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Non-linear Optics Experiments with Rubidium-filled Fibre-optics Structures

Ada A. UMIŃSKA

supervised by

Dr hab. Szymon PUSTELNY

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Uniwersytet Jagielloński

Oświadczenie

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Abstract

This thesis presents fundamental studies on atom-photon interaction inside a hollow-core photonic-crystal fibre (HC-PCF), which was filled with alkali-metal vapour. Linear and non-linear spectroscopy was performed in rubidium atoms introduced into the fibre.

The alkali vapour introduced into the fibre adsorb to the inner-fibre surfaces forming agglomerates. The atoms from the agglomerates can be released by strong non-resonant light coupled into the fibre. Herein, the dynamics of the desorbed atoms was studied with continuous-wave (CW) and pulsed off-resonant desorbing beam, which can be used to induce stable and high atomic densities inside the fibre. A detailed study on atom dynamics inside the HC-PCF was performed.

The cascade-type energy-level system in rubidium atoms was used to perform studies on two-photon absorption (TPA) and electromagnetically induced transparency (EIT). These two phenomena support both fast- and slow-light media within the same energy-level scheme. They also enable dynamic control of the refractive index of the medium.

Negative refractive index changes of $\Delta n \approx -2.37 \times 10^{-7}$ and $\Delta n \approx -6.73 \times 10^{-8}$ were obtained in the TPA system. Additionally, the two-photon damped Rabi oscillations, with two-photon Rabi frequency of $\Omega_R = 15.526(24)$, were observed and characterised.

The effects of EIT and Autler-Townes splitting (ATS) were studied in upper-state driven cascade- and V-type energy level systems. Both revealed broad transparency in a dense atomic medium. In addition, the observed signal was significantly affected by the atoms desorbed from the inner-fibre walls. For the cascade system, the 641(1) MHz and $\sim 60\%$ of contrast temporal state splitting could be

observed.

The use of optical waveguides as an experimental platform enhances the interaction between light and atoms confined within the core. This made them promising hosts for integrated all-optical switches.

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For My Dad

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Chapter 1

Introduction

1.1 Non-linear Optics

Non-linear optics studies interaction between a medium and light, when the optical properties of a material depend on the electromagnetic-field intensity. In this sense, non-linear optics covers broad range of areas of modern optics including: optical parametric and optical harmonic effects, quantum optics, quantum electronics, laser physics, and electro-optics [1, 2]. Non-linear effects can be observed in different media: liquids, gases, solids, i.e., atomic and molecular ensembles, which also offer various applications of the effects.

In this work, weak non-linearities induced in atomic vapours would be considered. Such media supports quantum optical and coherent effects, which will be discussed within the dissertation.

The principles of light-matter interactions for non-linear optics significantly differ from the principles in a linear regime. For example, the frequency of light can be altered while travelling in a non-linear medium by producing light with a summed frequency¹. In the vicinity of an atomic transition, light can be absorbed, which along with spontaneous emission may lead to a significant redistribution of atomic populations. This modifies the optical properties of a medium changing its absorption coefficient and refractive index. Since the refractive index of

¹First observation of non-linear effects (second-harmonic generation) was made in 1961 by P. A. Franken and co-workers [3].

a material starts to depend on light intensity, the speed of light travelling in the medium also depends on the intensity of the light. This provides control over the light propagation. In addition, strong-light interaction can cause the absorption of multiple photons with a total energy matching that of an atomic transition, without the necessity of intermediate states excitation. These main consequences of non-linear interactions draw a very important conclusion: interaction between photons can be induced through interaction with a medium under the non-linear regime.

Non-linear processes in atomic vapours are extensively studied in numerous research groups all over the world. In particular, the ability to modify one light beam by the other, through interaction with non-linear medium, open the possibility of realising all-optical switches and relays for quantum information systems [4], and buffering and regeneration lines for telecommunication applications [5]. Effects such as multi-photon absorption [6, 7], electromagnetically induced transparency (EIT) [8, 9] or four-wave mixing (FWM) [10] allow the modulation of the amplitude, phase, frequency and velocity of light travelling in a medium modified by another strong field.

1.1.1 Example of Non-linear Interactions: Slow and Fast Light

When the frequency of light matches the transition frequency of an atom it can be absorbed, putting the atom into an excited state. Coherent population trapping (CPT) [11, 12] and EIT [8, 9, 12–14] are coherent effects², which are based on quantum interference processes³ [12, 14]. The interference arises when the atoms are being excited by two light beams of different-frequencies in a case when two alternative excitation pathways are present in the system, each of which is characterized with complex probability amplitudes. The coherent nature of the pro-

²The coherent effects relate to the processes that arise from well-defined phase relations within atomic ensemble [15, 16].

³EIT requires strong coupling field on one of two transitions in the considered system, and CPT incorporates two equal-power beams to create coherence between states.

cess allows the two pathways to interfere, leading to the modification of the light absorption. For example, the interference may lead to the cancellation of each excitation resulting in the reduction of the light absorption coefficient. The reduction can be seen as generation of a transparency window for a normally opaque medium.

In the vicinity of an atomic transition, light experiences changes in the refractive index of the medium and those changes strictly depend on light's frequency (*dispersion relation*). For reduced absorption, the medium experiences positive change in the refractive index (*normal dispersion*). This has very important consequences, as the *group velocity* v_g strongly depends on the amplitude of the changes, as well as their sign. Thus, for the gain medium which reflects normal dispersion properties, the light will travel slower than speed of light in vacuum ($v_g < c$). For narrow gain features, obtained with the EIT process, the group velocity becomes significantly reduced and it is even possible to stop the light pulse [17].

In a similar manner, narrow absorption resonances can lead to a significant increase of the group velocity compared with the speed of light⁴. In the most extreme case, the group velocity becomes negative. (A negative group velocity means that the pulse of light will appear on the output of the system at the same time as at its input, as the envelope of the electromagnetic field travels in the opposite direction than the phase⁵). This is caused by the *anomalous dispersion* properties of the absorptive media, i.e., the changes in the refractive index are negative.

There are several applications for slow- and fast-light media, one of them is interferometry. Spectral sensitivity of an interferometer can be significantly enhanced by introducing a slow-light material into one of the interferometer's arms [5]. In the case of a fast-light material being introduced in the interferometer, the spectral sensitivity decreases, however, the sensitivity to changes in the end mirror position is strongly enhanced [19].

⁴This does not mean that the information is carried faster than c , which is explained in Ref. [18]

⁵It can be also seen as a backwards propagation of a pulse in the negative group-index region.

Significant changes in refractive index causes an equivalent phase shift of the interacting light. Those changes can be easily controlled by the optical power of an electromagnetic field, and incorporated as an optical phase shifter [6]. In general, the slow- and fast-light properties of a medium open the possibilities for quantum logic gates [20], optical switches [6, 21, 22] and relays for quantum information applications, and in telecommunication systems for buffering and regeneration of the signal lines [5].

1.1.2 Communication and Quantum Information Applications

Photons travelling in a vacuum do not interact with other photons. At the same time, the electromagnetic field can carry a wide range of frequencies, which make photons good carriers of information. However, processing of the information carried by the photons, i.e., manipulating and encoding the information, can be introduced by interactions in a non-linear media. The non-linear media can be seen as a node of a quantum-computing network, where the computation processes are performed on the information-carrying photons [17].

The interaction of atomic ensembles with laser light are a promising platform for realising quantum logic gates. Coherent effects, like CPT, EIT, and multi-photon absorption, can be used as potential quantum computing tools [17, 23–25]. Information carried by a photon can be read in by careful atomic state engineering, stored in a prepared atomic state, and read out using a control field [17].

The idea of quantum computing comes from Richard Feynman, who proposed calculations on the basic quantum principles during his talk at MIT in 1981 [26]. Since that time, a great interest in building such machines followed the theoretical prediction of the numerous advantages from exploiting quantum principles in logic operations.

The fundamental unit of the information in conventional computing is a *bit*. The state of a bit can be specified by one number at a time, 0 or 1. In traditional computers this can be realised by the transistor, which can be in an *off* or *on* state.

Thus, every number or symbol can be represented by long strings of zeros and ones. The basic operation of classical computer is storing those sequences of numbers in memory (*registers*) and performing simple calculations on them like summing or subtracting. More complex operations are fulfilled by introducing *algorithms*. The calculation is performed in circuits called *logic gates*, which compare numbers stored in registers. An algorithm itself is a sequence of a logic gates, where the output from one gate is fed into the input of the next one.

Quantum information is a next step in computing systems and technology [27]. Numerous research groups are trying to solve technical problems to build systems which will allow performing data operations using superposition states, quantum parallelism, interference, and entanglement [28]. Quantum computers will still have corresponding features to classical machines like: bits, registers, logic gates and algorithms. The difference lies in the ability of perform multi-dimensional computing⁶ [29]. For instance, a classical algorithm can only be run in a sequence, since one bit can be only in one of two states. A two-level particle, following quantum-mechanical principles, can be in both states simultaneously (superposition states). Thus, such quantum systems will play the role of a quantum bit, the so-called *qubit*.

Theoretical predictions assume that quantum computing will have a great potential to beat classical machines in some applications (number factorization [30], database searches [31]) and bring breakthroughs for many still intractable problems in computation and cryptography⁷ [28]. However, there are still many constraints and complexities limiting a quantum systems performance. The main challenge is that the quantum states must be isolated from any interactions with the environment to maintain its state and allow for performing read in, calculation, and read out processes, without demolishing the state itself⁸. Thus, the performance of quantum computers and their advantages over traditional machines are yet unknown. However, the temptation to develop new powerful computing systems,

⁶Multi-dimensional computation would make them unbeatable in factorisation.

⁷Quantum computers will be able to implement the Shore's algorithm [30], a key technology for breaking public-key encryption.

⁸Measurement of the particles state causes a collapse of the entanglement.

has caused numerous research groups and the biggest industrial companies⁹ turn their interest to quantum computers [32–34].

There are many candidates and technologies being considered to realise a qubit. The qubit could be photons enclosed in optical cavities [35], quantum-scale particles like quantum dots and dopants in solids [36], molecules in liquids [37], atoms and trapped ions [38], or superconductors [33, 34]. A 49-qubit quantum computer based on superconducting circuits was introduced by Google last year [34], however, it is not yet determined which technology will become the most efficient and exploited in future quantum computing. Photons seem to be promising, as they are fast and powerful carriers of quantum states (i.e., state of polarization, phase, frequency), however photons themselves are hard to store and manipulate. They require incorporating techniques involving ensembles of atoms for light manipulation and storage. To realise simple operations, like read in, read out, and the storage of information, the atom must usually be prepared in so called *coherent state*.

In this context, the key challenge is to enhance the atom-photon interaction to induce meaningful and controllable changes in the photons' state. Thus, the goal is to induce non-linear interactions of the photon with a coherently prepared atom, which may preserve the full quantum information carried by the photon. To manipulate at the level of a single photon; performing such interactions at very low light level is of crucial importance [4].

Numerous studies were carried out with a single photon interacting with a single atom in high Q-factor optical cavities [27, 35]. Further development of the atom-photon interaction platforms lead to the miniaturisation of the optical cavities [39], and the integration of optical waveguides with the atomic ensembles. The introduction of microstructured optical fibres [40–42], nano-fibres [43–45] and micro-waveguides [46] enabled strong light confinement and its enhanced interaction with the atoms. It is the latter approach that is perused within this dissertation.

⁹Among many: Google, IBM, D-wave systems

1.2 Microstructured Optical Fibres

Breakthroughs in strong atom-photon interactions came with the development of microstructured optical fibres (MOFs). Their unique properties and geometry allows for filling them with a foreign media to enhance the interaction between the medium and guided light. This enables the observation of non-linear effects at the single-photon level [47, 48]. Furthermore, the small size of the fibres makes them suitable for integrated elements (i.e, quantum logic gates, all-optical switches) for quantum-information networks.

MOFs are a novel [49] and still developing area of photonics and atomic optics (when filled with gasses). Their unique optical properties, like endless-single mode guidance [50], high non-linearity [51], anomalous dispersion over a wide range of frequencies [52], and birefringence [51], that can all be modified by changes in their geometry, lead to various commercial and scientific applications [47, 53–55]. In general, MOFs are divided into two main groups. The first consists of solid-core fibres (SCFs), in which a solid core is surrounded by a system of large air-channels along its length. The second group are hollow-core photonic-crystal fibres (HC-PCFs) with an air-core surrounded by a crystal-like structure of air-channels. While the first type transmits light through the effect of *modified total internal reflection* [51], the second supports light guidance based on an interference phenomenon (*photonic bandgap* and *inhabited coupling*). In terms of spectroscopic research, the extraordinary and significant feature of both classes is the possibility to fill the air channels with liquids or gases, and observe the interaction between the filling media and guided light. The high intensity of guided light in the core and a long interaction distance allows ultra-sensitive detection of low concentration substances (at the level of nanomoles) in SCF filled with nanoliter volumes of liquid [56].

In terms of quantum information applications, HC-PCFs have become a promising platform for enhanced atom-photon interactions. Light confined within a small diameter air-core ($5\text{--}50\mu\text{m}$) is focused over long distances and interacts with atoms over entire fibre length. Further studies on the prospects of gas-filled

HC-PCFs for introducing coherent based effects like EIT [57, 58] for slow light and storage light applications [55, 59–61]. This made them promising hosts for integrated quantum-information systems, like all-optical switches [20], phase shifters [6], frequency standards [7, 62] and all-optical delay lines and light storage based on HC-PCF-filled with thermal or cold atomic ensembles.

1.3 Thesis Overview

The great potential of HC-PCFs used for atomic-based coherent interactions with light opens a new area of research. This project undertakes fundamental studies on linear and non-linear light interactions with an alkali vapour confined in a HC-PCF for potential quantum information applications. Unlike previously reported works¹⁰, the main idea was to incorporate a cascade-type energy level system for TPA- and EIT-based studies. Incorporation of those two phenomena allows for studying both fast- and slow-light media within the same energy-level scheme. Thus, the dynamic control and switching between superluminal- and subluminal-light propagation is possible.

Besides the scientific complexity, the experiments described herein are technically difficult. Particularly, loading a μm -diameter fibre core with alkali vapours is challenging. Therefore, the kagome-latticed large-core HC-PCF was used as the experimental platform. Furthermore, to enhance the atom-light interaction while keeping a large core diameter, the hypocycloid-core kagome-structured HC-PCF was exploited [63, 64]. The hypocycloid-core kagome-type fibre has a much smaller effective mode diameter ($\sim 30\ \mu\text{m}$) in comparison to its core diameter ($\sim 50\ \mu\text{m}$). This means that the optical power required for non-linear interactions between guided light and the alkali vapours is significantly reduced due to the strong light confinement. At the same time, the large core area enables more efficient atoms loading into the fibre, and a reduction in light attenuation (this is strictly connected to fibres geometry) [63].

In addition, the proposed project pursues detailed studies of the atomic vapours

¹⁰All the reported studied incorporate V- or Λ -based systems [9, 13].

dynamics inside the HC-PCF, for better understanding and control of the atomic density. Since the inner-surfaces of the HC-PCF are covered with the atomic layer, the introduction of a strong off-resonant optical field causes the release of the adsorbed atoms and a dramatic change in the optical density inside the fibre. This is followed by a variation of the experimental condition during the measurements. However, it can be also use to precisely control the atomic density, and significantly increase the number of interacting atoms. This process of light-induced atomic desorption (LIAD) is well known [65, 66], however, the dynamic control of the atomic densities inside the fibre with a pulsed desorbing laser beam has never been explored.

In this thesis, I describe the fundamental research on atom-photon interaction inside an optical-fibre host. A kagome-type hollow-core photonic-crystal fibre was filled with thermal rubidium vapour and used as a platform for TPA- and EIT-based studies.

This dissertation is organised as follows. Chapter 2, *Spectroscopy of Gases*, describes the theoretical background of linear and non-linear (saturated absorption spectroscopy) two-level atom absorption spectroscopy and combines the theoretical description and the measured rubidium atomic spectra. Further, the description of the HC-PCF chosen for the carried experiment is provided. The vacuum system and optical setup built for the purpose of this project is also described. The second part of the Chapter discusses basic linear spectroscopy in the vacuum chamber and the kagome-latticed fibre. The saturation intensity for the atoms inside the fibre was estimated.

Chapter 3, *Light Induced Atomic Desorption*, provides a detailed description on the LIAD process. The long-term exposure of the atoms inside the kagome-latticed HC-PCF to desorbing light was observed (Section 3.1). The second part of this Chapter, *Pulsed Light Induced Desorption* (Section 3.2), describes the dynamic control over optical depth of the atoms confined within the fibre.

Chapter 4, *Non-linear Spectroscopy*, provides a detailed description of the two-photon absorption (TPA) spectroscopy inside HC-PCF. It shows the theoretical description of non-linear third-order interactions (Section 4.1), as well as the study

of the fast-light medium (Section 4.2). The results of fast light propagation inside a rubidium-filled kagome-latticed HC-PCF are provided (Sections 4.3 and 4.4). In addition, two-photon Rabi oscillations were observed and are shown. Theoretical descriptions of single- and two-photon Rabi oscillations and damped Rabi oscillations are described in Section 4.5.

In Chapter 5, *Coherent Effects in a Three-level Atom*, the effect of EIT in a cascade energy-level system is discussed. The theoretical description of EIT in such energy scheme is followed by the presentation of experimental results obtained in the experimental system.

The whole dissertation is summarised in Chapter 6, *Conclusions*.

The results presented in Chapter 2 and Section 3.1 were obtained in the Department of Photonics of the Institute of Physics, Jagiellonian University in Krakow. In Section 3.2 and Chapters 4 and 5, experimental results acquired during the author's scholarship in the Precision Measurement Group at the Institute for Photonics and Advance Sensing (IPAS) at the University of Adelaide in Australia are presented.

Chapter 2

Spectroscopy of Gases

2.1 Optical Properties of the Atomic Spectra

Linear optics concerns phenomena that occur when the optical properties of a material do not depend on the power of the optical field traversing through the medium [1, 67]. In general, the electric field of light can be defined as

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}) \cos(\omega t + \phi), \quad (2.1)$$

where $\mathbf{E}(\mathbf{r})$ is the spatially-dependent electric field of light, ω is the frequency of the light, and ϕ its phase. This definition can be expressed in terms of positive and negative frequency

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) &= \mathbf{E}(\mathbf{r}) \frac{\exp(i\phi)}{2} \exp(i\omega t) + \mathbf{E}(\mathbf{r}) \frac{\exp(-i\phi)}{2} \exp(-i\omega t) \\ &= \mathbf{E}^{(+)}(\mathbf{r}) \exp(-i\omega t) + \mathbf{E}^{(-)}(\mathbf{r}) \exp(i\omega t), \end{aligned} \quad (2.2)$$

where the $\mathbf{E}^{(+)}$ component is the position-dependent amplitude associated with the positive frequency values ω , and $\mathbf{E}^{(-)}$ is the amplitude associated with the negative frequencies $-\omega$. For further convenience, consider the interaction of the atom in a given location, which allows to eliminate the spatial dependence of $\mathbf{E}$

$$\mathbf{E}(t) = \boldsymbol{\epsilon} E_0 \exp(-i\omega t) + c.c., \quad (2.3)$$

where E_0 is the amplitude of the electric field, and $\boldsymbol{\epsilon}$ is a unit vector determining the light's polarization. The response of light within a medium can be described by

the polarization vector $\mathbf{P}(t)$. This depends on the light according to the relation:

$$\mathbf{P}(t) = \varepsilon_0 \chi \mathbf{E}(t), \quad (2.4)$$

where the proportionality constant χ is the linear susceptibility of the material and ε_0 is the vacuum permittivity. In linear medium, the susceptibility does not depend on light intensity, so that $\chi \approx \chi^{(1)}$ and Eq. (2.4) can be written in the form

$$\mathbf{P}(t) = \varepsilon_0 \chi^{(1)} \mathbf{E}(t). \quad (2.5)$$

In non-linear optics, the response of the medium depends on the lights electric field, so the susceptibility χ is given in the form

$$\chi = \chi^{(1)} + \chi^{(2)} E + \dots, \quad (2.6)$$

yielding

$$\begin{aligned} \mathbf{P}(t) &= \varepsilon_0 \chi^{(1)} \mathbf{E}(t) + \varepsilon_0 \chi^{(2)} \mathbf{E}^2(t) + \varepsilon_0 \chi^{(3)} \mathbf{E}^3(t) + \dots \\ &\equiv \mathbf{P}^{(1)}(t) + \mathbf{P}^{(2)}(t) + \mathbf{P}^{(3)}(t) + \dots, \end{aligned} \quad (2.7)$$

where the constants $\chi^{(2)}$ and $\chi^{(3)}$ are the second- and third-order non-linear optical susceptibilities, respectively [67]. Depending on the light intensity, the strength of the light-medium coupling, and the specific orders of symmetry of the medium, particular orders of the susceptibilities need to be considered. In this Chapter, only linear absorption, and thus linear approximation [Eq. (2.5)] will be considered.

2.1.1 Lorentz Model

The interaction of light with a dielectric medium, such as an atomic vapour or non-metallic solid, are well described within the Lorentz model¹¹ [1, 16]. This model treats atoms as classical driven harmonic oscillators with a damping force. A one-dimensional equation of motion of the electron due to the electric field of light is given by

$$m\ddot{\mathbf{x}} + m\gamma\dot{\mathbf{x}} + m\omega_0^2\mathbf{x} = -\epsilon q E_0 \exp(i\omega t), \quad (2.8)$$

¹¹The Lorentz model is valid for the stationary atoms. The non-stationary case will be further discussed.

where m is the reduced mass of the electron¹², γ is the damping rate, and q is the unit charge. The damping force of the form $m\gamma\dot{x}$ models the classical analogue of spontaneous emission and collisions with the other atoms. Assuming that the general solution of Eq. (2.8) is given by $\mathbf{x}(t) = \epsilon x_0 \exp(-i\omega t)$, one can determine the amplitude of the electron displacement due to the field $\mathbf{E}$:

$$x_0 = \frac{eE_0/m}{\omega^2 - \omega_0^2 + i\gamma\omega}, \quad (2.9)$$

where $-e$ is the charge of an electron. The dipole moment of the atom $\mathbf{d} = -e\mathbf{x}$ is induced by the electric field. If the displacement of the electron (phase shift) is $\delta = \arctan[\gamma\omega/(\omega_0^2 - \omega^2)]$ and the polarization is the dipole moment per unit volume, thus, for atomic vapour with density N the polarization equals:

$$\mathbf{P} = N\mathbf{d} = \epsilon \frac{e^2 N E_0 / m}{\omega^2 - \omega_0^2 + i\gamma\omega}. \quad (2.10)$$

Recalling that the polarization of the medium in a linear regime is defined as $\mathbf{P}(t) = \epsilon_0 \chi^{(1)} \mathbf{E}(t)$, one can rewrite the susceptibility of a medium as

$$\chi = \frac{Ne^2}{m\epsilon_0} \frac{1}{\omega^2 - \omega_0^2 + i\gamma\omega}. \quad (2.11)$$

The total refractive index is a complex number described by the susceptibility in the following manner:

$$\eta(\omega) = \sqrt{1 + \chi} = n(\omega) + i\kappa(\omega). \quad (2.12)$$

The real part of the complex refractive index $\eta(\omega)$ describes dispersion $n(\omega)$ of the dielectric medium (*phase index*) and the imaginary part its absorption $\kappa(\omega)$ (*absorption coefficient*). Further, the refractive index can be rewritten as:

$$\eta = \sqrt{1 + \chi} = \sqrt{1 + \chi' + i\chi''}, \quad (2.13)$$

where χ' corresponds to the real and χ'' the imaginary part of the susceptibility. Expanding $\sqrt{1 + \chi}$ as a power series with respect to χ ,

$$\sqrt{1 + \chi} \approx 1 + \frac{\chi}{2}, \quad (2.14)$$

¹²In a centre-of-mass coordinates in respect to the nuclear mass.

and one obtains that

$$n = 1 + \frac{\chi'}{2}, \quad (2.15)$$

$$\kappa = \frac{\chi''}{2}. \quad (2.16)$$

Finally, substituting Eq. (2.11) into Eqs. (2.16) and (4.8) one gets

$$\eta(\omega) = 1 + \frac{Ne^2}{2m\varepsilon_0} \frac{\omega^2 - \omega_0^2}{(\omega^2 - \omega_0^2)^2 + \gamma^2\omega^2} + i \frac{Ne^2}{2m\varepsilon_0} \frac{\gamma\omega}{(\omega^2 - \omega_0^2)^2 + \gamma^2\omega^2}, \quad (2.17)$$

where

$$n = 1 + \frac{Ne^2}{2m\varepsilon_0} \frac{\omega^2 - \omega_0^2}{(\omega^2 - \omega_0^2)^2 + \gamma^2\omega^2}, \quad (2.18)$$

$$\kappa = \frac{Ne^2}{2m\varepsilon_0} \frac{\gamma\omega}{(\omega^2 - \omega_0^2)^2 + \gamma^2\omega^2}. \quad (2.19)$$

Now the spatial dependence of the electric field can be recalled. For light propagating in the medium with a phase factor:

$$E(z) = E_0 \exp(ikz) = E_0 \exp(i\eta k_0 z) = E_0 \exp(i\Re[\eta]k_0 z) \exp(-\Im[\eta]k_0 z), \quad (2.20)$$

where k is the wavevenumber of the electric field in the medium, and k_0 is the wavenumber in a vacuum. Therefore,

$$n(\omega) = \Re[\eta], \quad (2.21)$$

represents the phase shift of the propagating wave, and

$$\alpha(\omega) = 2k_0 \Im[\eta] \quad (2.22)$$

is a *intensity absorption coefficient*, which describes the attenuation of the field.

The intensity absorption coefficient α is defined by the Lambert-Beer law:

$$\frac{dI}{dz} = -\alpha I. \quad (2.23)$$

Integrating Eq. (2.23) over z , one gets:

$$I = I_0 \exp(-\alpha z). \quad (2.24)$$

Significant absorption occurs for small detunings, where $|\omega_0 - \omega| \ll \omega_0$, thus $\omega_0^2 - \omega^2 \approx 2\omega(\omega_0 - \omega)$. As a result, the phase index n , and the absorption coefficient α , can be expressed in terms of the susceptibility in the following manner:

$$n(\omega) \approx 1 + \frac{Ne^2}{2m\epsilon_0} \frac{(\omega^2 - \omega_0^2)/2\omega}{(\omega - \omega_0)^2 + (\gamma/2)^2} \quad (2.25)$$

$$\alpha(\omega) \approx \frac{Ne^2}{m\epsilon_0 c \gamma} \frac{(\gamma/2)^2}{(\omega - \omega_0)^2 + (\gamma/2)^2}. \quad (2.26)$$

The classical absorption coefficient is expressed as a Lorentzian absorption profile with a full width at half maximum (FWHM) γ and a resonant absorption coefficient $\alpha_0 = Ne^2/m\epsilon_0 c \gamma$. The phase index gives the dispersion profile, i.e., the dependence of the refractive index on the light frequency. Both absorption and dispersion profiles are presented in Fig. 2.1.

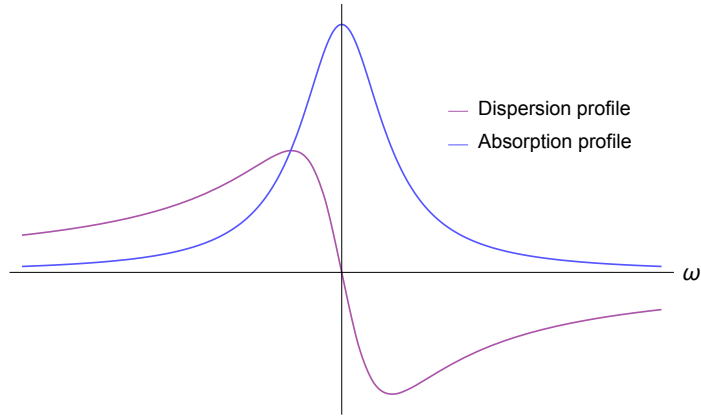


Figure 2.1: Shape of the absorption coefficient and phase index profiles described by Eqs. (2.25) and (2.26). The absorption is derived from the real part, and the phase index from the imaginary part of Lorentzian profile.

2.1.2 Doppler Broadening

The actual linewidth of a given transition of a gas observed in laser spectroscopy is broadened due to inhomogeneous Doppler broadening. This broadening is caused by the Maxwell velocity distribution of the atoms.

If an atom is irradiated by the laser frequency ν_L , within its moving frame the

atom will experience a Doppler-shifted laser frequency

$$\nu = \nu_L \left(1 - \frac{v_z}{c} \right), \quad (2.27)$$

related to the z -component of the atom's velocity v_z (where z is light propagation direction). Thus, if the laser is scanned across an atomic transition of the frequency ν_0 , the class of atoms with velocity v_z will absorb the light if the following condition is fulfilled

$$\nu_L = \frac{\nu_0}{1 - \frac{v_z}{c}}. \quad (2.28)$$

The velocity distribution of atoms in vapours is described by the Maxwell-Boltzmann statistics:

$$p(v_z) = \sqrt{\frac{m}{2\pi k_B T}} \exp \left(-\frac{mv_z^2}{2k_B T} \right), \quad (2.29)$$

where k_B is the Boltzmann constant, T is the absolute temperature of the vapours, and m is the atomic mass. In turn, the relative number of atoms which are resonant with the laser frequency follows the normal distribution:

$$N = N_0 \sqrt{\frac{m}{2k_B T}} \exp \left[-\frac{mc^2}{2k_B T} \left(\frac{\nu_0 - \nu_L}{\nu_0} \right)^2 \right], \quad (2.30)$$

where N_0 is the total number of atoms. The FWHM of the above Gaussian distribution related to the Doppler broadening, is given by:

$$\Delta\nu_D = 2\frac{\nu_0}{c} \sqrt{2 \ln 2 \frac{k_B T}{m}}. \quad (2.31)$$

To include Doppler broadening in the theoretical model, the actual linewidth is described by the convolution of Lorentz and Gaussian functions (as each atom experiences homogeneous broadening given by a Lorentz profile and a Doppler frequency shift characterised by a Gaussian distribution for atomic ensemble). This convolution is called a Voigt profile, which includes both: the natural linewidth and any inhomogeneous broadening.

2.2 Rubidium Vapours

Alkali metals are found in group *one* (s-block) of the periodic table. They possess one outermost electron in the s-orbital. This makes them similar to the hydrogen atom, and thus easily theoretically approximated by a hydrogen-like solution. Rubidium's hydrogen-like approximation, and absorption in a convenient spectral range, enabling their manipulation with light (and other fields), make them attractive in spectroscopic research.

The single valence electron makes alkali vapours highly reactive at room temperature. Thereby they readily lose their valence electrons to form salts. Thus, they are not abundant as free elements in nature, which means that spectroscopic research that utilises alkali requires the use of glass cells or the construction of vacuum systems filled only with desired vapours (or potentially other inactive gases such as noble gases or molecular nitrogen).

Rubidium atoms, i.e., the alkali used in research presented within this dissertation, have the atomic number 37 and an electron configuration $[\text{Kr}]5s^1$. There are two naturally occurring stable isotopes of rubidium; 85 (with abundance 72.17 %) and 87 (abundance 27.83 %). The valence electron in either of the isotopes can be addressed with near-infrared (NIR) radiation with a frequency of ~ 800 nm. In spectroscopic nomenclature, the D line is the transition between the ground state and the first excited state in every alkali atom. Due to the fine splitting in rubidium, one must distinguish the D_1 and D_2 lines. The D_1 line corresponds to the transition from the ground state $5^2S_{1/2}$ to the $5^2P_{1/2}$ state, and D_2 from the $5^2S_{1/2}$ to $5^2P_{3/2}$ state. Their wavelengths are 795 nm and 780 nm, respectively.

In this work, excitation at the rubidium's D_2 line, in both naturally abundant isotopes is considered. A detailed energy-level scheme of the D_2 line and a corresponding saturated absorption spectra are shown in Fig. 2.2.

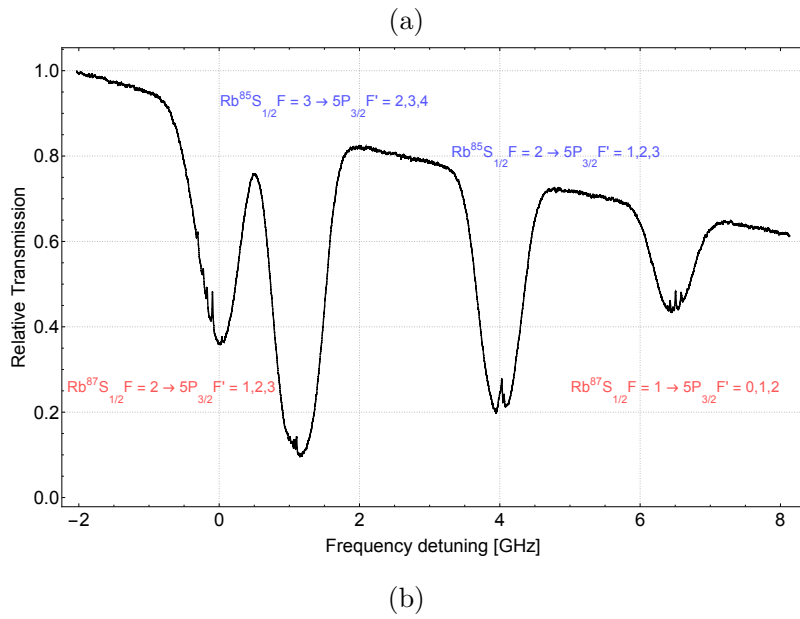
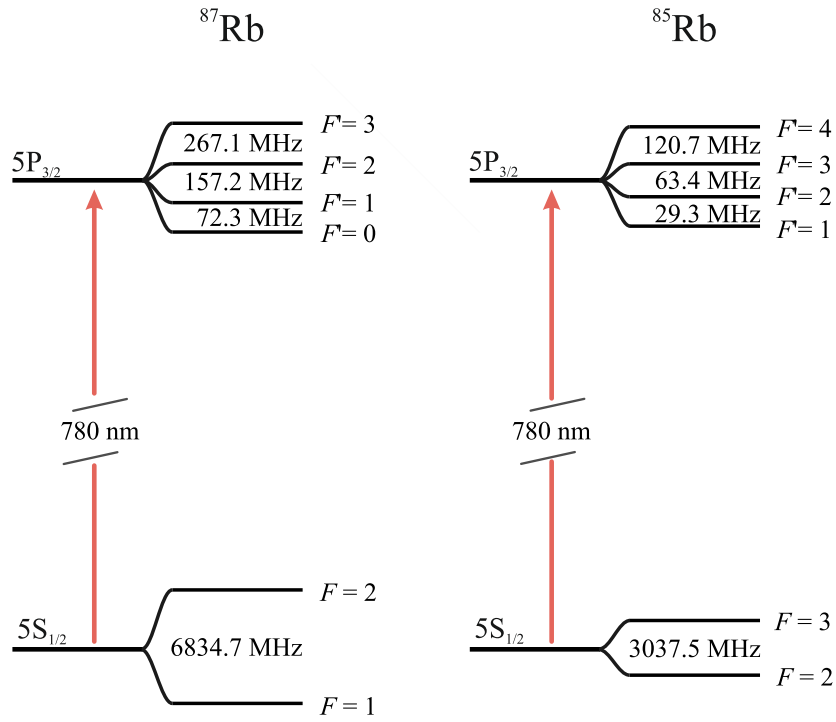


Figure 2.2: (a) Energy level structure and (b) corresponding saturated absorption spectrum of the rubidium D_2 line of two stable isotopes, ^{87}Rb and ^{85}Rb . Red and blue labels mark ^{87}Rb and ^{85}Rb transitions, respectively.

Saturated Absorption Spectroscopy

Saturated absorption spectroscopy (SAS) is a Doppler-free non-linear technique, where two counter-propagating laser beams, strong and weak, at identical frequencies, interact with the atom ensemble. If one of the beams interacts with the atoms of particular velocity v_z , the other beam will interact with the group with velocity $-v_z$. However, at resonance, both beams interact with the same velocity-class atoms ($v_z = 0$). Since the strong beam is depleting atoms from the ground state, the weak probe beam experiences decrease in its absorption. This is seen as a *dip* in the otherwise Doppler-broadened spectrum (see Fig. 2.2b). Some of the dips come from the so-called cross-over resonances, when the frequency of the laser is set exactly half-way between two transitions [68, 69].

The natural width of the D₂ absorption line is 6.066 MHz for both isotopes. The absorption spectra is Doppler broadened, as the non-stationary atoms experience the Doppler effect (Fig. 2.2b). In turn, the transitions are seen as four Voigt profiles, where each of those profiles consist of three hyperfine transitions. In the presented spectra, the hyperfine structures of the 5P_{3/2} state can be seen as a ‘holes’ in the broadened spectra as the SAS technique was applied (see Fig. 2.2b).

2.3 Microstructured Optical Fibres

Microstructured optical fibres (MOFs) were first introduced by P. S. J. Russell et al. in 1996 [49]. Their unique structure possesses inclusions formed by air-channels introduced along their entire length¹³ (Fig. 2.3). The MOFs are divided into two main groups. The first group is solid-core fibres (SCFs), where light is confined in a solid-core surrounded by large air-channels forming the fibres cladding [49, 51, 53]. An example of SCF is shown in Fig. 2.3a. The second group consists of fibres with a cladding structure of air-channels surrounding an air-core (called hollow-core photonic-crystal fibres (HC-PCFs)). An example of HC-PCFs is presented in Figs. 2.3b to 2.3d. Both MOF types exhibit novel and interesting

¹³There is also another group of MOFs, all-solid microstructured optical fibres, where different index-of-refraction glass is used to form inclusions instead of air-channels [70].

properties including, endlessly-single mode guidance [50], high non-linearity [71], anomalous dispersion [72], easily generated large birefringence [73], all strictly related to their geometry. Another important feature of the MOFs is the possibility of filling their air channels with foreign media (gases or liquids) [47, 53]. This enables studies of light-atom interaction under unique experimental conditions. Below, different types of MOFs, used in spectroscopic measurements of gases and liquids, are reviewed.

Suspended-Core Fibre

Suspended-core fibres possess small (diameter of approximately $1\text{ }\mu\text{m}$) solid cores, which are suspended on thin (width of 100 nm) struts and surrounded by large air-channels (see Fig. 2.3a). These types of MOFs support a wide range of wavelengths (in general, white light). Spectroscopic measurements can be performed by using the guided light that penetrates into the air channels in the form of an evanescent field. This makes them a perfect platform for a variety of applications in biological and chemical sensing [53, 56, 74]. They also have a high non-linear index, which can be increased by doping the glass, and customizable zero-dispersion region [52]. These properties make them perfect for non-linear optics applications such as laser amplifiers [75], and as a highly non-linear medium for super-continuum generation [54].

Hollow-Core Photonic-Crystal Fibre

The light guidance mechanism for HC-PCFs is based on an interference phenomenon. A photonic-crystal structure, which is formed by air-channels in a fibre cladding (see Fig. 2.3b), can be approximated as a two-dimensional Bragg mirror. They allow for selective reflection, where the reflected wavelengths interfere destructively inside the cladding structure and constructively on the incident core side. As a result, reflected wavelengths are confined inside the air-core, despite the fact it has a lower refractive index than the cladding. This type of guidance is called photonic-bandgap guidance (PBG).

