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Electromagnetically Induced Transparency and Squeezed Light

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Abstract

This thesis presents the experimental demonstration of novel methods for generating and interacting squeezed states of light with atomic ensembles. For the first time, three different techniques have been put together: narrow band non-classical light generation at atomic wavelengths using optical parametric amplifiers, electromagnetically induced transparency interaction in the quantum regime, and full state reconstruction after the atomic interaction using time domain homodyne tomography.

We used this approach to realize the first proof of principle experiment of storage of squeezed light. Given the nature of our experiment, these results can be extended to arbitrary quantum states of light, therefore achieving a universal toolbox for quantum optical memory.
A mi madre Rosa, la razón de todo mi esfuerzo.

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Chapter 1

Introduction

Light is a robust and reliable carrier of quantum information, and it can be used to perform quantum information processing. However, every computer, quantum or classical, needs memory: a means of interim and long-term storage of the information to be processed. Following this definition, we can think of a quantum memory as a device that stores quantum information. If we intend to design a light-based quantum computer we need to think of a medium that would allow storage of the quantum information carried by light.

A very promising candidate for this role is atomic vapor. Atoms cannot transport quantum information well, but they are very good at storing it, and, most importantly, interact strongly with light. Generally speaking, we need to develop an interface that would allow quantum information to be transferred between optical and atomic media.

The solution to the atom-light interface problem could be given by the recently developed technique of storage of light by means of electromagnetically-induced transparency (EIT) [7]. EIT is a quantum interference effect that permits light to propagate through an otherwise opaque medium. Observed in gaseous atomic media, EIT is associated with high linear dispersion which leads to a tremendous reduction in the group velocity of light [8]. Reducing this velocity to zero will stop, and store, the light pulse in the medium. The process can be reversed and the light pulse regenerated in its original quantum state, thus implementing a quantum memory cell for light.

Considerable experimental effort has been given to achieve quantum memory. Seminal work in the storage of pulses of coherent light [9] was followed by the realization of EIT in the quantum regime [10] and the storage of single photons produced in atomic ensembles.
Nevertheless, since it is not possible to define the phase of a single photon (at least not in the classical sense), experiments where the quantum fluctuations in both the field strength and the phase are stored in an atomic coherence became the next step. For these experiments to be realized, squeezed light is a very good candidate since it provide us with a state where the quantum fluctuations are phase dependent. Moreover, if after the storage we can completely reconstruct the state, we could in principle repeat the experiment for any non-classical state, therefore achieving a universal test-bed for quantum memory.

In order to achieve this goal, work in three branches of this topic needed to be done: a) characterization of the decoherence mechanisms that destroy EIT and the subsequent storage, b) implementation of sources of light with non-classical properties in order to bring these processes to the quantum regime, and c) implementation of protocols useful for quantum communications that take advantage of the atom-light interaction. Work in these directions will be addressed in the following chapters of this thesis.

In Chapter 2, I make a brief review of the main theory of electromagnetically induced transparency and its use for storing optical states in atomic media. I will also review the state of the art experiments that have been performed over the past five years as well as their relation to our results.

In Chapter 3, I review our experimental study of decoherence of the ground energy levels of $^{87}$Rb atoms in vapor cells. The decoherence of the ground state was measured using three different methods: measuring the decay constant of the storage of light in atomic vapor, the decay rates of transient coherence oscillations of the ground state, and the width of the electromagnetically induced transparency resonances. All the measurements showed decoherence rates on the scale of $10^4 \text{ s}^{-1}$; this was the first time such agreement was achieved using completely different methods.

The main result in this section was the characterization of electromagnetically induced
transparency resonances in the D1 line of $^{87}$Rb under various experimental conditions. The dependence of the EIT linewidth on the power of the pump field was investigated at various temperatures for the ground states of the lambda-system associated with different hyperfine levels of the atomic $5S_{1/2}$ state, as well as magnetic sublevels of the same hyperfine level. Strictly linear behavior was observed in all cases. A theoretical analysis of our results shows that dephasing in the ground state is the main source of decoherence, in which population exchange plays a minor role.

