations of up to π rad with a signal power of 25 μW, corresponding to a nonlinear Kerr coefficient, n2, of 0.8 × 10−6 cm/W, or 1.3 × 10−6 rad per photon.

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Photons are a promising vehicle for processing [1–7] and storing [8–14] quantum information. They are particularly attractive because of their weak interaction with the environment, ensuring long-lived quantum states. This very feature, however, implies that it is difficult to engineer deterministic interactions between photons, necessitating strong interactions between light and matter [15–17]. The best-studied light-atom interaction in this regard is the cross-Kerr effect, where an effective interaction between a control and probe field is mediated by a nonlinear medium [16]. The interaction is characterized by observing a phase shift on the probe field which varies linearly with the power of the control field. The largest cross-phase modulation observed to date is 0.2 rad per photon in microwave waveguides, using a single transmon qubit as the nonlinear medium [18]. At optical frequencies, nonlinear optical fibers with cross-Kerr shifts have been directly measured at the level of 10−7 rad per photon [19,20]. Recent experiments using vapor-filled hollow-core photonic crystal fiber (HC-PCF) inferred shifts up to 10−6 rad per photon [21]. Furthermore, such systems have also been shown to be highly effective all-optical switches [22,23].

The single-pass operation of cross-phase nonlinearities is conceptually and technologically alluring. Recent theoretical [24,25] and experimental [18] studies have shown that the extension to the single-photon regime involves subtleties about the dynamics of the nonlinear medium itself, making extrapolation to the single-photon regime difficult. Fan et al. [25] showed that for traveling waves, the interplay between quantum noise and the intrinsic saturation of the nonlinear medium ensure that single-photon-induced phase shifts are always too small to be reliably resolved shot-to-shot. Indeed, data presenting cross-Kerr shifts at optical frequencies have alluded to this being the case [19,20]. This situation can be overcome by embedding the nonlinear interaction within a resonant cavity, however, the efficiency of such is dependent on the loss of the nonlinear medium. Hence, the critical physics of this architecture is captured in the ratio of the nonlinearity to loss. Previous work using vapor-filled HC-PCF did not address this aspect. Here we present a systematic study of cross-phase modulation, atomic saturation, and loss for an HC-PCF filled with an Rb vapor. By demonstrating a large phase shift with low loss, we show the possibility of a path to a promising noncryogenic architecture for scalable quantum information processing.

The coupling between light and a collection of dipoles can be maximized by matching the transverse dimensions of both the optical field and dipoles. In practice, engineering the atomic dipole moment is difficult, however, the advent of HC-PCF enables constriction of the transverse dimensions of the optical field to several microns over arbitrarily long distances [26–28]. In our experiment we achieve an extended, and strong, light-atom interaction using an HC-PCF to confine both an optical field and Rb vapor within the fiber’s 45-μm-diameter hollow core [29]. The fiber’s kagome lattice cladding [the cross section shown in Fig. 1(b)] provided low-loss guidance from 600 to 1600 nm [30]. The fiber was mounted between two vacuum chambers, one of which contains a dense Rb vapor. Fluorescence measurements confirmed that over half of the 40-cm fiber was filled with Rb. The Rb density within the fiber was elevated by heating the vacuum chamber and fiber to ~110°C.

The $5S_{1/2}(F=3)\rightarrow 5D_{5/2}(F’=1–5)$ two-photon transition of $^{85}$Rb is used as the basis of the nonlinear interaction. The atomic energy level scheme, along with decay routes and driving lasers, is depicted in Fig. 1(a). The two-photon transition strength was resonantly enhanced by the use of a small detuning from the intermediate $5P_{3/2}$ state: this requirement set the wavelengths of the driving lasers at 780 and 776 nm, respectively. For the rest of this paper the ground ($5S_{1/2}$) state will be labeled $|g\rangle$ while the intermediate ($5P_{3/2}$) and excited ($5D_{5/2}$) energy levels are labeled $|i\rangle$ and $|e\rangle$, respectively, with associated rates $\Gamma_i$ and $\Gamma_e$. The frequency detuning from the intermediate state is given by $\Delta_i = \omega_{ie} - \omega_{80}$, and the two-photon detuning $\Delta_e = \omega_{sh} - \omega_{80} + \omega_{776}$, where $\omega_{ij}$ denotes the $|i\rangle \rightarrow |j\rangle$ transition frequency.

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The $|g\rangle \rightarrow |e\rangle$ transition was excited using a Doppler-free configuration [31], providing both strong light-atom interaction (absorption >70% for on-resonance pump laser powers >5 µW) and a narrow linewidth ($\Gamma_0 \approx 10$ MHz) [29]. These attributes make this transition ideal for cross-phase modulation experiments, as one can operate at a small detuning which provides simultaneous high interaction but small absorption. The lineshape of the transition is well described by a Voigt function with a full-width at half-maximum (FWHM) dominated by transit-time, residual Doppler, and magnetic field broadening [29].

Figure 1(c) shows the optical setup and detection scheme. The 780-nm radiation was supplied by a Ti:sapphire laser. The lasers were coupled into opposite ends of the HC-PCF, enabling Doppler-free spectroscopy of the two-photon transition within the trapped vapor. To maximize the meter power detected, the polarizations of the two lasers were aligned orthogonally, allowing their separation after the fiber using polarizing beam splitters. A diffraction grating further rejects any reflected signal beam from the input HC-PCF. This avoided saturation of the photodiode.

Finally the phase shift per atom $\phi_{\text{atom}}$ is

$$
\phi_{\text{atom}} = \phi_{\text{met}} / (\rho L A).
$$

Importantly it can be seen that, in the absence of atomic saturation, $\phi_{\text{met}}$ does not depend on the meter beam power.

To directly measure the phase shift induced by the signal beam, two separate meter beams of equal power $P_{\text{met}}$, but different frequency, were coupled into the fiber. The second meter beam was generated using an acoustic optic modulator (AOM) and was frequency offset by 80 MHz, see Fig. 1(c). This frequency separation is larger than the transition manifold width $\sim 32$ MHz [33], which ensures that only one beam interacts with the transition at a time. The noninteracting meter beam provided a phase reference while the second beam experiences the cross-Kerr phase shift. A beat-note between the two meter beams was detected, both before and after the fiber, Fig. 1(c). The former mixing product provided an RF phase reference which was compared to the output beat-note phase using an RF lock-in amplifier.
This approach thus directly measures the cross-phase shift in the optical phase of the meter signal. When compared to cross-phase measurements based on polarization rotation [21], this approach is immune to unwanted birefringence changes in the fiber that may result from vibration or temperature changes generating both short and long term noise. Furthermore, this technique automatically rejects any self-phase modulation of the meter beam because the two beams composing the meter would suffer an equal phase shift.

A typical spectrum of the phase shift and absorption as the 780-nm laser was scanned through the two-photon transition is shown in the top panel of Fig. 2. In this example a phase shift of up to $\pi$ radians was observed for $P_{\text{sig}} \approx 25 \mu W$. Asymmetry in the measured phase shift arises from the asymmetric absorption profile due to the individual excited state hyperfine components, whose positions and absorption strengths [33] are marked by vertical lines in Fig. 2. The bottom panel shows the ratio between the phase shift and absorption which is found to increase with increasing $|\Delta\nu|$, as expected from a two-level atomic model [34]. It is clear that operation at high detunings from the two-photon resonance can deliver reasonable phase shifts with exceedingly small absorption.

The sensitivity of the cross-phase modulation to both signal and meter powers was explored by varying each by over two orders of magnitude. In each measurement, 5 to 10 spectra were taken to reduce statistical uncertainty on the measured phase shift. For each spectra recorded, the measured dispersion curve was fitted and the phase shift calculated from this fit.

Knowing this, and that the data from Fig. 3 show an effective phase shift of $3.6 \times 10^{-6}$ rad/photon for $P_{\text{sig}} \approx 25 \mu W$, we use Eqs. (2) and (3) and find phase shifts of $\phi_{\text{ph}} \approx 1.3 \times 10^{-6}$ rad/photon and $\phi_{\text{atom}} \approx 2.9 \times 10^{-9}$ rad/atom. Such phase shifts correspond to a cross-Kerr, nonlinear index of...
The spectral density of the phase noise floor of our meter was $7 \times 10^{-5} / (\sqrt{P_{\text{met}}/\mu W}) \text{rad}/\sqrt{\text{Hz}}$ as directly measured at the output of the lock-in amplifier measuring the meter. This noise level was consistent with that calculated from the photon shot-noise of the meter beam, and its origin was verified by varying the meter power and observing the expected improvement in the sensitivity with the square-root of the power. This sensitivity could be improved substantially by using a detector with a higher quantum efficiency for IR radiation than the one used here (4%).

This work is a demonstration of the potential of this new platform for exhibiting strong photon-photon interaction while simultaneously showing low absorption. Furthermore, Eqs. (1) and (2) suggest several routes to improve performance. First, reducing the core diameter to 5 μm improves atom-light coupling by a factor of ~80. This has negligible effect on induced phase shifts as long as the exciting optical pulses are shorter than the average transit time for an atom across the fiber mode [21]. Second, the use of light-induced atomic desorption (LIAD) can increase the Rb density by a factor of > 200 [35–37], giving a consequent benefit in the cross-phase sensitivity. A final factor can be gained through increasing the effective atom-light interaction length by a factor of 10. This can be achieved by either filling a longer length of HC-PCF, or using slow-light techniques [38]. By using high quantum-efficiency detectors [39] and the aforementioned techniques, the extrapolated sensitivity can approach > 0.2 rad/photon. In this regime we will be able to resolve the controversy between the predictions of the classical Kerr theory and the new quantum Kerr theory outlined in Ref. [25], and lay the foundation of a scalable photonic architecture for quantum information processing.