Due to the nature of guidance, HC-PCFs exhibit narrow transmission band-

widths (~ 70 THz) and low attenuation, typically 200 dB/km at 800 nm. In addition, strong interactions between the guided light and a filling medium over long distances can be achieved. This makes them potential platforms for hosting gases, particularly atomic vapours, and applications in atomic optics [76], such as non-linear spectroscopy [47] and frequency metrology [77]. HC-PCFs open new possibilities for many photonic and atomic optics applications. However, their small air core (typically $6 - 8 \mu\text{m}$) makes loading the fibres with a low-pressure atomic vapour (e.g., rubidium) slow and difficult [78]. Furthermore, the large surface to volume area enhances the atom-surface interaction, i.e., adsorption of the alkali atoms to inner-fibre walls, highly limiting the atomic spectroscopy performance.

Kagome-structured HC-PCF

Kagome-structured HC-PCFs are another class of HC-PCFs, introduced in 2002 [79]. Figures 2.3c and 2.3d present two types of kagome-structured HC-PCFs: a 7-cell hypocycloid-core, and a large-pitch single-cell, respectively. The great advantage of kagome-structured fibres over PBG HC-PCFs, is their large air-core (greater than $35 \mu\text{m}$) and broadband guidance (typically $600 - 1700$ nm) [63, 80]. Unlike PBG HC-PCFs, kagome-latticed fibres exhibit a low density of photonic states over a broad bandwidth [64], this means that no bandgaps are present. All cladding modes, where the effective refractive index n_{eff} is close to the core-mode refractive index value are localized mainly in the glass struts where they exhibit rapid field decay at the silica-air surface, followed by a fast phase oscillation. The fast oscillation of the transverse-field component does not match the slowly phase-varying field of the core mode of the same n_{eff} value [64]. Geometrically perfect cladding, for low loss and broadband guidance, would require a thin glass web with no sharp bends to increase the transverse mismatch between core and cladding field¹⁴. The described guidance mechanism is based on an effect called *inhabited coupling* [63, 64], which was first described by J. von Neumann and E. Wigner (*Neumann-Wigner bound states in continuum*) [81].

¹⁴Typically, the thickness of the lattice glass webs are approximately 300 nm, which gives the limitation on shorter wavelengths (cut-off at the 600 nm wavelength).

The large air-core of kagome-latticed HC-PCFs and their broadband light guidance makes them more suitable for atomic spectroscopy applications than the PBG HC-PCF. Furthermore, the introduction of hypocycloid-core kagome-structured HC-PCF, allowed for lowered attenuation limitations of 70 dB/km over a spectral range of 750 – 850 nm [78]. The light in a hypocycloid-core kagome-latticed fibre is confined between the most inward cups of the cladding (see Fig. 2.3c), thus, the interaction between core and cladding modes is diminished [63]. This means that the effective mode diameter is narrowed with respect to the core diameter (see Table 2.1). This reduction of the effective mode area not only allows for significant guidance-loss reduction, but also leads to an enhancement of light-vapour interaction for large-core fibre.

All the above mentioned properties make the kagome-latticed HC-PCFs the most suitable MOFs for non-linear effects in rubidium-filled HC-PCF. In the work presented in this dissertation, both types of kagome-structured HC-PCFs, depicted in the Figs. 2.3c and 2.3d, were utilised.

In this Chapter, I will only concentrate on 7-cell hypocycloid kagome latticed fibre¹⁵ presented in Fig. 2.3c. For further convenience it will be called *Fibre 1* (optical properties of both fibres are provided in Table 2.1).

Light Coupling

A simple experimental setup was built to examine light-coupling conditions to Fibre 1 and to find suitable coupling lenses. The light was provided from an external-cavity diode laser (ECDL) operating at 780 nm. Two 3-axis micrometer-precision stages¹⁶ were used to couple light to and from the fibre. Transmitted light was observed on an optical power meter to estimate the transmission through the fibre, and CCD camera beam profiler¹⁷ to check the coupling quality.

Light coupling to Fibre 1, i.e., the fibre with relatively large core diameter and small numerical aperture (NA) (see Table 2.1), requires a proper lens or microscope

¹⁵GLOphotonics: PMC-C-650/20007C

¹⁶Thorlabs: MBT616D/M

¹⁷Thorlabs:BC106N-VIS/M

	7-cell hypocycloid kagome Fibre 1	Single-cell kagome Fibre 2
Core Diameter (μm)	49	45
Mode Diameter (μm)	31(1)	–
M^2	1.30(5)	–
Numerical Aperture	0.02	–
Spectral Range (nm)	650 – 1400	600 – 1700

Table 2.1: Properties of the kagome-type HC-PCFs used in the experiment [63, 80]. Some of the values for the single-cell kagome-latticed HC-PCF were not known or determined. M^2 is a beam quality factor, which represents the degree of variation of a laser beam from the perfect Gaussian shape [67].

objective to achieve high efficiency. The highest efficiency of coupling (60%) was achieved with 5x microscope objective¹⁸, which has low NA (0.1) and long focal length (25.4 mm). From the point of view of this project, the large operating distance of a microscope objective is of high importance in terms of coupling light to the fibre through the vacuum viewport.

The far-field mode profile of Fibre 1 is presented in Fig. 2.4. The 2D and 3D far-field images of light leaving Fibre 1 when placed in a vacuum chamber, are depicted in Figs. 2.4b and 2.4c, respectively. In free space the mode image is clear (see Fig. 2.4a), in the vacuum chamber additional cladding air-gap coupling occurred. This is due to the deformation of the light mode by the vacuum viewport. Great care was taken to reduce any additional coupling, however, it was impossible to fully cancel it. Therefore, during the experiment the imaged area was reduced to the core image with an iris.

¹⁸Newport M-5X microscope objective

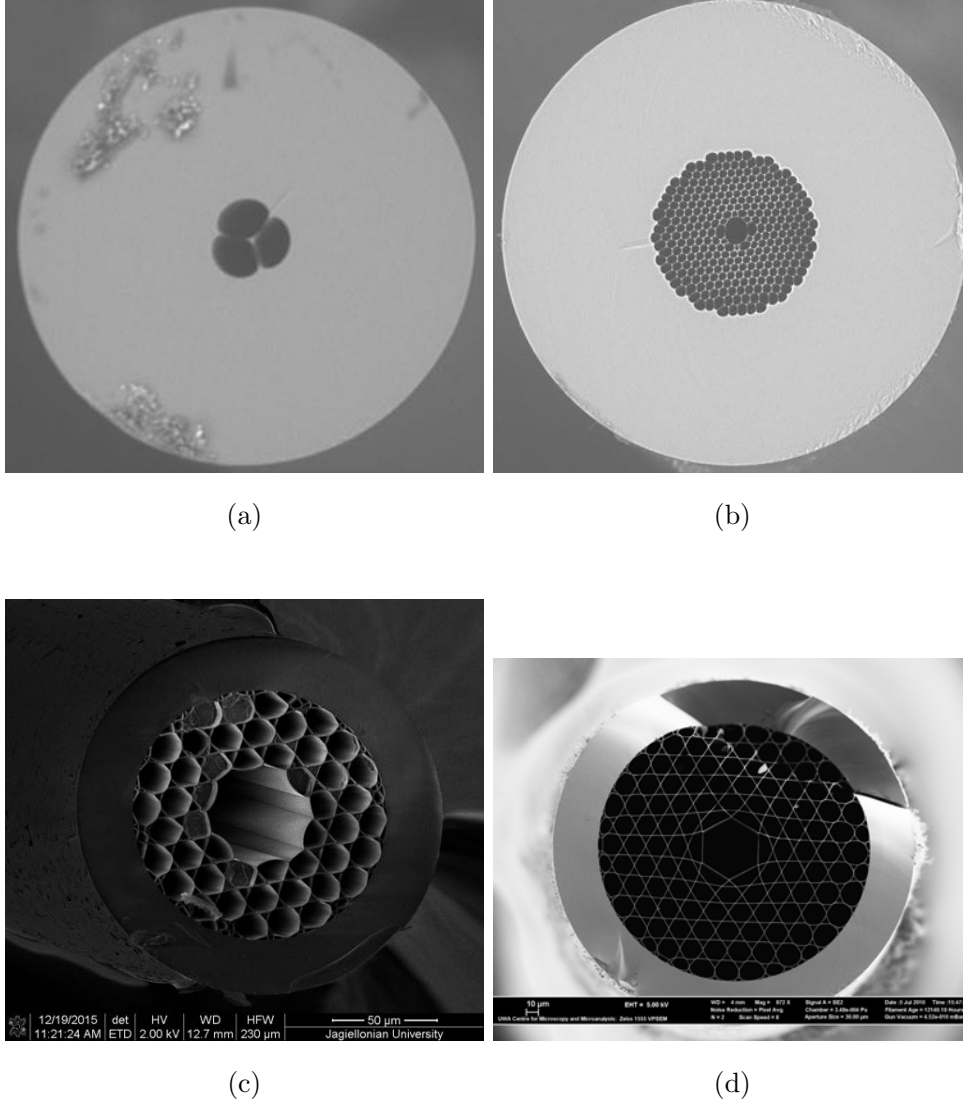


Figure 2.3: Scanning-electron micrographs of the different types of MOFs: (a) a solid-core fibre or suspended-core fibre, in which the small solid core (diameter $\sim 1 \mu\text{m}$) is suspended on thin glass struts (width of 100 nm) and surrounded by large air-channels forming a cladding, (b) a hollow-core photonic-crystal fibre, whose cladding, formed by air inclusions, encloses discrete wavelengths of light inside the air core (diameter of $6 \mu\text{m}$), (c) a 7-cell 3-ring hypocycloid-core kagome-structured HC-PCF (Fibre 1), and (d) a single-cell kagome-structured HC-PCF (Fibre 2) the latter two were used in experiments. The core diameters are $\sim 49 \mu\text{m}$ and $\sim 45 \mu\text{m}$, respectively.

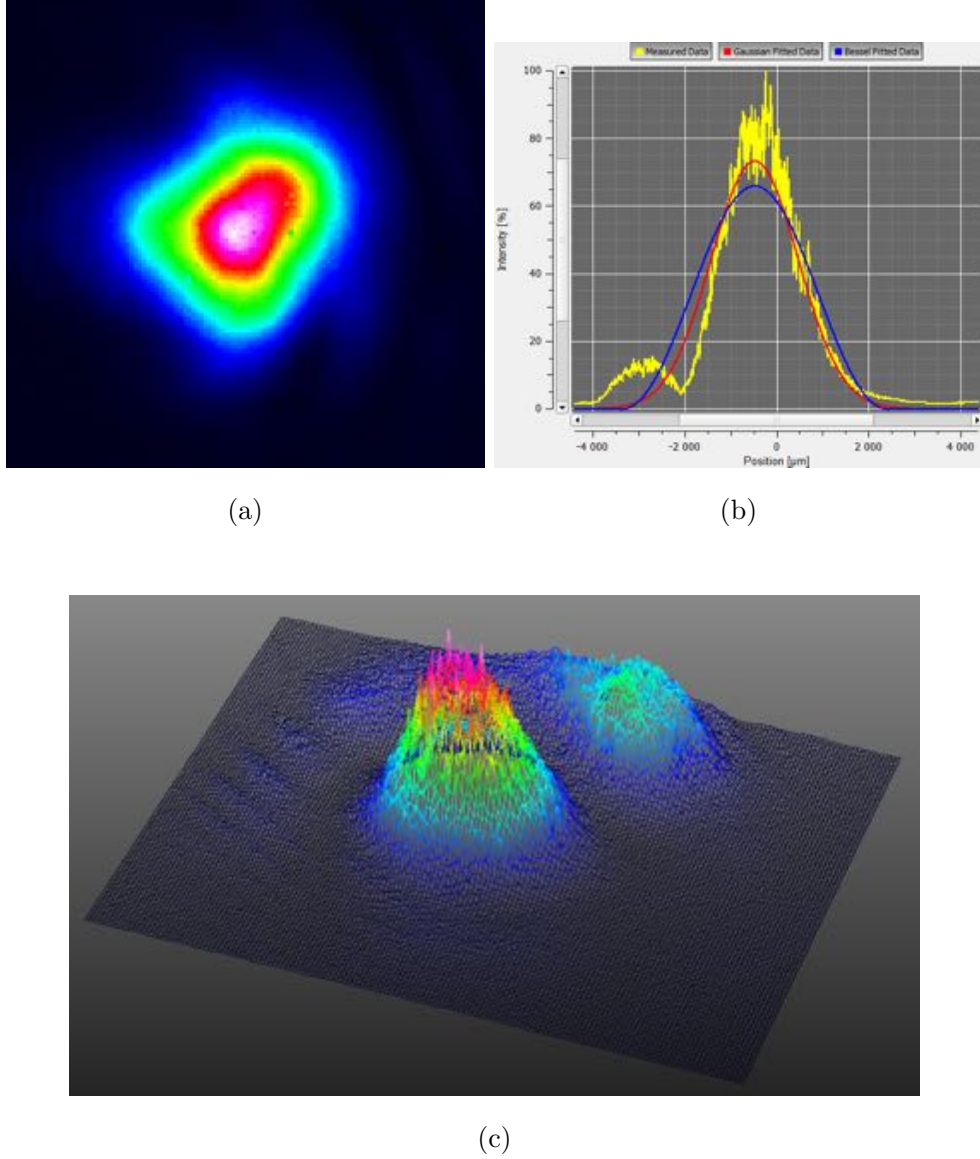


Figure 2.4: CCD camera beam-profiler images of the far-field mode profile of a 7-cell 3-ring hypocycloid-core kagome-structured HC-PCF (Fibre 1). (a) The fundamental-mode profile observed in free space. The colours indicate the intensity of light. (b) The x -projection of the profile of the fundamental-mode, observed in vacuum, with Gaussian and Bessel function fits. (c) A 3D profile of the fundamental and additional cladding modes in the vacuum chamber, where light is coupled through the glass viewport. The obtained mode profiles exhibit a Gaussian shape, however, it was easy to accidentally excite additional modes in the cladding air-channels while placed in the vacuum chamber.

2.4 Experimental Challenges

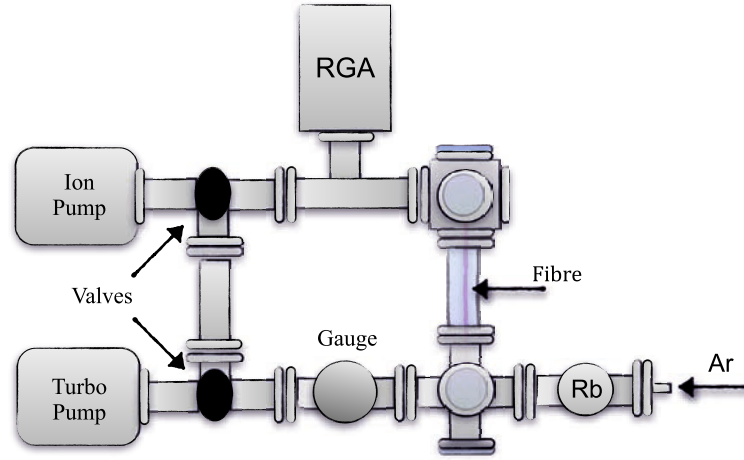
2.4.1 Vacuum Setup Design

The use of rubidium vapour requires an high vacuum environment. This is implied by the high reactivity of rubidium. Therefore, a significant part of this PhD was dedicated to redesigning and reconstruction of experimental setup existing in the Department of Photonics at the Institute of Physics at the Jagiellonian University, and the replacement of previously used fibre within a new waveguide. The previous setup is described in the PhD dissertation of Michał Grabka [82]. Since a lot of details regarding the setup is described in Ref. [82], herein I will only focus on the parts that were replaced or rebuilt for studies using rubidium vapour.

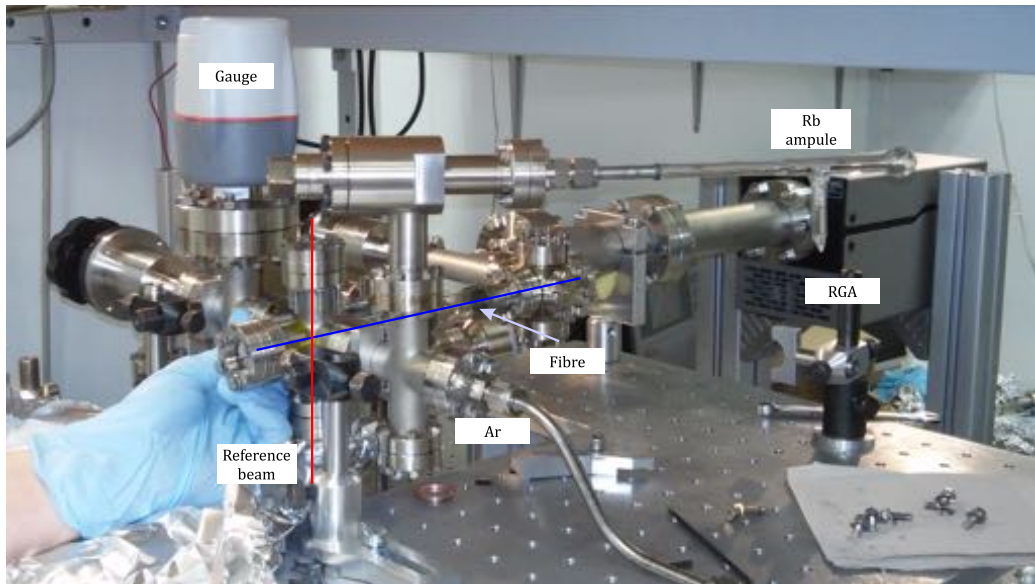
The initial setup was designed for operation with long (~ 30 cm) standard HC-PCF [82]. The original fibre had a core of $8\text{ }\mu\text{m}$ diameter. Unfortunately, with such a fibre no rubidium atoms were observed inside the air-core. This was the main reason for redesigning the vacuum chamber and changing the fibre to the kagome-type HC-PCF with a larger core.

The scheme of the vacuum system is depicted in Fig. 2.5. The rubidium-host fibre was the main part of the setup. It was placed between two independent vacuum chambers. The only connection between the chambers was a fibre air-core. The waveguide was placed inside a glass-tee to protect it from any damage. The glass-tee also allows for external fibre observation. The first of the chambers (*Chamber 1*), built with CF16 flanged six-way cross, was connected to a rubidium ampoule and a pure (99.995%) noble-gas reservoir (Ar). The rubidium ampoule was placed in a specially prepared glass cell [82], connected to the chamber through an all-metal valve¹⁹. The argon was used as a buffer gas during the experiment. A noble gas was provided, from the gas bottle, to the first chamber through a *Swagelok* valve system, connected with non-magnetic soft stainless steel tubes. The averaged residual oxygen value in 99.995% pure argon varies between 5-10 ppm, which is enough to oxidise desired vapour pressures ($10^{-6} - 10^{-5}$ mbar at $\sim 50^\circ\text{C}$).

¹⁹Hositrad: Valve All Metal Flange NW16CF



(a)



(b)

Figure 2.5: (a) Schematic of the vacuum setup. The kagome-type HC-PCF is glued into the flange, which separates the two chambers. Rubidium atoms and a buffer gas (Ar) were introduced only to the first chamber. Both chambers were equipped with viewports enabling light coupling (decoupling) to (from) the fibre. The second chamber was connected to the residual gas analyser (RGA). Both chambers were connected to turbomolecular and ion pumps. (b) A picture of the vacuum setup.

For that reason, additional molecular and gas²⁰ filters were used (see Fig. 2.6). The valve system was introduced to reduce the gas pressure provided from the bottle. The metering valve allowed for a precise gas dosing. The wide range gauge in the first chamber was calibrated to measure the pressure of the argon gas.

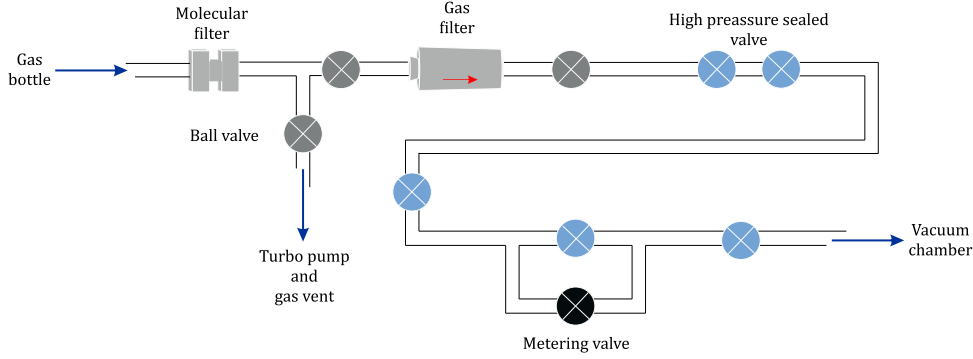


Figure 2.6: Noble gas setup. The initial argon pressure (right after the bottle) was controlled by the gas reductor. Further, the gas could be either directed to the turbomolecular pump, or released from the metal tubes through a ball valve. Then the gas was purified with molecular and gas filters. To ensure that the proper gas flow was provided, two valves before and after the gas filter were introduced. Next, high pressure valves were used to reduce the gas pressure before introducing argon into the vacuum chamber. Argon could be dose with a meter valve [82].

Both of the chambers were equipped with a set of viewports to enable light coupling (decoupling) to (from) the fibre, and for reference spectral measurements. To analyse and diagnose any impurities or leakages, the residual gas analyser (RGA)²¹ was placed directly into the second chamber. RGA is a compact mass spectrometer, mainly used to examine the quality of vacuum systems. The analyser ionizes molecules and measures them according to their molecular mass [83]. Both chambers were connected to the turbomolecular pump station²², with a pumping speed 61 l/s²³. The pump station was equipped with an active wide range gauge²⁴

²⁰Saes Pure Gas: MC1-902F

²¹SRS: RGA 200 amu

²²Edwards: T-Station 75

²³measured for the nitrogen

²⁴Edwards: WRG-S NW25

for pressure determination. The ion pump²⁵, with a pump speed 20 l/s²⁶, was connected close to the second chamber, and when all the valves were opened it allowed for pumping both chambers simultaneously. Since the ion pump is able to attain low pressures, this configuration permitted lowering the pressure to desired 10^{-7} mbar.

Standard CF16 flanged stainless steel parts with copper gaskets were used to build the vacuum setup. All the metal parts were washed in isopropanol using an ultrasonic machining process. During construction of the setup, the due care was taken to reduce all possible impurities (e.g., finger prints) that might caused further out-gassing in the system.

2.4.2 Preparation of the Fibre

The kagome-structured HC-PCF, Fibre 1, was stripped of its polymer coating at both ends and precisely cleaved with a diamond cleaver to approx. 24 cm. Next, the fibre was flushed with isopropanol and a streamed a noble gas, and glued to the flange using ultra-high vacuum adhesive glue (see Fig. 2.7a). The prepared fibre was then placed in the oven at 150°C for three days. Once the glue stopped out-gassing and hardened, the fibre was placed in the vacuum setup. The cleaved ends of the fibre were supported by specially designed holders, which are visible in Fig. 2.7b [82].

The whole vacuum setup was flushed with a noble gas and preheated for almost two weeks at 150°C to release all remaining impurities from the inner walls of the vacuum elements and fibre coating. For the first few days, only the turbomolecular pump was operating. When the chamber pressure reached value of approximately 10^{-6} mbar, the ion pump was turned on. When the pressure of the chamber stabilised and did not change for a few days, the temperature was lowered to 60°C. After that, pressure in the setup, according to the gauge in the first chamber and the ion pump indication, dropped to about 7×10^{-8} mbar. At this point, the first chamber was closed, i.e., both pumps were only pumping the second chamber,

²⁵Gamma vacuum: Titan 25S

²⁶The ion pump was still able to pump noble gas, but with a lower efficiency.

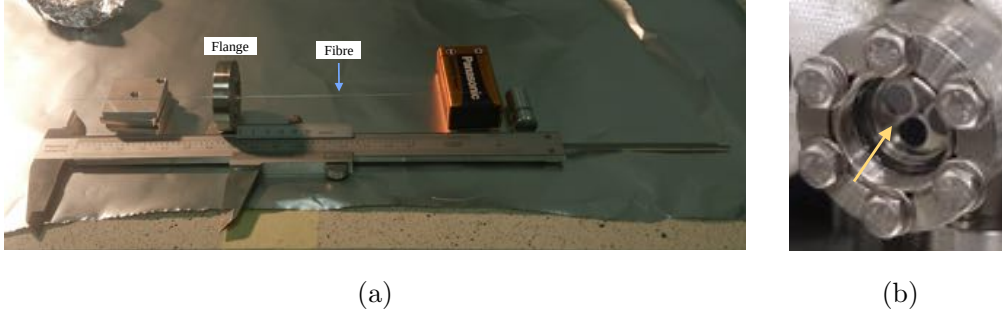


Figure 2.7: (a) Picture of the kagome type HC-PCF glued to the vacuum flange. (b) Fibre holder mounted inside the vacuum chamber (indicated with the arrow).

and the rubidium ampoule was open. To create a large atom density, the glass part with the ampoule was held at 90°C , while the whole chamber was held at approximately 60°C . To prevent rubidium from fully escaping from the ampoule, the alkali atom reservoir was kept at a lower temperature.

2.4.3 Experimental Setup

Lasers

There were two diode lasers used in the experiment. A commercial ECDL²⁷, and a home-built high-power laser.

Light from the ECDL was scanned over the rubidium D_2 line (780 nm). The side output of the laser, delivering 10% of the total optical power, was sent to a SAS reference cell and a wavelength stabilisation setup (mini-dichroic atomic vapour laser lock [84]). The main output of the laser was split into two beams on a beam splitter (BS). One of them (a probe beam) was coupled to Fibre 1. The second fraction of the light was directed to the side viewports of the vacuum chamber to perform SAS reference spectroscopy inside the chamber. The reference beam was reflected back and detected using home-build Si photodetector with a fixed gain. The probe beam was double passing an acousto-optic modulator (AOM)²⁸ for further phase-sensitive detection performance and coupled

²⁷Sacher Lion

²⁸Isomet: 1205C-2

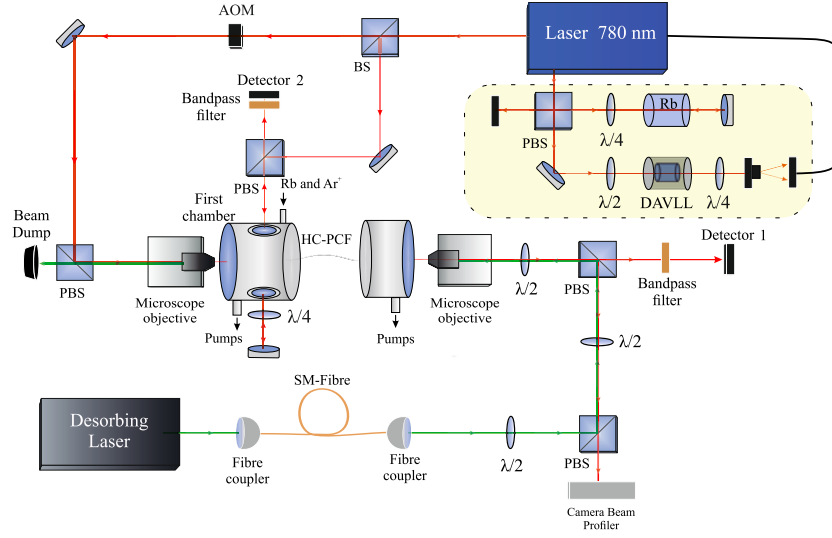


Figure 2.8: Schematic of the optical setup for spectroscopy of the rubidium vapour filling Fibre 1. The main fraction of light was split into two beams on the beam splitter (BS), one of the beams was sent to the vacuum chamber. This beam worked in a retro-reflecting mode, such that the arrangement could be used for SAS of the gas filling the vacuum chamber. The other fraction of the light, after passing an acousto-optic modulator (AOM), was coupled to the HC-PCF using the $5\times$ microscope objective. At the other end of the fibre, the light was decoupled with an identical microscope objective. The half-wave plate ($\lambda/2$) was used to maximize the intensity of light directed to the photodiode (Detector 1). Signal measured at Detector 1 were sent to a lock-in amplifier (not presented here). Some fraction of the light was divided onto a polarising beam splitter (PBS) and sent to a CCD Camera Beam Profiler for fibre-mode observation. An additional diode laser was used to desorb atoms from the inner fibre walls. Desorbing light was coupled to a single-mode fibre (SMF) for spatial filtration of the mode, and further coupled to the Fibre 1 in a counter-propagating manner (with respect to the main beam). Separation of the desorbing and probe beams was maximised with adjustment of the counter polarisation ration. A bandpass filter was used to prevent any reflection of the desorbing beam towards the photodiode. Light from the external-cavity diode laser (ECDL) was scanned over the D_2 line, while part of the light was directed to the SAS reference cell and laser locking setup (mini-DAVLL).

(decoupled) to (from) the Fibre 1 with the Newport 5 $\times$ objective microscope mounted on a 3-axis micrometer-precision stage. Transmission through the fibre was $\sim 60\%$ in a free space which did not change after placing the fibre into the vacuum chamber. Decoupled light was detected by a Si amplified photodetector²⁹. To ensure that the fundamental mode of the fibre was excited, a CCD Camera Beam Profiler was used.

A home-built high-power (100 mW) diode laser³⁰, operating at 800 nm, was used as a desorbing light source. Since it was not necessary to narrow the lasing bandwidth, an external cavity was not introduced. The laser beam was collimated and coupled to the SMF to spatially filter the beam shape and provide high efficiency coupling into Fibre 1. A commercial current and temperature controller³¹ was used.

The desorbing laser was coupled into Fibre 1 in a counter-propagating manner with respect to the probe laser. Beam separation was performed by setting their polarisations orthogonal. In addition, to prevent Detector 1 from measuring any additional reflections from the high power desorbing laser, a bandpass filter (centred at 780 nm wavelength)³² was used.

Dichroic Atomic Vapour Laser Lock

The dichroic atomic vapour laser lock (DAVLL) was used to precisely stabilize the light emitted from the ECDL at a desired wavelength. This system exploits magnetically-induced circular dichroism of atoms to create an error signal crossing zero in the vicinity of an atomic transition [84]. The mini-DAVLL setup consisted of a small rubidium cell (~ 20 mm long) placed between two magnets, which generated a relatively homogeneous magnetic field. For minimizing the leakage of the magnetic field outside of the system, the cell was placed in a μ -metal shield [84]. Linearly polarized light traverses the cell along the magnetic field (Fig. 2.8). Due to the selection rules, each of the circular components of the linearly polar-

²⁹Thorlabs: PDA100A

³⁰Roithner Lasertechnik: RLT830-150GS

³¹Thorlabs: ITC502 - Benchtop Laser Diode and Temperature Controller ± 200 mA/16 W

³²Thorlabs: FB780-10

ized light induces transmission with different changes of the angular momentum projection. At a non-zero magnetic field, the transitions are frequency shifted leading to a dichroism of the vapour. To detect this dichroism, a quarter-wave plate ($\lambda/4$), a Wollaston polariser, and a two-channel detector were used. One of the photodetector channels was reversed and subtracted from the other, producing dispersion-like error signal. An additional waveplate in front of the setup was used to change the orientation of the incident linearly polarized light with respect to the Wollaston polariser, and thus control the locking point of light (in addition to the $\lambda/4$ plate, which also provided such limited tunability).

Measurement Technique

A small core diameter is responsible for creating a region of high intensity of the guided light, even for a low-power beam. To avoid saturating the atoms, and thus enabling the measurement of very low signals, phase-sensitive detection (PSD) was performed. This technique is widely used to measure low-level signals and to improve the signal-to-noise ratio [21].

The PSD requires two oscillating signals at its input, a modulated signal to be measured (V_{exp}) and a reference (V_{ref}):

$$V_{\text{exp}} = V_p \sin(\omega_p t + \varphi_p), \quad (2.32)$$

$$V_{\text{ref}} = V_r \sin(\omega_r t + \varphi_r). \quad (2.33)$$

Those two signals are multiplied giving the following result:

$$V_{\text{PSD}} = \frac{V_r V_p}{2} \{ \cos[(\omega_r - \omega_p)t + (\varphi_r - \varphi_p)] - \cos[(\omega_r + \omega_p)t + (\varphi_r - \varphi_p)] \}. \quad (2.34)$$

Low-pass filtering of the obtained product rejects the oscillating terms. However, if the two input signals oscillate at the same frequency, a DC signal proportional to a V_{exp} will be observed:

$$V_{\text{PSD}} = \frac{V_p V_r}{2} \cos(\varphi_r - \varphi_p). \quad (2.35)$$

Therefore, the filter will reject noise while the observed signal will be amplified. The level of discrimination depends on the steepness of the applied low-pass filter.

The lower the filters corner frequency, and steeper the filter, the more selective the technique is and hence less noise contribute to the signal.

In the experiment, the probe signal was amplitude modulated using an AOM. The modulated signal measured on Detector 1 was sent to a digital lock-in amplifier³³, where it was recombine with the reference signal from a frequency generator. The lock-in amplifier amplifies the measured signal while rejecting the noise.

2.5 Absorption Spectroscopy in MOFs

2.5.1 Vacuum Chamber Spectroscopy

When the vacuum system reached the desired pressure of less than 10^{-7} mbar, the rubidium atoms were introduced into the first chamber. The temperature of the glass part with the ampoule was set to 90°C and the first chamber was held at approximately 57 - 60°C in the coolest spots. It took two days to passivate the inner chamber walls with rubidium atoms. After this time, the atomic density reached a measurable level. Spectra from the vacuum chamber, along with a theoretical model fit³⁴, is presented in Fig. 2.9. The atomic concentration obtained from the theoretical model was $1.55(2) \times 10^{11}$ atoms/cm³, which corresponds to the temperature of 55°C. This agrees with the temperature measured on the chamber walls. The vapour pressure for this temperature was approximately 7.5×10^{-6} mbar.

³³Zurich Instruments: HF2LI

³⁴Wolfram Mathematica: ADM package by Simon Rochester

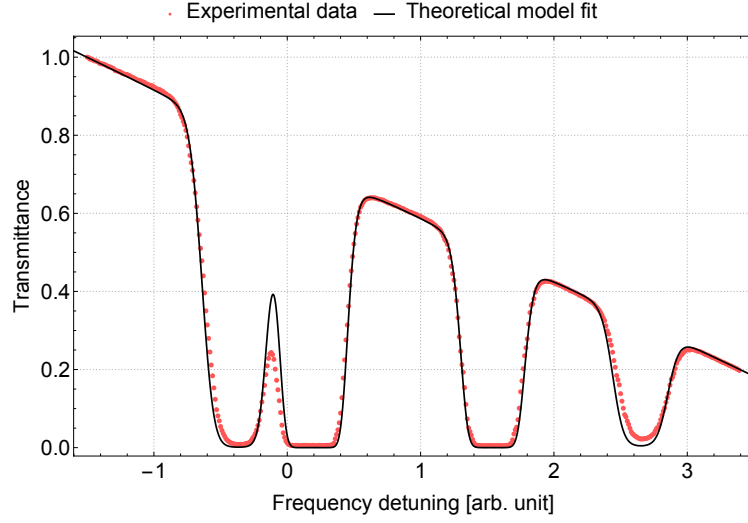


Figure 2.9: Spectroscopy of rubidium vapours in the first vacuum chamber. The concentration of the atoms obtained from the fit: $1.55(2) \times 10^{11}$ atoms/cm³.

2.5.2 Rb-filled Hollow-Core-Fibre Spectroscopy

After a few days (up to a week) of keeping the fibre in a vapour environment, it was possible to observe rubidium spectra through the fibre (Fig. 2.11a). To determine the origin of the observed signal, in particular whether or not it is due to atoms present in the chamber, the atomic concentration for the distance between vacuum viewport and the fibre end-face (approximately 0.69 cm) was calculated. To do so, a red theoretical curve was fitted to the experimental blue points, and a concentration of 1.6×10^{11} atoms/cm³ was determined for the optical power of $0.5 \mu\text{W}$.

The actual optical intensity of light inside the fibre was 37 mW/cm^2 . This is much higher than the theoretical saturation intensity value for this transitions (for the D₂ line and π polarized light $I_{\text{SAT}} = 2.50399(52) \text{ mW/cm}^2$ [85]). The saturation intensity is defined as the intensity of light, for which the saturation parameter drops to half its original value. This is related to optical pumping of the atomic population into an uncoupled ground state. However, the short transit time inside the fibre significantly affects this interaction, and the pumping process is substantially reduced. To estimate the I_{SAT} for Fibre 1, the intensity-dependent absorption coefficient $\alpha(I)$ was measured (see Fig. 2.10).

The expected saturation absorption dependence is given by [68]:

$$\alpha(I) = \frac{\alpha_0}{\sqrt{1 + I/I_{\text{SAT}}}}, \quad (2.36)$$

where α_0 is the ‘zero power’ absorption coefficient and I is the laser intensity. The expected saturation intensity for the HC-PCF is $18(2) \text{ mW/cm}^2$, which is much higher than the value in the bulk³⁵.

Values of the atomic concentrations extracted for different intensity-dependent coefficient values are plotted in Fig. 2.11b. Comparing reference atomic concentration from the vacuum chamber [$1.55(2) \times 10^{11} \text{ atoms/cm}^3$] and the one measured in the fibre for low power ($2.52 \times 10^{11} \text{ atoms/cm}^3$). It could be tentatively concluded that the atoms were loading into the fibre host.

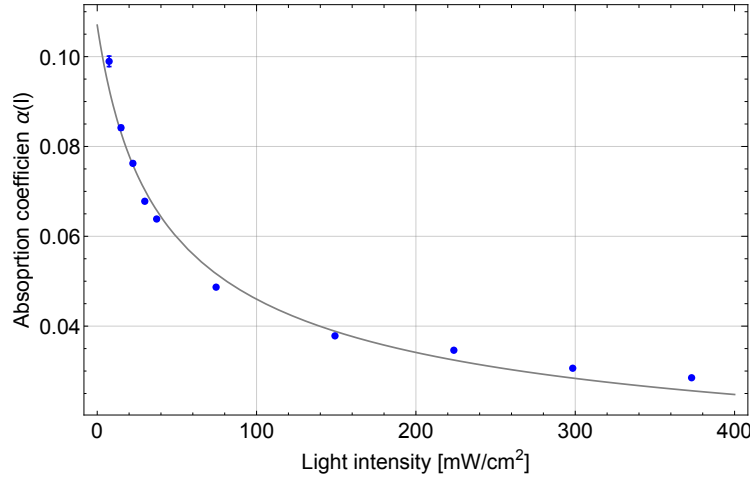


Figure 2.10: Intensity-dependent absorption coefficient measured in Fibre 1.

³⁵Saturation intensity for Fibre 2 is calculated in Ref. [86].