In the last section of Chapter 3, I briefly mention a quantum communication protocol that enables frequency conversion of quantum optical information. The protocol is based on electromagnetically induced transparency in systems with multiple excited levels. The proof-of-principle experiment was performed using the hyperfine levels of the $^{87}$Rb D1 line.

In Chapter 4, I briefly describe the design and implementation of a source of non-classical light. The optical parametric amplifier (OPA) type source, after several modifications, is capable of producing about 4 dB of squeezing tuned to the D1 line of $^{87}$Rb.

As the first proof of the capabilities of our source, we determined the effect of the electronic noise of the detector in an optical homodyne tomography experiment and showed it to be equivalent to an optical loss if the detector is calibrated by measuring the quadrature noise of the vacuum state. An explicit relation between the electronic noise level and the equivalent optical efficiency is obtained and confirmed in the experiment.

In Chapter 5 I study the propagation of squeezed light under EIT conditions. In the first section we analyze the propagation of continuous-wave (CW) squeezed light through atomic vapor together with a control field forming a lambda configuration.

In the next section I analyze the propagation of pulses of squeezed light under EIT conditions. The behavior of slowdown and the amount of squeezing preserved after different experimental conditions is analyzed.
A theory for the losses in the squeezing due to the interaction with the atomic ensemble is constructed and, for the first time a theory is presented that agrees with the measurements of EIT in the full quantum regime for both the continuous variable regime and the pulsed configuration.

In Chapter 6 I present the chronological development of our quantum memory and the first proof of principle results storing squeezed light pulses. We produced a pulse of squeezed vacuum at 795 nm in an optical parametric amplifier and stored it in a rubidium vapor cell for 1 µs using electromagnetically induced transparency. The recovered pulse is analyzed using time-domain homodyne tomography and exhibits up to 0.28 ± 0.05 dB of squeezing.

In Chapter 7, I present the current state of the project together with an outlook for the future. I present our preliminary results on decoherence of the quantum memory for squeezed light. The storage times for the squeezed vacuum are compared both to the storage time of classical pulses and to the times for strongly attenuated coherent states. I elaborate overall conclusions of the state of the project and provide some perspective into future experiments and possible direction of this project and the field of quantum memory in general.
Chapter 2

Introduction to Electromagnetically Induced Transparency

Before tackling our atom-light interface problem towards building our memory, we review the tools we have to use in order to achieve our goals. According to my perspective, the most important breakthrough that allowed several of the seminal experiments in this area is the electromagnetically induced transparency effect (EIT), reported for the first time in the early 1990’s by Prof. Stephen Harris at Stanford University [13].

In this effect, we take advantage of the properties induced in atomic media by simultaneously applying two optical fields which couple two ground states of an atomic ensemble to a common excited state, a configuration known as lambda (See Fig. 2.1). In this situation, we actually create a coherence between the two ground states and, by doing this, we prevent the atoms from moving to the excited state, thus eliminating absorption in a narrow spectral region. This is a very similar situation to the coherent population trapping (CPT) technique [14].

It is not my intention to review all the theoretical results in this field in detail, but rather to briefly address the main physical effects important for our purposes. For an actual detailed presentation, I recommend the reviews by M. Fleischhauer [8] and T. Halfmann [15].

2.1 Brief description of existent theory

2.1.1 Electromagnetically induced transparency

To understand the physical properties of an EIT medium, we have to consider an ensemble of atoms in the mentioned lambda configuration (See Fig. 2.1). An excited state $|A\rangle$ is
Figure 2.1: Level-scheme of an atom with a \( \Lambda \) configuration of the coupling fields. The excited state \( |a\rangle \) is coupled to two ground states \( |b\rangle, |c\rangle \) by a weak signal field with Rabi frequency \( \Omega_b \) and detuning \( \delta \) and a stronger driving field with Rabi frequency \( \Omega_c \) and detuning \( \Delta \). \( \Gamma_b \) and \( \Gamma_c \) are rates of spontaneous emission into the respective states \( |b\rangle \) and \( |c\rangle \) whereas \( \gamma_{bc} \) describes the decay rate of coherences between these states.