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This chapter introduces and presents the peer-reviewed publication “Frequency evaluation of collimated blue light generated by wave mixing in Rb vapour”, published in the journal *Journal of Physics B: Atomic, Molecular and Optical Physics*.

11.1 Overview

Single-photon sources are integral components for optical quantum computing, hence there are many different implementations to generate single-photons, as discussed in section 1.3. One proposal is to use four-wave mixing in a diamond configuration within a warm atomic vapour to generate two entangled photons, one of which can be used as a herald-photon, the other as a single-photon. Here, two pump fields coherently drive a two-photon transition, generating two coherent photon through four-wave mixing, returning the atoms to its ground state. Such a four-wave mixing scheme has been demonstrated, although not at the single-photon regime. In these demonstrations the Rb $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition was pumped at 780 nm and 776 nm to efficiently produce 420 nm collimated blue light (CLB) [179–182]. If such a four-wave mixing process could produce single-photons reliably it would be ideally suited for the quantum memories discussed in section 1.3. This is because most of these memories are based on warm Rb vapours, hence single-photons generated in a Rb vapour would already be tuned to the Rb resonances due to phase matching conditions. Furthermore, as two photons are produced in the four-wave mixing process, entanglement of the two photons may be possible. To explore these possibilities a collaboration with Swinburne University of Technology was formed, where experimental work based on four-wave mixing in Rb vapours was already being conducted.

Similar to the experiments described in chapters 7 - 10, the optical transition used
was the Rb two-photon $5S_{1/2} \rightarrow 5D_{5/2}$ transition as described in section 3.1.2. This transition was excited using two laser of different wavelengths such that the detuning from the intermediate $5P_{3/2}$ energy could be controlled and was typically $< 1$ GHz. Amplified spontaneous emission at 5.23 $\mu$m on the $5D_{5/2} \rightarrow 6P_{3/2}$ enables four-wave mixing to occur. This produces an up-converted laser beam on the decaying $6P_{3/2} \rightarrow 5S_{1/2}$ transition.

The experiments described in this paper were conducted at Swinburne University of Technology, and hence was a different experimental set-up to those described in chapters 7 - 10. However, there are some similarities to the reference cell set-up described in chapter 6. The two lasers were arranged to be in a co-propagating configuration through the Rb cell so that the laser fields could be phase matched. Dichroic filters were used to separate the pump beams from the 420 nm CBL. Typically $\approx 10 \mu$W of CBL was produced using this method as described in the paper.

The resulting CBL was spectrally examined using a Fabry Pérot optical cavity showing that generate CBL cavity resonance had a width of 2.4 MHz showing that the CBL was temporally coherent. The CBL’s optical frequency was determined by comparison to a 420 nm laser tuned to the $5S_{1/2} \rightarrow 6P_{3/2}$. A beat note between these two laser sources was not possible because of the low output power of the blue laser. Hence a combination of Rb spectroscopy and the optical cavity were used to determine the CBL frequency as discussed in this paper. Using these techniques, the frequency was CBL absolute frequency was determined to be centred on the $5S_{1/2}(F=3) \rightarrow 6P_{3/2}(F’=4)$ transition of Rb.
Frequency evaluation of collimated blue light generated by wave mixing in Rb vapour

Alexander Akulshin¹, Christopher Perrella², Gar-Wing Truong², Russell McLean¹ and Andre Luiten²,³

¹ Centre for Atom Optics and Ultrafast Spectroscopy, Swinburne University of Technology, Melbourne, Australia
² School of Physics, University of Western Australia, Nedlands 6009 WA, Australia
³ Institute of Photonics and Advanced Sensing and the School of Chemistry and Physics, The University of Adelaide, Adelaide 5005 SA, Australia

E-mail: aakoulchine@swin.edu.au

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Abstract
An evaluation of the absolute frequency and tunability of collimated blue light (CBL) generated in warm Rb vapour excited by low-power cw laser radiation at 780 nm and 776 nm has been performed using a Fabry–Perot interferometer and a blue diode laser. For the conditions of our experiments, the CBL tuning range is more than 250 MHz around the resonant frequency of the $^8$Rb $5S_{1/2} (F = 3) \rightarrow 6P_{3/2} (F' = 4)$ transition. A simple technique for stabilizing the power and frequency of the CBL to within a few per cent and 10 MHz, respectively, is suggested and demonstrated.

1. Introduction

The technique of frequency conversion of low-power cw radiation of diode lasers into collimated blue and far-infrared light in Rb and Cs vapours, first demonstrated by Zibrov et al [1], continues to be an active area of research [2–7]. Potential applications of this approach include tunable coherent light sources, quantum information processing [8] and underwater communication [9]. Key spectral characteristics of collimated blue light (CBL) such as linewidth and tuning range are important for most of these applications. Previous investigations demonstrated high temporal coherence of the CBL using a Fabry–Perot cavity and two-slit diffraction [1, 2], but did not determine the absolute frequency in detail. CBL results from four-wave mixing in atomic media with a diamond-type energy level configuration. An alkali vapour driven by laser radiation tuned close to strong optical transitions in a ladder-type configuration, which in the case of Rb atoms are the $5S_{1/2} \rightarrow 5P_{3/2}$ and $5P_{3/2} \rightarrow 5D_{5/2}$ transitions (figure 1(a)), can produce an optical field at 5.23 μm through amplified spontaneous emission on the $5D_{5/2} \rightarrow 6P_{3/2}$ transition. Mixing of this field and the laser fields produces optical radiation at 420 nm in the direction satisfying the phase-matching relation $k_1 + k_2 = k_{IR} + k_{BL}$, where $k_1$, $k_2$, $k_{IR}$ and $k_{BL}$ are the wave vectors of the radiation at 780, 776, 5230 and 420 nm, respectively. The wave mixing origin of CBL is supported by observations of the unidirectional generation of the CBL [3] and the transfer of the combined orbital angular momentum of the applied laser fields to the coherent blue light [7]. But the possibility of incoherent optical pumping to the $6P_{3/2}$ level contributing to the observed CBL should not be discounted, particularly given the variations in a number of experimental parameters such as pumping rates and atomic densities made in the present study.

In contrast to conventional optical parametric oscillation, the new field generation occurs without an optical cavity. Rb atoms provide not only high Kerr nonlinearity, but also set the resonant conditions for the blue and far-IR radiation.

From the phase matching condition, which is crucial for the parametric FWM and CBL generation, for co-propagating 780 nm and 776 nm beams the blue light frequency is

$$\nu_{BL} = \left( n_1 \nu_1 + n_2 \nu_2 - n_{IR} \nu_{IR} \right) / n_{BL}. $$

(1)
where $n_1$ and $n_2$ are the refractive indices seen by the light fields at 780 nm and 776 nm, $n_{IR}$ and $n_{BL}$ are the refractive indices at 5.23 μm and 420 nm, respectively, while $v_s$ is the corresponding frequency of the optical field at each wavelength. It is clear that the absolute frequency of the CBL must lie close to the $5S_{1/2}(F = 3) \rightarrow 6P_{3/2}$ transition frequency. This is readily confirmed by the observation of isotropic blue fluorescence from Rb vapour excited only by CBL in a heated auxiliary cell. However, given that the Doppler broadening of the transition is approximately 1 GHz, this observation does not preclude a substantial frequency detuning of the CBL from the $5S_{1/2}(F = 3) \rightarrow 6P_{3/2}$ transition. Taking into account the unknown absolute frequency of the far-IR radiation and that the refractive indices depend on the optical frequencies, intensities and polarizations of the applied laser fields, as well as the atomic density $N$, the precise CBL frequency is a complex function of all these parameters. In this paper, we undertake an experimental study of the absolute frequency and frequency tuning range of the CBL generated by parametric wave mixing in Rb vapour.

2. Absolute frequency evaluation

We evaluate the absolute frequency and tuning range of the CBL generated in $^{85}$Rb vapour with high resolution by measuring the frequency difference between the CBL and reference radiation of known frequency. An obvious source of suitable reference radiation is a narrow-linewidth extended-cavity blue diode laser locked to a sub-Doppler feature of the $5S_{1/2} \rightarrow 6P_{3/2}$ absorption line produced by a standard nonlinear Doppler-free spectroscopic technique [10]. This would readily provide a frequency accuracy of approximately 1 MHz and we could measure the frequency difference between the CBL and this radiation by comparing the spectral positions of Fabry–Perot interferometer (FPI) transmission peaks of the laser light and the CBL. Unfortunately the output power of the available diode laser at 420 nm is insufficient to allow a standard saturation absorption spectroscopy arrangement and we instead implement a modified scheme for sub-Doppler spectroscopy of Rb atoms on the $5S_{1/2} \rightarrow 6P_{3/2}$ transition.

While in a conventional scheme of Doppler-free spectroscopy both the probe and saturating beams are produced from the same laser, we use the 420 nm radiation as a probe while strong optical pumping radiation is provided by a 795 nm laser tuned to the Rb D1 absorption line. This radiation produces velocity selective ground-state optical pumping of the atoms. Distinctive features of Doppler-free spectroscopy have previously been observed when using independent lasers tuned to different absorption lines [11–13].