Optical Depth

Optical depth (OD) is a more convenient and accurate quantity to describe atomic concentrations inside the fibre than the number of atoms per volume. Optical depth is defined as a logarithm of the fraction of the incident light intensity (I_I) to the transmitted light (I_T) intensity:

$$\text{OD} = \log \left(\frac{I_I}{I_T} \right). \quad (2.37)$$

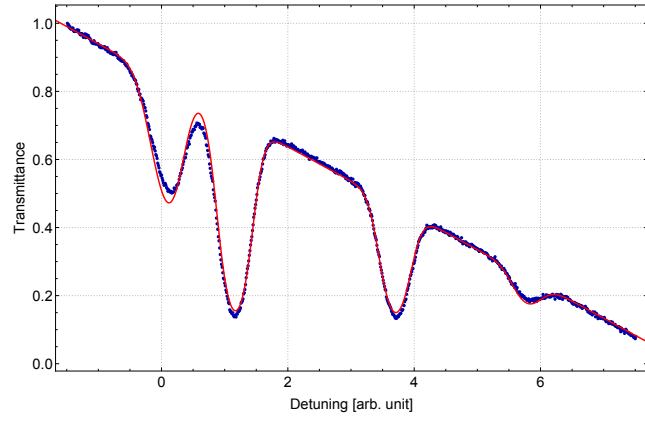
Remembering that the absorption coefficient is defined as: $I = I_0 \exp(-\alpha z)$ (see Section 2.1), OD can be expressed in terms of the absorption coefficient in the following way:

$$\text{OD} = \alpha(\omega)l, \quad (2.38)$$

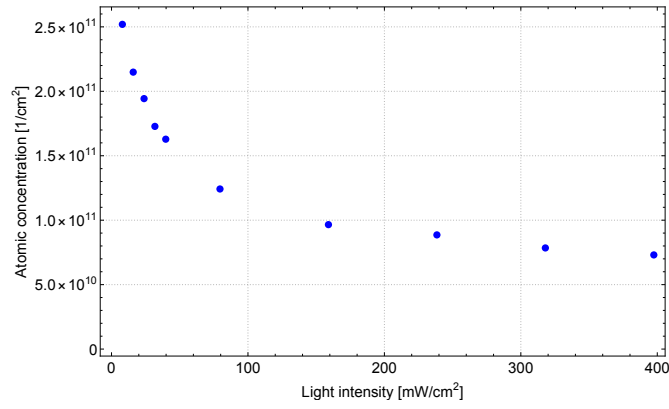
where l is a length of the fibre. OD is a dimensionless quantity, which brings an easier and clearer approach to describe atomic concentrations in a μm -dimension fibre core. It already contains information about the fibre length³⁶, and it is straightforward to compare the fibre OD to the OD value from the reference vacuum chamber (and other HC-PCFs from different experiments).

The OD inside the vacuum chamber (8.36 cm long) is approximately 18(2). In comparison, the OD obtained in the fibre is plotted in Fig. 2.11c (note that these values were predicted for the distance between the end-face of the fibre and a coupling viewport ~ 0.69 cm). The OD is much higher in the fibre direction than for the short length between a viewport and the fibre end-face ($\text{OD} \sim 80$), this indicates a higher atomic concentration inside the fibre. The actual value of the OD, for the entire fibre length (24 cm), was 2.5, measured at the lowest-power of the probe beam.

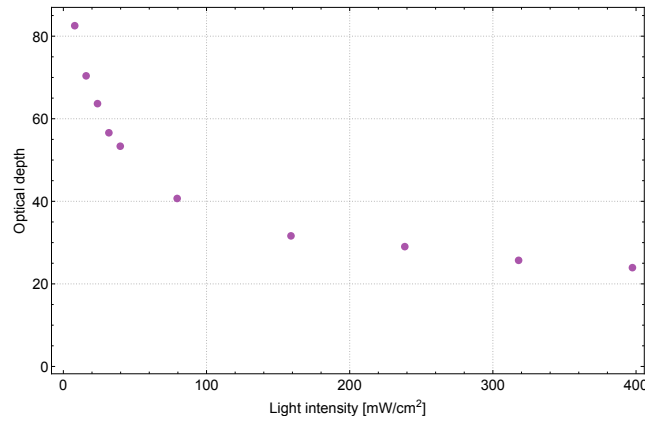
³⁶Assuming that the whole fibre is filled with atoms.



(a)



(b)



(c)

Figure 2.11: Rubidium spectroscopy in Fibre 1. (a) Spectra measured in the fibre (blue points) for light intensity of 40 mW/cm^2 , with the theoretical model fit (red line). (b) Atomic concentration and (c) optical depth measured for various light intensities.

2.6 Summary

In this Chapter, the theoretical model of the optical properties of the atomic spectra was provided for both stationary and non-stationary atoms. The model was compared with spectra observed with rubidium atoms in a glass cell. The measurements were performed using SAS.

Next, a detailed discussion of different types of MOFs was provided. It was shown that the most suitable fibres for atomic-based spectroscopy are kagome-structured HC-PCFs. Their advantages are their large air-core, enabling comparatively easy filling with alkali vapour, and a broad range of supported wavelengths. Two types of the kagome-latticed HC-PCF were chosen for the experiment: a 7-cell hypocycloid-core and a large-pitch single-cell.

A large part of this Chapter described the vacuum-system design and preparation of the experimental setup. This was also an significant part of the PhD project described in this dissertation. A detailed discussion of the procedure for fibre preparation and the procedure for filling the fibre with atomic vapours were provided.

Finally, spectroscopic measurements of rubidium vapours in a vacuum chamber and inside the fibre were described. Particularly, it was shown that rubidium vapours are filling the fibre host, and providing additional absorption of light. The experimental value of the saturation intensity I_{SAT} for alkali vapours filling Fibre 1 was determined and the discussion of the measured value, with respect to the I_{SAT} observed in a bulk system, was described. This result demonstrates differences between a microscopic and a bulk system.

Chapter 3

Light Induced Atomic Desorption

Atomic vapours are highly reactive, and readily adsorb to dielectric porous surfaces. An amorphous structure of silica glass allows its free electrons to pair with alkali atoms. The adsorbed atoms form atomic layers and nanoparticles, which can be desorbed and modified via interaction with light. Therefore, the interacting light may cause the atoms' movement from clusters to nanoparticles and other way around [66].

A process of light-induced atomic desorption (LIAD) [87] is well known in bulk materials, such as cells with paraffin coating, and pure silica glass [65, 66]. LIAD is considered as a non-thermal desorption process that induces detachment of the atoms formed on the surface (atomic clusters), and their diffusion through silica nanopores causing further growth of nanoparticles [66]. The origin of the LIAD effect is not fully defined, however, the oscillation of the electrical charge at the silica-surface defects could be a possible explanation [66]. LIAD occurs for non-resonant light but still reveals a dependence on the wavelength of the desorbing light and its power [65, 66]. Particularly for shorter wavelengths, i.e., higher photon energies, where the effect is stronger. Similarly, the effect is stronger for more intense light. The fact that LIAD depends on optical power, and does not require resonant light, provides a method of control over the concentration of atoms in a vapour. In particular, this effect may be used to control the OD of a vapour inside a fibre.

The other effect that causes atomic desorption are surface-plasmon oscillations,

surface-plasmon induced desorption (SPID), which is a non-thermal desorption process of nanoparticles [66]. The SPID effect requires the excitation of a plasmon resonance, which is a charge-density oscillation at the interface of an alkali metal and a dielectric silica glass [88]. The SPID process desorbs atoms from nanoparticles and causes the formation of atomic clusters.

The two desorption processes, LIAD and SPID, are competing processes if they occur simultaneously. The plasmon resonances, for Rb nanoparticles inside porous silica, are located in NIR region [66]. This gives the possibility that the two effects, LIAD and SPID, will occur at the same time. However, there are no studies on Rb nanoparticle resonances in silica fibres which gives convincing proof if the SPID effect actually take place in such systems.

Non-linear optical experiments require higher optical powers of their control beams which, according to the LIAD and SPID processes, additionally affect the number of atoms inside a fibre host. Therefore, the observed non-linear effects might be influenced by a variation in OD. In particular, the LIAD effect was used to enhance the atom-photon interaction inside the fibre by increasing the number of atoms. An exploration of atom dynamics inside a fibre, and a way of controlling OD inside alkali-filled optical waveguides, appeared as an important challenge in terms of the non-linear optics in such system. Numerous studies showed that the way to control OD in alkali-filled hollow-core fibres, is by desorbing atoms from the fibre walls using non-resonant high-intensity light [89–91].

The dynamics of the atomic vapours inside the fibre host are much different than the behaviour of the atoms in a bulk. The surface-to-volume ratio is much larger in fibres than for glass cells, so interactions of atoms with a silica surface (i.e., adsorption of the atoms) and light with an inner-fibre-core surface are significantly enhanced. Thus, every light beam coupled into the fibre will interact with the atoms adsorbed to the cores inner surface. This results in a strong dependence of the OD inside the fibre on the lights intensity. It was already shown in Ref. [91] that extremely large OD values (> 2000) can be generated inside a HC-PCF. In fact, such large values have never been observed in any bulk system. This provides a useful mean of enhancing the atom-photon interaction for non-linear optics and

photonics applications. However, large OD values could only be generated for a finite time, i.e., the increase in atomic density is followed by an abrupt decay process (Fig. 3.1).

3.1 Long-term Exposure to Desorbing Light

Considering that the LIAD process is releasing atoms from the porous surface of silica glass, which is manifesting an increase of OD, it can be used to determine the presence of atoms inside a fibre. The increase of the atomic concentration that contributes to the signal from the HC-PCF but does not affect the reference signal from the vacuum chamber, will provide the proof for presence of atoms inside the fibre. The long-time scale LIAD experiment was performed to investigate the presence and atom behaviour inside Fibre 1.

3.1.1 Experimental Setup

To perform LIAD experiment in vapour filled HC-PCF, the temperature of the vacuum setup was lowered to approximately 36°C, which reduces the background vacuum chamber concentration. Prior to this operation the fibre has already been loading ('baked' in rubidium atmosphere) for a few weeks, which should provides enough diffusion time for the vapour to penetrate inside the fibre.

The experimental setup used to examine long time-scale LIAD processes was described in Section 2.4.3. The ECDL was used as a weak probe-beam source. Light from the laser was split into two beams. One of them was measuring the absorption signal from the fibre, while the other was used to monitor concentration in the vacuum chamber (reference spectrum). The probe beam (~ 12 nW) was scanned over the rubidium D₂ line and its absorption was continuously monitored (the absorption spectra was measured over time). The high-power diode laser was used as the source of desorbing light. The strong (3.3 mW) desorbing beam was introduced into the fibre for one hour. After which it was turned off for 30 minutes, to allow for the recovery of the desorbed atoms, and turned on again for another 30 minutes.

3.1.2 Results

The spectra from the fibre and vacuum chamber were captured at the same time, a ~ 1 s delay between successive measurements. A theoretical model of the full spectrum³⁷ was fitted to reveal the OD. The long-term OD value was determined based on the measurements shown in Fig. 3.1. The top graph shows the OD changes in the vacuum chamber. Shaded regions mark the presence of the desorbing beam inside the fibre. The general trend of the decrease of OD in the chamber indicates that the temperature is still stabilizing. As shown in the chamber, there was no change in the atomic concentration due to the presence of the desorbing beam. At the same time, the OD in the fibre varies due to the illumination of the desorbing light. At the beginning of the measurements, the OD also decreases in a similar manner as the OD in the reference measurement. However, after turning on the LIAD beam, the optical depth (concentration) suddenly rises and then starts decreasing exponentially. This is a LIAD behaviour known in bulk [65] and HC-PCF [89, 91].

The first illumination cycle lasts about 1 hour, after which the desorbing beam was turned off for 30 minutes and then turned on again for another 30 minutes. Interestingly, after turning off the LIAD beam, the OD level was lower than the initial OD. This is caused by the depletion of rubidium atoms and their further penetration into the porous silica glass. The OD level increased again during the recovery of the system, after the desorbing beam was turned off. This indicates that the atoms did not escape the fibre, as the process of loading vapours into the fibre core was long and unnoticeable over such a short time frame (it took a few days to start observing a signal from the fibre). The second cycle of the desorbing-beam illumination again caused a rise in the OD value, but this time to a lower level than the initial one. This agrees with the studies described previously in Refs. [89–91], where a strong desorbing beam caused a temporarily high level of OD (~ 2000 for a fully filled 30-cm long fibre), then followed by an exponential decay in the concentration. The temperature from the measured OD was

³⁷Wolfram Mathematica: ADM package by Simon Rochester

approximately 30°C, which agrees with the temperature measured on the vacuum elements (approximately 36°C).

Changes in the atomic concentration in the HC-PCF strongly depends on the presence of the desorbing beam. At the same time, there is no influence of the desorbing light on the OD in the vacuum chamber. This indicates that the fibre was loaded with rubidium atoms, and that the increase in the OD was induced by the atoms desorbed from its walls.

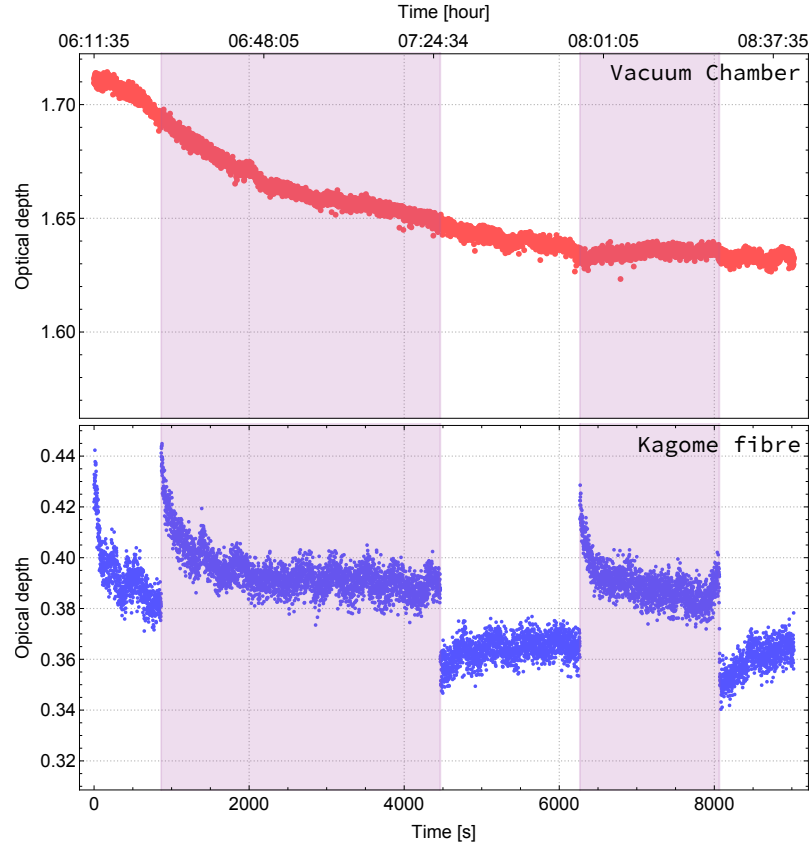


Figure 3.1: Optical depth in the vacuum chamber (top) and kagome-latticed HC-PCF (bottom) over time under LIAD. The shaded regions indicate the presence of the desorbing beam in the HC-PCF. The OD inside the fibre varies due to the presence of the desorbing beam, whether there are no OD changes inside the chamber. The temperature from the measured OD was approximately 30°C, which agrees with the temperature measured on the vacuum elements (approximately 36°C).

3.2 Pulsed-Light Induced Desorption

To carefully examine atom dynamics inside the fibre host, a short pulse rather than a CW desorbing beam was used. The pulsed-LIAD experiment can explain the dynamics and nature of the desorbing processes [90]. The dependence on light intensity, pulse duration, and pulse energy can be compared and analysed. Furthermore, by adjusting the pulse duration and pulse energy, the pulsed-LIAD experiment enables dynamic control over the atomic concentration by introducing a feedback signal to the desorbing source of light.

3.2.1 Experimental Setup

Kagome-structure HC-PCF

The kagome-structured HC-PCF used in the pulsed-LIAD experiment is described in Section 2.3, as *Fibre 2*. It is a large-pitch single-cell kagome-structured HC-PCF with core diameter of $45\text{ }\mu\text{m}$, that was entirely filled with rubidium atoms. The experimental setup is depicted in Fig. 3.4. The approximately 30-cm long piece of HC-PCF was placed in the vacuum chamber filled with dense rubidium vapours. The temperature of the vacuum system was kept at $70 - 80^\circ\text{C}$. The transmission through the fibre was $\sim 25\%$ from each side.

Lasers

The lasers used in the experiment were: a home-built ECDL emitting at 780 nm used as a probe, and a commercial tunable Ti:Sapphire laser³⁸ used as a desorbing-light source.

The ECDL was frequency stabilized to another laser (‘a frequency standard’), to allow for precise tuning of the probe laser around the transition frequency. The ‘frequency standard’ laser was stabilized to the $5S_{1/2} F = 3 \rightarrow 5P_{3/2} F' = 4$ transition in ^{85}Rb . The scheme of the locking setup is shown in Fig. 3.2. A beat-note of the probe laser and phase-locked standard laser was detected by a photodetector,

³⁸M Squared: SolsTiS

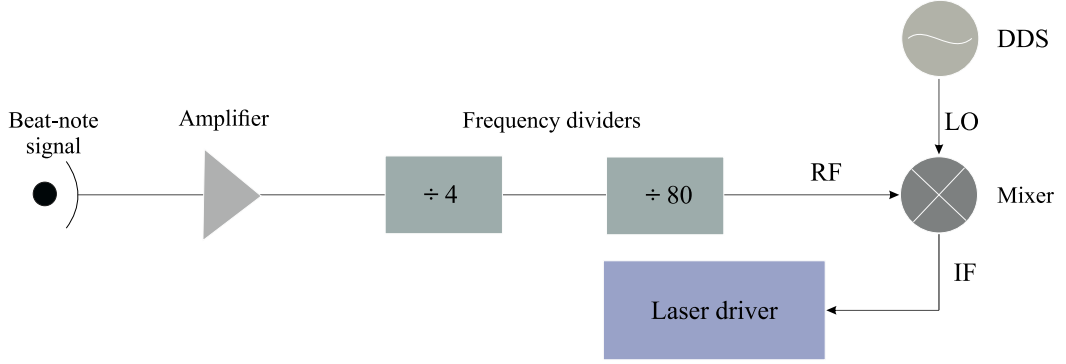


Figure 3.2: Probe-laser frequency-locking setup. The error signal is provided by the multiplication of a beat-note signal, generated by superimposing probe light with frequency standard laser, with a local oscillator (LO) reference provided by a direct digital synthesizer (DDS).

which transforms an optical signal into an electrical one. The measured beat-note signal was amplified, multiplied, then mixed with a local oscillator (LO) to create an error signal. The error signal provided feedback to a laser driver. In this way, it was possible to precisely tune the locking point by adjusting the frequency from the direct digital synthesizer (DDS). The desired optical frequency could be set in the following manner:

$$\left| f_{\text{LO}} - \frac{1}{320}(\nu_{\text{standard}} - \nu_{\text{probe}}) \right| = 0, \quad (3.1)$$

where ν_{standard} and ν_{probe} are the optical frequencies of the frequency standard and probe laser, respectively. The f_{LO} is a LO frequency provided by the DDS.

The probe laser was also coupled to the optical cavity providing 300 MHz frequency markers and passing the SAS reference cell to provide the absolute frequency reference (see Fig. 3.3).

The probe signal from Fibre 2 was measured by phase-sensitive detection (Section 2.4.3). The probe beam was modulated by the AOM, and the signal exiting Fibre 2 was measured on an avalanche photodiode (APD), which was then sent to a digital lock-in amplifier³⁹ (see Fig. 3.4).

Light emitted from the desorbing Ti:Sapphire laser was stabilised to two built-

³⁹Stanford Research Systems: SR810/SR830

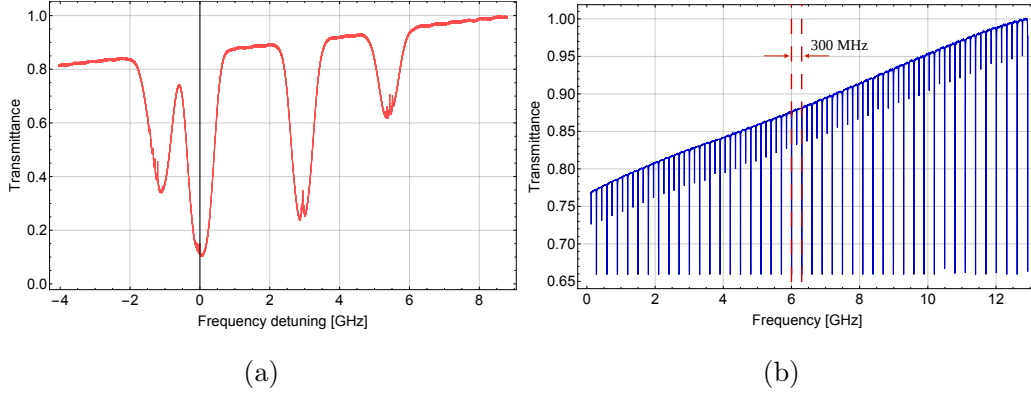


Figure 3.3: (a) SAS from the reference cell, and (b) signal from the reference optical cavity providing 300-MHz frequency markers (red dashed line).

in cavities. The wavelength was set to 771 nm, which is far detuned from any transition in rubidium atoms. Two photodiodes, placed before⁴⁰ and after⁴¹ the HC-PCF, were monitoring if there were any power fluctuations on the desorbing beam (see Fig. 3.4). Both of the detectors were small-area fast photodiodes suitable for measuring short light pulses. The pulses on the desorbing beam were formed by the AOM receiving a modulated driving signal. The pulsed signal from the function generator, together with a 80-MHz modulation signal, was fed to the AOM.

The polarisations of the probe and desorbing lasers were set orthogonal to facilitate separation of the beams. To prevent any additional reflections of strong desorbing light on the APD responsible for measuring the probe signal, a diffraction grating was used.

⁴⁰Thorlabs DET100A/M

⁴¹Thorlabs DET10A/M

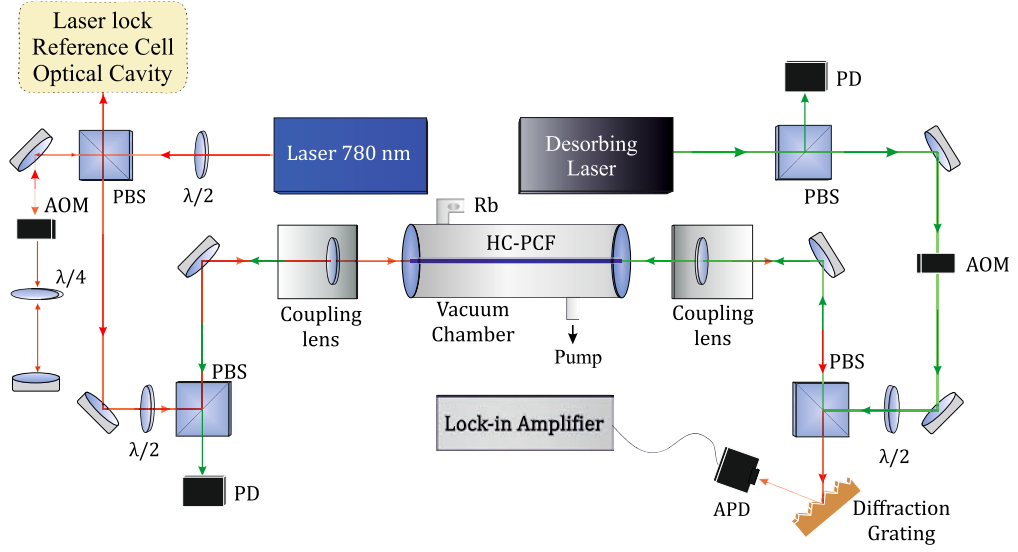


Figure 3.4: Experimental setup for pulsed LIAD. The probe beam was tuned to the D_2 transition of rubidium atoms. Fractions of the light were sent, to the SAS reference cell, the optical cavity, and the laser-locking setup. The majority of the light was coupled into Fibre 2 and measured after the fibre with the APD. The probe light was modulated by an acousto-optic modulator (AOM), and detected by a lock-in amplifier. The Ti:Sapphire laser was used as a source of desorbing light. The power of the desorbing beam was monitored before and after the HC-PCF, which ensured that no power fluctuations occurred during the measurement on the two small-area photodiodes (PDs). The AOM formed the desorbing pulses using a modulated drive signal.

3.2.2 Results

The reference spectrum presented in Fig. 3.5b was obtained from Fibre 2 using weak ($\sim 17\text{ nW}$) CW probe light, while no desorbing beam was present. The theoretical model fit (blue line) was obtained by the summation of two Doppler-broadened transitions of the D_2 line in rubidium vapour (see Fig. 2.2b). The Maxwell distribution of the atoms velocities was modelled by the Gaussian function. Thus, the convolution of the Lorentzian profile, which reflects the natural width of the transition, and the Gaussian broadening, is defined by the Voigt profile or

by the real part of the Faddeeva function [Eq. (3.2)]:

$$V(\omega, \omega_0, \gamma, \Gamma_D) = \Re \left[\exp \left[- \left(\frac{\omega - \omega_0 + i\gamma/2}{\Gamma_D} \right)^2 \right] \operatorname{erfc} \left[-i \frac{\omega - \omega_0 + i\gamma/2}{\Gamma_D} \right] \right], \quad (3.2)$$

where γ is the FWHM of the Lorentzian profile (natural width of the transition), Γ_D represents the Doppler width, and ω_0 is the transition frequency. To correspond to the experimental data the function was normalised to unity,

$$V_{\text{norm}}(\omega, \omega_0, \gamma, \Gamma_D) = \frac{V(\omega, \omega_0, \gamma, \Gamma_D)}{V(\omega_0, \omega_0, \gamma, \Gamma_D)}, \quad (3.3)$$

so that the OD in the following equation represents the actual optical depth. The absorption function Abs , reproducing the experimental spectra, can be written in a following form

$$Abs(\omega, \omega_0, \gamma, \Gamma_D) = \text{OD} \left(\sum_i a_i V_{\text{norm}}(\omega, \omega_0 + \omega_i, \gamma, \Gamma_D) \right), \quad (3.4)$$

where ω_i is the frequency separation between hyperfine transition i and the transition $5S_{1/2} F = 3 \rightarrow 5P_{3/2} F' = 4$, and a_i is the relative intensity of the i -th transition. The absorption function Abs is plotted in Fig. 3.5a, where the Doppler width was set to be below the natural linewidth, and all the hyperfine transitions are visible.

The experimental data for the reference spectra, with fit according to the model [Eq. (3.4)] is presented in Fig. 3.5b. The OD yields the value of 17.5(5), the Doppler width was estimated to be 336(2) MHz. This gives an approximate temperature of the vapours of 77°C, which agrees with the approximate temperature of the vacuum setup.

The actual experimental data, when the pulsed desorbing beam was introduced, was obtained with an optical power of the probe of ~ 150 nW. During the pulsed LIAD experiment, the probe laser was locked to several frequencies in the vicinity of the $5S_{1/2} F = 1 \rightarrow 5P_{3/2} F' = 0, 1, 2$ transition in rubidium 87, marked as blue points in Fig. 3.6a. When pulsed desorbing was introduced to the fibre it caused a temporal increase in the OD, followed by its decay toward the initial level after the beam was turned off. The exemplary OD response to the presence of the desorbing pulse is depicted in Fig. 3.6b. Here, the 2-ms desorbing-light pulse was

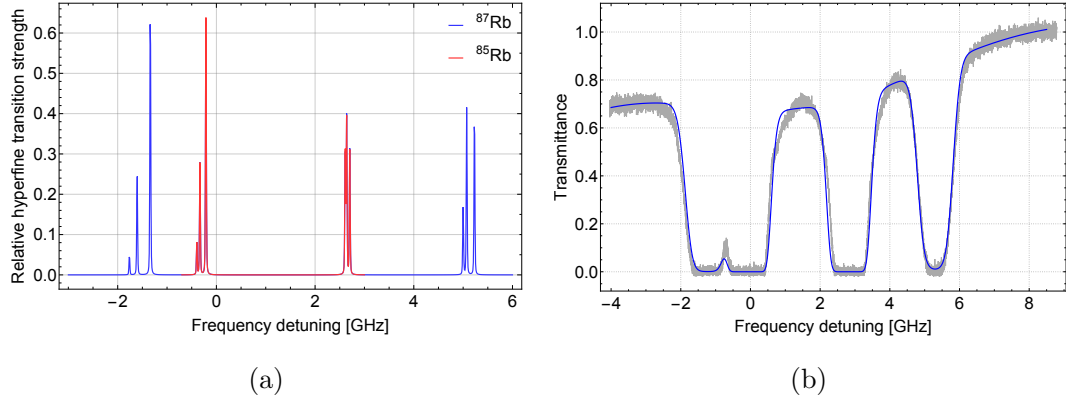


Figure 3.5: (a) Theoretical absorption function for all the transitions of the rubidium D_2 line for both stable isotopes (^{87}Rb blue and ^{85}Rb red line) for the Doppler-free case. (b) A reference spectrum for the pulsed-LIAD experiment, obtained with a CW probe beam, with a theoretical model fit. Fitting parameters: $\text{OD} = 17.5$, $\omega_0 = 5.09 \text{ GHz}$, $\Gamma_D = 336 \text{ MHz}$.

introduced marked as a shaded region. An optical power of the desorbing beam of 40 mW was measured before the fibre. The induced OD changes were measured for different optical powers and pulse durations of the desorbing light for different frequency detunings around the transition.

To reveal the level changes in the OD and decay time, exponential functions were fitted to both the increasing and decreasing slopes (see Fig. 3.6b):

$$\text{OD}(t) = \mp A \exp\left(-\frac{t}{T_1}\right) + \text{OD}_{\text{final}}, \quad (3.5)$$

where T_1 is the rise (decay) time, A is the amplitude, and OD_{final} is the final OD asymptotic value. The OD_{final} indicates the upper and lower level of the OD after turning the desorbing beam on and of, respectively. This approach is motivated by a similar data analysis in bulk systems [65, 92].

The obtained upper OD_{final} values are compared in Fig. 3.7 for different desorbing light conditions. Each of the graphs represents the OD changes induced for the same averaged energy delivered to the atoms, but with different pulse durations and optical powers. In Fig. 3.7a pulses with an average energy of 20 mJ and time durations of 4, 2, and 1 ms are presented. Figure 3.7b shows pulses with an averaged energy of 40 mJ and pulse durations of 8, 4, 2, and 1 ms. Figure 3.7c

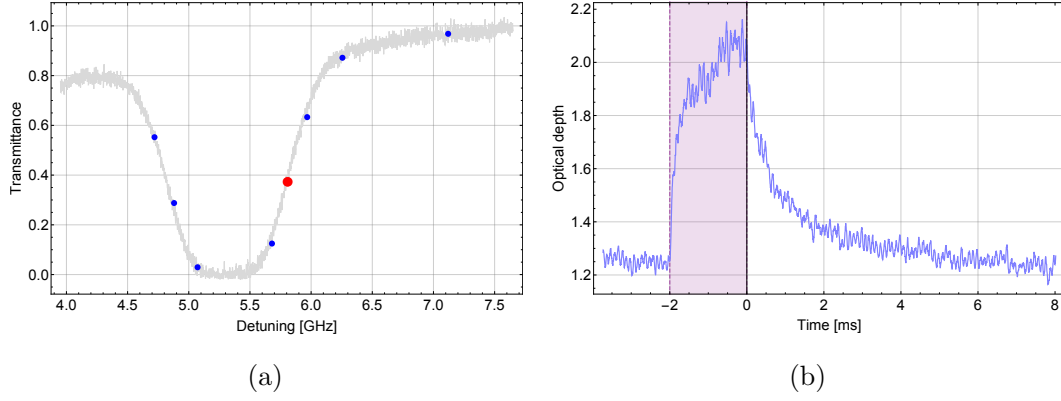
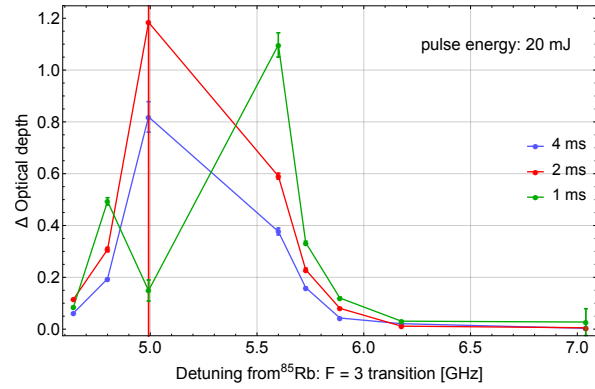


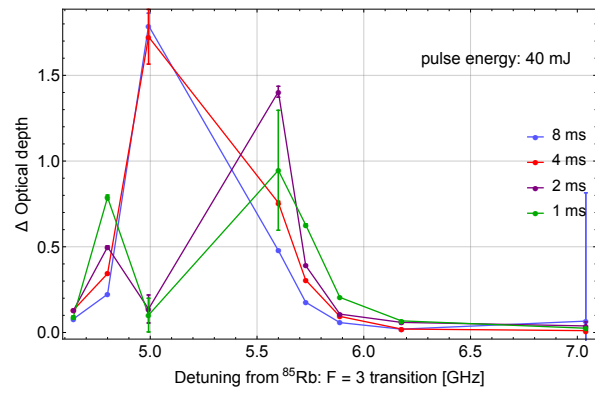
Figure 3.6: (a) Absorption spectrum of the ^{87}Rb : $5S_{1/2} F = 1 \rightarrow 5P_{3/2} F = 0, 1, 2$ transition. The grey line represents the reference spectrum, the blue dot positions are where the probe laser was locked and changes in transmission due to the presence of the desorbing light were measured. (b) Exemplar signal, in OD, for the point marked as a red circle. The optical power of the desorbing beam was 40 mW and the time duration of the pulse was 2 ms. The shaded region indicates the presence of the desorbing light and dashed lines mark beginning and the end of the desorbing pulse.

shows pulses with a pulse energy of 80 mJ and pulse durations of 8, 4, and 2 ms. The points measured close to the resonance, i.e., frequency detuned between 5 and 5.55 GHz, have a high uncertainty. In particular, it can be observed for 1-ms pulses carrying 20 and 40 mJ energy, and 2-ms pulse with an energy of 80 mJ. The reason is that, for those points, the measured signal approaches zero transmission ‘immediately’ after the introduction of the desorbing beam.

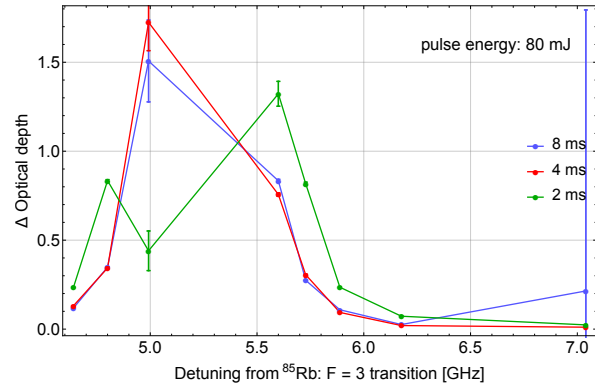
The presented results show that higher energy pulses (Fig. 3.7c) caused greater changes in the OD. Furthermore, shorter pulses, carrying a higher optical intensity, induced greater changes than the longer pulses carrying the same averaged energy. This is non-trivial result, which suggests that the response of the system is definitely non-thermal interaction between atomic agglomerates and nanoparticles with light, and depends on the temporal light intensity rather than total energy delivered to the system.



(a)



(b)



(c)

Figure 3.7: Changes in the OD for various tunings of the probe beam, the pulse length, and the energy of the desorbing light. The panels represent (a) 20-mJ, (b) 40-mJ (c) 80-mJ of the desorbing-pulse light. The power of the probe beam was ~ 150 nW.

In Fig. 3.8 the rise and decay times of the OD changes for the 20- and 40-mJ energy pulses were plotted⁴². The times remain the same for the same optical power but vary with the pulse duration, which is indicated by the same colour of points in Fig. 3.8, i.e., pulses of different duration but carrying the same optical power are indicated by the same colour. The rise times are significantly shorter for short pulses, which carry higher light intensities (see Fig. 3.8). Since the OD changes are greater for shorter pulses, this result highly agrees with the data plotted in Fig. 3.7, which shows the dependence of the OD changes on the light intensity. However, the same behaviour for the decay time (T_1), is not as intuitive. The decay time is significantly shorter for short pulses and higher intensities, which suggests that the depletion of the atoms can also be caused by extra processes for the pulses with high intensities. This process could be associated with LIAD, more complex behaviour of the agglomerates formed by the adsorbed atoms or the presence of the SPID process [66, 90].

A stable level of the OD of alkali vapours in silica waveguides can be achieved by the introduction of a sequence of desorbing pulses with adjustable energy and duration. This could allow for precise and dynamic control of the optical depth, and to keep a stable value during an experiment.

⁴²The number of points in this figure was reduced due to large error bars for the points (see Fig. 3.6a) measured exactly on resonance and when far-detuned (frequency detuning greater than 6 MHz).

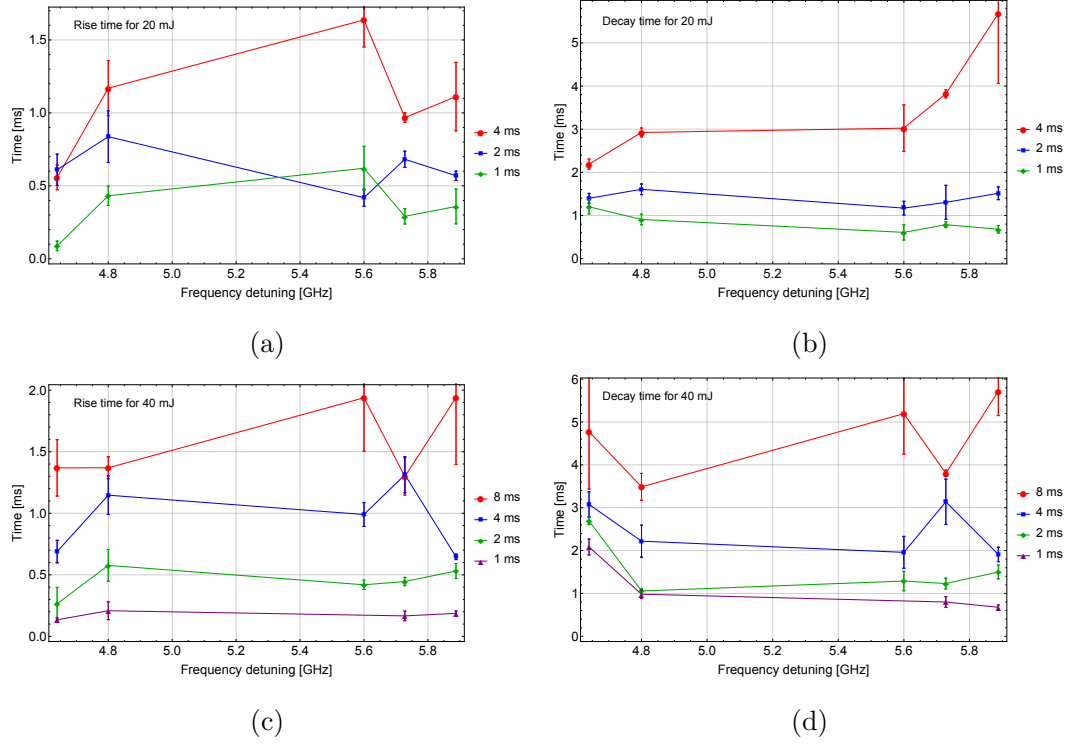


Figure 3.8: Rise and decay times of the OD change induced by pulsed LIAD. (a) and (b) is the rise and decay time for the pulse energy 20 mJ, respectively. (c) and (d) is the rise and decay time for pulse energy 40 mJ, respectively. The colours of the lines and points indicate the optical intensity: red - 5 mW, blue - 10 mW, green - 20 mW and purple - 40 mW.

3.3 Summary

In this Chapter, the process of light-induced atomic desorption (LIAD) in two types of kagome-latticed HC-PCF was examined. The LIAD experiment was performed in two regimes: long time-period LIAD using CW light and pulsed-LIAD. Both of them show strong dependence of the OD on the presence of a desorbing beam and revealed the information about the dynamics of the atoms inside the fibre.

The long exposure of the atoms inside the fibre to the high-intensity light caused a temporal increase of the atomic concentration followed by a depletion of the atoms and their further penetration into the silica-matrix. The detailed

studies on the LIAD process in Ref. [66] shows that the desorbed atoms diffuse through the silica nanopores causing further grow of nanoparticles. Furthermore, a long time-period LIAD process was performed to determine the origin of the absorption signal in Fibre 1. This showed that Fibre 1 was being successfully filled with rubidium atoms.