Coupled to a ground (or metastable) state \( |C\rangle \) by a strong driving field (known in the literature as the control field) with Rabi frequency \( \Omega_C \) and detuning \( \Delta \). A weak signal field with Rabi frequency \( \Omega_B \) and detuning \( \delta \) probes the \( |A\rangle \leftrightarrow |B\rangle \) transition (this field is also consistently called probe field).

We can construct the Hamiltonian between a single atom and the fields. It is common to work in the frame of the moving atoms. Under these assumptions, the Hamiltonian takes the form (considering zero detunings):

\[
\hat{H} = -\hbar \left( \Omega_B |A\rangle \langle B| + \Omega_C |A\rangle \langle C| + \frac{\delta}{2} |B\rangle \langle B| + \frac{\Delta}{2} |C\rangle \langle C| + \text{H.c.} \right). \tag{2.1}
\]

An important thing to notice is the fact that the state

\[
|\text{dark}\rangle = \frac{1}{\sqrt{\Omega_B^2 + \Omega_C^2}} \left( \Omega_C |B\rangle - \Omega_B |C\rangle \right) \tag{2.2}
\]

is an eigenstate of the Hamiltonian with eigenvalue 0, when \( \Delta = \delta = 0 \). In other words,
if an atom is driven into this state, no absorption is possible [16]. This can be explained in terms of the interference between two different paths that an atom could take to emit a photon in the A to B transition. For example, one atom in $|C\rangle$ could be driven by the control field to $|A\rangle$ and then decay to produce a photon, or an atom in $|B\rangle$ could be driven by the signal field to $|A\rangle$, decay to $|C\rangle$, interact with the control field to get back to $|A\rangle$ and then finally decay to $|C\rangle$. It is, in principle, possible to prove that the probability amplitudes of these two events interfere destructively.

Knowing the Hamiltonian, we can use the Liouville equation to calculate the pertinent elements of the density matrix of the system in the steady state that will allow us to estimate the physical properties of the media. Also important to notice, decays of the states have to be added. Most commonly, the Lindblad formalism [17] is used to construct a more realistic density matrix. The element of the density matrix that determines the susceptibility of the medium with respect to the signal field has the form (as long as the Rabi frequency of the signal is small compared to the Rabi frequency of the control [14]):

$$\rho_{AB} = \Omega_B \frac{1}{\Delta - \delta_2 + \frac{|\alpha_G|^2}{\Gamma_{BC} + \delta_2} - i\frac{\Gamma}{2}}.$$ 

(2.3)

where $\Gamma = \Gamma_B + \Gamma_C$ is the inverse lifetime of the excited state and $\delta_2 = \Delta - \delta$ is the two-photon detuning.

To describe the measurable properties of the medium, we have to relate our coherence term to its polarizability. This is done by using the Maxwell relation that states this term to be proportional to both the dipole of the media and the coherence term. This allows us to formulate an expression for the complex valued refractive index for light that is close to the resonance:

$$\tilde{n} = \sqrt{1 + \chi} = \sqrt{\rho_{AB}\rho_{AB} + 1} \approx 1 + \frac{\rho_{AB}}{2} \frac{1}{\Delta - \delta_2 + \frac{|\alpha_G|^2}{\Gamma_{BC} + \delta_2} - i\frac{\Gamma}{2}}.$$ 

(2.4)
where, \( \mathcal{g}_{AB} = \frac{|D_{AB}|^2}{\hbar \omega_0} \). Here \( D_{AB} \) is the matrix element of the atomic dipole and \( \omega \) is the atomic density. Since the refractive index is a complex number, we can extract information about the dependence of the dispersion and the absorption of the signal field with respect to its detuning from the main resonance.