Dramatic changes in the efficiency of CBL generation by simultaneously driving the D1 transition have been demonstrated in [6], and here we use a similar idea for producing a sub-Doppler frequency reference for the blue laser.

3. Experimental set-up

Our experimental setup, schematically illustrated in figure 1, is similar to that used in our previous experiments [3, 6]. Radiation from extended cavity diode lasers (ECDLs) at 780 and 776 nm drives stepwise and two-photon excitation of Rb atoms. The frequency of the 780 nm ECDL can be swept across the $D2$ absorption line or stabilized using a modulation-free technique based on a sub-Doppler dispersion shaped polarization resonance obtained on the $^{85}$Rb $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ transition in an auxiliary Rb cell. The 776 nm laser is also swept across the $^{85}$Rb $5P_{3/2} \rightarrow 5D_{5/2}$ transition or side-locked to a low-finesse tunable confocal etalon. In the frequency stabilized regime, typical values of the standard deviation of the error signal correspond to frequency fluctuations in the range 200–300 kHz over a 1 s time interval for both lasers. The laser linewidth estimated from optical heterodyning of two lasers is approximately 1 MHz.

Radiation from both ECDLs is combined to form a bichromatic beam. The powers of the components at 780 and 776 nm are typically 12 and 6 mW, respectively. The bichromatic beam is weakly focused into a 5 cm long heated Rb cell containing a natural mixture of Rb isotopes with no buffer gas. The cross section of the beam inside the cell is about 0.5 mm². The temperature of the cell is set within the range 50–100 °C meaning that the density $N$ of Rb atoms varies from $1.5 \times 10^{11}$ to $6 \times 10^{12}$ cm⁻³.

An additional Rb cell heated to 60 °C and two ECDLs tuned to the $^{85}$Rb D1 line and to the $5S_{1/2} \rightarrow 6P_{3/2}$ transition at 420 nm are employed to determine the absolute frequency reference for the CBL. Colour filters with optical density approximately 0.5 and 4.0 at 420 nm and 780 nm, respectively, are used to spectrally select the CBL and isotropic blue fluorescence, both of which are detected by photomultipliers. A μ-metal shield is used to reduce the ambient magnetic field in the Rb cell to a few milligauss.

The CBL spectral purity and linewidth are explored using a tuneable concave mirror FPI of length $L = 14.5$ cm and having high finesse in the blue spectral region. The
spatial distribution of the blue light transmitted through the interferometer is monitored with a CCD camera to ensure that the radiation is mainly coupled into the fundamental TEM$_{00}$ interferometer mode by a combination of lenses.

4. Results

4.1. Spectral properties of the CBL

Figure 2(a) shows CBL spectral profiles as a function of the 776 nm laser frequency taken at different atomic densities in the cell with the 780 nm laser locked to the 5S$_{1/2}(F = 3)\rightarrow 5P_{3/2}(F' = 4)$ transition. Just above the threshold atomic density, which is about $N \approx 1.5 \times 10^{11}$ cm$^{-3}$ in these experiments, the profile has a shape close to Gaussian. At higher Rb density the CBL doublet structure, which is both laser intensity and frequency dependent becomes evident. This doublet structure has been discussed previously [3], but is still not properly understood. The frequency range of the 776 nm laser over which the CBL generation occurs is less than the Doppler width of the 5P$_{3/2}$→5D$_{3/2}$ transition at this temperature.

Figure 2(b) demonstrates the blue light transmission through the scanned FPI. Narrow, high-contrast transmission resonances for various FPI tunings over the entire CBL profile confirm the single-frequency narrow-linewidth spectrum of the CBL. The spectral interval between the two highest peaks, which are due to coupling of the CBL to TEM$_{00}$ interferometer modes, corresponds to the free spectral range (FSR) of the interferometer, approximately 1034 MHz. Smaller transmission peaks are due to coupling to higher order transverse modes of the FPI and provide a convenient finer frequency scale. A single TEM$_{00}$ mode transmission resonance is shown in figure 2(c). The resonance is fitted by a Lorentzian profile of width 2.4 MHz (FWHM). This value is quite insensitive to both the applied laser intensity and the atomic density. Results of our investigation of the temporal coherence of the CBL will be presented elsewhere.

From the phase matching condition (1), frequency tuning of the 776 nm laser causes tuning of the CBL, but it is important not to assume the two frequencies will change identically. Reasons for a deviation from a direct one-to-one correspondence in the detunings ($\delta \nu_2$ for the 776 nm laser and $\delta \nu_{BL}$ for the CBL) include dispersion in the warm Rb vapour, and frequency variations of the far-IR radiation at 5.23 $\mu$m, which cannot be detected in our experiment. Spectrally selective refractive index changes have been reported in warm Rb vapours due to light-induced coherences ($\Delta n \sim 10^{-4}$ for vapours with complete transparency due to ground-state EIT [14] and $\Delta n \sim 0.1$ in cascade configuration at high atomic density $N \sim 10^{15}$ cm$^{-3}$ [15]). Simultaneous observations of the CBL intensity variations and FPI transmission resonances as a function of the 776 nm laser frequency detuning such as in figure 3(a), allow a quantitative relation between $\delta \nu_2$ and $\delta \nu_{BL}$ to be established. For this purpose, $\delta \nu_{BL}$ is estimated using the low-finesse confocal etalon, while $\delta \nu_{BL}$ is measured using the frequency scale provided by the higher order modes of the blue FPI. The length of the blue FPI is adjusted so that transmission peaks of its fundamental axial modes approximately coincide with one of the intensity maxima of the CBL profile. With this technique we find that $\delta \nu_{BL}/\delta \nu_2 \approx (1.00 \pm 0.03)$. The precision is estimated from the nonlinearity in the 776 nm laser frequency tuning, so that within this
atomic density dependent just above the CBL threshold \((N < 6 \times 10^{11} \text{ cm}^{-3})\). We have noted previously that there is an atomic density threshold for generation of CBL as well as a strong CBL intensity dependence on atomic density [3]. The atomic density directly influences parameters that are important for parametric FWM such as how far the various beams propagate through the cell before being absorbed. This makes a theoretical estimation of the CBL tunability difficult. Under present experimental conditions the atomic density dependence saturates at \(N > 7 \times 10^{11} \text{ cm}^{-3}\), while the dependence on the 780 nm laser power reveals almost constant growth.

### 4.2. Blue laser frequency evaluation

We now describe how the velocity-selective optical pumping technique can be used in evaluating the absolute frequency of the blue ECDL (figures 1(a), (c)).

If the blue laser with fixed frequency \(\nu_L\) is tuned to the peak of the inhomogeneously broadened fluorescence line on the \(^{85}\text{Rb}\) \(5\text{S}_{1/2}(F = 3) \rightarrow 6\text{P}_{3/2}(F' = 2, 3, 4)\) transitions, then the main contribution to the isotropic fluorescence comes from atoms excited on the \(F = 3 \rightarrow F' = 4\) cycling transition and having longitudinal velocity \(\nu_z = 2\pi (\nu_L - \nu_{34})/k_{\text{BL}}\), where \(\nu_{34}\) is the resonant frequency of the cycling transition. Two other resonant velocity groups which interact on the weaker open transitions \(F = 3 \rightarrow F' = 2, 3\) are significantly depopulated due to spontaneous decay to the \(5\text{S}_{1/2}(F = 2)\) level.

The population of the \(\nu_j\) velocity group in the \(5\text{S}_{1/2}(F = 3)\) level could itself be modified by hyperfine optical pumping. If, for example, counter-propagating radiation at 795 nm having frequency \(\nu_{D1}\) is detuned by \(\delta \nu_{D1} = (\nu_{34} - \nu_{D1}) = k_{D1}\nu_z/2\pi\) from either transition from the \(^{85}\text{Rb}\) \(5\text{S}_{1/2}(F = 2)\) level, where \(\nu_{34}\) is the frequency of the \(5\text{S}_{1/2}(F = 2) \rightarrow 5\text{P}_{1/2}(F = j)\) transition, this results in a sub-Doppler width enhancement of the blue fluorescence. Through a comparison of the spectral positions of the sub-Doppler fluorescence peak and saturated absorption resonances observed simultaneously on the \(5\text{S}_{1/2}(F = 3) \rightarrow 6\text{P}_{3/2}\) and \(5\text{S}_{1/2}(F = 2) \rightarrow 5\text{P}_{1/2}\) transitions in the auxiliary Rb cell as the 795 nm laser is scanned, the frequency \(\nu_L\) of the blue laser can be evaluated. Due to the different Doppler shifts the detunings differ by a factor equal to the ratio of the wavelengths:

\[
\delta \nu_{D1} = (\nu_L - \nu_{34})(k_{\text{BL}}/k_{D1}) \approx 1.89 \times \delta \nu_L. \tag{2}
\]

Figure 4 shows normalized fluorescence at 420 nm produced by the fixed-frequency blue laser at \(\nu_L\) and plotted as a function of the optical pumping laser frequency \(\nu_{D1}\) as it is swept across the inhomogeneously broadened \(^{85}\text{Rb}\) \(5\text{S}_{1/2}(F = 2) \rightarrow 5\text{P}_{1/2}(F = 2, 3)\) transitions. In figure 4(a) the fluorescence is enhanced above the no-optical pumping level at four different frequencies of the pumping laser rather than the expected two. The two smaller peaks arise from optical pumping produced by 795 nm light back-reflected from the cell window. The D1 line saturated absorption resonances observed in the auxiliary Rb cell provide absolute frequency references for the optical pumping laser. The blue laser detuning \(\delta \nu_L\) is estimated using equation (2) from the offset of the fluorescence resonances from these saturated absorption resonances. For
example, the frequency offset shown in figure 3(a) suggests that the blue laser is tuned approximately 60 MHz above the $^{85}$Rb $5S_{1/2}(F = 3) → 6P_{3/2}(F' = 4)$ transition frequency. If the laser is tuned precisely on resonance, the large and small fluorescence peaks merge and spectrally coincide with the D1 line saturated absorption resonances, as shown in figure 4(b). In this case the 420 nm laser is resonant with the atoms that have zero axial velocity. Using this method the frequency of the blue laser can be tuned to the $^{85}$Rb $5S_{1/2}(F = 3) → 6P_{3/2}(F' = 4)$ transition with approximately ±5 MHz accuracy. Figure 4(b) also illustrates power broadening of the sub-Doppler resonances. It is possible to obtain better resolution with lower pumping power.