The pulsed-LIAD experiment, performed in Fibre 2, showed interesting results. Shorter pulses carrying a higher optical power induced greater OD changes than the longer-duration pulses of the same average energy but lower optical power. This indicates that from the point of view of LIAD, it is intensity not power of the desorbing light that controls the amplitude of the process and hence enhancement of atomic concentration. The time scale of the rise and decay times of the LIAD process were also examined and revealed a strong dependence on the light intensity.

In general, the LIAD process can be used to generate a large and stable vapour density inside a fibre [90, 91]. Since the optical depth strongly depends on the intensity of light coupled into a fibre, control of the atoms concentration is of high importance for non-linear experiments performed in any HC-PCF.

Chapter 4

Non-linear Spectroscopy

4.1 Non-linear Optical Susceptibility

Non-linear optics studies phenomena that occur in the presence of strong light, which modifies the optical properties of a medium. The non-linearity depends on the light intensity and sometimes its wavelength.

As shown in Chapter 2, the response of a medium to light can be characterised by the polarization vector:

$$\mathbf{P}(t) = \varepsilon_0 \chi \mathbf{E}(t), \quad (4.1)$$

where ε_0 is the vacuum permittivity and χ is the medium's susceptibility. In the non-linear case, the polarization vector can be expressed by a power series of the light-field amplitude

$$\mathbf{P}(t) = \varepsilon_0 \chi^{(1)} \mathbf{E}(t) + \varepsilon_0 \chi^{(2)} \mathbf{E}^2(t) + \varepsilon_0 \chi^{(3)} \mathbf{E}^3(t) + \dots, \quad (4.2)$$

where $\chi^{(1)}$ is the linear (first order) susceptibility and the quantities $\chi^{(2)}$ and $\chi^{(3)}$ are the second- and third-order non-linear optical susceptibilities. In general, the non-linear susceptibilities are second- and third-rank tensors, respectively.

One can also write Eq. (4.2) using $\mathbf{P}^{(2)} = \varepsilon_0 \chi^{(2)} \mathbf{E}^2(t)$ and $\mathbf{P}^{(3)} = \varepsilon_0 \chi^{(3)} \mathbf{E}^3(t)$, the second- and third-order non-linear polarization vectors. Importantly, physical processes that occur according to the second-order polarization are distinct from the ones that occur according to third-order polarization. Second-order polarization phenomena can only take place in non-centrosymmetric crystals (i.e., that do

not posses inversion symmetry). Since gases, including atomic vapours, posses an inversion symmetry, so in such media the second-order susceptibility $\chi^{(2)}$ vanishes [1] and the lowest order polarization vector responsible for non-linear processes is $\mathbf{P}^{(3)}$.

In this Chapter, the focus will be on two-photon absorption (TPA), i.e., the process which is a third-order non-linear phenomenon occurring in multi-level atomic systems. TPA permits an interaction of different-frequency photons through the non-linear properties of atomic media. On more practical grounds, TPA reveals sub-Doppler resonant features, which gives the possibility of a strong modification to the optical properties of a host material. Such strong changes are crucial for processes such as slow and fast light.

The Kramers-Kronig formula relates the real and imaginary parts of frequency-depended quantities. In particular, these allow for determining the imaginary part of the electric susceptibility from the knowledge of the real part. In other words, it enables determining the dispersion properties of the medium based on the knowledge of its absorption profile.

4.1.1 Kramers-Kronig Relations in Linear Optics

The Kramers-Kronig relations can be straightforwardly applied to linear optics, but only in some cases can they be formulated for non-linear interactions [1, 93]. In general, for Kramers-Kronig to apply, one must ensure that $\chi(\omega)$ is a regular analytic function in the positive imaginary plane of a complex frequency ω .⁴³ Thus, poles do not exist and the contour integral over the upper half-plane is equal to zero.

The polarization induced in a material with a linear response to the electric field can be written in the following form [93]:

$$\mathbf{P}^{(1)}(t) = \varepsilon_0 \int_0^\infty R^{(1)}(\tau) \mathbf{E}(t - \tau) d\tau, \quad (4.3)$$

where $R^{(1)}(\tau)$ is a linear response function, which provides a contribution to the polarization at time t with respect to the electric field applied at time $t - \tau$. To

⁴³This is a purely mathematical concept to examine some properties of susceptibility $\chi(\omega)$ [1].

ensure the causality law, i.e., the observed polarization at time t , follows the earlier interaction with electric field (at time $t - \tau$), $\mathbf{R}^{(1)}(t) = 0$ for $\tau < 0$. Therefore, the linear susceptibility can be expressed as:

$$\chi^{(1)}(\omega) = \int_0^\infty \mathbf{R}^{(1)}(\tau) \exp(i\omega\tau) d\tau. \quad (4.4)$$

Considering that the frequency is a complex value $\omega = \omega' + i\omega''$, the integral from Eq. (4.4) can be rewritten in the following manner:

$$\chi^{(1)}(\omega) = \int_0^\infty \mathbf{R}^{(1)}(\tau) \exp(i\omega'\tau) \exp(-\omega''\tau) d\tau, \quad (4.5)$$

since $\mathbf{R}^{(1)}(\tau)$ is finite everywhere, the term with the imaginary part of the frequency ensures the convergence of the integral for $\omega'' > 0$. On the real axis, where $\omega'' = 0$, the convergence of the integral is ensured by the properties of the susceptibility $\chi^{(1)}(t)$, which is a measurable physical quantity. The Kramers-Kronig relations are valid if the linear susceptibility is an analytical and continuous function, which is ensured by the convergence of the integrand [Eq. (4.4)] in the upper half-plane of a complex frequency.

The Kramers-Kronig relations for the linear susceptibility can be written as [1, 93]:

$$\begin{aligned} \chi'(\omega) &= \frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\omega' \chi''(\omega')}{\omega^2 - \omega'^2} d\omega', \\ \chi''(\omega) &= \frac{2\omega}{\pi} \mathcal{P} \int_0^\infty \frac{\chi'(\omega')}{\omega^2 - \omega'^2} d\omega', \end{aligned} \quad (4.6)$$

where $\mathcal{P}$ denotes the Cauchy principal value.

Now, consider the wavevector of an electromagnetic field travelling in weakly absorbing medium:

$$k = \frac{\omega}{c} = k_0 \sqrt{\varepsilon\mu} = k_0 \sqrt{1 + \chi}, \quad (4.7)$$

where ε and μ are the electric permittivity and magnetic permeability of the medium, respectively. Decomposing the wavevector into its real and imaginary parts: $k = \beta + i\alpha/2$, where β is the propagation constant and α describes the attenuation of propagation in the material, and remembering that (see Section 2.1):

$$n(\omega) = 1 + \frac{\chi'}{2}, \quad \kappa(\omega) = \frac{\chi''}{2}. \quad (4.8)$$

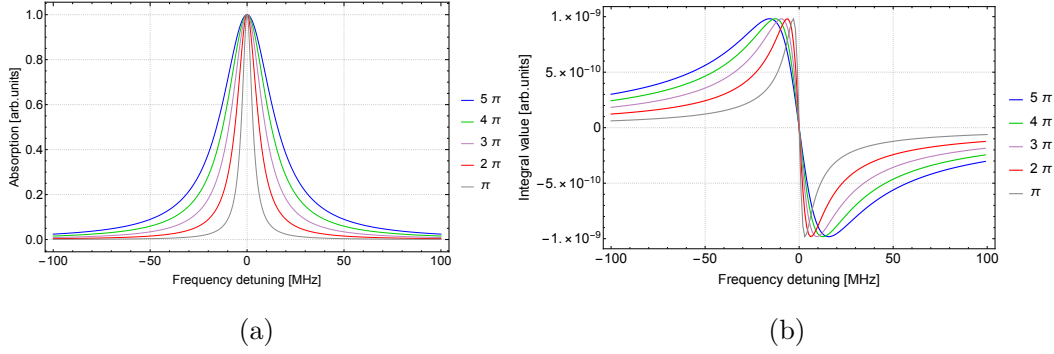


Figure 4.1: (a) Absorption profile modelled by the Lorentz function [Eq. (4.11)] and (b) results of the calculation of the integral from Eq. (4.12) for frequency detuning $(\omega - \omega_0)$ and various decay constants $(\Gamma = \pi, 2\pi, 3\pi, 4\pi, 5\pi)$.

the relation between the absorption coefficient and the imaginary part of the refractive index κ is obtained as

$$\alpha = 2\kappa \frac{\omega}{c}. \quad (4.9)$$

Based on Eqs. (4.6), (4.8) and (4.9) the relation between the refractive index and the absorption coefficient of the medium α can be written by:

$$n(\omega) = 1 + \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\alpha(\omega')}{\omega^2 - \omega'^2} d\omega'. \quad (4.10)$$

Finally, the dispersion profile for the Lorentzian model can be obtained by inserting the explicit form of the absorption coefficient (Fig. 4.1a)

$$\alpha(\omega) = \frac{A\Gamma^2}{(\omega - \omega_0)^2 + \Gamma^2}, \quad (4.11)$$

into Eq. (4.10):

$$n(\omega) = 1 + \frac{2c}{\pi} \mathcal{P} \int_0^\infty \frac{\Gamma^2}{[(\omega' - \omega_0)^2 + \Gamma^2] (\omega^2 - \omega'^2)} d\omega'. \quad (4.12)$$

The results of the integral given by Eq. (4.12) calculated for various decay constants are presented in Fig. 4.1b. For a positive absorption coefficient, anomalous dispersion is expected. Alternatively, anomalous dispersion accompanies absorption features, whereas a normal dispersion profile is observed when there is gain.

4.1.2 Kramers-Kronig Relations in Non-linear Optics

For non-linear processes, the Kramers-Kronig relations cannot be straightforwardly applied. The situation changes since the non-linear absorption coefficient $\bar{\alpha}$ begins to depend on the light intensity $I = \frac{1}{2}c\epsilon_0\langle\mathbf{E}^2\rangle$.

Saturated Absorption Spectroscopy

Saturated absorption spectroscopy was already described in Section 2.2, and is also a third-order non-linear process. It is seen as a decrease of absorption in the presence of strong light. The non-linear absorption coefficient is expressed as:

$$\bar{\alpha} = \frac{\alpha}{\sqrt{1 + I/I_{\text{SAT}}}}, \quad (4.13)$$

where α is the low-intensity absorption coefficient (linear regime). I_{SAT} is a parameter known as the saturation intensity, and is 2.503(3) mW/cm² for the D₂ transitions in rubidium atoms under normal conditions (i.e., inside evacuated buffer-gas-free vapour cell).

Two-Photon Absorption

In the case of the TPA in gases, the polarization vector can be rewritten into the following form:

$$\mathbf{P}(t) = \epsilon_0\chi^{(1)}\mathbf{E}(t) + \epsilon_0\chi^{(3)}\langle\mathbf{E}(t)^2\rangle\mathbf{E}(t) = \epsilon_0\chi_{\text{eff}}\mathbf{E}(t), \quad (4.14)$$

where $\chi_{\text{eff}} = \chi^{(1)} + \chi^{(3)}I$ is the effective electric susceptibility. Remembering that the complex refractive index is strongly related to the electric susceptibility of a material $\eta(\omega) = \sqrt{1 + \chi}$ (Section 2.1), in the non-linear regime, the effective susceptibility χ_{eff} should be considered. The complex refractive index can be expressed as a power-series expansion of the electrical susceptibility

$$\begin{aligned} \eta &\approx 1 + \frac{\chi_{\text{eff}}}{2} \\ &= 1 + \frac{\chi^{(1)}}{2} + \frac{\chi^{(3)}}{2}I \\ &= \eta_0 + \bar{\eta}I, \end{aligned} \quad (4.15)$$

where η_0 is the linear and $\bar{\eta}$ is the non-linear complex refractive index.

Therefore, the two-photon absorption coefficient $\bar{\alpha}$ can be expressed in the following form:

$$\bar{\alpha} = \alpha + \beta I, \quad (4.16)$$

where α corresponds to linear absorption and β is the TPA non-linear absorption coefficient. In this case, the refractive index is also intensity dependent and it can be written as:

$$n = n_0 + \bar{n}I, \quad (4.17)$$

where n_0 is the linear and $\bar{n}$ is the TPA-induced non-linear refractive index. The dependence of the refractive index on light intensity (i.e., scales quadratically with the electric field) is known as the *AC Kerr effect*.

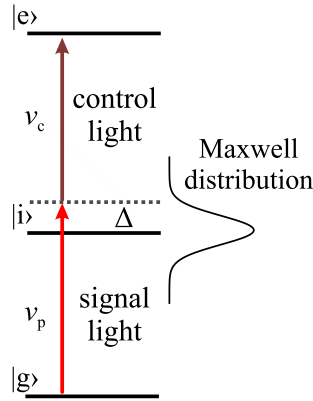


Figure 4.2: Simplified energy-level scheme for two-photon absorption with schematically marked atomic motion.

Now consider the dependence of the effective susceptibility [Eq. (4.14)] on the light frequency for the case depicted in Fig. 4.2. Herein, the sum frequency of weak probe beam of the frequency ω_p and strong coupling beam of frequency ω_c matches two-photon transition. In this case, the form of the absorption coefficient [given by Eq. (4.16)] does not guarantee the existence of the dispersion relation since the non-linear complex refractive index depends on two frequencies (ω_p and ω_c).

In such a case, the Kramers-Kronig relations cannot be applied. However, it is possible to redefine the problem using the perturbation formalism and try to find the form of the resulting dispersion.

It is straightforward to apply the linear Kramers-Kronig relations to the linear part of Eq. (4.15) [as shown in Section 4.1.1]:

$$n_0(\omega) = 1 + \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\alpha(\omega')}{\omega^2 - \omega'^2} d\omega'. \quad (4.18)$$

However, in the non-linear case, one has to make sure that no poles of the integral exist in the upper-half of the complex frequency plane for the Kramers-Kronig relations to be valid (i.e., the dispersion relations exist) [1, 93].

In the general case, the Kramers-Kronig relations can be modified by adding some external perturbation, thus the change in refractive index can be calculated from the change in the absorption coefficient due to some external perturbation [93]:

$$\Delta n(\omega; \zeta) = \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\Delta \alpha(\omega'; \zeta)}{\omega^2 - \omega'^2} d\omega', \quad (4.19)$$

where ζ denotes the perturbation, and the frequency ω' is independent from the perturbation. In the non-linear formalism, the perturbation must be constant on both sides of the relation and must not vary over the integral calculation, i.e., it must be a constant physical value. The change in the optical properties of the material is caused by a change in excited-state population, thus the excited-state population can be considered as a perturbation itself. It is important to ensure that the excited-state population stays constant, which can be achieved using a fixed frequency of the strong coupling light and a low power of the probe light in the experimental scheme. This ensures the assumption for the non-linear susceptibility is an analytical and continuous function over the upper part of the imaginary plane of the complex frequency is valid. For TPA, the control electric field ω_c can be a perturbation itself. In this case, the changes in refractive index $\Delta \bar{n}$ caused by the non-linear term can be described as [93]:

$$\Delta \bar{n}(\omega_p; \omega_c) = \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\beta(\omega'; \omega_c)}{\omega^2 - \omega'^2} d\omega', \quad (4.20)$$

where ω_c is the perturbation parameter, i.e., the frequency of a control laser, and ω_p is the probe-laser frequency.

4.2 Slow- and Fast-Light Media

The solution of integral given by Eq. (4.20), reveals a well-known dispersion profile. For an absorptive feature, a medium exhibits anomalous dispersion properties, while for gain, it exhibits normal dispersion. The refractive-index dependence determines the propagation of light pulses through the material (group velocity), when the conditions of strong dispersion may lead to very unusual light propagation condition (subluminal and superluminal light)

The speed of an electromagnetic wave propagating through a medium, is defined by various velocities. Particularly, the *phase velocity* v_p , which describes the rate the phase propagates through medium, is given by

$$v_p = \frac{\omega}{k}, \quad (4.21)$$

where ω is the angular frequency of the electromagnetic wave and k is the wavenumber. Therefore, the refractive index, which characterizes the group velocity, is nothing other than the ratio between the speed of light in a vacuum c and the phase velocity v_p :

$$n = \frac{c}{v_p}. \quad (4.22)$$

The *group velocity* v_g defines the speed at which the envelope of the wave, e.g., a light pulse, travels through space. It is defined as the derivative of frequency ω over wavenumber k :

$$v_g = \frac{d\omega}{dk} = \frac{c}{n(\omega) + \omega \frac{dn(\omega)}{d\omega}} = \frac{c}{n_g}, \quad (4.23)$$

which enables one to introduce the *group refractive index* n_g :

$$n_g = n(\omega) + \omega \frac{dn(\omega)}{d\omega}. \quad (4.24)$$

It is now clear that if the derivative of the refractive index over frequency is greater than the refractive-index value $\omega dn(\omega)/d\omega \gg n$, the lights group velocity

is significantly reduced with respect to its phase velocity. This situation appears for normal dispersion, and is known as the *slow-light* effect. Similarly, for anomalous dispersion, the group velocity becomes greater than the phase velocity, and can even be negative [94]. The fractional or negative values of group refractive index results in superluminal group velocities, i.e., *fast light*.

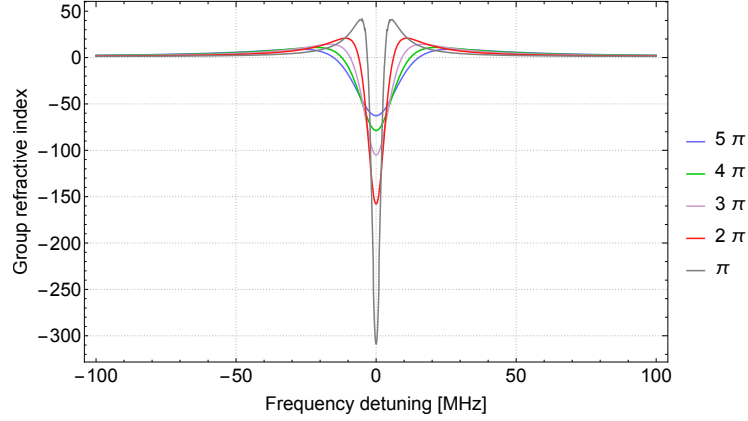


Figure 4.3: Group refractive index n_g simulated for the various widths of the transition but the same strengths. Refractive index was calculated with the data shown in Fig. 4.1b. The group refractive index becomes significantly different than unity for narrow resonance widths.

A simulation of the group-refractive-index values based on the refractive-index values calculated for Fig. 4.1b are shown in Fig. 4.3. The presented graph reveals negative-group-refractive-index values associated with a strong absorption in the centre of the atomic transition. A light pulse in a negative-group-refractive-index medium propagates backwards, thus, the pulse itself starts to appear at the output of the medium before it actually fully enters it [5, 94]. This agrees with the intuition and standard meaning of group velocity v_g . Backward propagation of a light pulse was experimentally demonstrated in Ref. [95]. However, it is not peak-to-peak propagation, a singularity point along which information travels, does not exceed the speed of light c [96].

The pulse in slow- or fast-light media become spatially compressed or expanded by a factor determined by the group-refractive-index value, respectively. At the same time, the pulses' peak energy changes by the same factor, so the pulse

intensity remains constant. Slow and fast light are complementary phenomena, and they are ruled by the same principals, and in this sense they are the same processes.

Therefore, fractional ($n_g < 1$) and negative values of group refractive index are associated with absorption in the medium. In atomic ensembles it was observed for *electromagnetically induced absorption* [97] and between two gain lines [98]. Slow light, observed for large values of n_g , appears for gain media. The narrow, sub-Doppler suppression of the absorption can produce significant delays of pulsed light. The largest reduction of group velocity (8 m/s) was achieved in coherently prepared atomic ensembles [99].

4.3 Two-Photon Absorption Spectroscopy in HC-PCF

Two-photon absorption is one of the most powerful Doppler-free spectroscopic methods. It allows for generation of the narrow sub-Doppler absorption resonances of high amplitude. This opens possibilities for numerous applications, as it allows for inducing strong interactions between two light beams through an atomic environment. It offers A platform for all-optical switching [6, 20], quantum logic devices [100], and quantum-memory devices [101].

A simplified energy-level scheme for TPA is shown in Fig. 4.2. TPA can be observed between ground $|g\rangle$ and excited $|e\rangle$ states, when atoms interact with both, the probe and control beams at respective frequencies of ν_p and ν_c . The two-photon transition can be achieved by two-step absorption in two step process by independent absorption, this means that each of the two of photons matches the energy of the two single-photon transitions ($|g\rangle \rightarrow |i\rangle$ and $|i\rangle \rightarrow |e\rangle$). However, the two-photon transition can be fulfilled when the sum energy of two photons matches the transition between the ground and excited states, without incorporating an intermediate state. The second case describes the situation when a so-called *virtual state* is incorporated in the TPA instead of a real intermediate one, and two

photons are simultaneously absorbed. In turn an otherwise forbidden transition can be excited. The process of TPA is enhanced if the virtual state lies close to the real intermediate state.

One may consider counter-propagating beams of equal frequencies ($\nu_p = \nu_c = \nu_L$). In the rest frame, each atom with velocity v_z interacts with two Doppler-shifted frequency waves:

$$\nu_p = \nu_L \left(1 - \frac{v_z}{c} \right), \quad (4.25)$$

$$\nu_c = \nu_L \left(1 + \frac{v_z}{c} \right). \quad (4.26)$$

The atoms in the ground state $|g\rangle$ can reach an excited state $|e\rangle$ by absorbing two photons. The resonant condition for an atom absorbing two counter-propagating photons is described by the difference between energy levels:

$$h\nu_e - h\nu_g = h\nu_L \left(1 - \frac{v_z}{c} \right) + h\nu_L \left(1 + \frac{v_z}{c} \right) = 2h\nu_L. \quad (4.27)$$

As shown, the Doppler terms cancel, and as a consequence the resonance condition is fulfilled for all atoms, irrespective of their velocity. This leads to significant enhancement of the absorption strength with respect to a single-photon absorption for a given ν_p . Under the condition of $\nu_p = \nu_c$, the linewidth of Doppler-free TPA is predicted to be Lorentzian, with the natural linewidth of excited state $|e\rangle$ [69].

However, if the frequencies of the two lasers are not identical, residual Doppler broadening will occur:

$$E_e - E_g = h(\nu_p + \nu_c) + h(\nu_p - \nu_c) \frac{v_c}{c}. \quad (4.28)$$

The Doppler broadening of a two-photon transition is narrower than for a single-photon transition, while simultaneously showing significantly enhanced absorption (Fig. 4.4). Additional enhancement of the TPA amplitude can be achieved if the virtual state lies close to an existing intermediate state $|i\rangle$.

A particular system supporting TPA are rubidium vapours, where the ground state $|g\rangle$ is the state $5S_{1/2}$; the intermediate state $|i\rangle$ is $5P_{3/2}$ and the excited level $|e\rangle$ can be the $5D_{5/2}$ state. This system is used for TPA studies within this dissertation.

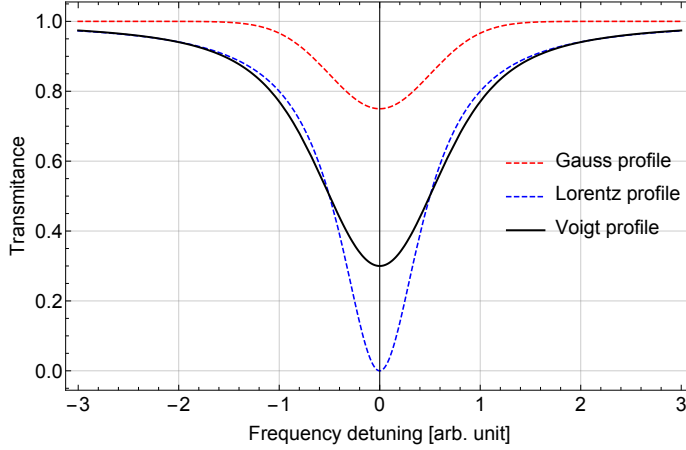


Figure 4.4: Simulation of a TPA profile for two-colour counter-propagating beams [Eq. (4.28)]. A narrow Lorentzian profile (red dashed line) is convolved with a broad Gaussian function (blue dashed line).

4.3.1 Two-Photon Absorption in Rubidium Atoms

A detailed energy-level scheme of rubidium atom is presented in Fig. 4.5. As shown, the transition frequencies between the $5S_{1/2}$ and $5P_{3/2}$ levels and the $5P_{3/2}$ and $5D_{5/2}$ levels are 384 THz and 386 THz, respectively, which allows for significant reduction of Doppler broadening.

Taking into account that the TPA coefficient depends on light intensity [Eq. (4.16)], and that all the atoms take part in TPA, regardless of their velocity, a fibre host seems to be well suited for non-linear experiments exploiting TPA.

4.3.2 Experimental Setup

The experimental setup utilised in the fast-light and TPA experiments is presented in Fig. 4.6. A detailed description of the apparatus follows.

Kagome-latticed HC-PCF

The kagome-latticed HC-PCF used in the TPA experiment was described previously in Section 2.3 and was utilised in the pulsed-LIAD experiment (Section 3.2). It is the large-pitch kagome-structured HC-PCF (Fibre 2) with a core diameter of $45\ \mu\text{m}$. Transmission of coupled light was around 25%.

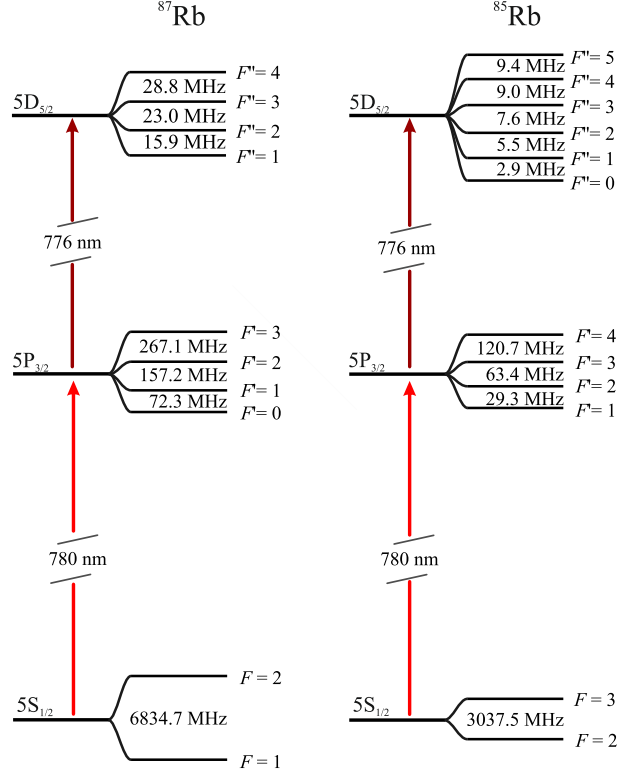


Figure 4.5: Energy-level scheme of rubidium 87 and rubidium 85.

Lasers

The lasers used in the experiment were two home-built ECDLs, one of which is presented in Fig. 4.7. One of the lasers (used as a control beam) was constructed for the purpose of this project. An anti-reflection coated high-power diode⁴⁴ operating over a wide range of wavelengths (765 - 805 nm) was used in the laser. The diode could only lase when an external feedback (cavity) was provided. The external cavity was designed using a diffraction grating in the Littrow configuration, where the feedback is via the first-order reflection from the grating. The laser output was obtained via the zeroth order of reflection. The cavity could be adjusted with three adjustment screws (two of them are visible in Fig. 4.7). Fine tuning was provided via a piezoelectric element. The laser diode was thermally stabilised by a Peltier cooling module placed underneath the diode. The laser was driven by

⁴⁴Toptica: LD-0790-0120-AR-1

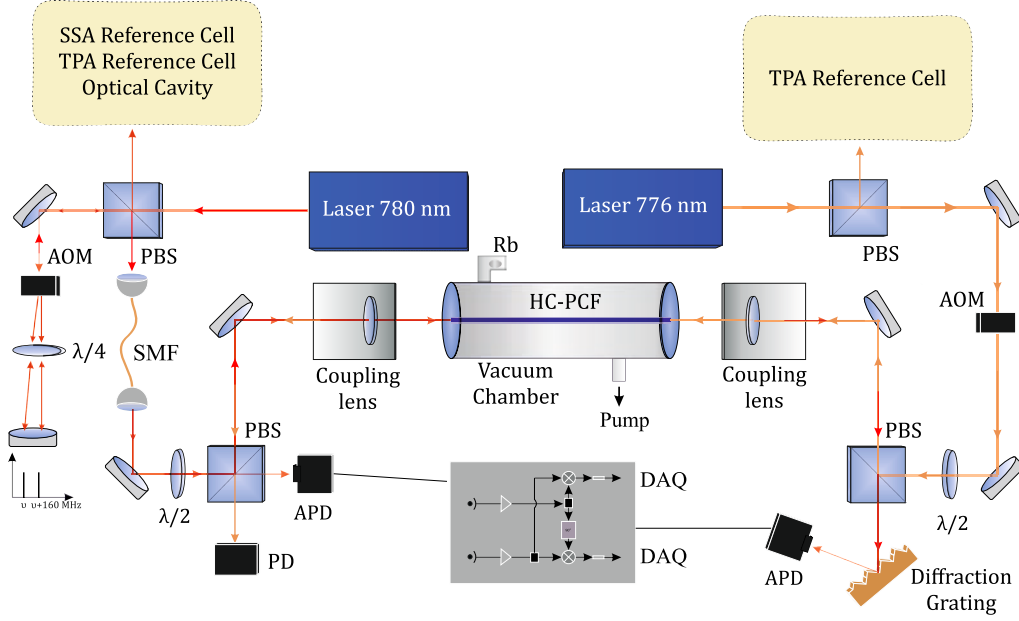


Figure 4.6: Experimental setup used for two-photon absorption and fast light experiment. PBS is polarising beam splitter, SMF is a single-mode fibre, and DAQ is a data acquisition setup.

a commercial controller⁴⁵. This laser, due to its wide tunability, could operate at 776 nm, which corresponds to the transition from $5P_{3/2}$ to $5D_{5/2}$ in rubidium atoms.

A second ECDL (used as the probe laser) was tuned to the D_2 transition in rubidium (780 nm) and its wavelength was monitored with the SAS system. To ensure the stable operation of both lasers, external optical isolators were introduced.

Laser Locking System

Laser locking systems were provided to stabilise the wavelength of both lasers, and prevent for their frequency from drifting during the measurements. The probe laser was locked to the stable frequency-standard laser present in the laboratory. A detailed description and setup of the locking method was provided in Section 3.2.1. The second laser was stabilised using the two-photon transition. The locking setup

⁴⁵MOGLabs: External Cavity Diode Laser Controller DLC202

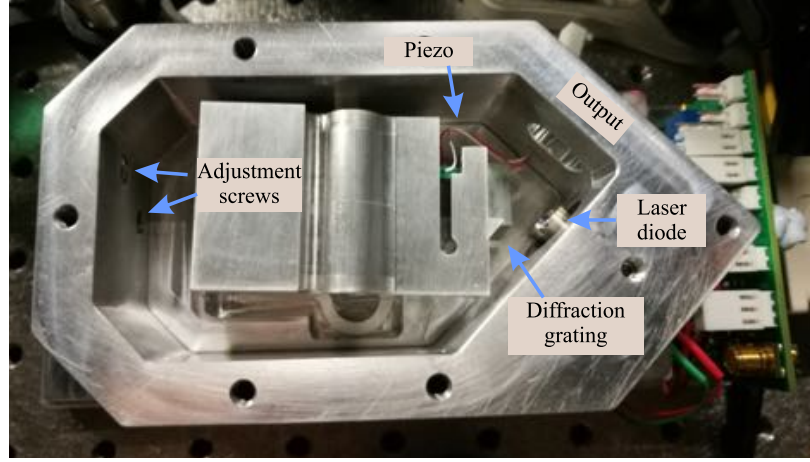


Figure 4.7: External-cavity diode laser used for 776-nm light generation (control light).

is presented in Fig. 4.8. The locking signal was obtained from the TPA reference cell (heated to approximately 80°C), where both the probe and control lasers were used to illuminate the atoms. A photo-multiplying tube (PMT) was used to detect a fluorescence signal at a wavelength of 420 nm produced when atoms were coupled to the $5D_{5/2}$ state i.e., the two-photon resonance. The blue-light fluorescence was generated when atoms decayed from the metastable $6P_{3/2}$ state to the $5S_{1/2}$ ground level (Fig. 4.8a). This fluorescence signal was used to generate an error signal for the laser feedback (Fig. 4.8b). This was achieved by frequency modulating 776-nm light delivered to the TPA reference cell by double-passing an AOM driven with 80-MHz electric signal generated by a frequency generator. The signal from the PMT was mixed with the modulating signal, the sum frequency was filtered out with a low-pass filter, and the difference signal was sent to the laser controller. The feedback was provided to the current module as well as to the piezo-module for fine adjustment. In addition, the wavelength of the 776-nm laser was also monitored on a wavemeter.

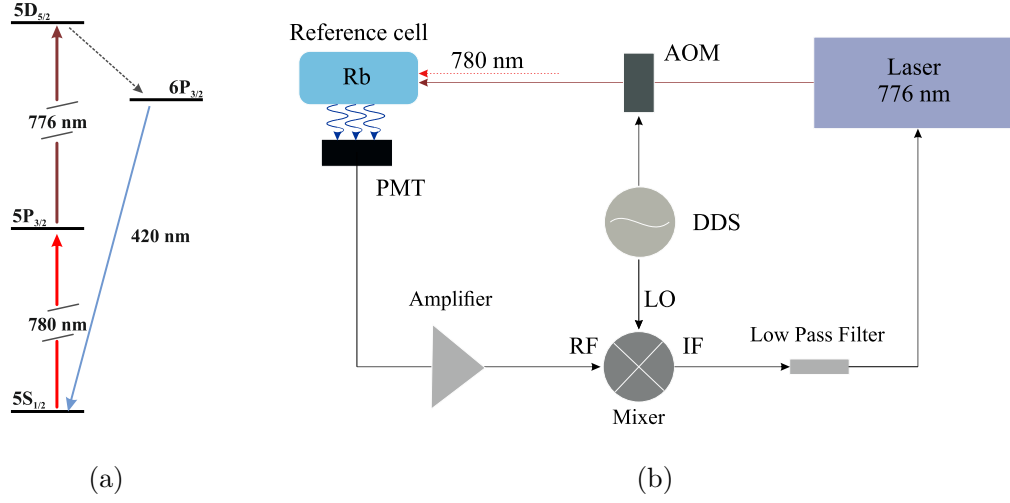


Figure 4.8: (a) Energy-level scheme for rubidium atoms with relaxation channels of the level populated with the two-photon excitation. (b) The scheme of the locking setup for 776-nm laser.

Measurement Techniques

To investigate the absorption and dispersion in a TPA medium, a heterodyne optical detection method [7] was introduced. The light from the probe laser was sent to an AOM, where it was split into two beams of equal optical power directed into zeroth and first order of refraction. Both beams were retro-reflected so that they double passed through the AOM, and produced a frequency shift of 160 MHz between them (see experimental setup shown in Fig. 4.6). This leads to a beat-note signal. The large frequency separation between the produced beams (160 MHz) ensured that only one of them would interact with the control light, and the other would act as an unperturbed reference, i.e., the separation is larger than the width of the $5D_{5/2}$ manifold. Thus, the phase changes experienced by both beams were different, e.g., only one of them would be absorbed in the TPA process. Further, both beams were coupled to a single-mode fibre (SMF) to ensure a spatial overlap. The beat-notes of the beams were measured before and after the rubidium-filled fibre on identical APDs. The initial beat-note signal was used as a reference signal, while the beat-note measured after the fibre (experimental signal) was combined with the reference signal to produce two quadratures X and Y (Fig. 4.9).

A phase-shift in one of the interfering beams resulted in a phase shift of the beat-note signal. The amplitude of the beat-note is proportional to the absorption amplitude. In this manner the complex refractive index could be measured. The absorption and phase-shift can be calculated in the following way:

$$\alpha = \sqrt{X^2 + Y^2}, \quad (4.29)$$

$$\Delta\phi = \arctan\left(\frac{Y}{X}\right). \quad (4.30)$$

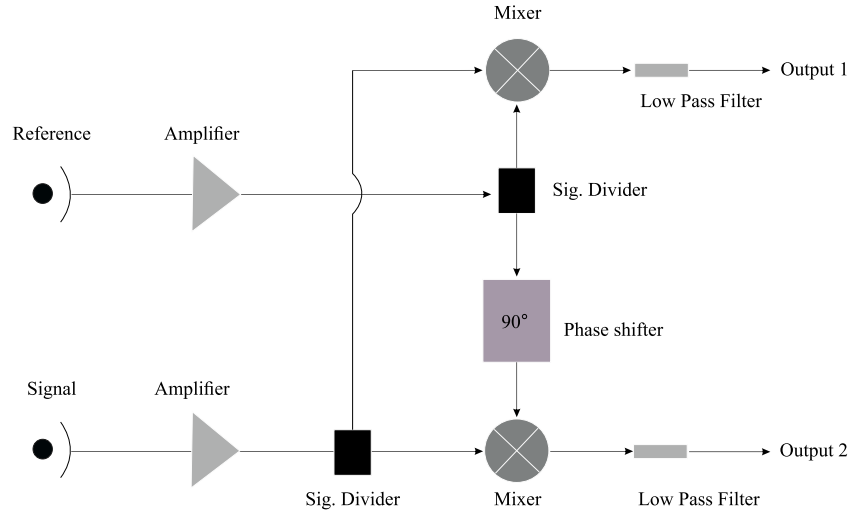


Figure 4.9: Schematic of the signal-processing setup. The reference and experimental interference signals were measured on identical APDs and then amplified. The reference is split into two signals, one of them is directly down-mixed with the experimental beat-note, while the other is shifted by 90° on the phase shifter and they are used as a LO signals to generate two quadratures X and Y . The high frequencies are filtered out with identical low-pass filters (48 MHz), and the two output signals were measured on an oscilloscope.

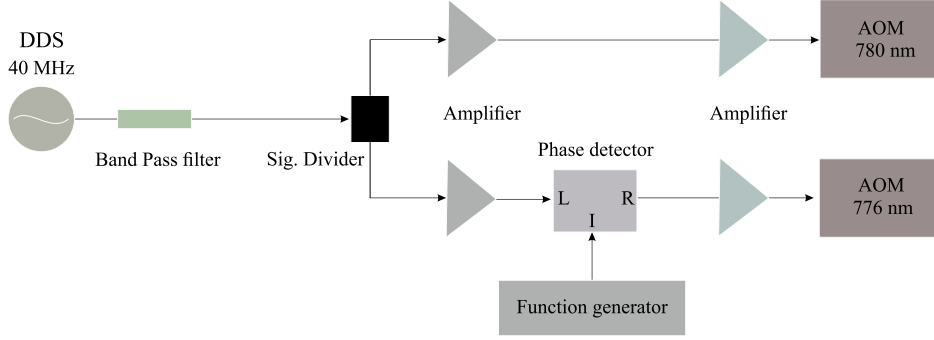


Figure 4.10: Scheme of the AOM driving setup. The amplitude modulated signal on the 776 nm AOM enabled pulse formation. The modulating square-wave signal comes from a function generator, which is fed to the AOM driving the control laser.

The changes in the absorption and dispersion of the atomic medium filling the fibre were measured by introducing $0.5\text{-}\mu\text{s}$ square pulses on the control field. To ensure fast switching of the AOM responsible for pulsing the light, a bespoke electronic setup was built (Fig. 4.10). The 80-MHz signal was amplitude modulated with a square signal from a function generator and fed to the AOM, varying the control-laser intensity. A non-modulated version of the signal was amplified and fed to the AOM driving the probe laser.