In Fig. 2.2 we can see the plots of both components of the refractive index. The first important characteristic to notice is the transparency dip created where otherwise absorption should exist, and also the huge slope in the dispersion curve near the resonance. These two results are the basis of the interface that we will address in the following chapters. The first time such plots were experimentally observed was in a seminal paper by Harris et al. in strontium [13].

2.1.2 Doppler effects in a cloud of atoms

Since we have to deal with experiments, here we have to modify our model to make a more realistic approximation. The previous results were calculated for non-moving atoms. Normally that is not the case, unless you can cool them to submicro-Kelvin temperatures [18].

As stated before, we work with ensembles, meaning that the atoms are moving, at a high velocity and thus, perceive the light with different frequencies, depending on their speed and direction of motion. For the particular case where the driving fields are co-propagating and the energy gap between \( |A\rangle \) and \( |B\rangle \) is much larger than the gap between \( |B\rangle \) and \( |C\rangle \), the Doppler shifts are almost identical. The two-photon detuning \( \delta_2 \) is independent of the individual atom's motion while the driving field detuning \( \Delta \) is not.

In order to consider the influence of the Doppler effect in our predictions, we have to average the susceptibility of the medium over all atoms using a proper velocity distribution,
Figure 2.2: Refractive index close to EIT conditions for an ensemble of resting atoms. Depicted are $\Re \tilde{n}(\omega)$ (the dispersion) and $\Im n(\omega)$ (the absorption) for the cases with and without control field. The Rabi Frequency of the control field is 1 MHz.
\[
\chi_B = \frac{\mathcal{P}_{AB}}{\Omega_B} \int p(\Delta) \rho_{AB} d\Delta. \tag{2.5}
\]

where \(\Delta\) is the detuning from resonance.

The velocity distribution is of Maxwell-Boltzmann form. This explains why the absorption lines in a Rubidium cell look convoluted with a Gaussian when we perform spectroscopy experiments in glass cells (See Appendix. A).

In order to calculate the average integral, Lee and Javan [19] suggested that a change from the Boltzman distribution:

\[
p(\Delta) = p_{\text{gauss}}(\Delta) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{1}{2}(\frac{\Delta}{\sigma})^2}, \tag{2.6}
\]

where \(\sigma = \frac{2\pi}{\lambda} \sqrt{\frac{k_B T}{m_{\text{atom}}}}\), to a more integral-friendly Lorentzian of equal Full Width Half Maximum (FWHM) and same maximum:

\[
p_{\text{lorentz}}(\Delta) = \frac{1}{\sqrt{2\pi}\sigma} \frac{1}{1 + \left(\frac{2\Delta}{W_d}\right)^2}, \tag{2.7}
\]

with \(W_d = 2\sigma \sqrt{2 \log 2}\) being the FWHM of the Doppler broadening, would help with the analysis.

This one is indeed not a thermal distribution, but it can be proven to closely resemble the Gaussian in the region of interest. Using the Lorentzian distribution we can calculate the average integral using Complex Analysis techniques. This gives the new susceptibility:

\[
\chi = \frac{i\gamma_{bc} + \delta_2}{[W_d + \frac{\gamma}{2} + i\delta](\gamma_{bc} - i\delta_2) + (\Omega_C)^2}. \tag{2.8}
\]

From here we can determine the dispersion \(\Re(\chi)\) and the absorption \(\Im(\chi)\) of our
Figure 2.3: Refractive index close to EIT conditions for an ensemble of Doppler broadened atoms. Depicted is $\Im n(\omega)$ (the absorption) for the same physical conditions as in Fig. 2.2 for both cases, with and without Doppler broadening.

Doppler-broadened media in terms of the detunings of the fields.