Figure 5 shows FPI transmission resonances for the blue laser and CBL generated in the Rb cell and the 420 nm laser tuned to the $^{85}$Rb $5S_{1/2}(F = 3) → 6P_{3/2}(F' = 4)$ transition, using the procedure described above. The separation of the peaks is easily changed by tuning the frequencies of the 776 and 780 nm lasers around their corresponding transitions. In this way we are able to confirm that the CBL frequency is centred on the $^{85}$Rb $5S_{1/2}(F = 3) → 6P_{3/2}(F' = 4)$ transition, and that it could be tuned over a range of 250 MHz in this experiment.

4.3. CBL frequency stabilization

For the CBL to be used effectively as a narrow-band light source, it is desirable to have a means of stabilizing its frequency and amplitude. As has been discussed above, the frequency of the CBL depends primarily on the frequencies of the two applied laser fields. Assuming the other factors are not varying, stabilizing the frequencies of the two applied laser fields can stabilize the CBL frequency and to some extent the CBL power. While the 780 nm laser can be locked routinely to the strong $^{85}$Rb $5S_{1/2}(F = 3) → 6P_{3/2}(F' = 4)$ cycling transition with standard Doppler-free spectroscopic techniques, this is not the case for the 776 nm laser as it is tuned to a transition between excited states. Although one could stabilize this laser to a reference cavity with high long-term stability, a more elegant solution is to lock the 776 nm laser directly to the CBL profile. Here we demonstrate that both peak and side locking can be used to control the step-wise excitation of Rb atoms and, consequently, the CBL frequency and intensity, with the error signal derived from the CBL intensity itself.

For the peak locking, the dither voltage at 10 kHz from a lock-in amplifier modulates the 776 nm laser frequency. Curve (i) in figure 6(a) shows the intensity profile of the CBL as the 776 nm laser is scanned across the $5P_{3/2} → 5D_{5/2}$ transition, with an accuracy of ±5 MHz. The increased error signal in the 2 MHz CBL cycling transition is due to servo system gain adjustments while searching for the 776 nm laser. The 776 nm laser is scanned across the $5P_{3/2} → 5D_{5/2}$ transition, with the 780 nm laser locked to the $^{85}$Rb $5S_{1/2}(F = 3) → 6P_{3/2}(F' = 4)$ transition. Curve (ii) represents the lock-in amplifier output, which is proportional to the first derivative of the CBL spectral profile, and is used as an error signal.

Active stabilization of the CBL is shown in figure 6(b). The 776 nm laser is initially scanned manually, then tuned to the higher peak of the CBL profile and the locking loop closed at $t = 0$. The increased error signal in the $2s < t < 5s$ interval is due to servo system gain adjustments while searching for optimum values of the locking parameters.
and error signal temporal variations recorded with slow manual frequency scanning of the 776 nm laser (regime (776 nm laser scan while the 780 nm laser is locked to the 5S1/2→5P3/2(F = 4) transition). The error signal is derived from the CBL profile itself and used to stabilize the power and frequency of the CBL, in which an error signal to noise ratio of the error signal and long-term CBL frequency locking, as the CBL reference line is relatively broad. The CBL self-locking is very robust due to the large recapture range provided by the relatively wide reference line. The considerably improved long-term frequency and intensity stability demonstrates the effectiveness of this simple method.

We have also demonstrated that the 776 nm laser can be locked to the side of the CBL resonance, eliminating the need to dither the 776 nm laser frequency and preserving the intrinsic linewidth of the CBL. Even higher values are obtained for the relative frequency stability of the CBL (ΔνBL/νBL ≈ 6 × 10^{-10} for 10 s); however, the coupling between CBL power and frequency variations and sensitivity to beam alignment impose limits on the long-term stability achievable with this approach. Side locking is preferable if high coherence of the CBL is required. We note also that a similar arrangement for locking the sum frequency of two lasers to Doppler-free fluorescence resonances that result from two-photon excitation of Rb atoms on the 5S1/2→5D3/2 transition was implemented in [16]. In that work the reference resonances are much narrower, but more importantly their absolute frequency is determined by the separation between the Rb energy levels, while the CBL frequency is a complex function of the parametric FWM process.

5. Conclusion

We have investigated the absolute frequency of collimated blue light generated by a parametric four-wave mixing process in atomic Rb vapour. The frequency of the CBL has been compared with that of a blue diode laser using a tunable Fabry–Perot interferometer. The blue laser frequency has, in turn, been determined using a modified sub-Doppler spectroscopy technique where velocity-selective pumping radiation has been applied on the Rb D1 line. This has allowed the absolute frequency of the CBL to be evaluated with approximately ± 5 MHz precision, limited primarily by the precision with which we are able to determine the frequency of the laser. The CBL frequency is found to be centred on the 5S1/2→6P3/2(F = 4) transition, and can be tuned over a range of more than 250 MHz around this frequency by tuning the frequencies of the driving lasers. Atomic density and laser power dependences of the CBL tuning range have been examined.

Finally, we have proposed and demonstrated a method for stabilizing the power and frequency of the CBL, in which an error signal is derived from the CBL profile itself and used to lock the frequency of the 776 nm laser involved in the wave mixing process.

Acknowledgments

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References


This chapter summarises the work presented in this thesis regarding the technology of a Rb filled HC-PCF, while also presenting future pathways to extend its use into other fields. Section 12.1 ties the work presented in this document together, focussing on the results that have been achieved using the Rb filled HC-PCF. Section 12.2 discusses future avenues for research on vapour filled HC-PCFs, including: more efficient techniques for loading HC-PCF, extensions of the applications presented here, and several new applications.

12.1 Conclusion

The work presented in this thesis is based on loading a HC-PCF with a Rb vapour. Important applications for this technology can be found in the fields of metrology and quantum optics, as outlined in chapter 1. These include: a HC-PCF based frequency standard, discussed in section 1.2; and a platform to produce efficient photon-photon interactions, measure photon numbers, and produce single-photons, all of which are discussed in section 1.3. Furthermore, the small size and robustness of HC-PCF allows this device to potentially be miniaturised for commercial and industrial applications.

The different styles of HC-PCF used were discussed in chapter 2. Specific details regarding the particular fibres used in the experimental work presented were discussed in section 2.4. Techniques to load a HC-PCF with a Rb vapour, along with experimental data demonstrating their effectiveness was presented in section 6.3.

The energy level structure and physical properties of Rb were discussed in chapter 3, which outlined the $D_1$, $D_2$ and two-photon Rb transitions which were used in the experimental work presented in chapters 7 - 11. Using these transitions, the effect of the fibre’s confined geometry upon the atomic spectra was investigated. Extent-
sive theoretical modelling, both analytic and numerical, was conducted to identify the major broadening mechanisms present, as described in chapters 4 and 5. To investigate these effects, atomic spectra were measured with the experimental set-up described in chapter 6 with the results presented in chapters 7 and 8.

Transit-time broadening was found to be a major source of spectral broadening within the HC-PCF for both the hole-burning and two-photon experiments, with contributions of $1.0 \pm 0.7 \text{ MHz}$ and $5.61 \pm 0.05 \text{ MHz}$ respectively. The marked difference in transit-time broadening contribution is due to a difference in effective mode area within the fibre for each experiment. For the hole-burning experiment, the mode diameter was $36 \mu\text{m}$, while for the two-photon experiment the effective mode area was $13 \pm 1 \mu\text{m}$. The small two-photon mode area is due to the multi-mode nature of the HC-PCF generating poor overlap of the two driving laser beams as discussed in chapter 8.

It was demonstrated in chapters 7 - 10 that a Rb filled HC-PCF provides excellent light-atom coupling as both the atoms and light were confined within the same volume. This was particularly evident on the Rb $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition which exhibited complete extinction of the driving laser beams when on two-photon resonance. Such absorption was orders of magnitude larger than that possible with free-space optics due to the excellent light-atom coupling provided by the HC-PCF. This two-photon transition excited within the HC-PCF was extensively characterised in chapter 8, which showed it behaved as expected, in agreement with the theoretical models developed in sections 4.4.2 and 5.1.2. Using the two-photon transition, three applications were investigated and discussed.