Since the AOM shifts the frequency of the beam by 160 MHz, the laser locking beam also had to be shifted by the same amount to produce TPA at the same locking point. A summary of all the beams from the probe and coupling lasers are shown in Fig. 4.11.

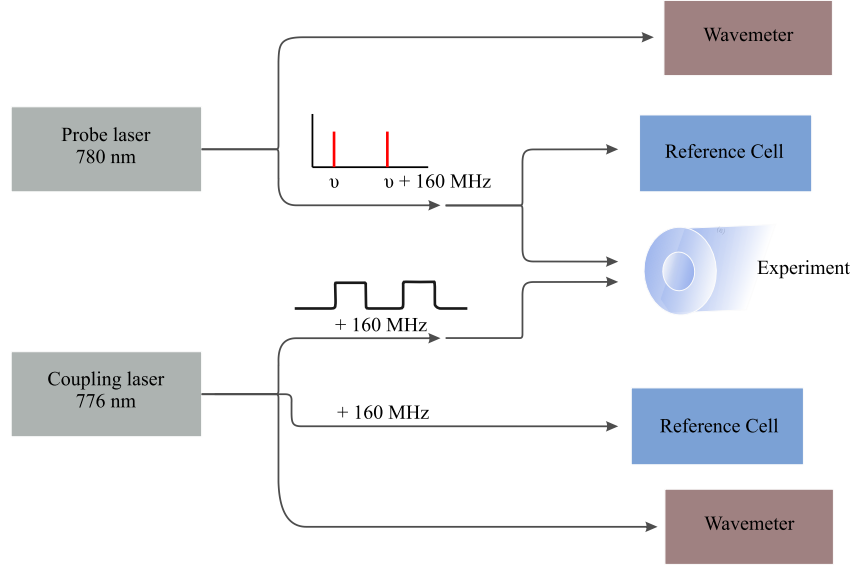


Figure 4.11: Schematic of the probe (780 nm) and control (776 nm) laser-beam layouts in the TPA experiment. The probe beam is divided into two equally-strong beams, separated by 160 MHz, and sent to both the experiment and reference cells. The control beam is divided into two beams, one of which is used to produce pulses with a frequency shift of 160 MHz while the other is a CW beam, also shifted by 160 MHz, and sent into the reference cell to provide wavelength stabilization. Some fraction of each of the lasers was sent to a wavemeter.

4.3.3 Results

The two-photon absorption-experiment was realised in rubidium 85. The weak (~ 20 nW) probe beam was scanned over the ^{85}Rb : $5S_{1/2} F = 3 \rightarrow 5P_{3/2} F'' = 2, 3, 4$ transition to obtain a reference measurement. The OD for a single-photon transition was so high that the transmittance was equal to zero in the vicinity of a resonance centre.

The CW control beam was detuned from the two-photon transition by 770 MHz. The scheme of the energy levels taking part in this transmission are marked as solid black lines in Fig. 4.12c. The spectra obtained in the presence of the control beam is plotted in Fig. 4.12a. TPA resonances come from two probe beams separated by 160 MHz. The middle feature appears exactly half-way between two transitions, which is a crossover resonance [68]. Figure 4.12b shows a zoomed ver-

sion of TPA, where the blue points mark the measured data and the solid grey line is a theoretical fit.

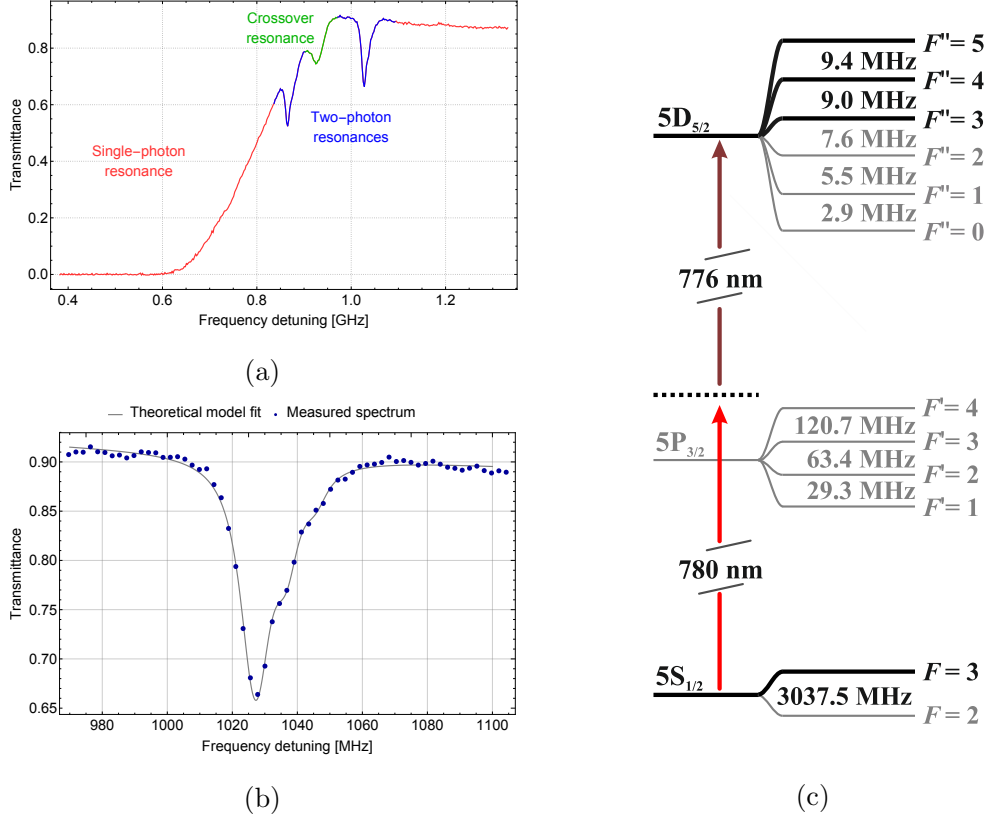


Figure 4.12: Two-photon absorption in the rubidium-filled HC-PCF. (a) Spectra from the fibre. The light red points show the transition from $5S_{1/2}$ $F=3$ to $5P_{3/2}$ $F=2,3,4$ with the TPA transition on the side (light blue points). The two dips come from two beams separated by 160 MHz and the middle feature is a crossover (green). (b) Zoomed TPA with a theoretical model fit. (c) The energy-level scheme of ^{85}Rb used for the TPA. The black thick lines indicate the states taking part in TPA.

The linewidth of TPA for two-colour laser spectroscopy is reflected by the Voigt profile. The hyperfine-level structure of the $5D_{5/2}$ manifold is visible in the acquired spectrum (see Fig. 4.12b). Therefore, the sum of the Voigt functions,

normalised to unity height, were fitted to obtain the broadening value:

$$T = \sum_i \alpha_i V(\nu, \gamma_i, \Gamma_D), \quad (4.31)$$

where T is the transmittance, α_i is the optical depth of i -th transition, $V(\nu, \gamma_i, \Gamma_D)$ is the Voigt function (the full function was shown in Section 3.2.2), Γ_D is the Doppler width and γ_i is the Lorentzian width of the i -th transition (FWHM). The distances between transitions were fixed to their theoretical values [102] (Fig. 4.12c). The results of fitting the function [Eq. (4.31)] to the observed spectrum are presented in Table 4.1.

The precisely determined lifetime of the $5D_{5/2}$ state in bulk is 238 ns [103]. This gives a natural linewidth of the level of 668.7 kHz (FWHM). However, there are various contributions to the TPA resonance broadening, one of them has already been carefully discussed, which is Doppler broadening. The other contributions were carefully measured and discussed for Fibre 2 in Ref. [77]. Particularly, the broadening results from the intermediate-state detuning, transit time of the atoms between the fibre walls, atom-surface interaction, and ambient magnetic field. The intermediate-state detuning seems to play a significant role, since the laser is close to the broadened single-photon resonance ($\Delta = 770$ MHz). The lifetime of the excited state of the atom inside the fibre is determined by the time of flight across the fibre core, predicted to be 123(3) ns, i.e., a broadening of 5.61(5) MHz [86]. For comparison the Doppler width is predicted to be only 1.9 MHz at 350 K, and this is the fixed value used in the theoretical model.

It should be noted that the TPA resonance amplitudes of transitions do not agree with the theoretical values plotted in Fig. 4.13. It is believed that this is caused by saturation of the stronger transitions (despite an extremely low probe light power of ~ 20 nW).

A simple pulsed measurement was performed to estimate the saturation value and the dependence of the absorption coefficient on the control-beam intensity. This is also the simplest realisation of all-optical switch, where one beam can be amplitude modulated by another. For this measurement, the lasers were locked at the exact TPA transition, and the 0.5- μ s square pulses of the control light, carrying

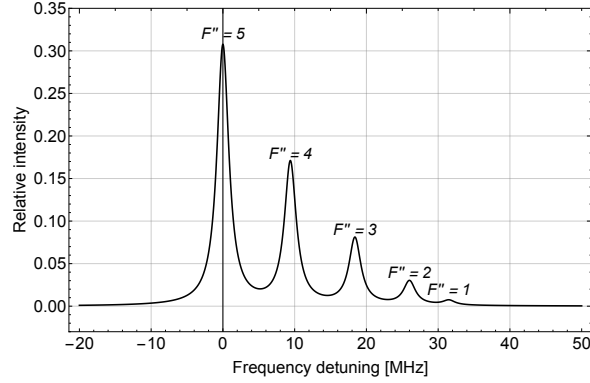


Figure 4.13: Modelled spectrum (Lorentz profiles) of the TPA spectrum in ^{85}Rb with all hyperfine transitions of the $5D_{5/2}$ state. The strength and separation of the lines are theoretical values from Refs. [102, 103].

α_5	α_4	α_3	γ_5 [MHz]	$\gamma_{4,3}$ [MHz]	Γ_D [MHz]
0.305(6)	0.097(6)	0.031(5)	9.1(3)	8(1)	1.9

Table 4.1: Coefficients extracted from fitting the TPA spectrum with the function given by Eq. (4.31). The theoretical amplitude ratio is 1:0.5:0.25 and the natural linewidth is 668.7 kHz for all hyperfine levels.

different optical powers, were introduced. Since the probe beam was detuned from the single-photon resonance, without the control beam its transmission was equal to unity. In the presence of the control beam, the probe was absorbed due to TPA. This was seen as a decrease in the transmission, Fig. 4.14a shows pulses from the probe light absorption measured in OD. The dependence of the OD on the control-beam intensity is plotted in Fig. 4.14b. The theoretical assumption [Eq. (4.16)] predicts that the non-linear TPA coefficient depends linearly on light intensity. This dependence is shown in Fig. 4.14c. Intensities above 1.6 W/cm^2 saturates the transition, and is seen as a plateau. For further experiment, two intensities were selected: 0.16 W/cm^2 and 0.64 W/cm^2 , marked as solid red and blue lines in Fig. 4.14a, respectively.

All-optical switching requires strong atom-photon interaction. This is shown by the absorption of the probe beam modulated by the control laser tuned to the TPA resonance.

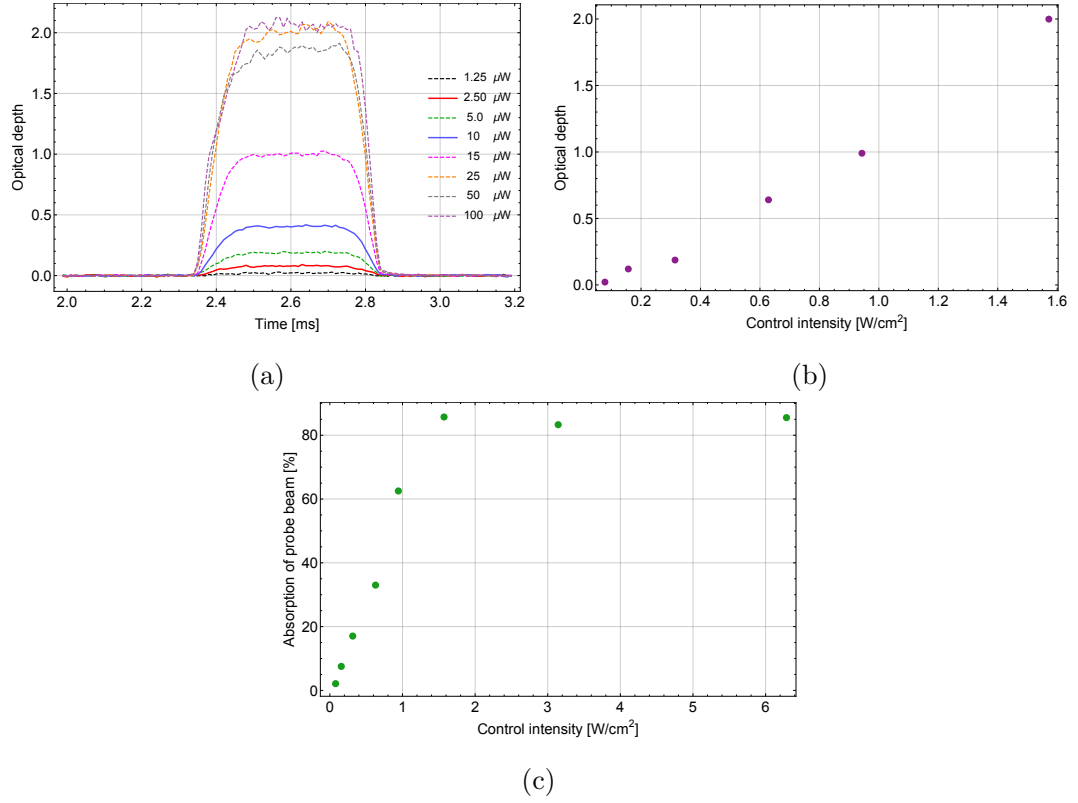


Figure 4.14: Dependence of the probe-beam absorption due to TPA, recalculated into optical depth. (a) The transmission of the probe beam over time induced by the presence of the control field. (b) The dependency of the OD (shortened to linear values), (c) and the absorption percentage on the control-beam intensity.

4.4 Fast Light in HC-PCF

The following studies presented in this dissertation concentrate on controlling the changes of the refractive-index and group-velocity obtained within the vapour-filled HC-PCF. This is based on the fact that the absorptive profile of a two-photon transition is associated with an anomalous dispersion profile, which can be examined by introducing light pulses and measuring their advancement. The amplitude of the dispersion can be easily controlled by altering the intensity of the interacting beams (Fig. 4.14). This opens the possibility for realising a tunable fast-light all-optical switch.

A fast-light medium allows for the realisation of various experiments and ap-

plications. The most important are: the storage of light [101], large cross-phase shifts per photon [6], and four-wave mixing [104].

4.4.1 Results

To obtain absorption and dispersion profiles of rubidium vapours inside of Fibre 2, the probe laser was locked to 770 MHz off the centre of a single-photon absorption resonance. This light (~ 20 nW, $2.83 \mu\text{W}/\text{cm}^2$ inside the fibre) was coupled to the fibre, where it interacted with rubidium. The control laser was locked to the TPA transition. The AOM, forming $0.5\text{-}\mu\text{s}$ pulses of the control field, was used to precisely tune the control field around the two-photon resonance. In the presence of the control field, the probe beam was absorbed.

The absorption spectra and dispersion profiles were obtained for two control beam powers $0.16 \text{ W}/\text{cm}^2$ and $0.63 \text{ W}/\text{cm}^2$ (intensities inside the fibre). The absorption spectra are presented in Figs. 4.15a and 4.15b (the red and blue points are for $0.16 \text{ W}/\text{cm}^2$ and $0.63 \text{ W}/\text{cm}^2$, respectively). There are three transitions (to $5D_{5/2} F'' = 5, 4, 3$)⁴⁶ visible in the plots. Their theoretical positions are marked as vertical lines, which also indicate the relative intensity of the transitions (11/36, 1/6, 7/90 and 1/36) [102]. The theoretical model, which is a sum of three Voigt profiles [Eq. (4.31)], was fitted to the experimental data. The received fitting coefficients are presented in Table 4.2. The observed amplitudes roughly correspond to the theoretical values giving an amplitude ratio of 1:0.3:0.1. In the same way as for the CW TPA experiment (Section 4.3.3), the observed transitions are much broader than the theoretically predicted values. The main reasons for that are: transit-time broadening, and intermediate-state detuning. An increase in the control power by a factor of 4 leads to an exact 4-fold increase in the observed resonance amplitudes.

Changes in the refractive index are related to the phase shift in the following

⁴⁶Transitions to $5D_{5/2} F'' = 2$ and 1 are located further than the scan range. For the control power of $0.63 \text{ W}/\text{cm}^2$ the fourth transition can be seen as a deviation of the point at 24 MHz frequency detuning.

manner:

$$\Delta\phi = k(\omega)l, \quad (4.32)$$

$$k(\omega) = \frac{\Delta n(\omega)\omega}{c}, \quad (4.33)$$

where $k(\omega)$ is the wavevector, l is the length of a medium, and Δn is the refractive-index change.

The imaginary part of the Voigt function was used to fit the refractive index signal (the solid line in Figs. 4.15c and 4.15d). The fitting coefficients are summarised in Table 4.2. The dashed lines in Figs. 4.15c and 4.15d are the results of a calculation of the refractive index using the absorption coefficient and the Kramers-Kronig relations (Section 4.1).

Fitting parameters of absorption spectra.						
	α_5	α_4	α_3	γ_5 [MHz]	$\gamma_{4,3}$ [MHz]	Γ_D [MHz]
cw spectra	0.305(6)	0.097(6)	0.031(5)	9.1(3)	8(1)	1.9
0.63 W/cm ²	0.65(1)	0.20(1)	0.034(22)	7.2(3)	6(1)	1.9
0.16 W/cm ²	0.164(2)	0.047(3)	0.016(3)	8.9(2)	6.0(7)	1.9
Fitting parameters of dispersion function.						
	α_5	α_4	α_3	γ_5 [MHz]	$\gamma_{4,3}$ [MHz]	Γ_D [MHz]
0.63 W/cm ²	0.51(1)	0.12(2)	0.027(13)	6.7(5)	6(1)	1.9
0.16 W/cm ²	0.138(2)	0.021(5)	0.005(2)	8.5(4)	7(1)	1.9

Table 4.2: Coefficients resulting from fitting Voigt profiles to the measured spectra. The $\alpha_{5,4,3}$ coefficients represent the strength of the transitions to the $5D_{5/2}$ $F'' = 5, 4$ and 3 hyperfine levels, respectively, γ is the width of the Lorentzian profile (natural width) and Γ_D is the Doppler width.

The negative refractive index change for a 0.63 W/cm² control beam is $\Delta n \approx -2.37 \times 10^{-7}$, and $\Delta n \approx -6.73 \times 10^{-8}$ for a light intensity of 0.16 W/cm². This is an order of magnitude lower than the refractive index change reported for anomalous dispersion between two gain lines in thermal cesium vapours [98]. However, in a TPA scheme, dynamic control over the refractive index can be achieved by

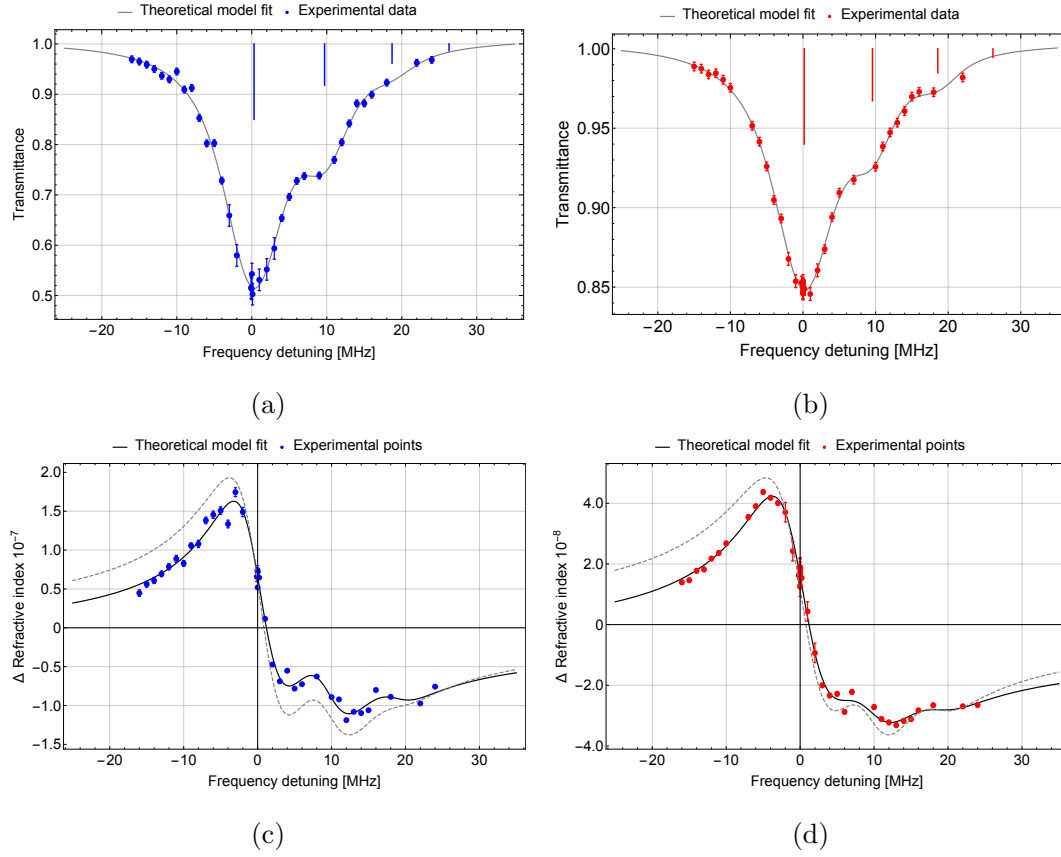


Figure 4.15: Experimentally measured two-photon transition spectra along with a theoretical fit for a control-beam intensity of (a) 0.63 W/cm^2 and (b) 0.16 W/cm^2 . (c) and (d) present the corresponding refractive-index change along with the fit (solid line). The dashed lines in (c) and (d) represent the curve based on the absorption data. The positions of the resonances are marked with vertical lines, whose lengths correspond to the relative transition strengths.

modulating the control light. In addition, the use of the fibre as a host lowered the light power necessary to generate significant changes in the dispersive properties of the medium (inducing gain lines in atomic vapours requires a high optical power).

Negative-refractive-index changes lead to a negative group velocity, corresponding to a negative pulse delay in the medium. In practice, a negative pulse delay means an advancement of the pulse with respect to the normal propagation of the same pulse in a vacuum. The group velocity can be calculated by taking derivation of the refractive index over frequency (see Section 4.2). Further, the

pulse advancement in the fibre can be calculated as follows

$$t_{adv} = t_{vac} - \frac{l}{v_g}, \quad (4.34)$$

where t_{vac} is the travel time of the pulse in vacuum⁴⁷, l is the length of the fibre, and $v_g = c/n_g$ is the group velocity. The group refractive index n_g and pulse advancement for the predicted group velocity is shown in Fig. 4.16⁴⁸. The light grey shaded area indicates the region of negative group refractive index, where the pulse is in the fast-light regime. Positive values of group refractive index leads to the slow-light effect, which is marked with dark grey.

To intuitively understand the pulse delay or advancement in the system, where a CW probe is used, one has to imagine the situation where the CW beam is formed by a sequence of pulses. One of these pulses is affected by the refractive-index change. Therefore, the pulse will travel with a group velocity dictated by the temporal change of the group refractive index caused by the presence of coupled light.

The red and blue points represent experimental data measured with control beams of 0.16 W/cm² and 0.63 W/cm², respectively. Pulse advancement was observed as a time difference between the appearance of the pulse on the photodiode with respect to the signal generating pulse. There is good agreement between the measured advancement and the one theoretically predicted for the lower power control beam (0.16 W/cm²), however, the points have large error bars and are widely spread. The experimental data for the higher control-light intensity (0.63 W/cm²) is in high disagreement with the predicted advancement. It appears greatly broadened, and the side peaks of advancement, which come from the weaker hyperfine transitions, are shifted. The most suggestive reason is that the pulse is being reshaped in the dispersive media, which will be of further consideration.

⁴⁷The refractive index of the fundamental mode is ≈ 1 .

⁴⁸Group refractive index values are coincidentally similar to the pulse advancement values due to the fibre length being ≈ 30 cm.

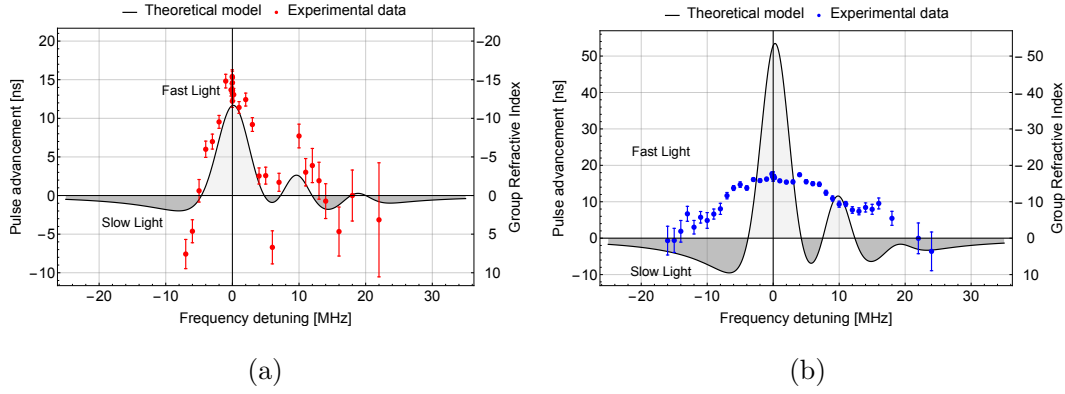


Figure 4.16: Pulse advancement (left axis) and group refractive index (right axis) for the vapour-filled fibre for two different optical intensities (a) 0.16 W/cm^2 (red points) and (b) 0.63 W/cm^2 (blue points). A larger advancement was observed with the higher control beam intensity, which agrees with the theoretical model. The light grey shaded region represents fast-light propagation and dark shaded regions represent slow-light propagation.

4.4.2 Pulse Reshaping in a Fast-light Medium

A major problem with pulse propagation in a fast-light medium is the strong pulse-reshaping effect. This reshaping arises from the dependence of the group-velocity index on the spectrum of the pulse. Due to this effect, different frequency components experience distinct refractive indices, which results in the pulse chirping and deforming. The amount of chirp imposed on the pulse can be estimated by the group velocity dispersion (GVD) [67, 94, 105].

The spectral width of a $0.5\text{-}\mu\text{s}$ pulse, used in the experiment, is 318 kHz. This seems to be quite narrow, however the Fourier transform of the square pulse into the frequency domain gives a $\text{sinc}(\omega)$ function⁴⁹. Thus, the actual square shape of the pulse is highly complicated, and requires solving complicated integrals. To simplify the considerations, GVD for a Gaussian pulse will be estimated first to predict the scale of the pulse reshaping.

⁴⁹The sinc function is defined as $\text{sinc}(x) = \frac{\sin(x)}{x}$ for $x \neq 0$.

Group Velocity Dispersion for Gaussian Pulse

A Gaussian pulse in the time domain can be expressed as:

$$E(t) = A \exp\left(\frac{-t^2}{4\sigma^2}\right) \exp(-i\omega_0 t), \quad (4.35)$$

where σ is its temporal pulse width, ω_0 is its central angular frequency, and A is its amplitude. A Fourier transform of the pulse in the angular frequency domain gives following spectrum of the pulse

$$E(\omega) = A' \exp\left(-\frac{(\omega - \omega_0)^2}{\sigma^2}\right). \quad (4.36)$$

The phase accumulation, gained by the pulse after passing through a dispersive medium, is $\Delta\phi = k(\omega)l$, where $k(\omega)$ is the wavevector and l is the length of the medium. After passing the medium, the pulse is described by

$$E(\omega) = A' \exp\left(-\frac{(\omega - \omega_0)^2}{4(\frac{1}{2}\sigma)^2}\right) \exp(ik(\omega)l). \quad (4.37)$$

The wavevector k can be expressed in terms of the refractive index, $k(\omega) = \frac{n(\omega)\omega}{c}$, and approximated by its Taylor expansion around the central frequency ω_0 in the following manner:

$$\begin{aligned} k(\omega) = \frac{n(\omega)\omega}{c} &= \frac{n(\omega_0)\omega_0}{c} + \left(\frac{n(\omega_0) + n'(\omega_0)\omega_0}{c}\right)(\omega - \omega_0) + \\ &+ \frac{1}{2} \left(\frac{2n'(\omega_0) + n''(\omega_0)\omega_0}{c}\right)(\omega - \omega_0)^2 + \dots \end{aligned} \quad (4.38)$$

The first term in Eq. (4.38) is the refractive index, i.e., the refractive index associated with non-linear interaction. The second term arises due to the group refractive index, and the third stems from the group velocity dispersion (GVD). The GVD calculated for the a 0.64 mW/cm² pulse is presented in Fig. 4.17.

The ‘amount’ of reshaping can be calculated as a multiplication of the GVD, the pulse bandwidth and the mediums length, and hence the total bandwidth of the pulse after passing the medium is described by [105]:

$$\sigma_{chirp} = \sqrt{\sigma^2 + \left(l \cdot \text{GVD}(\omega_0) \cdot \frac{1}{2\sigma}\right)^2}. \quad (4.39)$$

For the 0.5- μ s pulse, the expected pulse duration after passing the medium is $\sigma_{chirp} = 0.500324 \mu\text{s}$, which seems negligible. The value of the GVD for a control

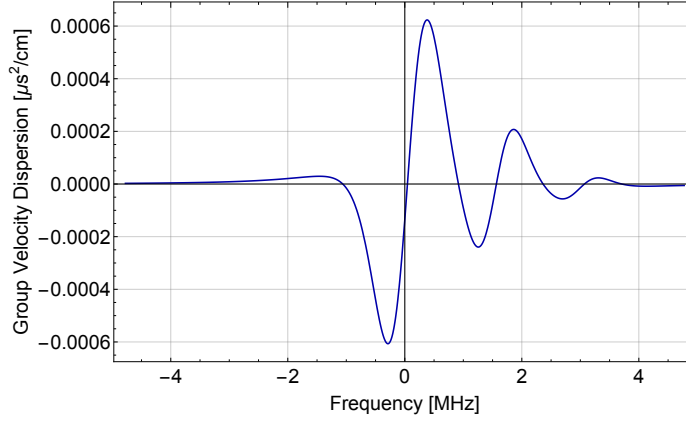


Figure 4.17: Group velocity dispersion of a 0.64 mW/cm^2 -pulse in the fibre system.

beam of intensity 0.16 W/cm^2 is between -0.00012 and $0.00012 \mu\text{s}^2/\text{cm}$. It has even less influence on the pulse shape in a dispersive media. Therefore, the GVD for a square pulse will be considered.

Group Velocity Dispersion for Square Pulse

The actual amount of chirp for a square pulse can be calculated with a numerical simulation. A square pulse in the frequency domain is represented by a sinc function, which contains a large amount of harmonics. Calculations for a square pulse are the same as for a Gaussian pulse, except for the pulse shape difference. A square pulse in the frequency domain must be multiplied by a term which describes the phase accumulation after passing through a dispersive medium [Eq. (4.37)]. To calculate the pulse chirp, it must be transformed back to the time domain using an inverse Fourier transform. This is ‘nasty’ integral, and while it can be calculated numerically, it gives very uncertain results (see Fig. 4.19); these results show that for far detuned pulses, the integrals calculated for the sharp edges of the pulse do not converge properly (Fig. 4.18a). Pulses centred closer to the two-photon resonance are highly reshaped regardless of the density of points (each point in the Figs. 4.18a to 4.18c represents one integral calculated for this point). However, from the simulated shapes it can be tentatively concluded that the pulses do not experience significant expansion.

Position of the pulses converted back into the time domain reproduce the

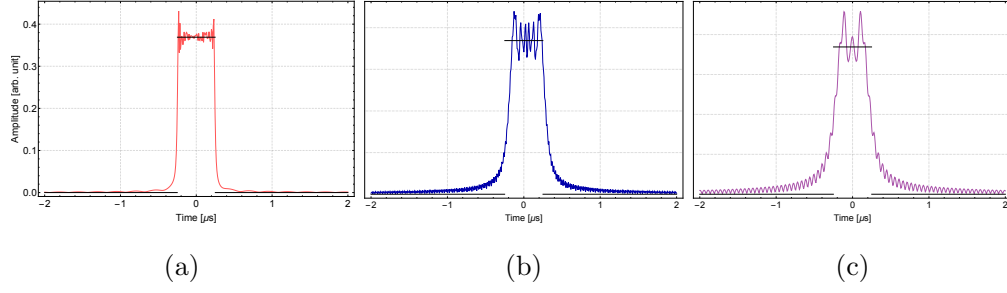


Figure 4.18: Square-pulse shapes calculated in atomic vapours under the TPA condition for a 0.64 W/cm^2 -intensity control beam for different central frequency detunings: (a) -30 MHz , (b) 1 MHz , and (c) 6 MHz . Black lines indicate the initial pulse in the time domain.

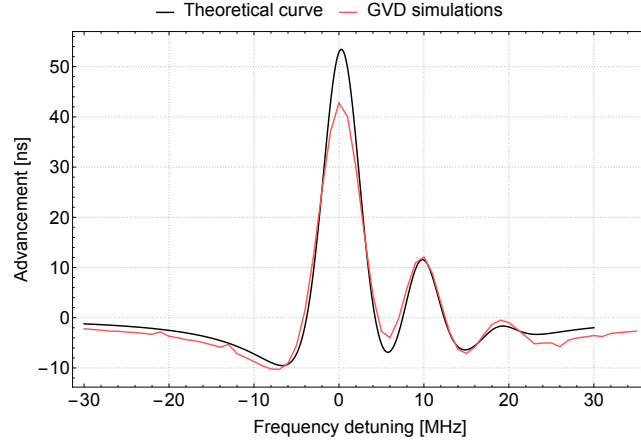


Figure 4.19: Simulated pulse advancement (light red line) compared with a calculated value of advancement from the Kramers-Kronig relations (black line).

dispersion curve calculated from the Kramers-Kronig relations (Fig. 4.16b), which is compared in Fig. 4.19. This is quite surprising result, since the calculated square-pulse shapes (see Fig. 4.18) seem unreliable.

The estimated pulse reshaping due to GVD for Gaussian pulses appear as negligible corrections to the pulse duration after passing the dispersive medium. However, the square-pulse reshaping seems to cause slight reduction of the advancement, this is shown in Fig. 4.19. Therefore, the dispersion of the group velocity is not clearly responsible for the discrepancy between the predicted pulse advancement and the measured one (see Fig. 4.16). This indicates presence of

additional effects that are not considered in the model.

4.5 Rabi Oscillations

The application of two light beams that couple the ground and excited states through TPA leads to the creation of a coherence between the states $5S_{1/2}$ and $5D_{5/2}$. As is typical for such a scheme, the coherence creation manifests through damped Rabi oscillations. These oscillations were observed on the rising edge of the absorption pulse for a stronger coupling beam (0.63 W/cm^2). This was carefully examined and included in the pulse appearance determination for the pulse advancement measurement. Below, the effect is investigated in more detail.

4.5.1 Two-level System Rabi Oscillations

In the presence of a driving field, a two-level atom can cyclically absorb and re-emit photons. This stems from coherent evolution, characterised by oscillatory population transfer. To describe this process semi-classical, a approach must be considered⁵⁰. Consider first a simple two-level system, with the ground level $|g\rangle$ of zero energy and an excited level $|e\rangle$ of energy $\hbar\omega_0$ (see Fig. 4.20), interacting with a classical electric field:

$$\begin{aligned}\mathbf{E}(\mathbf{r}, t) &= \boldsymbol{\epsilon} \frac{E_0}{2} \exp(-i\omega t) + c.c. \\ &= \mathbf{E}_0^{(+)} \exp(-i\omega t) + \mathbf{E}_0^{(-)} \exp(i\omega t) \\ &= \mathbf{E}_0^{(+)}(t) + \mathbf{E}_0^{(-)}(t),\end{aligned}\tag{4.40}$$

where $\boldsymbol{\epsilon}$ is the unit polarization vector of the field and the electric field has been decomposed into positive-, $\mathbf{E}_0^{(+)}$, and negative-, $\mathbf{E}_0^{(-)}$, rotating components.

The total Hamiltonian of the system is given by a sum of the atomic and interaction Hamiltonians:

$$H = H_A + H_{AF},\tag{4.41}$$

⁵⁰Classical approach of solving rate equations does not predict oscillatory population transfer.

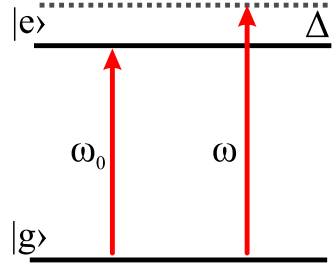


Figure 4.20: Two-level atom energy scheme considered for the Rabi oscillations. The frequency separation between the ground $|g\rangle$ and excited $|e\rangle$ levels of the atom is $\hbar\omega_0$. The electromagnetic field of frequency ω is detuned from the atomic transition by Δ .

where the atomic free-evolution Hamiltonian is given by:

$$H_A = \hbar\omega_0|e\rangle\langle e| \quad (4.42)$$

and the atom-field interaction Hamiltonian in the dipole approximation is described by:

$$H_{AF} = -\mathbf{d} \cdot \mathbf{E}, \quad (4.43)$$

with $\mathbf{d}$ being the reduced atomic dipole operator. Assuming that only one valence electron will interact with an external electric field, the dipole operator is given by:

$$\mathbf{d} = -e\mathbf{r}, \quad (4.44)$$

where $-e$ is the electron charge, and $\mathbf{r}$ is the electron-position operator.