These two quantities will be of great interest for our purposes in further chapters. Notice that equations 2.3 and 2.8 are very similar, we only need to replace $\Gamma/2$ for $W_d + \Gamma/2$. It can be proven then that the linewidth of the EIT resonance changes from $\frac{\Omega^2}{\Gamma}$ to $\frac{\Omega^2}{W_d}$, meaning that the Doppler broadening actually originates a narrowing of the EIT line. In Fig. 2.3 we can appreciate this effect.

2.2 Slow light

Once we have seen how EIT works, we turn our attention to more specific properties of the EIT medium. The theory I have reviewed in the previous sections is applicable to a monochromatic wave, but more interesting effects will appear if we send pulses of light with a defined bandwidth and a center frequency around resonance. To describe the propagation of these pulses, we have to make use of the group velocity $v_g = \frac{\partial \omega}{\partial k}$. This
together with the relation $k = \frac{n(\omega)\omega}{c}$, where $n(\omega)$ is the refractive index will give us the group index $n_g$ for a pulse centered around an angular frequency $\omega_0$,

$$n_g = \frac{c}{v_g} = n(\omega_0) + \omega_0 \frac{dn}{d\omega}|_{\omega_0}. \quad (2.9)$$

Now, we have to turn our attention to the derivative term in the above equation. As it can be seen, the group velocity of our pulse of light will strongly depend on how drastically the refractive index changes. As depicted in Figure 2.2, around resonance, the slope of the dispersion is enormous, leading to a considerable decrease in the group velocity when the pulse is propagating inside the EIT medium. Much experimental research has been devoted to explore this phenomenon in the years since the discovery of EIT. Of particular interest is that performed by the Hau Group [20] at Harvard, where the group velocity of pulses moving under EIT conditions in a Bose-Einstein condensate was proven to be in the order of $10 \text{ m/s}$, unsurprisingly baptizing this technique as “bicycling faster than light”.

Several years later, there is still research in this topic, particulary on how the bandwidth and shape of the EIT affects the propagation of the slowed-down pulse and how it can be used to process information [21].

2.3 Storage of light

Once we are able to couple a pulse of signal light under EIT conditions and it propagates with a reduced group velocity while in the media, the next question to address is whether, using an elegant technique, we can transfer the information encoded in the light to the atomic cloud.

Controlling the control beam in time allows storage of the information within the atomic sample. The storage procedure can be academically understood as follows. The
control beam prepares the atoms initially in the state $|B\rangle$ through optical pumping. When the signal pulse propagates in the medium, coherences are created between the two ground states of the atoms and acquire the information of the signal pulse during its compression inside the medium. After the compression, most of the signal field energy has been transferred to the control beam and leaves the cell. At this point, the atoms possess the frequency information of the signal within the transparency window, distributed in momentum space. When the control beam is switched back on, the probe beam is regenerated using photons from the coupling beam and leaves the medium while reading the spin state of the atoms.

This explanation would suffice for a panel of experimentalists, but since we have to fulfill the expectations of theoreticians as well, we have to dedicate the next section to showing the formal explanation of this phenomena. Again, it is not our intention to reproduce the entire calculations but rather highlight the main results that justify our experimental effort.

2.3.1 Theory of the Dark State Polaritons

In 2000 M. Fleischhauer and M. D. Lukin introduced the notion of “dark-state polaritons” to describe the interaction of weak light pulses with an EIT medium. In this section, I present its brief overview [22]. As will become clear later, the advantage of this formalism is that it allows us to describe the interaction of the atoms with quantum states of light. In other words, the quantum fluctuations of this field will be now coupled to the fluctuations of the atomic coherences inside the EIT medium. This combined system can be described by constructing a Hamiltonian for these quasi-particles (optical and atomic excitation combined). Such quasi-particles are commonly named Polaritons.