The first application was an optical atomic frequency standard which made use of the long atomic lifetime for the Rb $5D_{5/2}$ energy level. For this frequency standard the $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition was excited with two lasers at $780\text{ nm}$ and $776\text{ nm}$. As described in chapter 9, it was the sum frequency of these lasers which was the stabilised optical signal. Two different implementations of this frequency standard were demonstrated, both of which have the potential to be compact and robust, with applications in commercial and industrial environments. The first frequency standard was based on a HC-PCF loaded with a Rb vapour, in which modulation-transfer spectroscopy was used to provide a frequency discriminator. This resulted in a fractional frequency stability of $9.8 \times 10^{-12}$ at 1 s. The second scheme used a Rb bulk cell where detection of atomic fluorescence was employed to create a frequency discriminator. This produced a fractional frequency stability of $8 \times 10^{-13}$ at 1 s. The stability of both these frequency standards was measured by comparison to an optical frequency comb. Current performance limitations for both
standards were presented, along with a clear pathway to improving the performance of both standards by an order of magnitude.

The second application targeted the area of quantum optics where photon-photon interactions are keenly sought after. Here, the Rb filled HC-PCF was used to produce efficient photon-photon interactions using the Rb two-photon transition. Chapter 10 presented a systematic study of cross-Kerr non-linearities, and absorption, based around the two-photon transition excited within the HC-PCF. A self-heterodyne technique was used to measure the induced phase-shifts which characterise the strength of the photon-photon interaction. A maximum phase-shift of $\pi$ rad with a signal power of 45 $\mu$W was observed, corresponding to a non-linear Kerr coefficient, $n_2$, of $1.4 \times 10^{-6}$ rad cm$^2$/W, or $0.7 \times 10^{-6}$ rad per photon.

Finally, the Rb filled HC-PCF could potentially be used as a single-photon source by exploiting the two-photon transition and four-wave mixing processes. A preliminary investigation into this was carried out in collaboration with Swinburne University of Technology, where four-wave mixing using the Rb two-photon transition was carried out to produce a collimated beam of 420 nm light. The spectral coherence and absolute frequency of this up-converted beam was experimentally tested. A Fabry Pérot optical cavity was used to place an upper limit of 2.4 MHz on the 420 nm light’s linewidth. Furthermore, a 420 nm laser in conjunction with optical pumping spectroscopic techniques was used to determine the absolute frequency of the generated 420 nm light to be centred on the $5S_{1/2}(F=3) \rightarrow 6P_{3/2}(F'=4)$ transition of Rb.

12.2 Future Project Directions

The work presented in this thesis is the beginning of a whole host of opportunities that could be achieved with Rb loaded HC-PCF. This section aims to highlight new methods for loading HC-PCF with Rb vapours, along with extensions to the applications already presented. Furthermore, future applications not yet presented in this thesis will be discussed.

12.2.1 Future Loading Techniques

The current vacuum system for loading Rb vapour into a HC-PCF, discussed in section 6.2, is limited in a number of ways. Firstly, it is only capable of loading a single fibre with a Rb vapour at one time. Secondly, it relies on Viton seals to produce a high quality vacuum seal between the fibre and the vacuum system,
limiting the temperature that the vacuum system can be heated to, hence limiting the Rb density and HC-PCF loading rate. Thirdly, the HC-PCF is bent leading to higher loss and coupling to higher-order transverse optical modes. Finally the system is very slow to load a fibre with Rb vapour. Presented below are several approaches that aim to overcome these limitations.

Next Generation Vacuum System

One approach to increasing the loading speed of a HC-PCF is to increase the Rb density. As discussed in section 3.2.2, the vapour pressure of Rb is temperature sensitive. By increasing the vacuum chamber temperature from \( \approx 100^\circ C \) to \( \approx 150^\circ C \), the Rb density, and hence loading speed of a HC-PCF, increases by a factor of 15. Furthermore, by loading the HC-PCF from both ends of the fibre, an extra factor of 2 can be achieved, making the loading speed \( \approx 30 \) times faster. Based on this reasoning, a new vacuum system was designed as shown in figure 12.1.

To achieve this, the Viton seals present in the system, as discussed in section 6.2, was removed to allow higher baking and operating temperatures. The resulting vacuum system was constructed using only copper gaskets, capable of maintaining a vacuum seal at temperatures \( > 200^\circ C \). Furthermore, the whole fibre is contained within a single chamber, hence capable of loading the fibre from both ends.

To maximise the output of this vacuum chamber, it was designed so that 5 fibres could be loaded at a single time. These fibres are all presented in section 2.4 with their cross-sections and transmission/loss spectra shown in figure 2.3. A specially designed fibre holder was manufactured to hold the multiple fibres as shown in figure 12.1. This holder allowed access to the fibre ends for input-/output-coupling and observation of fluorescence from midway down the fibre’s length.
12.2. FUTURE PROJECT DIRECTIONS

Figure 12.2: Side-on microscope image (right) and cross section scanning electron microscope image (right) of a 7-cell Kagome fibre with its cladding collapsed.

To test the origin of the observed reduction in HC-PCF transmission during Rb loading, a fibre with collapsed cladding at each end was included in the vacuum system. Using a Vitron splicer, the ends of a 7-cell kagomé HC-PCF (shown in figure 2.3) were collapsed so that the kagomé cladding did not fill with Rb, as shown in figure 12.2. By observing the transmission of this HC-PCF as it loads with Rb, the importance of this aspect upon HC-PCF transmission will be ascertained.

Unfortunately, due to time constraints, this vacuum system was built but no experimental data was retrieved from it prior to the submission of this thesis. Future work involves conducting spectroscopy of the Rb vapour loaded within each fibre, to present clear results regarding transit-time broadening within a variety of HC-PCFs with varying core diameters.

Sealing the HC-PCF

Ultimately, for any device based on this technology to be compact and robust, the HC-PCF needs to be removed from the vacuum chamber all together. For this to occur, both ends of the HC-PCF need to be collapsed, or fused onto a standard SMF fibre, in order to seal the HC-PCF’s core from atmosphere or other containments. This has been demonstrated to be possible with molecular vapours which have pressures ranging between \( \approx 4 - 7500 \text{Torr} \) [116, 117], orders of magnitude larger than the vapour pressure of Rb. Nonetheless, there is no fundamental reason why such a process could not also be achieved with a Rb loaded HC-PCF.
12.2.2 Trapping Hot Atoms

The dominant spectral broadening source within HC-PCF is transit-time broadening, as described in chapters 7 and 8. To avoid this, there are two options: increase the HC-PCF’s core diameter, which has the unwanted effect of reducing the efficiency of the two-photon transition due to lower intensities; or reduce the atom’s average transverse speed. The first option is not favourable as many of the benefits of HC-PCF are lost as the core diameter is increased. Instead, the second option is favourable as light can be used to slow the atoms.

The enormous intensities achievable within HC-PCF, due to their confined geometry, lends itself toward dipole trapping and guiding atoms within the HC-PCF. This has been explored previously by guiding warm atoms through a capillary [183, 184], and cold atoms into a HC-PCF [185–187]. Using realistic values for optical powers and detuning from the $D_2$ transition ($P \approx 100\,\text{mW}$ and $\Delta \approx 30\,\text{GHz}$), a dipole potential depth of 1 K can be produced within a 7-cell defect bandgap HC-PCF [183]. This would result in 0.001% of the thermal atomic population being trapped within the HC-PCF’s optical mode, hence not colliding with the core walls and greatly extending light-atom interaction time. This would be of great benefit to all the applications previously described.

12.2.3 Optical Magnetometry

A novel application for Rb loaded HC-PCF is optical magnetometry, in which optical fields can be used to sense magnetic fields. Optical magnetometry has proved to be very successful with detectors reaching sensitivities better than 1 fT at 1 s averaging times [188]. This surpasses the sensitivity of commercial superconducting quantum interference devices (SQUID) which are commonly used to measure small magnetic fields [188]. However, SQUIDs are large and not easily portable devices, hence an alternative is an optical magnetometer based on a Rb filled HC-PCF.

An optical magnetometer based on a Rb filled HC-PCF would be able to probe weak magnetic fields in locations that SQUIDS cannot reach. Such an example would be to use an array of Rb filled HC-PCF optical magnetometers to probe the magnetic fields created by the human heart or brain, by placing them around the subject being measured [188]. Such a concept is feasible assuming long light-atom interactions can be achieved within a HC-PCF. One such method is to use an optical dipole trap as described in the previous section. An alternative is to use a buffer gas which produces atomic collisions to slow the atom’s transverse velocity. As describe in section 5.1.5, a background gas causes de-coherence of an
optically excited state, however these optical magnetometers rely on ground state coherence which is significantly less perturbed through atomic collisions. Indeed, a vast amount of optical magnetometers rely on buffer gases to produce extended light-atom interaction times [188]. Hence, a Rb filled HC-PCF, loaded with a buffer gas to increase light-atom interaction times could possibly produce a miniature optical magnetometer.
Part IV

Appendices
A List Of Publications

A.1 Journal Papers

Published


APPENDIX A. LIST OF PUBLICATIONS

Accepted


In Preparation

- C. Perrella, T. J. Milburn, P. S. Light, T. M. Stace, A. N. Luiten, ‘Sub-Doppler features within Doppler broadened two-photon spectra’

A.2 Conference Proceedings

Presenting authors are highlighted in bold.


and Quantum Optics’, Australian Institute of Physics (AIP), Melbourne, Australia.


Spectral Selection of Optical Comb Modes

This chapter introduces and presents the paper “Interferometric selection of frequency comb modes”, submitted to the peer-reviewed journal Applied Physics B: Lasers and Optics.