By applying the identity $|g\rangle\langle g| + |e\rangle\langle e| = 1$ to both sides of the operator $\mathbf{d}$, and defining $\sigma := |g\rangle\langle e|$ as the lowering operator, the dipole operator will become (the diagonal matrix elements vanishes $\langle g|\mathbf{d}|g\rangle = \langle e|\mathbf{d}|e\rangle = 0$):

$$\mathbf{d} = \langle g|\mathbf{d}|e\rangle(\sigma + \sigma^\dagger) = \mathbf{d}^{(+)} + \mathbf{d}^{(-)}. \quad (4.45)$$

The dipole operator $\mathbf{d}$ was decomposed into its positive- and negative-rotating components, in a similar manner as the electric field [Eq. (4.40)]. Thus, the $\mathbf{d}^{(+)} \sim$

σ and $\mathbf{d}^{(-)} \sim \sigma^\dagger$. The explicit decomposition of the dipole operator and electrical field gives the following atom-field Hamiltonian:

$$\begin{aligned} H_{AF} &= -(\mathbf{d}^{(+)} + \mathbf{d}^{(-)}) (\mathbf{E}^{(+)} + \mathbf{E}^{(-)}) \\ &= -\mathbf{d}^{(+)} \cdot \mathbf{E}^{(+)} - \mathbf{d}^{(-)} \cdot \mathbf{E}^{(+)} - \mathbf{d}^{(+)} \cdot \mathbf{E}^{(-)} - \mathbf{d}^{(-)} \cdot \mathbf{E}^{(-)}. \end{aligned} \quad (4.46)$$

The terms $(\mathbf{d}^{(\pm)} \cdot \mathbf{E}^{(\pm)})$ of the interacting Hamiltonian oscillate rapidly with a sum frequency of $\exp[\pm i(\omega + \omega_0)]$. Those frequencies could not be observed and the rotating-wave approximation (RWA) will be applied. Therefore, the fast oscillating terms vanish, and the H_{AF} becomes:

$$H_{AF} \approx -\mathbf{d}^{(+)} \cdot \mathbf{E}^{(-)} - \mathbf{d}^{(-)} \cdot \mathbf{E}^{(+)}. \quad (4.47)$$

Thus, the explicit time-dependent form of the interacting Hamiltonian can be written as

$$\begin{aligned} H_{AF} &= -\langle g | \boldsymbol{\epsilon} \cdot \mathbf{d} | e \rangle \left[E_0^{(-)} \sigma \exp(i\omega t) + E_0^{(+)} \sigma^\dagger \exp(-i\omega t) \right] \\ &= \frac{\hbar\Omega}{2} \left[\sigma \exp(i\omega t) + \sigma^\dagger \exp(-i\omega t) \right], \end{aligned} \quad (4.48)$$

where Ω is the *Rabi frequency* defined as (the phase of the $\mathbf{d}$ operator is chosen such that the Rabi frequency is a positive value):

$$\Omega := -\frac{2\langle g | \boldsymbol{\epsilon} \cdot \mathbf{d} | e \rangle E_0^{(+)}}{\hbar} = -\frac{\langle g | \boldsymbol{\epsilon} \cdot \mathbf{d} | e \rangle E_0}{\hbar}. \quad (4.49)$$

The time-dependent Schrödinger equation

$$i\hbar \partial_t |\psi\rangle = H |\psi\rangle, \quad (4.50)$$

together with the atomic-state vector, written as

$$|\psi\rangle = c_g |g\rangle + c_e |e\rangle, \quad (4.51)$$

where c_e and c_g are slow-varying amplitudes, leads to following equation:

$$\partial_t c_g |g\rangle + \partial_t c_e |e\rangle = -i\omega_0 c_e |e\rangle - i\frac{\Omega}{2} \exp(i\omega t) c_e |g\rangle i - \frac{\Omega}{2} \exp(-i\omega t) c_g |e\rangle. \quad (4.52)$$

Projecting $\langle g |$ and $\langle e |$ on Eq. (4.52) gives a pair of coupled differential equations:

$$\begin{aligned} \partial_t c_g &= -i\frac{\Omega}{2} \exp(i\omega t) c_e, \\ \partial_t c_e &= -i\omega_0 c_e - i\frac{\Omega}{2} \exp(-i\omega t) c_g, \end{aligned} \quad (4.53)$$

which can be solved for the atomic evolution.

It is convenient to transform the problem into the rotating frame of the laser field, by defining the slowly-varying amplitude of the excited state. This rotating frame approximation ensures that all the fast-oscillating terms at the exact resonance vanish:

$$\tilde{c}_e := c_e \exp(i\omega t). \quad (4.54)$$

The equations of motions can be rewritten in the following manner:

$$\begin{aligned} \partial_t c_g &= -i\frac{\Omega}{2}\tilde{c}_e, \\ \partial_t \tilde{c}_e &= -i\Delta\tilde{c}_e - i\frac{\Omega}{2}c_g. \end{aligned} \quad (4.55)$$

In the rotating frame, the atomic and interaction Hamiltonians become

$$\tilde{H}_A = -\hbar\Delta|e\rangle\langle e|, \quad (4.56)$$

$$\tilde{H}_{AF} = \frac{\hbar\Omega}{2}(\sigma + \sigma^\dagger), \quad (4.57)$$

recalling that $\Delta = \omega - \omega_0$. In the exact resonance, where $\Delta = 0$, the equations reduce to:

$$\partial_t c_g = -i\frac{\Omega}{2}c_e, \quad (4.58)$$

$$\partial_t \tilde{c}_e = -i\frac{\Omega}{2}c_g. \quad (4.59)$$

By differentiating the reduced equations

$$\partial_t^2 c_g = -i\partial_t \frac{\Omega}{2}\tilde{c}_e, \quad (4.60)$$

and substituting into the original Eqs. (4.58) and (4.59), uncoupled equations are obtained:

$$\partial_t^2 c_g = -\left(\frac{\Omega}{2}\right)^2 c_g, \quad (4.61)$$

$$\partial_t^2 \tilde{c}_e = -\left(\frac{\Omega}{2}\right)^2 \tilde{c}_e. \quad (4.62)$$

The general solution of the differential equation for $c_g(t)$ is given as:

$$c_g(t) = A \sin\left(\frac{1}{2}\Omega t\right) + B \cos\left(\frac{1}{2}\Omega t\right). \quad (4.63)$$

Now the initial conditions have to be considered. Setting $t = 0$ for the c_g solution, one gets $B = c_g(0)$. By differentiating the equation $c_g(t)$ and comparing it with the original equation of motion [Eq. (4.58)] one gets

$$\partial_t c_g(t) = \frac{1}{2}\Omega \left[A \sin\left(\frac{1}{2}\Omega t\right) + B \cos\left(\frac{1}{2}\Omega t\right) \right] = -\frac{i}{2}\Omega \tilde{c}_e(t). \quad (4.64)$$

Setting $t = 0$ gives $A = -i\tilde{c}_e$. Therefore, the general solutions of the equations become:

$$c_g(t) = c_g(0) \cos\left(\frac{1}{2}\Omega t\right) - i\tilde{c}_e(0) \sin\left(\frac{1}{2}\Omega t\right), \quad (4.65)$$

$$\tilde{c}_e(t) = \tilde{c}_e(0) \cos\left(\frac{1}{2}\Omega t\right) - i c_g(0) \sin\left(\frac{1}{2}\Omega t\right). \quad (4.66)$$

For the atom initially in the ground state where $c_g(0) = 1$ and $\tilde{c}_e = 0$, the ground and excited state populations are

$$P_g(t) = |c_g(t)|^2 = \cos^2\left(\frac{1}{2}\Omega t\right) = \frac{1}{2}(1 + \cos \Omega t), \quad (4.67)$$

$$P_e(t) = |\tilde{c}_e(t)|^2 = \sin^2\left(\frac{1}{2}\Omega t\right) = \frac{1}{2}(1 - \cos \Omega t). \quad (4.68)$$

The population oscillates between the ground and excited states at the angular Rabi frequency Ω . The oscillation of the atomic populations was simulated, and is shown in Fig. 4.21.

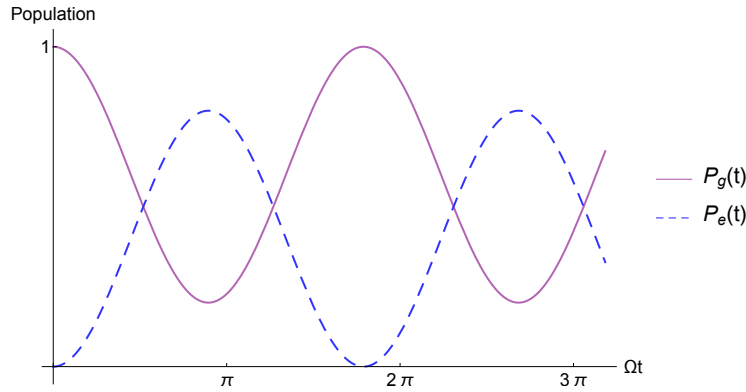


Figure 4.21: Rabi oscillations of atomic populations of the ground and excited states.

The above considerations lead to an idealistic model for two-level atoms at the exact resonance, with no relaxation or repopulating terms. For the non-resonant

case ($\Delta \neq 0$) the oscillations of the population are described by the generalised Rabi frequency

$$\tilde{\Omega} = \sqrt{\Omega^2 + \Delta^2}. \quad (4.69)$$

However, the two-level system gives good intuition of the atomic population behaviour and can be extended to a multi-level problem.

4.5.2 Three-level System Rabi Oscillations

In general, Rabi oscillations are not restricted to two-level systems where interaction with single photons is considered. They can also be observed in multi-photon absorption schemes, where intermediate levels are involved [106, 107]. If the electromagnetic field is sufficiently detuned from single-photon transitions, i.e., the intermediate levels stay unpopulated, a multi-level system can be effectively reduced to the two-level absorption scheme [107, 108].

The energy-level scheme for a two-photon transition is presented in Fig. 4.22. The electromagnetic field is tuned to match TPA with Δ detuning from the intermediate level $|i\rangle$ and a small δ detuning from the two-photon transition.

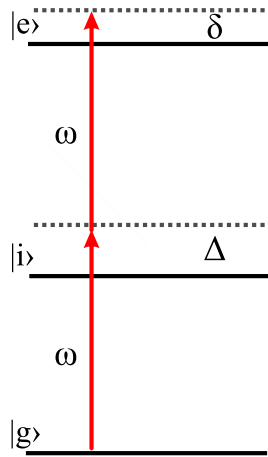


Figure 4.22: Energy-level scheme for two-photon Rabi oscillations. An electromagnetic frequency is coupling the two-photon transition, where Δ is the single-photon and δ is the two-photon transition detuning.

Consider the atom-field Hamiltonian for a three-level system, where the interaction

$$H_{AF} = H_{AF}^{gi} + H_{AF}^{ie} \quad (4.70)$$

is the sum of the interactions of two single-photon transitions with an electromagnetic field of the frequency ω . The interacting parts can be written as follows

$$H_{AF}^{gi} = \frac{\hbar\Omega_{ig}}{2} [\sigma_{gi} \exp(i\omega t) + \sigma_{gi}^\dagger \exp(-i\omega t)], \quad (4.71)$$

$$H_{AF}^{ie} = \frac{\hbar\Omega_{ei}}{2} [\sigma_{ie} \exp(i\omega t) + \sigma_{ie}^\dagger \exp(-i\omega t)], \quad (4.72)$$

where Ω_{gi} and Ω_{ie} are the single-photon Rabi frequencies for the $|g\rangle \rightarrow |i\rangle$ and $|i\rangle \rightarrow |e\rangle$ transitions, respectively. The lowering operators are defined as $\sigma_{ig} = |i\rangle\langle g|$ and $\sigma_{ei} = |e\rangle\langle i|$. The interacting Hamiltonian in the rotating frame of a laser can be expressed as:

$$\tilde{H}_{AF}^{gi} = \frac{\hbar\Omega_{ig}}{2} (\sigma_{gi} \exp(i\Delta t) + \sigma_{gi}^\dagger \exp(-i\Delta t)), \quad (4.73)$$

$$\tilde{H}_{AF}^{ie} = \frac{\hbar\Omega_{ei}}{2} (\sigma_{ie} \exp[i(\delta - \Delta)t] + \sigma_{ie}^\dagger \exp[-i(\delta - \Delta)t]). \quad (4.74)$$

The new state vector for a three-level atom is given by

$$|\psi\rangle = c_g |g\rangle + c_i |i\rangle + c_e |e\rangle, \quad (4.75)$$

where c_g , c_i , and c_e are slowly-varying amplitudes for the ground, intermediate, and excited states, respectively. In a similar way as for the two-level system, one can apply the RWA (since $\omega \approx \omega_{gi} \approx \omega_{ie}$) and transform the problem into the rotating frame of the laser field. In such a case, the time evolution of the slowly-varying amplitudes are given as:

$$\partial_t c_g = -i \frac{\Omega_{gi}}{2} \exp(-i\Delta t) \tilde{c}_i, \quad (4.76)$$

$$\partial_t \tilde{c}_i = -i \frac{1}{2} \{ \Omega_{gi} \exp(i\Delta t) c_g + \Omega_{ie} \exp[-i(\delta - \Delta)t] \tilde{c}_e \}, \quad (4.77)$$

$$\partial_t \tilde{c}_e = -i \frac{\Omega_{ie}}{2} \exp[i(\delta - \Delta)t] \tilde{c}_i. \quad (4.78)$$

The anticipated solutions for the coefficients can be expressed in the following

form

$$c_g = A \exp [\imath \lambda t], \quad (4.79)$$

$$\tilde{c}_i = B \exp [\imath (\lambda + \Delta) t], \quad (4.80)$$

$$\tilde{c}_e = C \exp [\imath (\lambda + \delta) t], \quad (4.81)$$

where λ represents the eigenvalue. This results in the following cubic eigenvalue equation

$$\lambda \left[(\lambda + \Delta)(\lambda + \delta) - \frac{1}{4} \Omega_{gi}^2 (\lambda + \delta) \right] = 0. \quad (4.82)$$

A general solution to this equation is intractable, but the equation can be solved for the exact resonance $\delta = 0$. Under this condition, the eigenvalues are:

$$\begin{aligned} \lambda_0 &= 0, \\ \lambda_{\pm} &= -\frac{1}{2} \Delta \pm \frac{1}{2} \sqrt{\Delta^2 + \Omega_{gi}^2 + \Omega_{ie}^2}. \end{aligned} \quad (4.83)$$

If the intermediate-state detuning Δ is sufficiently larger than the single-photon Rabi frequency, and hence the probability of the one-photon transition to the intermediate state is negligible. In turn, the following approximation can be applied:

$$\Omega_{gi}^2, \Omega_{ie}^2 \ll \Delta^2, \quad (4.84)$$

which allows Eq. (4.86) to simplify to the following form

$$\begin{aligned} \lambda_0 &= 0, \\ \lambda_+ &\approx \frac{\Omega_{ie}^2 + \Omega_{gi}^2}{4\Delta}, \\ \lambda_- &\approx -\Delta. \end{aligned} \quad (4.85)$$

For small two-photon detunings Δ the eigenvalues will not change significantly, as the large eigenvalue $\lambda_- \approx -\Delta$ and the approximation $\Omega_{gi}^2, \Omega_{ie}^2 \ll \Delta^2$ is still valid. Thus, Eq. (4.82) for the remaining eigenvalues (λ_0 and λ_+) becomes:

$$\lambda^2 + \lambda \left(\delta + \frac{\Omega_{ie}^2 + \Omega_{gi}^2}{4\Delta} \right) - \frac{\Omega_{gi}^2 \delta}{4\Delta} = 0. \quad (4.86)$$

The approximate solutions of the eigenvalue in Eq. (4.86) for ($\delta \ll \Delta$) are

$$\lambda_0 \approx \frac{1}{2} \left(\frac{\Omega_{ie}^2 + \Omega_{gi}^2}{4\Delta} - \delta - \Omega_2 \right), \quad (4.87)$$

$$\lambda_+ \approx \frac{1}{2} \left(\frac{\Omega_{ie}^2 + \Omega_{gi}^2}{4\Delta} - \delta + \Omega_2 \right), \quad (4.88)$$

$$\lambda_- \approx -\Delta, \quad (4.89)$$

where

$$\Omega_2 = \sqrt{\left(\delta - \frac{\Omega_{ie}^2 + \Omega_{gi}^2}{4\Delta} \right)^2 + \frac{\Omega_{gi}^2 \delta}{\Delta}}. \quad (4.90)$$

Introducing the eigenvalues [Eqs. (4.87) to (4.89)] to the general solutions of the time-dependent Schrödinger equation [Eqs. (4.79) to (4.81)], the slow-varying amplitudes c_g , $\tilde{c}_i$ and $\tilde{c}_e$ can be calculated. Thus, the time evolution of the populations can be obtained

$$\begin{aligned} P_g(t) &= |c_g(t)|^2 \approx \frac{\Omega_R^2}{\Omega_2^2} \sin^2 \left(\frac{1}{2} \Omega_2 t \right), \\ P_e(t) &= |\tilde{c}_e(t)|^2 \approx 1 - \frac{\Omega_R^2}{\Omega_2^2} \sin^2 \left(\frac{1}{2} \Omega_2 t \right), \\ P_i(t) &= |\tilde{c}_i(t)|^2 \approx 0, \end{aligned} \quad (4.91)$$

where

$$\Omega_R = \frac{\Omega_{gi} \Omega_{ie}}{2\Delta} \quad (4.92)$$

is the *two-photon Rabi frequency*, and

$$\Omega_2 = \sqrt{\Omega_R^2 + \left(\delta + \frac{\Omega_{ie}^2 - \Omega_{gi}^2}{4\Delta} \right)^2} \quad (4.93)$$

is the *generalised two-photon Rabi frequency* [108].

The Rabi oscillations occur between the ground $|g\rangle$ and excited $|e\rangle$ states, while the intermediate state $|i\rangle$ remains unpopulated. It can also be deduced that the one-photon Rabi frequency is proportional to the electric field amplitude [Eq. (4.49)], while the two-photon Rabi frequency is proportional to the square of electric amplitude, and thus proportional to power [Eq. (4.92)].

In general, the two-photon Rabi oscillation process, where the electromagnetic radiation is detuned from the intermediate level, is similar to the single-photon

process observed in a two-level system. The only difference is a power-dependent shift of the resonance frequency:

$$\delta_{ge} = \frac{\Omega_{ie}^2 - \Omega_{gi}^2}{4\Delta}. \quad (4.94)$$

This shift can be identified as an *AC Stark shift* of the two-photon transition [108].

4.5.3 Damped Rabi Oscillations in Two-level System

In real atomic systems, there are interactions with the external environment, which leads to decoherence and dissipation. This can be seen as damping on the oscillations of the atomic population. Since the equations of motion are difficult to solve under such conditions in multi-level system, the problem is discussed in a two-level system.

Let's consider the operator ρ as an operator describing, in general case, the state of two-level system (see Section 4.5.1). For a pure state it takes the form

$$\rho = |\psi\rangle\langle\psi|, \quad (4.95)$$

where $|\psi\rangle = c_g |g\rangle + c_e |e\rangle$ [Eq. (4.51)]. In an optical rotating frame, $\tilde{\rho}$ is a slowly varying state of a system for which the following relations are fulfilled

$$\begin{aligned} \tilde{\rho}_{gg} &= c_g c_g^* = \rho_{gg}, \\ \tilde{\rho}_{ee} &= \tilde{c}_e \tilde{c}_e^* = \rho_{ee}, \end{aligned} \quad (4.96)$$

where c_g^* and $\tilde{c}_e^*$ are the complex conjugations of the probability amplitude of the $|g\rangle$ and $|e\rangle$ states, respectively. In this notation, one can introduce slowly varying coherences between the ground and excited states

$$\tilde{\rho}_{ge} = c_g \tilde{c}_e^* = c_g c_e \exp(-i\omega t) = \rho_{ge} \exp(-i\omega t), \quad (4.97)$$

$$\tilde{\rho}_{eg} = \tilde{c}_e \exp(i\omega t) c_g = \rho_{eg} \exp(i\omega t). \quad (4.98)$$

The populations of the ground and excited states can be calculated by introducing ρ_{gg} and ρ_{ee} into the time-dependent Schrödinger equation [Eq. (4.52)]:

$$\partial_t \rho_{gg} = -\partial_t \rho_{ee} = -i \frac{\Omega}{2} (\tilde{\rho}_{eg} - \tilde{\rho}_{ge}), \quad (4.99)$$

$$\partial_t \rho_{ee} = \tilde{c}_e^* (\partial_t \tilde{c}_e) + c.c. = i \frac{\Omega}{2} (\tilde{\rho}_{eg} - \tilde{\rho}_{ge}). \quad (4.100)$$

In the same manner, the coherences can be calculated by introducing Eqs. (4.97) and (4.98) into Eq. (4.52):

$$\partial_t \tilde{\rho}_{ge} = c_g \partial_t \tilde{c}_e^* + \tilde{c}_e^* \partial_t c_g = -i\Delta \tilde{\rho}_{ge} - i\frac{\Omega}{2}(\rho_{ee} - \rho_{gg}), \quad (4.101)$$

$$\partial_t \tilde{\rho}_{eg} = \partial_t \tilde{\rho}_{ge}^* = i\Delta \tilde{\rho}_{ge} + i\frac{\Omega}{2}(\rho_{ee} - \rho_{gg}). \quad (4.102)$$

Equations (4.99) to (4.102) are called the *optical Bloch equations*. These four equations can be rewritten in a following form [16, 109]

$$\partial_t \tilde{\rho} = -\frac{i}{\hbar} [\tilde{H}_A + \tilde{H}_{AF}, \tilde{\rho}], \quad (4.103)$$

which is called the *Schrödinger-von Neumann equation*. It describes the dynamics of a many-level atomic system interacting with an electro-magnetic field, and their derivation is almost equal to the Rabi-oscillation problem discussed in Section 4.5.1.

The optical Bloch equations [Eqs. (4.99) to (4.102)], for the two-level atomic system, can be rewritten into the form of a vector. By introducing the Pauli operators:

$$\begin{aligned} \sigma_x &= \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} = \sigma + \sigma^\dagger, \\ \sigma_y &= \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} = i(\sigma - \sigma^\dagger) \\ \sigma_z &= \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} = |e\rangle \langle e| - |g\rangle \langle g| = \sigma^\dagger \sigma - \sigma \sigma^\dagger = [\sigma^\dagger, \sigma]. \end{aligned} \quad (4.104)$$

one can express the populations and coherences as

$$\begin{aligned} \langle \sigma \rangle &= \text{Tr}[|g\rangle \langle e| \rho] = \text{Tr}[\langle e| \tilde{\rho} |g\rangle] = \tilde{\rho}_{eg}, \\ \langle \sigma^\dagger \rangle &= \text{Tr}[|e\rangle \langle g| \rho] = \text{Tr}[\langle g| \tilde{\rho} |e\rangle] = \tilde{\rho}_{ge}, \\ \langle \sigma^\dagger \sigma \rangle &= \text{Tr}[|e\rangle \langle e| \tilde{\rho}] = \rho_{ee}, \\ \langle \sigma \sigma^\dagger \rangle &= \text{Tr}[|g\rangle \langle g| \tilde{\rho}] = \rho_{gg}. \end{aligned} \quad (4.105)$$

Now, introducing the following relations between the Pauli and ρ operators, one

obtains

$$\begin{aligned}
\langle \sigma_x \rangle &= \langle \sigma \rangle + \langle \sigma^\dagger \rangle = \tilde{\rho}_{eg} + \tilde{\rho}_{ge}, \\
\langle \sigma_y \rangle &= \imath \langle \sigma \rangle - \imath \langle \sigma^\dagger \rangle = \imath(\tilde{\rho}_{eg} - \tilde{\rho}_{ge}), \\
\langle \sigma_z \rangle &= \rho_{ee} - \rho_{gg}.
\end{aligned} \tag{4.106}$$

The Bloch-vector equations can be rewritten in the following form

$$\begin{aligned}
\langle \sigma_x \rangle &= \Delta \langle \sigma_y \rangle, \\
\langle \sigma_y \rangle &= -\Delta \langle \sigma_x \rangle - \Omega \langle \sigma_z \rangle, \\
\langle \sigma_z \rangle &= \Omega \langle \sigma_y \rangle,
\end{aligned} \tag{4.107}$$

where $\langle \sigma \rangle = \langle \sigma_x \rangle \hat{x} + \langle \sigma_y \rangle \hat{y} + \langle \sigma_z \rangle \hat{z}$ is the *Bloch vector* [16, 109].

Equations (4.99) to (4.102) and (4.107) do not incorporate any relaxation processes. To model relaxation processes, i.e., spontaneous emission, some additional terms must be added. Assuming, that the excited state population decays at rate Γ to the ground level, and the coherences also decay with a rate described by $\gamma_\perp$. The optical Bloch equations can be written in the following form

$$\begin{aligned}
\partial_t \rho_{gg} &= -\imath \frac{\Omega}{2} (\tilde{\rho}_{eg} - \tilde{\rho}_{ge}) - \Gamma \rho_{ee}, \\
\partial_t \rho_{ee} &= \imath \frac{\Omega}{2} (\tilde{\rho}_{eg} - \tilde{\rho}_{ge}) + \Gamma \rho_{ee}, \\
\partial_t \tilde{\rho}_{ge} &= -(\gamma_\perp + \imath \Delta) \tilde{\rho}_{ge} - \imath \frac{\Omega}{2} (\rho_{ee} - \rho_{gg}), \\
\partial_t \tilde{\rho}_{eg} &= -(\gamma_\perp - \imath \Delta) \tilde{\rho}_{eg} + \imath \frac{\Omega}{2} (\rho_{ee} - \rho_{gg})
\end{aligned} \tag{4.108}$$

Now, the Bloch vector can be also rewritten in the presence of damping

$$\begin{aligned}
\langle \sigma_x \rangle &= \Delta \langle \sigma_y \rangle - \gamma_\perp \langle \sigma_x \rangle, \\
\langle \sigma_y \rangle &= -\Delta \langle \sigma_x \rangle - \Omega \langle \sigma_z \rangle - \gamma_\perp \langle \sigma_y \rangle, \\
\langle \sigma_z \rangle &= \Omega \langle \sigma_y \rangle - \Gamma (\langle \sigma_z \rangle + 1).
\end{aligned} \tag{4.109}$$

The excited-state decay term Γ describes the relaxation of the $\langle \sigma_z \rangle$ component of the Bloch vector, thus it is called the longitudinal relaxation rate. The coherence damping term relates to the perpendicular components of a Bloch vector ($\langle \sigma_x \rangle$ and $\langle \sigma_y \rangle$), therefore $\gamma_\perp$ is called a perpendicular relaxation rate. In general, the transverse relaxation is longer than half of the longitudinal relaxation $\gamma_\perp \geq \Gamma/2$.

The damped Rabi oscillations can be obtained by solving the optical Bloch equations (4.109) using the Laplace transform [16, 109]. Introduction of the Laplace transforms changes the problem of solving set of coupled differential equations into an algebraic problem⁵¹ In this case, the general solution of the damped Rabi oscillations is given by

$$\langle \sigma_\alpha(t) \rangle = A \exp(-ct) + B \exp(-at) \cos(bt) + \frac{C}{b} \exp(-at) \sin(bt) + D, \quad (4.110)$$

where the first term represents the decay of the populations and coherences, the second and third represent exponentially damped Rabi oscillations, and the last term is the steady-state solution. The constants A, B, C and D depend on both, the initial conditions, and on the Bloch-vector components, and when a, b and c are the roots of the cubic polynomial, which is a solution for the Laplace transform of the Bloch vector [Eq. (4.109)].

In general, the model for the damped Rabi oscillations is of high complexity, and there are only three regimes where solutions could be identified: at the exact resonance (when $\Delta = 0$), when damping is defined that coherences decay rate is equal to the decay rate of the excited state $\gamma_\perp = \Gamma$, and for a strong excitation (i.e., $\Omega \gg \Gamma, \gamma_\perp$).

Here, the case for the exact resonance ($\Delta = 0$) will be considered. Also, let's assume a damping such that $\gamma_\perp = \Gamma/2$, and that an atom is initially in the ground state. Thus, the Bloch vector components are as follows

$$\langle \sigma_x(0) \rangle = \langle \sigma_y(0) \rangle = 0, \quad (4.111)$$

$$\langle \sigma_z(0) \rangle = -1. \quad (4.112)$$

The roots of the cubic polynomial for $\Delta = 0$, which is a general solution for a Laplace transform of a Bloch vector are equal to [16]:

$$a = 3\Gamma/4, \quad b = \sqrt{\Omega^2 - (\Gamma/4)^2}, \quad \text{and } c = \Gamma/2. \quad (4.113)$$

Now, the constants A, B, C and D need to be determined by considering the initial conditions of the system. The x -component for the resonant case vanishes ($\langle \sigma_x(t) \rangle = 0$).

⁵¹This method was introduced by H. C. Torrey [110].

The complete solution for the y -component is

$$\langle \sigma_y(t) \rangle = -\frac{\Omega\Gamma}{\Omega^2 + \Gamma^2/2} \left[1 - \exp\left(-\frac{3\Gamma}{4}\right) \left(\cos(\Omega_\Gamma t) - \frac{\Omega^2 - \Gamma^2/4}{\Gamma\Omega_\Gamma} \sin(\Omega_\Gamma t) \right) \right], \quad (4.114)$$

where

$$\Omega_\Gamma = \sqrt{\Omega^2 - \left(\frac{\Gamma}{4}\right)^2} \quad (4.115)$$

is the *Rabi frequency in the presence of damping* Γ . The solution for the z -component can be written as:

$$\langle \sigma_z(t) \rangle = -1 + \frac{\Omega^2}{\Omega^2 + \Gamma^2/2} \left[1 - \exp\left(-\frac{3\Gamma}{4}\right) \left(\cos(\Omega_\Gamma t) + \frac{3\Gamma}{4\Omega_\Gamma} \sin(\Omega_\Gamma t) \right) \right]. \quad (4.116)$$

Therefore, the Rabi oscillations of the excited state population can be calculated as:

$$P_e(t) = \rho_{ee} = \frac{\langle \sigma_z \rangle + 1}{2}. \quad (4.117)$$

Simulated Rabi oscillations with damping Γ are presented in Fig. 4.23.

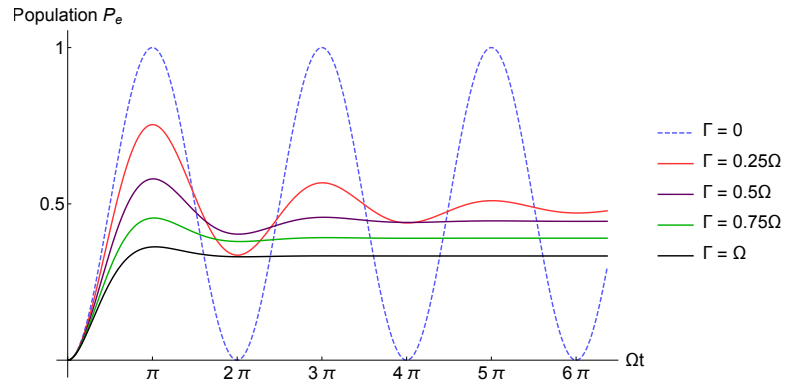


Figure 4.23: Simulated damped Rabi oscillations in a two-level system [Eq. (4.117)].

4.5.4 Results

The results from Section 4.5.2 show that for a large detuning and a low-power control field, the three-level system can be treated as a two-level system. The only difference is in defining the Rabi frequency for a two-photon transition, which is determined by the Rabi frequencies of two single-photon transitions [Eq. (4.92)]. In the same manner, it is valid to approximate the damped Rabi oscillations in a three-level atom by the solution in a two-level system.

Figure 4.24 shows pulses observed for a two-photon excitation of rubidium atoms inside the fibre using a control-beam power of $0.63 \mu\text{W}/\text{cm}^2$ (grey points). They were observed during the fast-light measurements described in Section 4.4. The pulse in Fig. 4.24a reveals a nearly symmetric profile. The rising edge of this pulse reflects the rise time of the AOM ($\sim 160 \text{ ns}$). The other pulse, presented in Fig. 4.24, was induced with a red-detuned control beam. The damped Rabi oscillations can be observed on the rising edge of this pulse. The oscillations were not observed for the lower-powered control beam, which because Rabi oscillations which strongly depend on optical power, indicates that they have been identified. To reveal more information about those oscillations, a theoretical function was fitted (blue line in Fig. 4.24).

For the measurements, the detuning from the intermediate state was 770 MHz, and the intensity of the coupling beam inside the fibre was $0.63 \mu\text{W}/\text{cm}^2$. This ensures that the two-level atom approximation can be applied. Therefore, the function fitted to the measured pulses consists of the damped Rabi oscillations solution [Eq. (4.116)] on the rising edge, and complementary error function [$\text{erfc}(x) = 1 - \text{erf}(x)$] modelling on the falling edge.

The oscillations were modelled using the excited state population [Eq. (4.117)]

$$\rho_{ee}(t) = \frac{1}{2} \frac{\Omega^2}{\Omega^2 + \Gamma^2/2} \left[1 - \exp\left(-\frac{3\Gamma}{4}t\right) \left[\cos\left(\sqrt{\Omega^2 - (\Gamma/4)^2}t\right) + \frac{3\Gamma}{4\Omega^2 - (\Gamma/4)^2} \sin\left(\sqrt{\Omega^2 - (\Gamma/4)^2}t\right) \right] \right], \quad (4.118)$$

where $\Omega_r = \sqrt{\Omega^2 + (\Gamma/4)^2}$ is the Rabi oscillation in the presence of damping, and $\Gamma/2$ is the damping rate of the oscillations. The function describing the excited

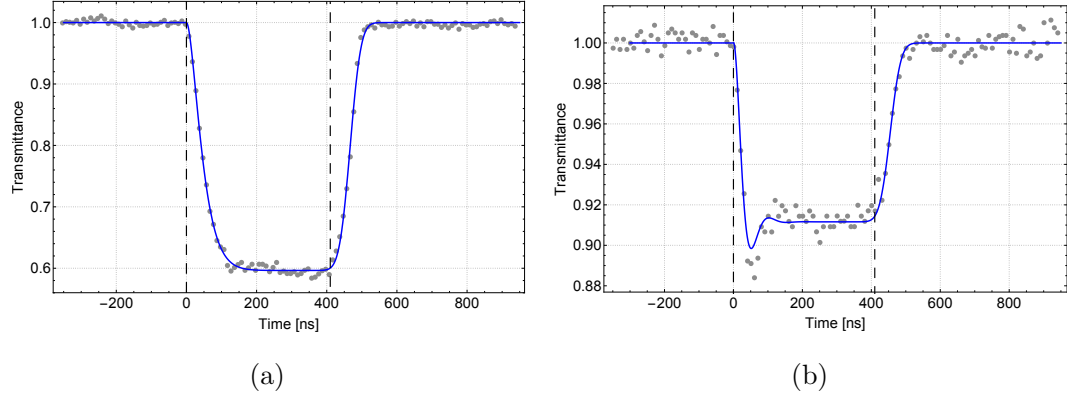


Figure 4.24: Probe-light pulse created by the presence of the control beam of the intensity 0.63 W/cm^2 . (a) Signal observed close to the two-photon resonance and (b) signal red detuned from the two-photon transition. The Rabi oscillation appeared on the rising slope of the pulse.

state population was obtained by introducing the solution from Eq. (4.116) into Eq. (4.117). This modelled the raising edge of the pulse, where the oscillations were observed. The falling edge was modelled by a complementary error function

$$g(t, t_0, \sigma) = \frac{1}{2} \text{erfc} \left(\frac{t - t_0}{\sigma} \right), \quad (4.119)$$

where t_0 describes the middle point of the falling edge and σ describes its steepness. The general form of the fitting function is given by

$$f(t, \Omega, \Gamma, t_0, t_1, A) = 1 - A \cdot \Theta(t - t_1) \cdot \rho_{ee}(t) \cdot g(t, t_0, \sigma), \quad (4.120)$$

where Θ is the Heaviside step function. The step function ensures the initial transmission is equal to 1.

For all red-detuned pulses, the Rabi frequency was determined and plotted in Fig. 4.25. The generalised Rabi frequency Ω depends on the frequency detuning δ from the two-photon resonance in the following way:

$$\Omega = \sqrt{\Omega_R^2 + \delta^2}, \quad (4.121)$$

where $\Omega_R = 15.526(24) \text{ MHz}$ is the zero detuning two-photon Rabi frequency estimated from the experiment. The black solid curve in Fig. 4.25 is a theoretical prediction of Ω . There is high agreement between theoretical prediction for a

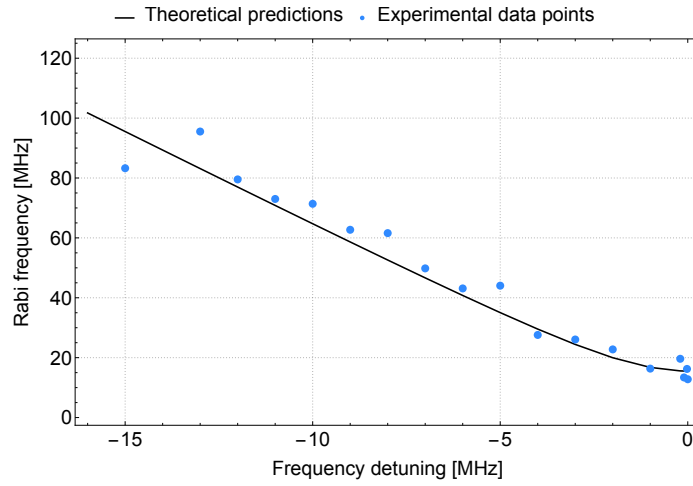


Figure 4.25: Rabi frequency for the red-detuned light pulses (blue points) compared with the predicted generalised two-photon Rabi frequency (solid line). The error bars for the measured points are smaller than the point size.

generalised Rabi frequency Ω and its estimated value from the theoretical fit. Damping of the oscillations was estimated to be 46(14) MHz.

4.6 Summary

In this Chapter the two-photon absorption experiment was presented. The theoretical description of the TPA linewidth, which reveals a significantly reduced Doppler width, was discussed. The major causes of linewidth broadening were the velocity distribution of the atoms in an intermediate state, the transit time inside the fibre, and the atom-surface interaction.

The absorption and dispersion properties of rubidium vapour filling the HC-PCF were obtained. It was shown that the absorption coefficient and non-linear refractive index for the TPA depends on the field strength. This allows for manipulation of a mediums properties by changing the control-field intensity, particularly giving a further opportunity to realise all-optical switching. Moreover, a fast-light switch was demonstrated and studied. The fibre filled with alkali atoms dramatically increased the measured (predicted) advancement time with negligible pulse reshaping.

The last part of the chapter discusses the coherences in two- and three-level atomic systems. A theoretical description of both systems was provided. It was shown that for a low-light intensities and significant frequency detuning from the intermediate state in a two-photon transition, the three-level system can be simplified to a two-level system. Rabi oscillations between the ground and excited states in a far-detuned three-level atom were observed. As shown, the measured generalised Rabi frequency scales properly with the frequency detuning.

The Rabi oscillations were implemented into the data analysis of the fast-light experiment. This correction for the measured pulse shape deepened the discrepancy between measured and theoretically predicted pulse advancement (Fig. 4.16b).

Coherent effects in three-level atomic system will be a matter for the next chapter.

Chapter 5

Coherent Effects in a Three-level Atom

It was shown in Section 4.5 that coherent effects in a two-level atom manifests in the presence of Rabi oscillations. A generalisation of this approach to a three-level system allows for generating more complex dynamics, leading to the rise of interesting phenomena such as coherent population trapping (CPT) [9, 17] and electromagnetically induced transparency (EIT) [9, 17, 24]. All these phenomena are based on quantum interference, which in a three-level system requires interactions with two electromagnetic fields. This opens the possibility to create a linkage between normally non-interacting atomic states and hence significantly affect the distribution of atomic populations [9].

A theoretical description of coherent processes requires the introduction of a formalism, which not only enables the description of such an interaction, but also the incorporation of dissipation processes (relaxation, decoherence) that may affect the dynamics of the evolution. This description can be achieved using the density-matrix formalism [15, 16].

The Density-Matrix Formalism

The state vector

$$|\psi\rangle = \sum_{\alpha} c_{\alpha} |\alpha\rangle, \quad (5.1)$$

where the summation runs over all eigenstates $|\alpha\rangle$ and c_α is the probability amplitude, represents the quantum state of the system. While the state vector offers the ability to calculate the evolution of the function in a relaxation-free system, a more general approach exploits the density matrix ρ . This allows one to describe the interaction between an external electromagnetic field and an atom in the presence of relaxation processes. For a pure state, the density operator is defined as⁵²

$$\rho := |\psi\rangle\langle\psi|. \quad (5.2)$$

Therefore, for a pure state, the density operator represents states in a coherent superposition of the eigenstates.

However, the definition of the density operator can be extended to a more general case, where it can also describe the incoherent sum of states

$$\rho = \sum_{\alpha} P_{\alpha} |\psi_{\alpha}\rangle\langle\psi_{\alpha}|, \quad (5.3)$$

where $|\psi_{\alpha}\rangle$ is the arbitrary state of particle α , with probability P_{α} of the system being in that state. The probability weights must obey:

$$\sum_{\alpha} P_{\alpha} = 1. \quad (5.4)$$

The definition of the density operator becomes evident if one calculates the density-matrix elements $\rho_{\alpha\alpha'}$ of the system with respect to the complete, orthonormal basis:

$$\rho_{\alpha\alpha'} := \langle\alpha|\rho|\alpha'\rangle. \quad (5.5)$$

For a pure state, the diagonal element $\rho_{\alpha\alpha}$ is obtained

$$\rho_{\alpha\alpha} = \langle\alpha|\rho|\alpha\rangle = |\langle\alpha|\psi\rangle|^2 = |c_{\alpha}|^2. \quad (5.6)$$

Thus, the diagonal elements represent the atomic populations. The off-diagonal elements (when $\alpha \neq \alpha'$) represent coherences between atomic states. The coherences give information about the relative phase between different components:

$$\rho_{\alpha\alpha'} = |c_{\alpha}c_{\alpha'}| \exp[i(\phi_{\alpha} - \phi_{\alpha'})]. \quad (5.7)$$

⁵²The definition of the density operator was already introduced in Section 4.5.3.