B.1 Overview

The ability to spectrally select the modes of an optical frequency comb allows for greater flexibility and usability. Commercially available fibre frequency combs typically have repetition rates of 100 – 250 MHz, resulting in optical mode spacings that are too narrow to be of use as they can yield ambiguous results in certain circumstances. Many different techniques have been implemented to circumvent this issue, which are discussed in the paper. In the approach presented here, an optical frequency comb was spectrally split into four daughter combs, each with a four times large repetition rate, and hence larger spacing between the optical modes.

To achieve this, a fibre-based Menlo optical frequency comb was used with a repetition rate of 250 MHz. The pulse output of the comb was increased by four times using fibre-based Mach-Zehnder interferometers to split, then interleave the optical pulses together. This approach readily increases the optical pulse repetition rate, however more care is required to spectrally rarify the optical comb. To accomplish this, the optical phase within each Mach-Zehnder interferometer needs to be controlled so that when the optical pulses are recombined, their phase difference is integer multiples of π.

Using this technique, the paper demonstrates the ability to spectrally split the comb into four “daughter” combs each with a repetition rate of 1 GHz, four times the original.
Interferometric selection of frequency comb modes

C. Perrella · P. S. Light · J. D. Anstie · F. N. Baynes · A. N. Luiten

Abstract We demonstrate a scheme to split an optical frequency comb into four separate frequency combs, each with four times the repetition rate of the original, but which are offset in frequency from each other. These spectrally rarified “daughter” combs are generated using fibre interferometers that are actively stabilised. We describe how these “daughter” combs can be used to resolve ambiguities that occur when comparing an arbitrary frequency continuous-wave signal against an optical frequency comb.

1 Introduction

Researchers have long dreamed of optical domain versions of coherent instrumentation such as the frequency synthesizer, arbitrary waveform generator, or network analyser. The optical frequency comb has been the breakthrough that has brought this dream close to reality, and one already sees important applications in high precision laser spectroscopy [1–3], frequency metrology [4–7] and arbitrary waveform generation [8, 9].

Nonetheless, the frequency comb remains a complex tool that is yet to be incorporated into widely available coherent instrumentation. To use the comb in this type of instrumentation requires three separate, but equally critical characteristics: (a) a compact and reliable comb source; (b) an inter-mode frequency spacing that is sufficiently high [10, 11] to identify each mode through conventional techniques [4], though not so high as to require extremely fast photonic techniques [12]; and (c) the means to unambiguously identify the difference in frequency between some arbitrary cw laser signal and the known comb mode frequencies [13, 14].

To meet requirement (a), the focus has been on fibre-based mode-locked lasers [15] and microresonators [16, 17]. Unfortunately, the mode spacing of conventional fibre-based mode-locked lasers is too narrow (100–250 MHz) to easily meet requirement, (b) and hence, a number of attempts have been made to increase this spacing by either modifying the laser itself [11, 18], or manipulating the pulse stream [19–25]. One sees that a simple mixing of a raw frequency comb and a cw laser source (i.e. the conventional technique to ascertain frequency differences between two cw laser signals) does not satisfy requirement (c) as this approach yield ambiguous results in certain circumstances. For example, when the cw laser has a frequency that matches a comb mode, or falls midway between two comb lines, then the standard frequency comparison process yields zero-frequency or near-degenerate mixing products [13, 14]. The low repetition rate of modern fibre-based mode-locked lasers exacerbates this problem as these “dead zones” are relatively closely spaced in frequency (50–125 MHz).

Many different techniques have been proposed to circumvent this ambiguity problem. For example, one can isolate a single comb mode and then generate a beat note between the cw signal and this isolated mode [26, 27]. The inherent challenge with this type of approach is the requirement to automatically isolate a comb mode that is...
sufficiently close to the optical cw signal. Alternatively, one could re-tune or step the comb [13, 28] to avoid entering a dead zone, however, there is unavoidable loss of phase during these switching events [14]. In addition, this approach is not amenable to situations in which multiple simultaneous measurements are performed using a single comb. The more general solution to this problem falls into two classes: either an elegant dual-comb solution [29] where a vernier approach provides the required unambiguous information, or the use of auxiliary signals at a known frequency offset from the incoming signal [14]. By the right choice of repetition rate difference between the two combs, or of the offset frequency of these auxiliary signals, it is possible to guarantee an unambiguous beat note over a wide range in input signal frequency [14, 29]. In contrast to those approaches, here, we make use of just a single comb and slice it into four constituent combs—each with a repetition rate four times the original, but where each “daughter” comb is offset from the other by the original repetition rate. As we will show below, this allows us to unambiguously derive the frequency relationship between the incoming signal and the original reference comb. In this article, we demonstrate this by developing a scheme that automatically stabilises the required interferometric elements in the correct phase. We also consider the effect of errors in the interferometer delays on the resulting modified optical combs. It is important to note that here, we are demonstrating more than just repetition rate multiplication—we are modifying the optical spectrum itself to generate a sparser optical frequency comb. It is this aspect which requires careful phase stabilisation and which would not be necessary if the objective was merely to multiply the repetition rate.

2 Theory

A frequency comb can be expressed in the form: $E(\omega) = \sum_m \tilde{a}_m \delta(\omega - m\omega_c - \omega_o)$ where $\delta(x)$ is a Dirac delta function, $\omega_c$ is the repetition rate frequency, $m$ is the mode number, $\omega_o$ is the carrier-envelope offset frequency, and where $\tilde{a}_m$ represents the spectral phase and amplitude of the $m$th mode. For definiteness, let us consider the case where $\tilde{a}_m = \exp(-((\omega - \omega_c)^2)/(4\Delta \omega^2)) \exp(-is(\omega - \omega_o))$, which represents a Gaussian amplitude envelope centred at a frequency of $\omega_c$, with an e-fold width of $\Delta \omega$ and which has allowed for a linear phase ramp with a slope of $s$. For a comb consisting of a large number of frequency modes (i.e. $m$ has a large range), the corresponding time-domain representation is a train of pulses with a Gaussian pulse envelope:

\[
E(t) \propto \sum_n \exp\left[-(t - nt_r - s)^2/\tau^2\right] \exp\left[i(\omega_n t + \phi_{\omega,n})\right]
\]

(1)

where $\tau = 1/\Delta \omega$ is the pulse duration, $t_r = 2 \pi/\omega_o$ is the inter-pulse time, $s$ sets a time offset of the pulse peak, and $\phi_{\omega,n} = n\omega_o t_r$ is the carrier phase offset for the $n$th pulse.

A standard approach to double the repetition rate of frequency combs [19, 23, 25, 30] is to inject the pulse train into an unbalanced Mach-Zehnder interferometer (MZI) where one arm of the MZI is designed to deliver a delay, $\tau_d$, that is close to an odd multiple of $t_r/2$ with respect to the other arm (as shown in the first box on the left hand side of Fig. 1). As we will show, in general, this approach does not automatically double the inter-mode spacing of the comb. The pulse train at one of the output ports of the MZI can be written as $\text{MZI}(t) = E(t) + BE(t - \tau_d) \exp(i\eta)$ where $B$ allows for inequality in the power of the two recombined combs, and $|\eta| < \pi$ accounts for the relative carrier phase (i.e. evaluated at $\omega_c$) of the two recombined pulse trains at the output of the recombining beamsplitter. This phase difference $\psi$ is set by the relative path length difference of the arms as well as the phase conditions at the input and output beamsplitters. We have chosen to express the recombined output in this way, splitting the time delay, $\tau_d$, that sets the location of the pulse envelope, from the rapidly varying optical phase component, $\psi$, as this allows us to emphasise the key differences between modifying the pulse rate and modifying the spectrum of the comb.

If the output of this unbalanced MZI is detected with a fast photodetector (as is simply done to generate a microwave comb [19, 23, 25, 30]), then we would detect a series of current pulses corresponding to the impulse response of the photodiode (because the optical pulse duration is much shorter than the detector response time). For an ideal photodetector, characterised by a simple response time of $\tau_{PD}$, the power spectral density of the detector output will be $|\text{PD}(\omega)|^2 \propto \sum_m \tilde{b}_m \delta(\omega - m\omega_c)$ where

\[
\tilde{b}_m = \left(1 + B^2 + 2B \cos[mt_r(\omega_o)]\right) \left(\frac{1}{4(1 + |m\omega_o \tau_{PD}|^2)}\right)
\]

(2)

We note that this expression is independent of $\psi$ and $\omega_o$, and we further see that if $B \sim 1$ and $\tau_d \sim (n + 1/2)t_r$ (where $n \in \mathbb{Z}$), then the even harmonics of the $\tau_{PD}$ rate will be enhanced while the odd harmonics are suppressed—effectively the pulse repetition rate of the comb has been doubled. The independence of the result in Eq. 2 on the relative carrier phase, $\psi$, shows why it has been unnecessary to stabilise $\psi$ in prior work aimed at repetition rate multiplication [19, 23, 25, 30].
In contrast, it is crucial to note that the optical spectrum of the interferometer output is sensitive to $\psi$. The power spectral density of MZI($t$) is proportional to

$$\sum_{m} \tilde{e}_m \delta(\omega - m\omega_r - \omega_s) \exp(-\tau^2(m\omega_r - \omega_s)^2)$$  \hspace{1cm} (3)$$

where $\tilde{e}_m = 1 + B^2 + 2B \cos[\psi + \tau_d(m\omega_r - \omega_s)]$. From an examination of Eq. 3, it is clear that to split the original comb into odd- and even-mode combs, with each separately exiting from one of the ports of the MZI, it will be necessary to set power balance between the two combined combs ($B \sim 1$) while $\tilde{e}_m$ must equal 0 for either odd or even $m$. It is clear that this only occurs when both $\tau_d \sim (n + \frac{1}{2})\tau_r$ for $n \in \mathbb{Z}$ and

$$\psi = \pi \frac{[2n + 1]\omega_r}{\omega_r} \mod \omega_r.$$  \hspace{1cm} (4)$$

To explain more completely the effect of $\psi$, let us consider the simple case in which $\tau_d = \frac{\tau_r}{2}$ and where $\omega_r$ is an exact multiple of $\omega_s$ (i.e. $\omega_s = 0$). Let us then set $\psi$ to be equal to zero as it must be according to Eq. 4. If we examine the time-domain behaviour of the output pulse train at B1, then we see that the output contains pulses delivered alternately from the upper and lower paths of the interferometer; however, the carrier phase of each output pulse is exactly the same (because $\psi = 0$). From the frequency domain view, this output comb has an inter-mode spacing of $2\omega_r$ with an $\omega_s$ of zero; that is, it is the even mode in the original input comb. If one examines the pulse stream at the B2 output (under the same circumstances), we note that the apparent carrier phase would be flipping by $\pi$ between adjacent pulses (this is enforced by the phase relations of a beamsplitter [32]). One thus sees the B2 output comb as having an inter-mode spacing of $2\omega_s$ with an offset frequency, $\omega_s$, equal to $\omega_r$ (i.e. exactly half the repetition rate of the output comb): this clearly corresponds to the odd modes of the input comb. We note also that this theory predicts a rather paradoxical situation: if $\psi$ is not set to one of the special values in Eq. 4, then the output comb will contain energy from all modes in the original comb (i.e. a mode spacing of $\omega_r$) although the measured inter-pulse time would be $t_r/2$ rather than the expected $t_r$.

If there is an overall error in the interferometer arm imbalance, that is, $\tau_d = (n + \frac{1}{2})\tau_r + \Delta$ where $\Delta$ represents the time error, then there is a resulting effect on both the spectrum of the pulses as well as on the optical spectrum. The pulse repetition rate spectrum will only show the desired suppression of the odd harmonics with respect to the even harmonics for modes up to an RF frequency of $1/(4\Delta)$. In the optical spectrum, we will only obtain a splitting of odd and even modes over a limited range of the spectrum even if the value of $\psi$ is held stable. The full frequency distance between the places where the odd and even comb modes becomes equal in amplitude (i.e. an estimate of the spectral distance over which the splitting is functional) is $1/(2\Delta)$. We demonstrate both of these effects experimentally. Further, we note that this technique is reasonably broadband since an arm length error of 10 μm can still lead to a bandwidth of 15 THz.

3 Experiment

The original comb is generated by a commercial 250 MHz repetition rate fibre mode-locked laser, which is frequency-doubled to 780 nm. The comb is injected into two cascaded fibre-based MZIs as shown in Fig. 1. The phase imbalance, $\psi$, in each MZI is detected by injecting a cw auxiliary signal into the input of the first stage and then modulating fibre stretchers that are placed in one arm of each of the two MZIs. The fibre stretchers are modulated at different frequencies (~2.5 and 3 kHz) resulting in dual synchronous modulation of the output power at the output of the second MZI. This signal is detected with two lock-in amplifiers to generate independent error signals, which are used to stabilise the relative phase, $\psi$, in both MZIs using the stretchers. No synchronous power modulation arises from the presence of the comb in the MZIs as there is no optical interference when the two time-displaced combs are recombined.
The output of the lockin amplifiers is zero when the auxiliary laser is in constructive or destructive interference at the monitored port. By tuning the frequency of the auxiliary laser, we can stabilise $\psi$ at any arbitrary value. To demonstrate the efficacy of the $\psi$ locking system, the output of the first MZI stage was coupled to both an optical cavity and a fast photodiode (see Fig. 2). The optical cavity has a free-spectral range of 29.4 GHz with a finesse of $\sim 300$. We used a monochromator to filter out $\sim 30$ GHz of the spectrum to prevent the confusion of overlapping cavity orders (although small overlaps are still visible at the extrema of Fig. 2a). It is the filtering effect of the monochromatic that is responsible for the overall envelope of the optical comb seen in the top panel of Fig. 2a—the actual comb itself is much broader than that displayed. The output from the optical cavity as it is scanned is shown in Fig. 2a while Fig. 2b shows the spectrum at the output of the fast photodiode detecting that optical comb. In the first row, we have adjusted the arm imbalance to the optimal value (i.e. $\Delta = 0$), while across the two lower rows, we have intentionally maladjusted the MZI by inserting free-space elements of 15 and 29 mm, respectively, into one arm ($\Delta = \Delta l/c \sim 5, 10 \times 10^{-11}$).

As predicted, the photodiode output spectra (i.e Fig. 2b) were seen to be independent of the value of $\psi$, that is, the spectra shown in Fig. 2b were stable in time and did not vary if $\psi$ was intentionally varied. In contrast, the optical spectrum viewed from one output port was seen to evolve from a purely odd through to an even mode comb if $\psi$ was not actively stabilised. However, if one locks $\psi$ to a particular value, then the interferometer can stably deliver either the even (or odd) modes of the input frequency comb to output port B1.

The lower two rows of Fig. 2 demonstrate the effect of maladjustment of the gross arm imbalance on the optical and microwave combs. Using the expressions given earlier, we predict that the suppression of the 250 MHz peaks on the microwave comb with respect to the 500 MHz peaks will disappear at an RF frequency of 5 and 2.6 GHz, respectively, for the presented arm imbalances. This is in good agreement with the measured values of $\sim 4.5$ and $\sim 2.5$ GHz derived from Fig. 2b.

The optical spectrum shows corresponding behaviour for these gross time delay errors: we see the production of side-lobes surrounding the central lobe where the output modes have the opposite parity to those in the central lobe. Once again, the experimental data support the analysis by showing a central lobe full width of 8.9 and 5.1 GHz respectively, which is in reasonable agreement with expected values of 10 and 5.2 GHz.

In Fig. 3, we show the measured optical spectrum of the comb through multiple MZI stages: the top panel displays the unmodified raw comb where we are just able to resolve the 250 MHz fundamental comb spacing. Once again the overall comb envelope seen here is associated with the monochromator filter (to prevent multiple order spectra from overlapping in the output) and is not a feature of the comb-splitting technique. In the middle panel, we observe the two outputs of the first stabilised MZI stage with 500 MHz comb spacing (positions B1 and B2 as red and blue respectively on Fig. 1). On the bottom panel, we display the two outputs from the second MZI stage with 1 GHz comb spacing (positions C1 and C2 as red and blue, respectively, on Fig. 1). Here, as on Fig. 2, one sees the effect of an error in the interferometer arm imbalances, which causes the modulation of the comb envelope as well

---

**Fig. 2** Measured optical spectra (a) and microwave spectra (b) at the output of a single MZI stage. The microwave comb is generated by detecting the optical comb with a photodiode while the optical comb is measured by its transmission through a high-resolution optical cavity (linear transmission units). **Top panel** is the output of an MZI with optimally set arm length imbalance ($\Delta = 0$ s) and $\psi = 0$ at position B1 (black) and B2 (green) with a 500 MHz repetition rate; **Middle panel** inserted a 15 mm free-space section to imbalance the interferometer ($\Delta \sim 5 \times 10^{-11}$ s); **Bottom panel** inserted 29 mm free-space section ($\Delta \sim 9.7 \times 10^{-11}$ s). The grey peaks on the optical spectra are auxiliary cw lasers marking a free-spectral range of the optical cavity. The dashed blue and red envelopes on the optical spectrum serve as a guide to the eye. The red and blue background on the photodiode spectra mark the odd and even $\psi$ harmonics, respectively.
as the switching of the odd and even harmonics between the two output ports. Nonetheless, the suppression of the unwanted modes exceeded 12 dB for this set-up—we were unable to measure the full suppression because of the noise floor of the measurement. By careful adjustment of the lengths of the arms (as shown in [22, 23]), it is possible to suppress this modulation. The outputs from the parallel second-stage MZI (positions C3 and C4 on Fig. 1) are offset by 250 MHz from those seen at C1 and C2.

To rotate $\psi$ through one full cycle (2$\pi$), it is necessary to tune the auxiliary laser frequency by one inter-mode spacing as measured at the output of the last interferometer stage (i.e. 1 GHz in our demonstration system). The frequency stability of the auxiliary laser in our case was around 10 MHz which was sufficient to maintain a stable comb output from one of the interferometer ports—we calculate that a 10 MHz error in the setting of this auxiliary laser frequency still allows more than 30 dB of suppression of the unwanted modes of the comb. If higher suppression is necessary, then it is possible to offset frequency lock [31] the auxiliary laser to one of the comb modes giving the approach indefinite stability.