The relations provided by Eqs. (5.6) and (5.7) correspond to pure states, thus the density operator fulfils the following relation $\text{Tr}[\rho^2] = \text{Tr}[\rho] = 1$.⁵³ For mixed states $\text{Tr}[\rho^2] < 1$, and the coherences are the sum of the complex numbers corresponding to different states [16].

The dynamics of an atomic system interacting with an external electromagnetic field can be described by differentiating the density operator for such a system and introducing the Schrödinger equation. The equation of motion for the density operator in a relaxation free environment can be written as:

$$\partial_t \rho = -\frac{i}{\hbar} [H, \rho], \quad (5.8)$$

where H is the system Hamiltonian, and is called the Schrödinger-von Neumann equation (see Section 4.5.3) or the *master equation*. If a relaxation is present, Eq. (5.8) needs to be modified to accommodate such a process. Under such conditions the master equation takes the form

$$\partial_t \rho = -\frac{i}{\hbar} [H, \rho] + \Gamma(\rho), \quad (5.9)$$

where $\Gamma(\rho)$ is the relaxation operator describing all (density-matrix dependent and density-matrix independent) processes.

5.1 Electromagnetically Induced Transparency

Electromagnetically induced transparency is a coherent optical effect occurring in three-level systems, when interacting with two electromagnetic fields. It is a quantum interference effect that can be observed for a weak probe beam tuned to an atomic transition, and propagated through a medium with a reduced absorption. The absorption is affected by the presence of a strong coupling electromagnetic field, operating at a linked transition. As a consequence, the reduction of absorption is seen as a sub-Doppler feature on the Doppler-broadened absorption resonance.

⁵³Where $\text{Tr}[\rho] = \sum_{\alpha} \rho_{\alpha\alpha}$ is a trace over the density matrix.

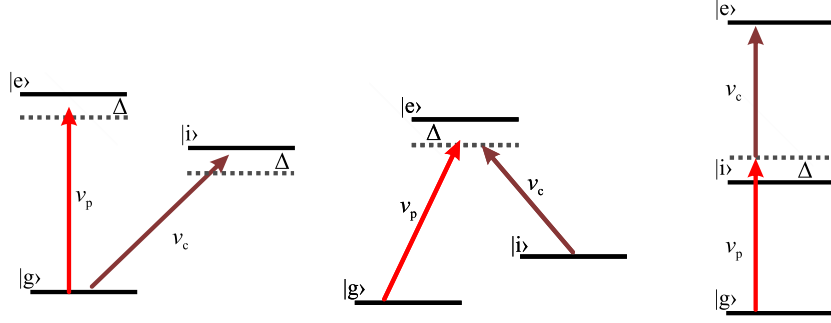


Figure 5.1: Energy level scheme used in the EIT experiment: (a) V , (b) Λ , and (c) cascade.

The process of inducing a transparency window in the absorption resonance was observed in various energy-level configurations: V ,⁵⁴ Λ , and cascade (see Fig. 5.1). While each of them is based on the same theoretical foundations, the difference in the energy structure leads to signals with different contrasts and widths of their transparency windows, which is related to the constraints of the system [13].

A theoretical description of the EIT process can be provided in a bare-state basis (the diagonalised basis of the atom plus field Hamiltonians without any interaction part) [16] or a dressed-state basis [9, 112, 113]. Dressed states are eigenstates of the complete atom-field Hamiltonian (including interaction) [114]. Regardless of the chosen basis, the theoretical description of EIT requires finding the solution of the master equation for the density operator. Hereafter, the dressed-state approach is utilized in the description of the process.

5.1.1 Dressed-state Approach: Autler-Townes Splitting

In the previous chapter, the semi-classical model, which assumes the interaction of quantised atom with an classical electromagnetic field, was considered. Here, the *Jaynes-Cummings model* will be utilised. It is a quantum theory model which describes the quantised two-level atom interacting with the quantized mode of a strong laser. Thus, the complete Jaynes-Cummings Hamiltonian contains the

⁵⁴In V configuration only the energy state splitting, due to strong coupling field, can be observed [14, 111], this will be explained later in this Section.

atomic Hamiltonian H_A , the external electromagnetic-field Hamiltonian H_F , and the atom-field interaction part H_{AF} [9, 114]:

$$H = H_A + H_F + H_{AF}. \quad (5.10)$$

The atomic free-evolution Hamiltonian H_A can be expressed as:

$$H_A = \hbar\omega_0|e\rangle\langle e|, \quad (5.11)$$

where $\hbar\omega_0$ is the energy of level $|e\rangle$, while the energy of the ground $|g\rangle$ is zero (see Fig. 5.2). The laser-field Hamiltonian is given by:

$$H_F = \hbar\omega_L a^\dagger a, \quad (5.12)$$

where a and $a^\dagger$ are annihilation and creation operators, annihilating and creating a photon of the energy $\hbar\omega_L$.

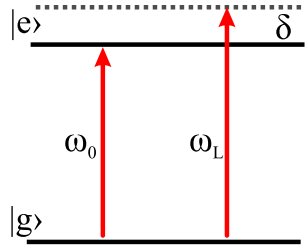


Figure 5.2: Energy-level scheme of two-level atom, with ground $|g\rangle$ and excited $|e\rangle$ states separated by frequency ω_0 in an electromagnetic field with frequency ω_L .

There are a few assumptions that have to be made for the strong laser mode. The strong laser field can be approximated by an optical cavity without losses, thus, it represents a laser mode. Due to the average photon number in one laser mode $\langle N \rangle \rightarrow \infty$, the energy density $\langle N \rangle/V$ inside the laser mode must remain constant, where V is the ‘volume’ of the mode. These assumptions mean that absorption and emission processes are negligible, and the number of photons in one laser mode remains infinite.

The eigenstates of the general Hamiltonian $H' = H_A + H_F$, neglecting the interaction part, are labelled by two numbers: the atomic quantum number g or e , and the number of laser photons N . Thus, the uncoupled eigenstates of H' can be

written as $|g, N + 1\rangle$ and $|e, N\rangle$. Near the resonance, i.e., for frequency detuning $\delta = \omega_L - \omega_0$ much smaller than the transition frequency $|\delta| \ll \omega_0$, the $|g, N + 1\rangle$ and $|e, N\rangle$ states are close to each other (see Fig. 5.3). The level $|g, N + 1\rangle$ lays above the level $|e, N\rangle$, if δ is positive, and the energy difference is $\hbar\delta$.

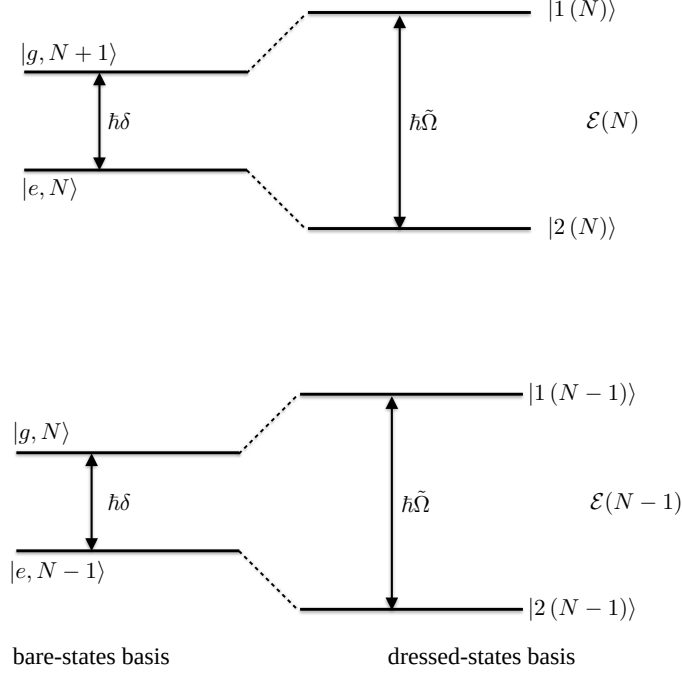


Figure 5.3: Energy-level scheme for an uncoupled atom with photons (left-hand side) and dressed states (right-hand side). The $\mathcal{E}(N)$ and $\mathcal{E}(N - 1)$ represents the manifolds with N and $N - 1$ number of photons, respectively.

The interaction Hamiltonian describes the interaction between the reduced atomic dipole operator $\mathbf{d}$ and an external electromagnetic field $\mathbf{E}$:

$$H_{AF} = -\mathbf{d} \cdot \mathbf{E}. \quad (5.13)$$

The dipole operator can be expressed by the lowering operator ($\sigma = |g\rangle\langle e|$), and rewritten in the form [114]:

$$\mathbf{d} = \langle g|\mathbf{d}|e\rangle (|g\rangle\langle e| + |e\rangle\langle g|) = \mathbf{d}_{ge}(\sigma + \sigma^\dagger). \quad (5.14)$$

Furthermore, the quantum-field operator is given by [114]:

$$\mathbf{E} = \sqrt{\frac{\hbar\omega_L}{2\varepsilon_0 V}} \boldsymbol{\epsilon}_L (a + a^\dagger), \quad (5.15)$$

where $\boldsymbol{\epsilon}_L$ is the polarisation of the laser mode, and ε_0 is the vacuum permittivity. Hence, the interaction Hamiltonian is given by

$$H_{AF} = g(\sigma + \sigma^\dagger)(a + a^\dagger), \quad (5.16)$$

where $g = -\boldsymbol{\epsilon}_L \cdot \mathbf{d} \sqrt{\hbar\omega_L/(2\varepsilon_0 V)}$ is the coupling constant. It is now clear that the interaction Hamiltonian couples each atom and field state in the following manner

$$\langle e, N | H_{AF} | g, N + 1 \rangle = g\sqrt{N + 1} \approx g\sqrt{\langle N \rangle}, \quad (5.17)$$

where the average photon number $\langle N \rangle \gg 1$ and the width of the distribution of the numbers of photons $\Delta N \gg 1$. The electromagnetic field can be treated as a semi-classical external field⁵⁵:

$$\mathbf{E} = 2\boldsymbol{\epsilon}_L \sqrt{\frac{\hbar\omega_L}{2\varepsilon_0 V}} \sqrt{\langle N \rangle}. \quad (5.18)$$

Therefore, resonant coupling can be expressed in terms of the Rabi frequency (see Section 4.5):

$$\hbar\Omega = 2\langle e, N | H_{AF} | g, N + 1 \rangle = 2g\sqrt{\langle N \rangle}. \quad (5.19)$$

Those atomic states with photons ($|e, N\rangle$ and $|g, N + 1\rangle$), so-called bare-states, can be rewritten to the new unperturbed *dressed-state* basis, which diagonalise the complete Hamiltonian H . The unperturbed dressed-states are separated by the generalised Rabi frequency:

$$\tilde{\Omega} = \sqrt{\delta^2 + \Omega^2}. \quad (5.20)$$

A scheme of the translation from the uncoupled basis to the dressed-state representation is presented in Fig. 5.3. In the presence of a strong electromagnetic field, the bare-states ($|g, N + 1\rangle$ and $|e, N\rangle$) are separated by energy $\hbar\delta$. In a new dressed-basis, the energy between the new eigenstates $|1(N)\rangle$ and $|2(N)\rangle$ is $\hbar\tilde{\Omega}$. The dressed states can be expressed by the bare atomic states in the following manner [114]:

$$\begin{aligned} |1(N)\rangle &= \sin \Theta |g, N + 1\rangle + \cos \Theta |e, N\rangle, \\ |2(N)\rangle &= \cos \Theta |g, N + 1\rangle - \sin \Theta |e, N\rangle, \end{aligned} \quad (5.21)$$

⁵⁵The laser mode can be in a coherent state $|\alpha \exp(i\omega_L t)\rangle$, where α is a real number [114].

where Θ is the mixing angle defined as:

$$\tan 2\Theta = -\frac{\Omega}{\delta}, \quad 0 \leq 2\Theta \leq \pi. \quad (5.22)$$

To obtain the full solution, which shows the coherences between non-interacting states, the master equation [Eq. (5.8)] in the dressed-state representation must be solved. However, the consequences of introducing the dressed-state basis can be already evaluated.

Consider a three-level energy system, where a strong coupling-laser field dresses the ground state $|g\rangle$ and intermediate state $|i\rangle$ with photons (Fig. 5.4). The states $|g, N+1\rangle$ and $|i, N\rangle$ are separated by energy $\hbar\delta$ (they are close to each-other in the presence of the field). A weak probe beam is coupling the unperturbed excited states $|e, N\rangle$ and $|i, N\rangle$. The frequency of this transition is sufficiently different than the frequency of the transition between the unperturbed states $|g\rangle$ and $|i\rangle$ ($\omega_0 \neq \omega'_0$). Under the influence of the coupling field, the states $|g, N+1\rangle$ and $|i, N\rangle$ can be transformed into two dressed states $|1(N)\rangle$ and $|2(N)\rangle$ separated by $\hbar\tilde{\Omega}$. This situation is presented in Fig. 5.4, where only the manifold with N photons ($\mathcal{E}(N)$) is presented (see Fig. 5.3).

If $\Omega \neq 0$, the weak probe laser will see two transitions: $|e, N\rangle \rightarrow |1(N)\rangle$ of frequency $\omega_0 - (\tilde{\Omega} + \delta)/2$ and $|e, N\rangle \rightarrow |2(N)\rangle$ of frequency $\omega_0 + (\tilde{\Omega} - \delta)/2$. Those two transitions are called an *Autler-Townes doublet* and can be seen as a reduction in the absorption between them. Therefore, the ATS and EIT processes share the same scheme, i.e., they are observed when a strong coupling field is applied between two states, and a weak laser beam is tuned to the linked transition, thus, they give similar results. However, there is a significant difference between those two processes, which is discussed in the following.

5.1.2 Difference between EIT and ATS

The ATS gives similar observations as for EIT, when both result in the suppression of absorption. Despite the similarities, these two processes are based on significantly different grounds.

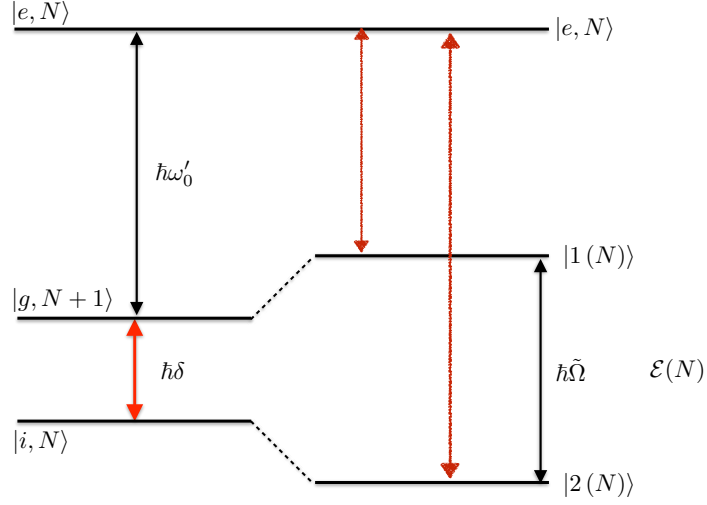


Figure 5.4: Energy-level scheme for Autler-Townes splitting. A strong laser beam is applied between ground $|g\rangle$ and intermediate $|i\rangle$ states (thick light red arrow). Only the $\mathcal{E}(N)$ manifold from Fig. 5.3 is presented. The splitting can be observed by introducing a weak probe beam to a linked transition from the dressed states $|1(N)\rangle$ and $|2(N)\rangle$ to some unperturbed state $|e\rangle$ (thin dark red arrows).

When a strong coupling field is applied between two energy levels, in a bare atomic basis, it splits each of the states in two. The splitting rate of those states is described by the generalised Rabi frequency $\tilde{\Omega}$. This situation, for an upper-state-driven cascade system, is depicted in Fig. 5.5. The strong field couples states $|i\rangle$ and $|e\rangle$, causing ATS. A weak beam probes the atomic transition between the ground and intermediate states, which results in reduction of absorption between the split states.

The suppression of absorption in an EIT process is based on destructive interference in a laser-dressed medium [9, 116]. There are two alternative routes for single- and two-photon absorption for the states split by the coupling field. These routes interfere destructively, opening a transparency window for a probe beam tuned exactly to the resonance [115]. This is shown in Fig. 5.5, where single- and two-photon transitions are indicated with red arrows.

The destructive interference is a *Fano-type interference* of two alternative pathways in an atomic transition [116, 117]. When the decay of two-photon coherence

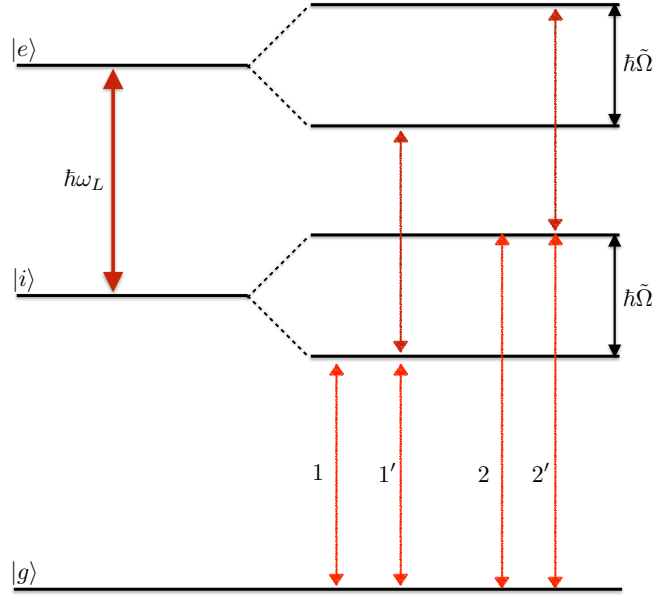


Figure 5.5: Energy-level scheme for electromagnetically induced transparency in the cascade system. The strong laser field $\hbar\omega_L$ is applied between states $|i\rangle$ and $|e\rangle$ splitting each of them into two states. The weak probe beam is scanning across the $|g\rangle \rightarrow |i\rangle$ and $|g\rangle \rightarrow |e\rangle$ transitions, which give rise to two routes of excitation for both single- and two-photon transitions. The interference between those routes give rise to EIT [115].

is negligible, the dressed-state approach provides an analogy between EIT and Fano-type interference. The dressed-states are equivalent to the presence of two levels, separated by the generalised Rabi frequency, and decaying to the common continuum level, hence interfering destructively [14]. In the general case, for an arbitrary decay rate, the theoretical approach requires the introduction of so-called *decaying dressed states*, which are the effective states of a system of atoms, a driving field, and a relaxation processes [14, 116].

Fano-type interference can only occur for a sufficiently small splitting of the dressed states, i.e., for a weak coupling field [14]. The contrast and width of the transparency depends on the rate of relaxation. For high-power coupling, the interference vanishes and the transparency window is due to a doublet structure of the absorption, i.e., ATS dominates and the interference between two pathways

cannot arise. Thus, EIT reveals a much narrower and higher contrast transparency window than laser induced state splitting (ATS).

The destructive interference can only occur in Λ and upper-state-driven cascade systems, in the weak coupling field limit [116]. This means that in V and lower-state driven cascade configurations, only ATS can be observed [14, 111, 116].

A theoretical description will be considered for the Λ system as a simplification, however, the conclusions drawn from the considerations can be directly applied to the upper-level-driven cascade system [116].

Decaying-dressed States: A Theoretical Approach

An atomic system driven by a strong coupling field can be seen as the superposition of two resonances separated by the generalised Rabi frequency $\tilde{\Omega}$. The *decaying-dressed states* can be considered as states corresponding to those two resonances. They allow for investigating of a weak-field coupling limit, when the relaxation processes start to play significant role. To show the difference between EIT and ATS, the dressed-state basis in a weak-coupling field limit must be considered.

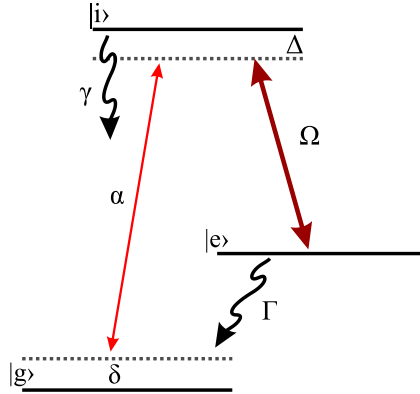


Figure 5.6: Energy-level scheme used for a decaying-dressed-state approach consideration. The strong laser field of the Rabi frequency Ω couples states $|i\rangle$ and $|e\rangle$. The field is detuned by Δ from the upper state. The weak probe beam, with the Rabi frequency α , is scanned across the $|g\rangle \rightarrow |i\rangle$ transition. The decay rate from the $|i\rangle$ to $|g\rangle$ state is γ and Γ is a decay rate from the $|e\rangle$ state to the $|g\rangle$ state.

Now, consider the energy-level scheme presented in Fig. 5.6. In this scheme, a strong coupling laser with frequency Ω drives the transition between the intermediate $|i\rangle$ and excited $|e\rangle$ states with frequency detuning Δ . A weak probe field with the Rabi frequency $\alpha = \langle g | \boldsymbol{\epsilon} \cdot \mathbf{d} | i \rangle / \hbar$ is detuned from the transition $|g\rangle \rightarrow |i\rangle$ by a frequency δ . The decay rates for the intermediate $|i\rangle$ and excited $|e\rangle$ states are γ and Γ , respectively.

In the bare-state basis (the semi-classical approach), the optical properties of a medium are described by the slow-varying coherence $\tilde{\rho}_{gi}$, i.e., a single-photon transition between the states $|g\rangle$ and $|i\rangle$, calculated with the density-matrix formalism [16]:

$$\tilde{\rho}_{gi} = \frac{\alpha}{2} \frac{\delta - i\Gamma}{\delta^2 + \delta [\Delta - i(\gamma + \Gamma)] - (\tilde{\Omega}/2)^2 - (i\Delta + \gamma)\Gamma}. \quad (5.23)$$

Equation (5.23) has two spectral poles of frequencies

$$\delta_{\pm} = \frac{-\Delta + i(\gamma - \Gamma) \pm \sqrt{[\Delta - i(\gamma - \Gamma)]^2 + \tilde{\Omega}^2}}{2}, \quad (5.24)$$

which give a resonant contribution to the atomic response. The poles can be considered as the effective states, with frequency and dephasing rates defined as their real and imaginary parts, respectively. The slow-varying optical coherence envelope can be presented as a sum of two contributions associated with the transition from the ground state to the corresponding dressed state [14]:

$$\tilde{\rho}_{gi} = \frac{\alpha}{2} \left(\frac{A_+}{\delta - \delta_+} + \frac{A_-}{\delta - \delta_-} \right), \quad (5.25)$$

where

$$A_{\pm} = \pm \frac{-\Delta + i(\gamma + \Gamma) \pm \sqrt{[\Delta - i(\gamma - \Gamma)]^2 + \tilde{\Omega}^2}}{2\sqrt{[\Delta - i(\gamma - \Gamma)]^2 + \tilde{\Omega}^2}}. \quad (5.26)$$

The two-photon coherence envelope $\tilde{\rho}_{ie}$ is defined similarly

$$\tilde{\rho}_{ie} = \frac{\alpha}{2} \left(\frac{B_+}{\delta - \delta_+} + \frac{B_-}{\delta - \delta_-} \right), \quad (5.27)$$

where

$$B_{\pm} = \frac{\tilde{\Omega}}{\delta_+ - \delta_-} = \frac{\tilde{\Omega}}{\sqrt{[\Delta - i(\gamma - \Gamma)]^2 + \tilde{\Omega}^2}}. \quad (5.28)$$

Therefore, if the Rabi frequency is significantly greater than the frequency detuning of the two-photon transition ($\tilde{\Omega} \gg \Delta$), and the difference between the decay rates is approximately zero ($|\gamma - \Gamma| \approx 0$), then the real values of the poles ($\Re[\delta_{\pm}]$) define the energy-level shift of the dressed states [see Eq. (5.24)]. The shift of each energy-level is half of the generalised Rabi frequency $\tilde{\Omega}/2$, with some small correction, which can be neglected. This agrees with the dressed-state approach, as will be discussed later. The imaginary parts of the poles $\Im[\delta_{\pm}]$ characterise the width of the resonances. In the dressed-state limit, the decay rate for both resonances is equal to the average dephasing rate $\Im[\delta_{\pm}] \approx (\gamma + \Gamma)/2 = \Gamma$.

However, if the decay rates are non-equal, $|\gamma - \Gamma| > 0$, and non-negligible when compared with the Rabi frequency $\tilde{\Omega}$, the real parts of $\delta_{\pm}$ do not reflect the usual dressed states [see Eq. (5.24)]. In other words, their splitting is not characterized by the generalised Rabi frequency. In addition, the width of the resonances, characterised by the imaginary parts of the poles ($\Im[\delta_{\pm}]$), become different from one another. Those states are called *decaying-dressed states*, and they define the atomic behaviour in the limit of significant dephasing rates [14].

Consider two dimensionless parameters:

$$x = \frac{\gamma - \Gamma}{\tilde{\Omega}}, \quad (5.29)$$

$$y = \frac{2\sqrt{(\tilde{\Omega}/2)^2 + \gamma\Gamma}}{\gamma + \Gamma}. \quad (5.30)$$

At the exact resonance ($\Delta = 0$) the amplitudes $A_{\pm}$ and the poles can be expressed by the parameters x and y in a following way

$$A_{\pm} = \frac{1}{2} \pm i \frac{x}{2\sqrt{1-x^2}}, \quad (5.31)$$

$$\delta_{\pm} = \frac{\gamma + \Gamma}{2} \left(i \pm \sqrt{y^2 - 1} \right). \quad (5.32)$$

Weak field coupling ($|x| \gg 1$).

In the weak-coupling-field limit ($|x| \gg 1$), the amplitudes A_+ and A_- correspond

to the one- and two-photon transitions:

$$A_+ = 1 + \frac{(\tilde{\Omega}/2)^2}{(\gamma - \Gamma)^2}, \quad (5.33)$$

$$A_- = -\frac{(\tilde{\Omega}/2)^2}{(\gamma - \Gamma)^2}, \quad (5.34)$$

respectively. The single-photon transition is always present, whereas two-photon transition begins to appear in the presence of weak coupling light. The amplitude of the one-photon transition A_+ is positive and greater than 1. The two-photon-transition amplitude A_- is negative (gain transition) and increases with the strength of the coupling field. The real parts of the poles are vanishing ($\Re[\delta_{\pm}] = 0$), thus, both of the resonances are centred on the $|g\rangle \rightarrow |i\rangle$ transition, and no splitting can be observed. In the absence of the coupling field, the one-photon decay rate is exactly equal the dephasing rate of the intermediate state $|i\rangle$, and for the two-photon transition it coincides with the decay rate of the excited state $|e\rangle$:

$$\Im[\delta_+] = \gamma - \frac{(\tilde{\Omega}/2)^2}{\gamma - \Gamma}, \quad (5.35)$$

$$\Im[\delta_-] = \Gamma + \frac{(\tilde{\Omega}/2)^2}{\gamma - \Gamma}. \quad (5.36)$$

If the weak coupling field is present, the resonances experience broadening or narrowing, and the observation of broadening or narrowing depends on the sign of the decay-rates difference in the denominator $(\gamma - \Gamma)$ [see Eqs. (5.35) and (5.36)]. Furthermore, the narrower transition is affected by the coupling field sooner than the other.

When $x > 1$, the broad one-photon resonance (Fig. 5.7a) is centred at the same position as the narrow two-photon gain transition (Fig. 5.7b). This is seen as a broad absorption profile with a narrow dip at the centre of the resonance. If the coupling field intensity fulfils the following restrictions:

$$\begin{aligned} 2\tilde{\Omega}^2 &\geq \gamma\Gamma, \\ 2\tilde{\Omega}^2 &< (\gamma - \Gamma)^2, \end{aligned} \quad (5.37)$$

EIT can be observed (Fig. 5.7). This is a different process than the energy-level splitting seen for a strong coupling beam. Here, the transparency window can be

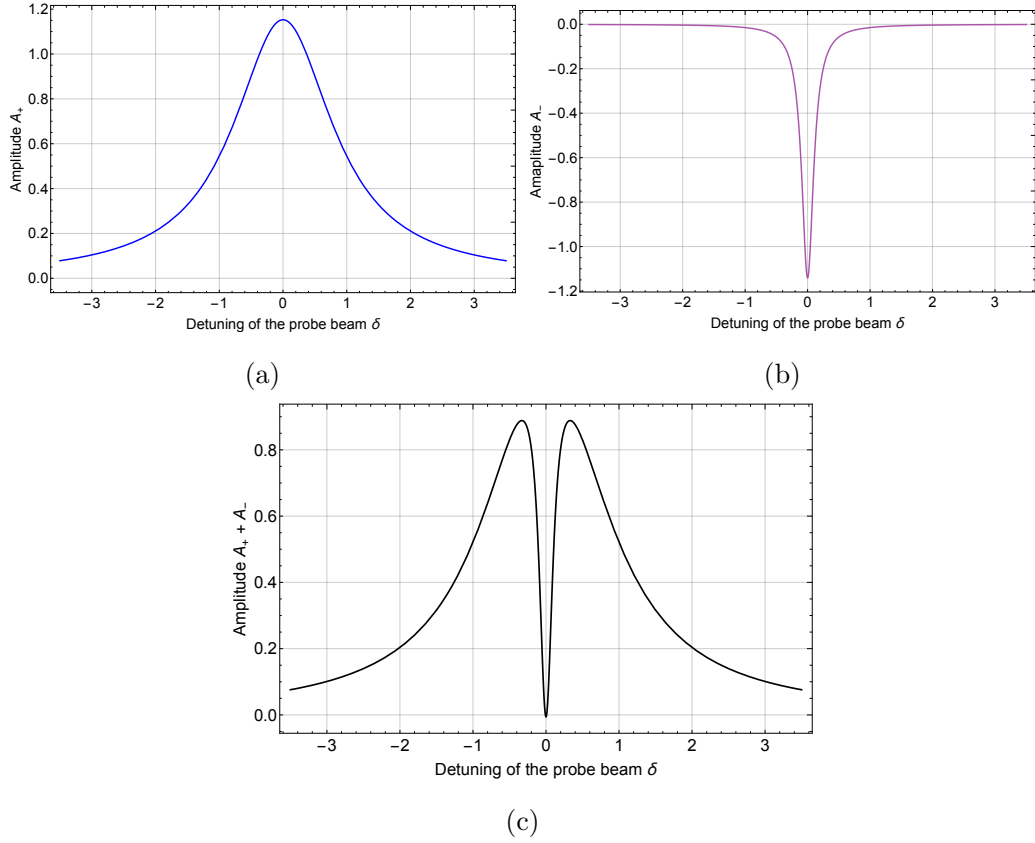


Figure 5.7: Simulated absorption and gain profiles in the weak-coupling field limit ($x > 1$) with the following parameters: $\gamma = 0.1$, $\Gamma = 1$, and $\Omega = 0.7\sqrt{\gamma\Gamma}$ ($x = 4.06$). (a) One-photon absorption with amplitude A_+ and width δ_+ described by Eq. (5.33) and Eq. (5.35), respectively. (b) A two-photon gain profile with amplitude A_- and width δ_- described by Eq. (5.34) and Eq. (5.36), respectively. (c) The sum of one- and two-photon transition profiles yielding EIT.

seen as the subtraction of a narrow gain Lorentz profile from the broad absorption profile, spectrally centred at the same frequency. Whereas ATS is seen as a summation of resonances separated by the generalised Rabi frequency $\tilde{\Omega}$.

Strong field coupling ($|x| < 1$).

Thus, for a strong driving field, i.e., $|x| < 1$, the amplitudes $A_{\pm}$ are equal to each other, and are real, positive values. The transitions from the ground level cannot be seen as one- and two-photon transitions anymore. Additionally, the dephasing

rate is equal to an average decay rate:

$$A_{\pm} = \frac{1}{2} \pm \frac{\gamma - \Gamma}{2\tilde{\Omega}}, \quad (5.38)$$

$$\delta_{\pm} = \pm \left(\frac{\tilde{\Omega}}{2} - \frac{(\gamma - \Gamma)^2}{4\tilde{\Omega}} \right) + i \frac{\gamma + \Gamma}{2}. \quad (5.39)$$

The position of resonances are $\pm\tilde{\Omega}/2$ with a slight correction from the dephasing rates, which agrees with the dressed-state approach (Fig. 5.8).

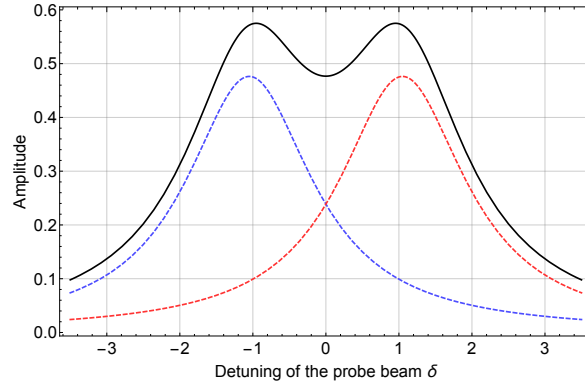


Figure 5.8: Simulated absorption profiles for a strong-coupling field ($|x| < 1$) for the following parameters: $\gamma = 1.1$, $\Gamma = 1$, and $\Omega = \gamma + \Gamma$ ($x = -0.095$). The blue and purple dashed lines indicate two resonances with amplitudes $A_{\pm}$, and widths $\delta_{\pm}$ described by the Eqs. (5.38) and (5.39). The black solid line represents the observed ATS.

Large decay rate ($\Gamma \gg \max\{\Delta, \tilde{\Omega}, \gamma\}$).

Now consider the case of a large decay rate of a two-photon resonance, i.e., $\Gamma \gg \max\{\Delta, \tilde{\Omega}, \gamma\}$. In this limit, only one shifted resonance can be observed:

$$A_- = 1, \quad A_+ = 0, \quad (5.40)$$

$$\delta_- = -\Delta + i \left(\gamma + \frac{(\tilde{\Omega}/2)^2}{\Gamma} \right). \quad (5.41)$$

Only the one-photon resonance is seen by the probe field and corresponding to the pole δ_- , positioned at $-\Delta$. Its width is defined by the decay rate of the single-photon transition power-broadened by the presence of a coupling field (see Fig. 5.9).

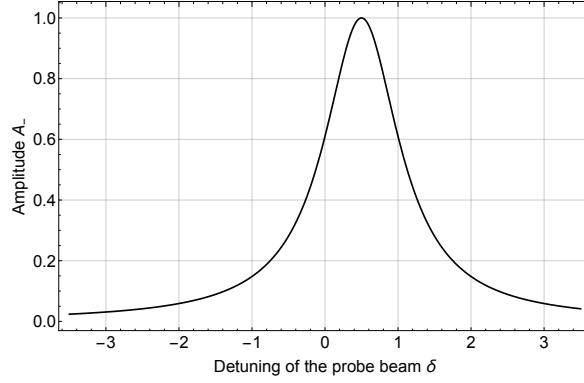


Figure 5.9: Simulated absorption spectra for large decay of two-photon resonance ($\Gamma \gg \max\{\Delta, \tilde{\Omega}, \gamma_j\}$) described by Eq. (5.40) for the following parameters: $\gamma = 0.5$, $\Gamma = 2$, $\Omega = 2\gamma$, and $\Delta = -0.5$

The theoretical description of decaying-dressed states shows a fundamental difference between EIT and ATS. These two processes occur for different conditions of the coupling field, and they reveal different properties (amplitude, width) of the induced transparency window. Therefore, in the weak-coupling limit, EIT can be observed. By increasing the strength of the coupling field, the Fano interference starts to vanish, and the state splitting (ATS) starts to dominate, i.e., there is a smooth transition between EIT and ATS [14, 116].

5.1.3 Dispersion Properties of a Medium Exhibiting EIT

The narrow resonance, seen as a suppression of absorption, is associated with a steep positive dispersion. Thus, both EIT and ATS processes are associated with a reduction of in the group velocity for light travelling through the medium. However the transparency window, produced by the quantum-interference process (EIT), exhibits narrower and higher contrast spectral features than ATS (see Figs. 5.7 and 5.8). This is associated with the long-lived coherences created in EIT.

For an atomic-vapour medium, exhibiting EIT, pulses of light slowed to 8 m/s [99], and even a complete stoppage of light [118] were demonstrated. Moreover, a stopped pulse could be released on demand. Slow and stored light in alkali vapours has a potential application in quantum-information processing as logic gates [17,

24, 59].

The EIT and ATS processes and the related pulse delays were previously observed in alkali-filled HC-PCFs for V and Λ energy-level configurations [20, 55, 89]. Within this dissertation, the processes were investigated in the upper-state driven cascade and V energy-level schemes.

5.2 Electromagnetically Induced Transparency in Cascade

5.2.1 Experimental Setup

The theoretical description of the dressed-state system can now be applied to a real atomic system. In rubidium, the ground, intermediate, and excited levels may be the $5S_{1/2}$, $5P_{3/2}$ and $5D_{5/2}$ states, respectively. In an upper-state-driven cascade system, the strong light couples the intermediate and excited states. Initially the upper states are not populated, and a weak probe beam does not effect this. Hence, there are no pumping effects populating the other hyperfine ground-energy level, i.e., the interaction with the weak probe beam is negligible. Furthermore, residual Doppler broadening is minimized by a counter-propagating beam alignment, and similar frequencies of both transitions in rubidium atoms (see Section 4.3).

The experimental setup was previously described in Section 3.2.1, and here only the most important elements will be recalled. The kagome-latticed Fibre 2, entirely filled with rubidium atoms and held in approximately 70°C, was used in the experiment. The ECDL was scanned across the rubidium D_2 line (the probe beam), and the tunable Ti:Sapphire laser was used as a coupling-light source. The coupling laser was tuned to the $5P_{3/2} \rightarrow 5D_{5/2}$ transition in rubidium 87. The coarse separation of the beams was done using the orthogonal polarization alignment. To make sure that no reflection from the coupling beam will affect the measurement, a diffraction grating was placed before the APD. The amplitude was modulated with the AOM, and the weak probe beam was measured using a phase-sensitive detector (see Section 2.4.3).