Finally, to demonstrate the effectiveness of our approach, we combined the output of the MZI-cascade with a continuous-wave (cw) laser to generate a heterodyne mixing signal (as shown on Fig. 1). This is displayed on Fig. 4 as a two-dimensional intensity plot where the darkness of a pixel represents the strength of the corresponding Fourier frequency. The five vertical lines on the two plots represent the repetition rate signals for the raw comb (upper panel) and the output of the two cascaded MZI stages. One notes that on the lower panel that the

1 GHz peak is now much stronger than the 250, 500 and 750 MHz signals as a result of the filtering of the cascaded MZIs. As the frequency of the cw laser is scanned (vertical axis), we see multiple beat notes due to the mixing products between the laser and the various modes of the combs. The red circles show areas where the beat note coalesces when $f_{\text{raw}} - mf_r + f_0 \sim nf_r/2$ (for $n \in \mathbb{Z}$). It is in these places where a simple frequency measurement of the difference frequency between the comb and cw laser cannot
easily yield the frequency of the cw laser. It is clear that this situation occurs 4 times less frequently in the rarified optical comb. However, much more important than that simple observation is the fact that our scheme gives us the means to completely avoid any of these degenerate or zero-frequency beat notes. If we observe the beat note between the comb and the cw laser falling within \( f/8 \) of \( f_r \), then we immediately flip the phase of \( \psi \) in the first stage by \( \pi \). This can be done simply by moving the auxiliary laser signal. In this case, the output comb shifts in frequency by exactly \( f/4 \) yielding a new beat note that has a unique interpretation. An alternative technique, which can avoid the need for this switching, is the use of two parallel second-stage MZIs, as indicated in Fig. 1. In this case, all four daughter combs are available continuously at C1–C4, and one just chooses the particular mixing product that provides an unambiguous output.

4 Conclusion

We have demonstrated that phase-stabilised MZIs are capable of separating an optical frequency comb into odd and even modes and delivering these “daughter” combs at separate ports. We have cascaded two of these stages and developed a technique to actively stabilise the MZIs. This approach delivers four daughter combs that are all offset in frequency from each other. We have furthermore explicitly demonstrated the effect of arm length errors on the optical comb and its consequence for microwave combs generated from these pulse trains, and have demonstrated the ability to measure arbitrary frequency signals using these divided combs.

Acknowledgments

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References

The atomic quantum numbers $L$, $S$, and $J$ are related to their quantum mechanical vector operator $\mathbf{L}$, $\mathbf{S}$ and $\mathbf{J}$ [130, 147]. The magnitude of these quantum numbers is given by $|L| = \hbar\sqrt{L(L+1)}$ for electrons orbital angular momentum [130, 147]. Similarly for the spin angular momentum $|S| = \hbar\sqrt{S(S+1)}$, and total electronic angular momentum $|J| = \hbar\sqrt{J(J+1)}$.

In transmission/absorption spectroscopic measurements, the fine energy level structure of the atoms energy levels can be observed. This fine structure is a result of coupling between the outer electrons orbital angular momentum $\mathbf{L}$ and spin angular momentum $\mathbf{S}$ [130] known as spin-orbit interaction. The interaction of the orbital and angular momentums can be thought of in the following classical picture. In the rest frame of the electron, the electron’s orbital motion is mapped onto the rest of the atom so that it appears to orbit the electron. The electric field of the atom produces a magnetic field in the rest frame of the electron. This magnetic field then interacts with the magnetic moment of the electron, caused by the electrons spin, which results in a shifting of the energy level depending of the relative orientation of $L$ and $S$ [147]. This shift is quantified in the total electronic angular momentum $\mathbf{J}$ is given by:

$$\mathbf{J} = \mathbf{L} + \mathbf{S} \quad (C.1)$$

The magnitude of the total electronic angular momentum quantum number $\mathbf{J}$ must lie in the range

$$|L - S| \leq J \leq L + S \quad (C.2)$$

Further energy level splitting occurs when the total electron angular momentum $\mathbf{J}$ interacts with the angular momentum of the nucleus, known as the nuclear spin $\mathbf{I}$. This interaction is called the hyperfine interaction and generates another quantum number called the total angular momentum quantum number $\mathbf{F}$. This splitting is approximately 2000 times smaller than the spin-orbit interaction due to the ratio
of proton to electron masses [147]. The total angular momentum $F$ can take the following possible values:

$$ F = J + I $$

(C.3)

The magnitude of the total angular momentum can vary between the values:

$$ |J - I| \leq F \leq J + I $$

(C.4)

The hyperfine energy level splitting within Rb is clearly visible in spectra of the singly and doubly excited states.
Transit-Time Broadening

The transit-time broadened spectra of an atom with decay, exposed to a uniform optical mode is governed by equation 5.24. The assumptions are made that the atomic populations are not greatly perturbed ($\rho_{g,g}(t) \approx 1$ and $\rho_{e,e}(t) \approx 0$). By using a similar substitution to that made in equation 5.17, a solution to the coherence of the atomic system can be found to be:

$\rho^\prime_{g,e}(t) = -\left(\frac{\gamma_{e,g}}{2} - i\Delta\right)\rho_{e,g}(t) + \frac{i\Omega}{2}(\rho_{e,e}(t) - \rho_{g,g}(t))$  \hspace{1cm} (D.1a)

$\Rightarrow \rho_{g,e}(t) = \frac{i\Omega}{\gamma_{e,g} - 2i\Delta} \left(1 - e^{-\frac{\gamma_{e,g}t}{2} + i\Delta t}\right)$  \hspace{1cm} (D.1b)

Using the derived coherence equations, the absorption coefficient is integrated over the atoms transit time $\tau$, as done in equation 5.15b, to give:

$\langle \alpha(\Delta) \rangle = \int_0^\tau \alpha(t, \Delta) dt$  \hspace{1cm} (D.2a)

$= N \omega h \int_0^\tau \Omega(t) \Im [\rho_{e,g}(t)] dt$  \hspace{1cm} (D.2b)

$= N \omega h \left(-\frac{2\Omega^2 (\gamma_{e,g}^2 - 4\Delta^2)}{\tau I (\gamma_{e,g}^2 + 4\Delta^2)^2} + \frac{\Omega^2 \gamma_{e,g}}{I (\gamma_{e,g}^2 + 4\Delta^2)}
\right.

$+ \left.\frac{2\Omega^2 e^{-\frac{\gamma_{e,g}t}{2}}}{\tau I (\gamma_{e,g}^2 + 4\Delta^2)^2} \left((\gamma_{e,g}^2 - 4\Delta^2) \cos[\tau\Delta] - 4\gamma_{e,g}\Delta \sin[\tau\Delta]\right)\right)$  \hspace{1cm} (D.2c)

As transit time approaches infinity, the spectral absorption should exhibit no transit-time effects. Taking the limit of equation D.2c as $t \to \infty$ results in $\langle \alpha(\Delta) \rangle \to \frac{\gamma_{e,g}^2}{\gamma_{e,g}^2 + 4\Delta^2}$, as expected from a two-level atom as described by the low power limit of equation 4.40 in chapter 4.3.1. This is a Lorentzian absorption profile with a FWHM of $\gamma_{e,g}$, the natural decay rate of the excited state.

For this system, absorption spectra for various transit times are shown in fig...
Figure D.1: Normalised absorption coefficient spectra of an atom passing through a uniform beam which can decay (a) for various transit times. Fractional error in the empirical fit to the FWHM numerical models (b) as a function of the unitless transit time parameter $\tau \gamma_{g,e}$.

There is a substantial difference between these spectra and that with no atomic decay, derived previously and shown in figure 5.1. This is due to the loss of coherence between the ground and excited states due to atomic decay. Hence, spectral artefacts caused by Rabi oscillations and fast intensity changes are less pronounced. It can be seen in figure D.1 that at long interaction times, $\tau = 10$, the spectra tends towards the transition’s natural Lorentzian lineshape.

The FWHM of the absorption profile can not be solved analytically due to the complex nature of equation D.2c. Hence, numerical methods were used to solve the spectral width over a wide range of transit times and natural decay rates. A fit was made to the resultant widths and an empirical formula for the FWHM was derived:

$$\Delta \omega_{\text{FWHM}} = \left( \gamma_{e,g}^{3/2} + \left( \frac{5.56}{\tau} \right)^{3/2} \right)^{2/3}$$

This is a non-linear summation of the FWHM for a uniform pulse with no decay, as derived in equation 5.16, and the natural linewidth of the transition, as defined by its decay rate $\gamma_{e,g}$. 
A Voigt profile is a convolution of between a Lorentzian and Gaussian functions [134]. This profile is commonly encountered when conducting spectroscopy of a thermal vapour. In this case, the Lorentzian component arises from the natural atomic transition, with the Gaussian component arises from the velocity dependent Doppler shifts that follow from the Maxwell-Boltzmann distribution [134].

A general expression for a Voigt profile is given by:

\[
V(\Delta; \gamma, \sigma) = \frac{2\gamma}{\pi^{3/2}\sigma} \int_{-\infty}^{\infty} \frac{e^{-\left(s/\sigma\right)^2}}{4(\Delta - s)^2 + \gamma^2} ds \tag{E.1}
\]

where \(\gamma\) is the Lorentzian’s FWHM and \(\sigma\) is the Gaussian’s \(1/e\) half width. Equation E.1 is defined such that its integral is normalised. Another common mathematical representation for the Voigt profile is:

\[
V(\Delta; \gamma, \sigma) = \Re \left( \text{Erfc} \left( \frac{\gamma - 2i\Delta}{2\sigma} \right) \exp \left( \frac{(\gamma - 2i\Delta)^2}{4\sigma^2} \right) \right) \tag{E.2}
\]

where Erfc is the complementary error function.

As the integral in equation E.1 cannot be evaluated, nor equation E.2 be analytically expanded, the FWHM of a Voigt profile does not have an analytic form. An empirical expression gives a good estimate of the spectral FWHM of the to the ±0.02% level. This is given by [157]:

\[
\Delta\omega_{\text{FWHM}} = 0.54\gamma + \sqrt{0.22\gamma^2 + \left( \frac{4.71}{\gamma_2\sigma} \right)^2} \tag{E.3}
\]


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