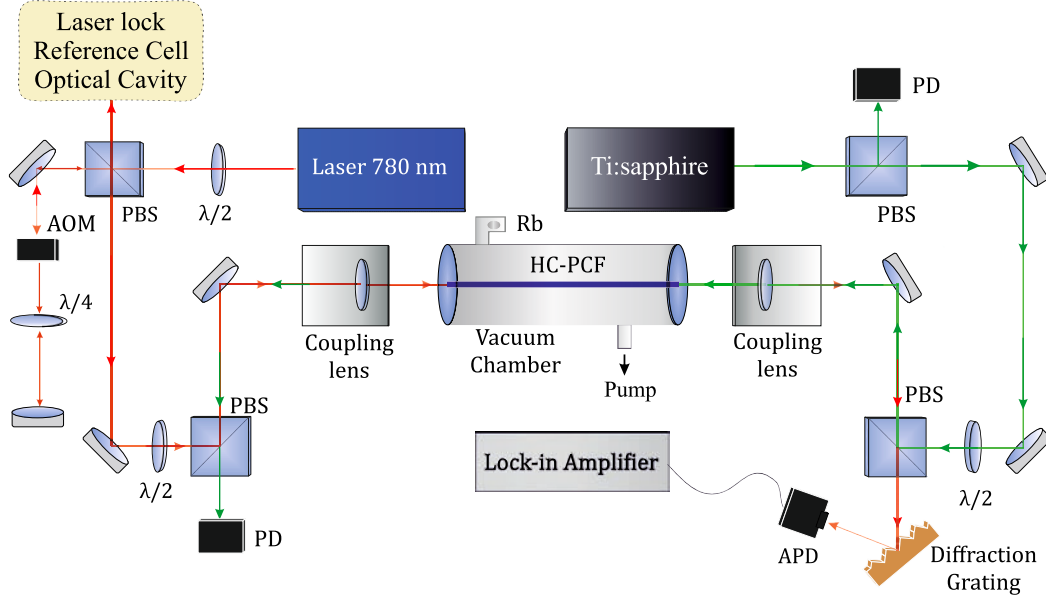


Figure 5.10: Experimental setup of the EIT experiment realised in the cascade energy-level scheme. The weak probe beam (100 nW) was provided from the ECDL (780 nm). The probe signal was modulated by the AOM and measured with a lock-in amplifier (phase-sensitive detection). The strong counter-propagating coupling beam (19 - 50 mW), working on the $5P_{3/2}$ to $5D_{5/2}$ transition, was delivered by the Ti:Sapphire laser (776 nm). To prevent any reflection of strong desorbing light on the APD measuring the probe signal, a diffraction grating was used.

5.2.2 Results

The theoretical value of the Rabi frequency for the coupled $5P_{3/2}$ and $5D_{5/2}$ states was first calculated to predict the ATS for the examined system. The calculated splitting values for various beam intensities are presented in Fig. 5.11. The computed Rabi frequencies were compared with experimental values presented in Ref. [115]. The theoretical and experimental values are in good agreement.

The experimental results of the ATS and EIT processes are presented in Fig. 5.12. Here, the weak probe beam (100 nW, corresponding to $1.3 \mu\text{W}/\text{cm}^2$) was scanned over the rubidium (780 nm) D_2 line. The reference spectra, in the absence of the coupling beam, is seen as a grey line. The strong beam (43 mW, corresponding to $540 \text{ W}/\text{cm}^2$ inside the fibre), coupling the $5P_{3/2}$ and $5D_{5/2}$ states

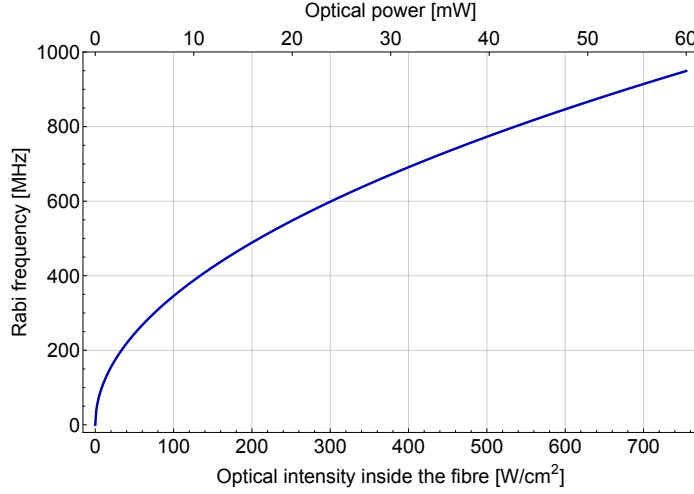


Figure 5.11: Theoretical predictions of the Rabi frequency for the $5P_{3/2}$ to $5D_{5/2}$ transition.

of ^{87}Rb , was slowly scanned in the vicinity of the two-photon resonance. The blue line corresponds to the spectra observed in the presence of the coupling field for four distinct coupling-beam tunings.

In the presence of the strong coupling beam, a broadening and shift of the transitions in the D_2 line, for both rubidium isotopes, were observed. The shift was caused by the presence of a high intensity electric field (Section 4.5.2). The broadening can be caused either by the shift of the levels or by desorption of the atoms from the fibre surface, and hence enhanced absorption and relaxation. Gaussian functions were fitted to the $5S_{1/2} F = 1 \rightarrow 5P_{3/2}$ transition in rubidium 87 to resolve information about the broadening for in reference spectra (grey line from Fig. 5.12), and when a detuned ($\delta = 815$ MHz) coupling field was present in the fibre (Fig. 5.13). The results of the fitting are summarised in Table 5.1. They show that the observed broadening is actually increased absorption caused by the change in the OD inside the fibre.

For the coupling field, detuned from the two-photon transition, a strong two-photon absorption profile can be seen on the side of the single-photon resonance. Predicted by the theory, 700 MHz ATS was not observed (Figs. 5.12a, 5.12b and 5.12d). The only sign of any splitting can be tentatively deduced from the feature marked in Fig. 5.12c with a red arrow.

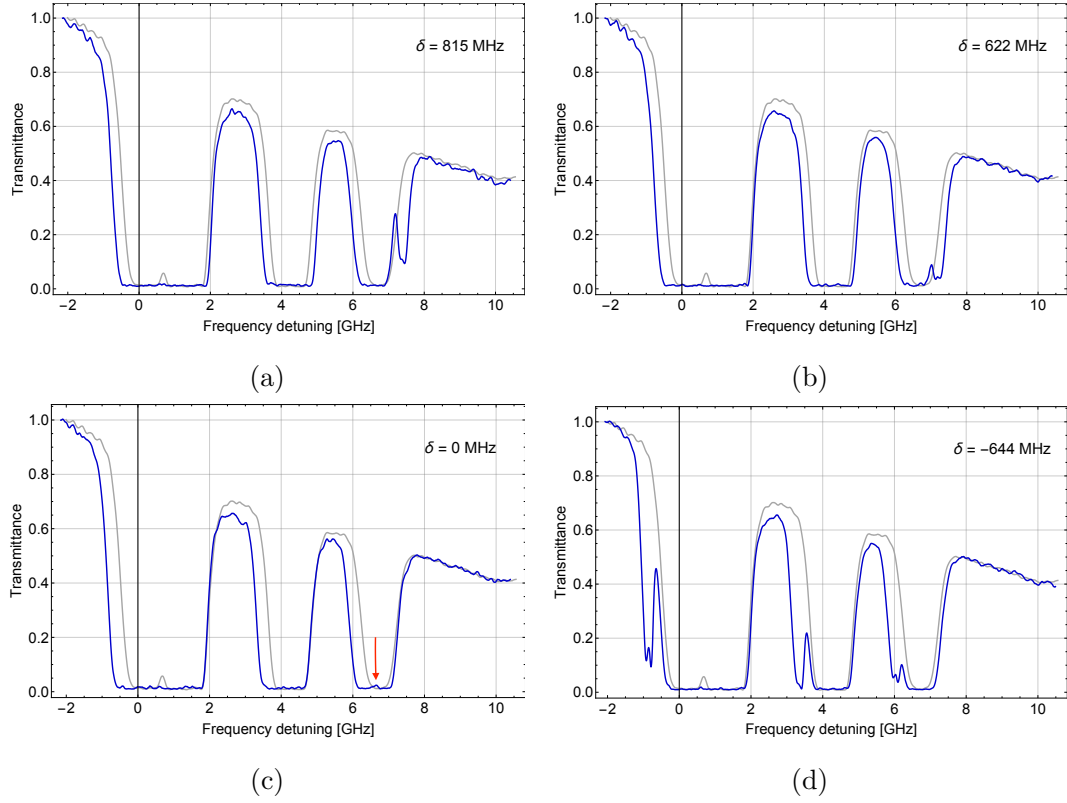


Figure 5.12: Observation of the EIT effect in the cascade system for a dense atomic vapour filling the fibre, by scanning the probe laser in the presence of the coupling beam. Each of the panels present different frequency detunings δ of the coupling field: (a) 815 MHz, (b) 622 MHz, (c) exactly on resonance, and (d) -644 MHz. Then exactly on resonance (c) a small feature, marked on with a red arrow, is visible. This may indicate state splitting. The grey line represents the reference spectra, when there was no coupling beam present.

The absence of level splitting is an interesting result, which indicates that neither ATS, caused by strong beam coupled to the upper-states of the cascade system, nor EIT are observed. The OD in the vapour-filled fibre was approximately 17.5(5), which seems to be the main problem affecting the EIT observations. The interaction between the two beams is significantly reduced, as the atoms fully absorb the probe beam when exactly on resonance. However, this does not fully explain, why ATS was not observed. The increased number of atoms in the presence of strong control field, and the resulting increased relaxation rate, could cause a

broadening of the split states. Therefore, a sign of the splitting could be the feature observed in Fig. 5.12c.

In the next step, the optical power of the probe beam was increased to $10 \mu\text{W}$ (corresponding to $126 \mu\text{W}/\text{cm}^2$). At this point, the D_2 transition was saturated and the shape of the Doppler- and power-broadened $5S_{1/2} F = 1$ to $5P_{3/2}$ transition was visible. This is shown in Fig. 5.14a as a solid black line, where the transmittance of the probe beam is approximately 60%. However, under such conditions the approximation of the non-populated intermediate and upper states, i.e., the weak probe beam, seems not to be valid. Additionally, the theoretical description of EIT and ATS processes may not be valid as well.

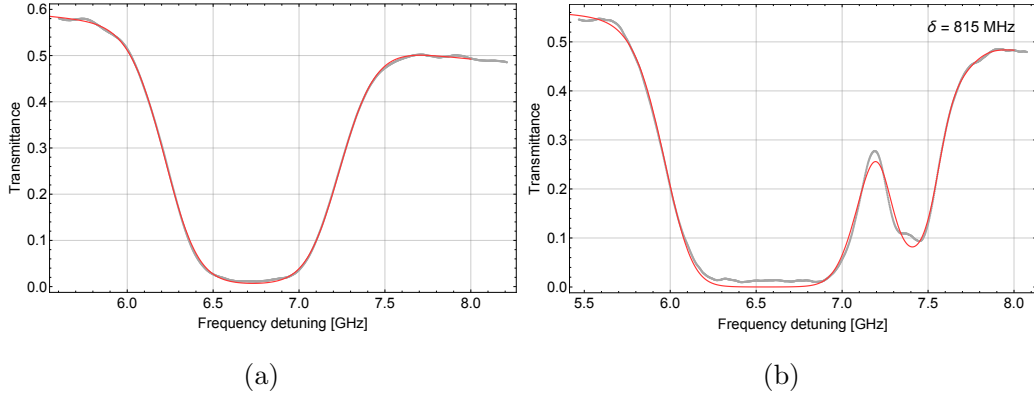


Figure 5.13: Spectrum of the $5S_{1/2} F = 1 \rightarrow 5P_{3/2}$ transition in rubidium 87 measured: (a) in the absence of the coupling field (reference spectrum) and (b) in the presence of the coupling field ($540 \text{ W}/\text{cm}^2$) detuned by 815 MHz, indicated as two-photon transition on the single-photon resonance. The red line indicates the theoretical fit.

A strong effect caused by the presence of the coupling beam is desorption of rubidium atoms from the inner-fibre walls. This was seen as increased absorption of the single-photon transition (see Fig. 5.14a). In addition, a temporal appearance of the transparency window in the resonance transition could be observed. This temporal splitting was immediately masked and vanished, which is plotted in Fig. 5.14a as a different coloured lines. Each line represents the spectra measured at: 0s, 5s, and 13s after the coupling beam was turned on. After a 13-s time

period, the splitting completely vanished, and only a small feature (see solid black line in Fig. 5.15) was visible. This feature looks similar to the one observed for the weak probe beam (see Fig. 5.12c).

	α	σ [MHz]
no coupling field	4.349(8)	267.5(2)
coupling field present	8.30(10)	266.9(8)

Table 5.1: Coefficients extracted from the Gaussian fit to the Doppler-broadened $5S_{1/2} \ F = 1 \rightarrow 5P_{3/2} \ F' = 0, 1, 2$ transition in rubidium 87 (Fig. 5.13), where α indicates the absorption strength and σ is the Doppler width (FWHM) of the transition.

Since there is a smooth transition between ATS and EIT, it is likely that the transparency window can be observed as the system approaches the weak-coupling field limit. A direct transition between the two processes makes them hard to distinguish [116]. However, the large relaxation of the two-photon coherence leads to the observation of a single broadened transition. Thus, the observed feature can be considered as a sign of EIT or ATS.

There are two intuitive reasons for the observed disappearance of the transparency window discussed in the following.

First, the sudden release of the atoms from the inner fibre walls results in a high OD, and causing near-zero transmission at the exact resonance for the probe beam. This is similar to the initial situation, where no splitting was observed for a non-saturated probe transition. The atoms exactly on resonance are not seeing both beams, as the probe beam is strongly absorbed. There, in the EIT regime, the high-contrast transparency window cannot be built, since there is no second path of the interference.

Second, the sudden increase of the optical density not only affects the absorption, but also changes the dephasing rate inside the fibre significantly. Since the dressed-state approach is considered in a secular limit, i.e., the driving field is greater than any relaxation processes ($\tilde{\Omega} \gg \gamma, \Gamma$), the increase of the dephasing

rate inside the fibre can make this approximation invalid. For a large-decay rate of the excited state $|e\rangle$, only the frequency shifted single-photon transition can be observed [14]. This was shown by the decaying-dressed state approach. However in this case, the dephasing rates for both (intermediate $|i\rangle$ and excited $|e\rangle$) states increased, which causes broadening of the split states and a decrease in the splitting value [see Eq. (5.39)]. If the one-photon decay rate starts to dominate over the coupling fields Rabi frequency, the system starts to approach weak-field coupling (EIT starts to dominate). However, the large relaxation of the intermediate state also affects the amplitude and width of the two-photon gain transition [Eqs. (5.34) and (5.36)], which means that the observed EIT would have a low contrast. Thus, the observed feature (see Fig. 5.12c) could be EIT.

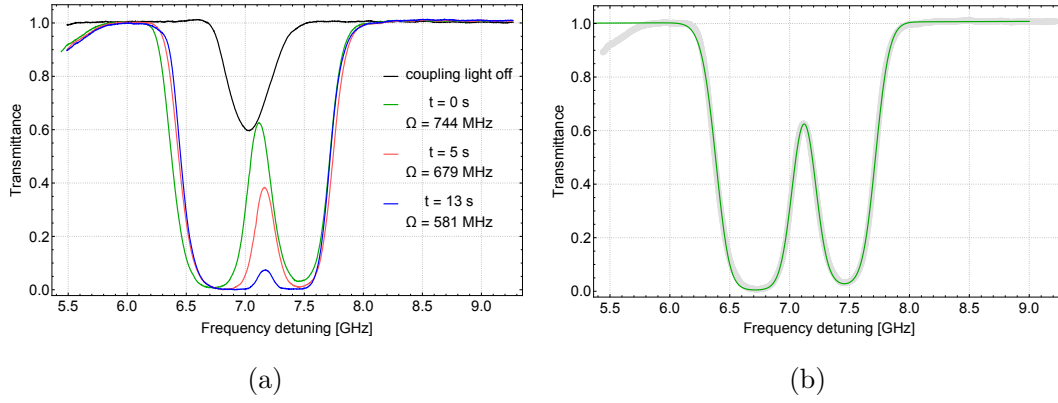


Figure 5.14: Rubidium 87 spectra measured under the condition of Autler-Townes splitting for the coupling beam of intensity 628 W/cm^2 . (a) The absorption spectrum with a clearly visible ATS feature for different times after switching on the coupling beam (different colours). The black curve is a reference spectra at the absence of the coupling beam. The other lines show the time-dependent vanishing of a splitting. (b) The theoretical model fit of the spectrum measured right after the the light switching on at $t = 0$ s.

To measure the experimental value of the splitting, two Gaussian functions were fitted to the obtained spectrum taken right after switching on the coupling laser, as shown in Fig. 5.14b. The splitting value is determined by the Rabi frequency (Section 5.1.1). Fitting to the data revealed that the separation between

the fitted functions was 744(2) MHz for the 50-mW coupling beam (628 W/cm^2), where the predicted value was 866 MHz. The same approach was implemented for various optical powers of the coupling beam, the results are summarize in Table 5.2.

optical power [mW]	19	23	46	50
light intensity [W/cm^2]	239	289	578	629
calculated Rabi freq. [MHz]	534	587	831	866
measured Rabi freq. [MHz]	472(1)	521.0(2)	738.3(5)	744.2(3)

Table 5.2: Theoretical and measured values of the Rabi frequency for ATS in the cascade scheme.

It is noteworthy that, all the measured values are lower than the predicted ones. This is most likely caused by absorption, i.e., an uncertainty in the actual maximum splitting obtained by the coupling light, before the desorbed atoms start masking the observed process.

In the presence of the coupling field, at much later times after turning the beam on, the probe light was strongly absorbed and the absorption resonances were broadened. After turning the coupling beam off, the depth of the transition was reduced to its initial value, which is shown in Fig. 5.15. The solid black line indicates an equilibrium situation, when the coupling beam has been present in the system over a long time period. The small feature around the centre of the broadened line indicates the residual splitting (or more likely EIT), similar to those observed for the non-saturated probe beam (see Fig. 5.12c). The green, red, and blue lines represent measurements after time delays of 5 s, 14 s, and 34 s, respectively, from when the control beam was turned off. The absorption of the single-photon transition, i.e., the $5S_{1/2} F = 1$ to $5P_{3/2} F' = 0, 1, 2$ transition, decreases, and the full Doppler- and power-broadened transition starts to be visible. This confirmed the assumption that the density of atoms is not allowing for the observation of energy-level splitting.

As the results show, EIT cannot be easily performed in vapour-filled HC-PCF

in the cascade system. It is difficult due to additional processes (auxiliary relaxation, absorption) that strongly contribute to observed signals. The interaction between two electromagnetic fields is significantly reduced due to the high optical depth inside the fibre and its counter-propagating configuration. For further examination of EIT in the fibre, the V energy-level scheme was used.

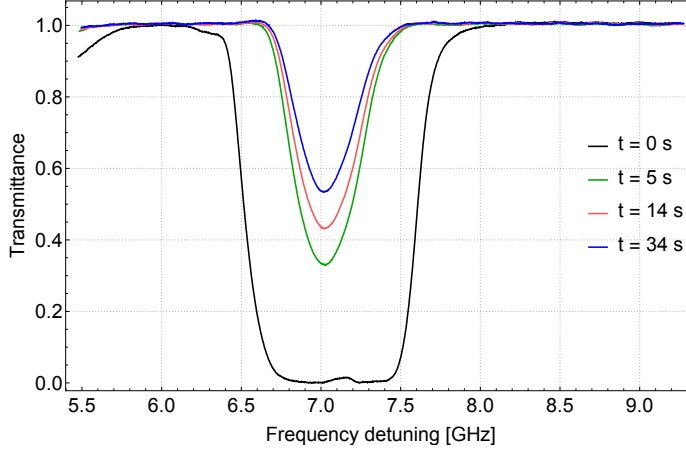


Figure 5.15: Absorption spectra of the probe beam operating at the $5S_{1/2} F = 1 \rightarrow 5P_{3/2} F' = 0, 1, 2$ transition at different times after switching off the control beam. The solid black line indicates the equilibrium situation for the desorption process (coupling beam switched on for a long time), and the blue line indicates the stable spectrum measured in the absence of the beam.

5.3 Autler-Townes Splitting in V

As discussed above, a transparency window can be observed in several energy-level configurations: Λ , V , and cascade. These schemes can be realised in various physical systems. For example, the V scheme can be realised in Rb atoms using the strong D_1 and D_2 lines.

In such a system, the smaller relaxation of the lower-energy state allows the energy-level splitting to be observed for a lower intensity of the coupling beam. It is noteworthy that atomic population will be pumped out of the system into the other hyperfine ground state, causing a decrease of the absorption. In general, this is a disadvantage, causing a low contrast in the observed transparency window

[13]. Since the pumping processes reduce the overall absorption, this also means that ATS could be observed regardless of the desorption processes.

The V energy-level scheme was chosen to further examine the possibility to generate a transparency window in a three-level system in the rubidium-filled fibre (Fig. 5.16).

5.3.1 Experimental Setup

A coupling field was applied at the D_1 line (795 nm), and tuned to the $5S_{1/2} F = 1 \rightarrow 5P_{1/2}$ transition in ^{87}Rb . A probe field was scanned across the D_2 line. Furthermore, the transparency window induced in the V scheme cannot be separated from the increased probe laser transmissions caused by saturation of the D_1 line, i.e., the presence of the coupling field causes a reduction of absorption seen by the weak probe beam.

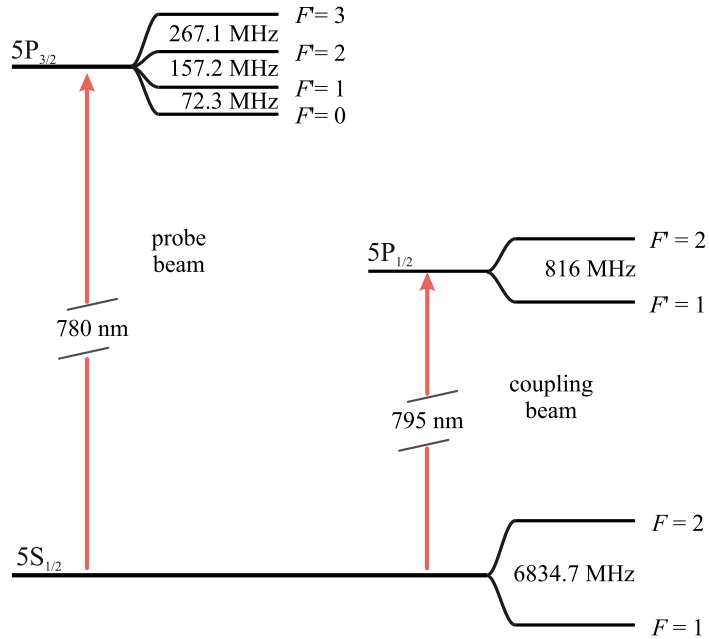


Figure 5.16: V -type energy-level scheme of rubidium 87 exploited for investigations of ATS.

The coherence dephasing rate is an average of the decay rates of the two upper states incorporated in the ATS [13]. The dephasing rate for this configuration is higher than for the cascade scheme. However, the lifetime inside the fibre is reduced due to the transit time, so the optical pumping rate is also reduced.

In the considered case, the residual Doppler broadening is different than for the two-photon case. Here it is given by

$$\nu_D = \sqrt{\frac{2k_B T}{m}} |(k_p - k_c)|, \quad (5.42)$$

where k_B is the Boltzmann constant, T is the temperature, m is the atomic mass, and k_p and k_c are the probe- and coupling-field wavenumbers, respectively [13]. If the beams are counter-propagating, one of the beams has a negative wavenumber and its contribution to Doppler broadening is enhanced. Therefore, to reduce the Doppler broadening, the beams were set in co-propagating manner. The experimental setup is shown in Fig. 5.17. The light from the ECDL and Ti:Sapphire lasers were coupled into a single-mode fibre coupler. The optical power of the probe beam was set to ~ 20 nW (corresponding to $0.25 \mu\text{W}/\text{cm}^2$), which is below the saturation level of the D_2 line in rubidium. The coupling laser power was set to 29 mW (corresponding to $356 \text{ W}/\text{cm}^2$). The polarisation of the lasers was set to be orthogonal for coarse beam separation. Moreover, to make sure that no coupling beam was seen by the APD, a diffraction grating was used. Otherwise, the setup was similar to the experimental setups described previously.

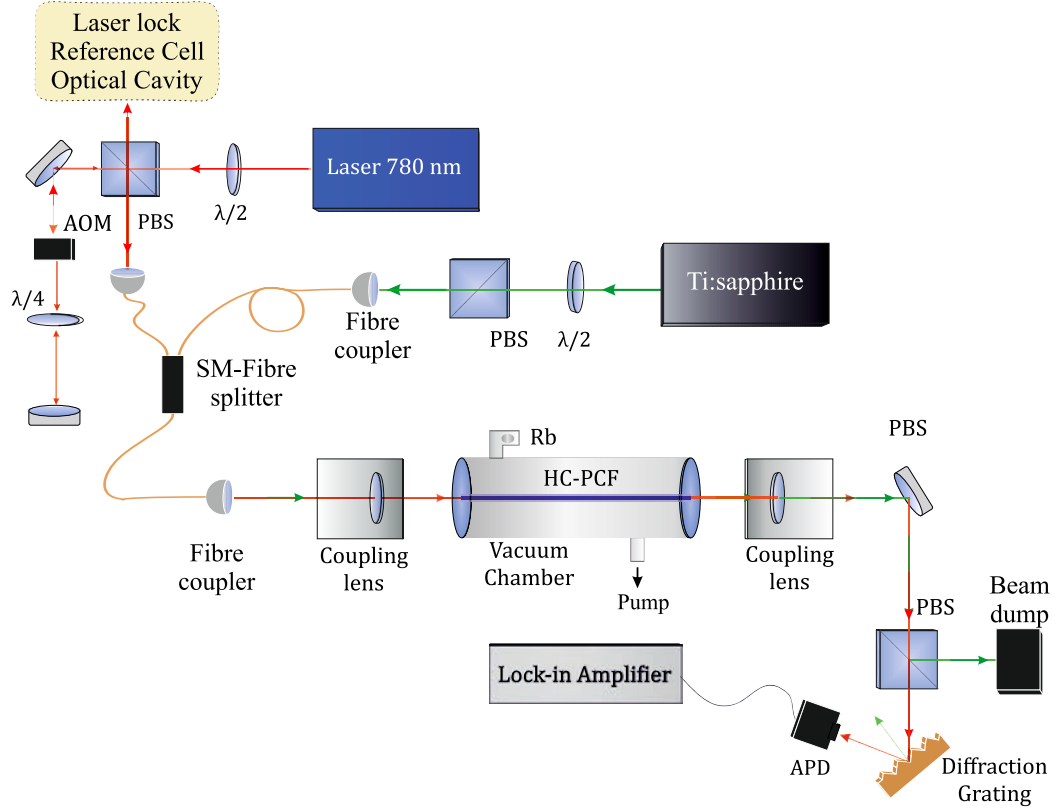


Figure 5.17: Experimental setup for ATS in a rubidium-filled HC-PCF in the V scheme. To reduce Doppler broadening, the coupling and probe beams were set in a co-propagating manner. They were coupled into an SMF coupler for a perfect spatial overlap. The probe beam was modulated with the AOM. The power of the weak probe beam was measured with an APD. The measured signal was sent to a lock-in amplifier, where phase-sensitive detection was used to reveal the spectra. The probe and coupling beams were separated by setting them to have orthogonal polarisations. Fine separation was provided by the diffraction grating.

5.3.2 Results

The results of ATS generated in the V scheme are shown in Fig. 5.18. The observed splitting of the states due to the presence of the strong coupling beam, reveals a low contrast suppression of the absorption. In addition, the overall absorption is highly reduced due to optical pumping of the population to the uncoupled $5S_{1/2}$ $F = 2$ state.

At the exact resonance, the presence of the coupling beam creates a transparency window in the absorption profile of ^{87}Rb : the $5S_{1/2}$ $F = 1$ to $5P_{3/2}$ $F' = 0, 1, 2$ transition. This is shown in Fig. 5.18a. Desorption processes, which accompany high-intensity electromagnetic radiation, do not significantly affect the observed transparency. Thus, the transparency window can be observed for the low-power probe beam. The transmission of the probe beam is significantly increased, leaving a broad transparency feature. This is caused by optical pumping and saturation of the D_1 transition by the strong coupling field.

If the coupling laser is detuned from resonance, it causes asymmetric splitting of the atomic states and pumping of the atomic population into the other hyperfine levels of the $5S_{1/2}$ state. This asymmetric splitting influences the routes of excitation to the upper-levels of the D_2 transition. This is shown in Figs. 5.18b to 5.18d. Therefore, the weak probe beam observes increased *and* reduced absorption due to saturation and optical pumping effects, as well as asymmetric splitting of the ground state induced by the strong coupling beam.

In the presence of the strong coupling beam, all the transitions for the D_2 line for both isotopes are shifted and broadened. This is a similar situation to the one observed in the cascade system, where the broadening was caused by desorption of atoms, and the asymmetric shift of the transition frequency is caused by the interaction with a strong electric field.

The transparency window due to Autler-Townes splitting in the V scheme was successfully observed with the weak probe beam. Therefore, as predicted, the desorption processes did not affect the observed transparency. However, the contrast of the induced transparency was significantly reduced due to optical pumping.

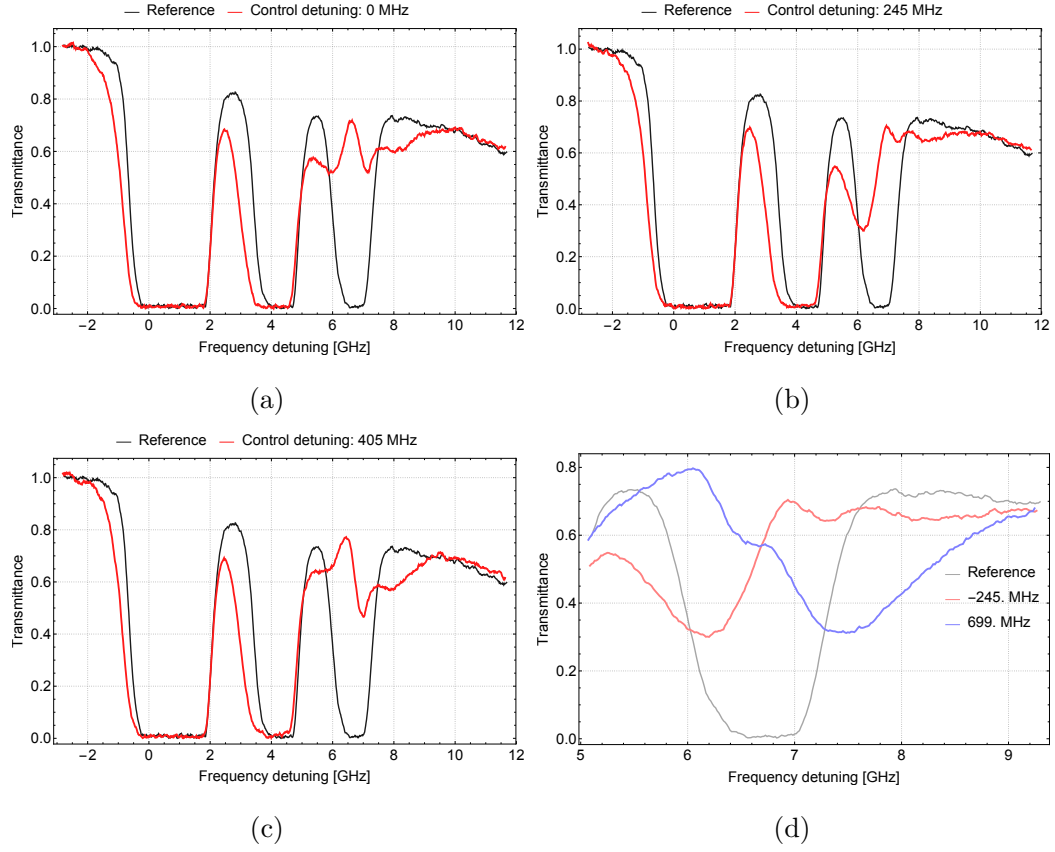


Figure 5.18: Absorption spectra observed in the V energy-level scheme under the condition of ATS for a different coupling beam tunings. Plots with a couplig field detuning of: (a) 0 MHz, (b) 245 MHz, (c) 405 MHz, and (d) -255 MHz (red line) and 700 MHz (blue line).

5.4 Summary

Coherent effects in a three-level atomic system inside a HC-PCF were examined. Studies on EIT and ATS phenomena were performed in the upper-driven cascade and V energy-level systems.

The temporal transparency window in the cascade system in an optically dense atomic medium (rubidium vapour), filling HC-PCF, was observed. It was possible to achieve a transparency window of spectral width of 641(1) MHz, with approximately 60% contrast. In the real experiment, the width value seems too large to obtain significant values of the group refractive index, which also indicates trans-

parency originating from energy-level splitting, rather than EIT.

The first problem in successfully realising a narrow-width high-contrast transparency window was the high density of the atomic medium, which was enhanced by the light-surface interaction with the strong coupling field. This led to the created splitting of the states disappearing due to an increase of the absorption and dephasing-rate values. A possible way to avoid this problem would be using a pulsed coupling field, which might reduce the influence of the desorption processes on the observed signal. Additionally, dynamic control of the atomic densities inside the fibre with a far-detuned desorbing laser, could be applied to avoid fluctuations and an uncontrollable increase of the atomic concentration during the measurement (see Section 3.2). The measured time scale of the desorption processes in Chapter 3, and the disappearance of ATS indicate that this approach would be possible for short pulses. The other possibility is to use kagome-latticed Fibre 1, where the light-surface interaction is significantly reduced, as the mode area is smaller than the core diameter (Table 2.1). Furthermore, the use of a shorter piece of the atom-filled fibre would improve the light transmission. However, it could still not be enough to avoid saturation of the probe transition. The proposed solutions might improve the observed transparency window, but it would act to ‘fight against’ the advantages of a dense atomic medium inside a fibre-host. This shows that a vapour-filled fibre is problematic platform to measure coherent effects, which require strong coupling beams.

In general, coherence experiments in an optically dense atomic media can be advantageous, and provide narrow spectral features for further optical switching and quantum logic gates [101, 119, 120]. It was shown by M. Lukin et al. [120] that narrow EIT features could be observed in fully opaque media for weak probe beams in the Λ energy-level configuration. A far-detuned four-level energy scheme would be a good choice to observe coherent effects, as was proposed by S. E. Harris and Y. Yamamoto⁵⁶ [121–123], and A. Joshi and Min Xiao⁵⁷ [22, 124].

To examine the three-level energy scheme for slow-light applications, the V

⁵⁶ N energy level scheme

⁵⁷Inverted- Y energy level scheme

type ATS was also explored. This energy-level scheme in rubidium vapours has the advantage of exploiting two strong transitions (the D_1 and D_2 line). The V configuration requires co-propagating beams to reduce Doppler broadening, and at the same time the interaction between both, the coupling and probe beams, remains at least in the first centimetres of the fibre. Thus, attenuation of the guided light do not play a significant role in this configuration. Furthermore, what is usually a disadvantage, the strong coupling beam optically pumps the second non interacting, ground level. This results in an increase in the transmission of the probe laser, but also causes a lower contrast of the observed transparency window. Transparency in this scheme was successfully observed and the desorption processes did not significantly affect the observed feature. However, this energy-level scheme is not suitable for achieving large values of group refractive index for travelling probe light.

There are many alternative ways to perform slow light and storage of light experiments, both of which open further possibilities in quantum information and communication systems. Atom-fibre hybrid platforms allow for realising all-optical switching [6, 20], logic gates [22, 101], metrology systems [77], and transmission and delay lines, in both thermal and cold atoms systems. One alternative is to incorporate TPA or other non-linear processes, which have advantages of high optical depths (ex., N or Y energy level schemes). This makes them promising future for all-optical devices.

Chapter 6

Conclusions

The aim of this project was to perform linear and non-linear spectroscopy in rubidium-filled HC-PCF. The main goal was to experimentally realise an all-optical switch based on the TPA and EIT processes, which allow for dynamic control over the group velocity and further storage methods of light. TPA and upper-driven cascade EIT share the same energy-level scheme, however, a strong coupling field can change the character of the observed effect, and produces either an absorption- or a gain-profile, respectively.

TPA results in the anomalous dispersive properties, which are associated with fractional or negative group-refractive index values. This gives rise to a superluminal group velocity ($c < v_g$), and hence propagation at faster than light speeds. Indeed, a negative refractive index and pulse advancement was observed in experiment. Both the amplitude of the TPA and the refractive index value were easily controlled by adjusting the optical power of light. The amount of advancement induced by low-intensity light agrees with theoretical predictions, however for larger light intensities the pulse advancement was distorted. Furthermore, two-level coherences (i.e., two-photon Rabi oscillations) in this three-level atomic system were observed and characterised, which could be the origin of the distortion. Future work on fast-light processes requires a deeper analysis of the origin of the discrepancy between predicted advancement values and the experimental data. This includes further studies on the induced Rabi oscillations in a three-level system. One solution would be to reduce the rise time of the pulse via faster switching. This

could be done by replacing the pulse driving AOM by an electro-optical modulator (EOM). This could allow for a detailed examination of spatial pulse expansion in a fast-light medium.

The EIT, as well as ATS, processes results in a gain resonance, which is associated with the normal dispersive properties of a medium, thus slow-light can be performed. Since an optically dense medium, such as a long piece of HC-PCF entirely filled with atomic vapours, allows for enhanced TPA, the EIT process could not be easily observed. Only temporal state splitting, caused by the presence of strong light, could be observed. However, this was further masked by the increasing number of atoms desorbed from the porous-silica surface of the fibre. The width of the observed transparency seems to be too large to obtain a significant reduction in the group velocity inside the HC-PCF. This requires further studies with different energy-level configurations, for which the high OD would be an advantage.

For the purpose of this project, an experiment consisting of a vacuum system for filling a fibre with alkali atoms and an optical measurement system were built. These were also a significant part of the performed experiment.

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Acronyms

AOM acousto-optic modulator. 30, 31, 34, 47–49, 71, 72, 74, 80, 102, 124, 125, 135, 142

APD avalanche photodiode. 47–49, 72, 73, 124, 125, 134, 135

ATS Autler-Townes splitting. v, 114–117, 121–131, 133–139, 142

BS beam splitter. 30, 31

CPT coherent population trapping. 2, 4, 107

CW continuous-wave. v, 46, 49, 51, 55, 75, 80, 83

DAQ data acquisition. 70

DAVLL dichroic atomic vapour laser lock. 30–32

DDS direct digital synthesizer. 47

ECDL external-cavity diode laser. 22, 30–32, 43, 46, 69–71, 124, 125, 134

EIT electromagnetically induced transparency. v, 2–4, 8–10, 107, 109, 110, 114–117, 120, 121, 123–125, 127–132, 137, 138, 141, 142

EOM electro-optical modulator. 142

FWHM full width at half maximum. 15, 16, 50, 77, 129

FWM four-wave mixing. 2

GVD group velocity dispersion. 84–87

HC-PCF hollow-core photonic-crystal fibre. v, 7–10, 19–27, 29–31, 36, 37, 39, 42–46, 48, 49, 55, 56, 68, 76, 79, 105, 124, 131, 135, 137, 141, 142

LIAD light-induced atomic desorption. 9, 41–46, 49–51, 54–56, 68

LO local oscillator. 47, 73

MOF microstructured optical fibre. 6, 7, 19, 20, 22, 24, 39

NA numerical aperture. 22, 23

NIR near-infrared. 17, 42

OD optical depth. 9, 37, 38, 41–45, 50–56, 75, 77–79, 126, 127, 129, 132, 139, 142

PBG photonic-bandgap guidance. 20–22

PBS polarising beam splitter. 31, 70

PD photodiode. 49

PMT photo-multiplying tube. 71

PSD phase-sensitive detection. 30, 33, 47, 125, 135

RGA residual gas analyser. 27, 28

RWA rotating-wave approximation. 90, 94

SAS saturated absorption spectroscopy. 9, 19, 30, 31, 39, 47–49, 61, 70

SCF solid-core fibre. 7, 19, 24

SMF single-mode fibre. 31, 32, 70, 72, 135

SPID surface-plasmon induced desorption. 42, 54

TPA two-photon absorption. v, 8, 9, 58, 61–63, 66–68, 70–72, 74–81, 87, 88, 93, 105, 139, 141, 142