The propagating signal should be described through an electric field operator $E(z,t)$. 
\[ \hat{E}(z, t) = \sum_k \tilde{a}_k(t)e^{ikz}e^{i\nu(z-ct)}. \]  

(2.10)

where the sum is over photonic modes with wave vector \( k \) and bosonic operator \( \tilde{a}(t) \), and \( \nu = \omega_{AB} \) is the carrier frequency of the optical field.

The field coupling \(|A\rangle \) with \(|C\rangle \) will still be described classically with a Rabi frequency \( \Omega_C \).

To describe the properties of the medium, it is common to define atomic operators

\[ \bar{\sigma}^{(j)}_{\alpha,\beta} = |\alpha\rangle_j \langle \beta| e^{i\omega_{\alpha,\beta} z_j} \alpha, \beta \in \{A, B, C\}, \]  

(2.11)

acting on the \( j \)-th atom located at position \( z_j \), with \( \omega_{\alpha,\beta} \) being the laser frequency.

Since we are dealing with collective effects (we will address many atoms at the same time), it makes sense to define collective atomic operators averaged over small volumes,

\[ \bar{\sigma}_{\alpha,\beta}(z) = \frac{1}{N} \sum_{j=1}^{N} \bar{\sigma}^{(j)}_{\alpha,\beta}, \]  

(2.12)

where \( N \) is the number of atoms in the small volume.

Given this situation, the operator \( \hat{P}(z, t) = \sqrt{N}\sigma_{B,A}(z) \) describes the atomic polarization oscillating at an optical frequency, and the operator \( \hat{S}(z, t) = \sqrt{N}\sigma_{B,C}(z) \) corresponds to a low frequency (compared to the optical frequency) atomic coherence (the spin wave we mentioned before). The atomic evolution can then be described by a set of Heisenberg equations: \( i\hbar \partial_t \hat{A}(z, t) = [\hat{A}(z, t), \hat{H}(z, t)] \), where \( \hat{H}(z, t) \) is the atom-field interaction Hamiltonian:
Here, $g = \varphi \sqrt{\frac{\nu}{3\hbar \omega}}$ is the atom-field coupling constant with $\varphi$ being the dipole moment between the $|A\rangle$ to $|B\rangle$ transition, $V$ the quantization volume, $L$ the length in the $z$ direction and $\tilde{A}(z, t) = \{\tilde{P}(z, t), \tilde{S}(z, t)\}$.

From these equations, one can derive the following system of coupled differential equations:

$$\tilde{P}(z, t) = -\frac{i}{\Omega} \partial_t \tilde{S}(z, t),$$

and

$$\tilde{S}(z, t) = -\frac{g\sqrt{N}\tilde{E}(z, t)}{\Omega}.$$

The evolution of the operator corresponding to the quantum field can be described by the propagating equation [22]:

$$\left(\frac{\partial}{\partial t} + c \frac{\partial}{\partial z}\right) \tilde{E}(z, t) = i\sqrt{N}g\tilde{P}(z, t).$$

The solution to the system of coupled equations can be obtained by introducing a new quantum field $\hat{\Psi}(z, t)$ that is a superposition of photonic and atomic components:

$$\hat{\Psi}(z, t) := \cos(\theta)\tilde{E}(z, t) - \sin(\theta)\tilde{S}(z, t)$$
where \( \cos(\theta) = \frac{\Omega_c}{\sqrt{\Omega_c^2 + N|g|^2}} \) and \( \sin(\theta) = \frac{\sqrt{N|g|^2}}{\sqrt{\Omega_c^2 + N|g|^2}} \). By changing the classical control field's intensity \( |\Omega_c|^2 \) the character of the polariton can be changed.

The field \( \hat{\Psi}(z, t) \) (our dark state polariton) now obeys the equation of motion:

\[
(\partial_z + c \cos^2(\theta) \partial z) \hat{\Psi} = 0,
\]

which is very similar to the paraxial wave equation. Its solution is:

\[
\hat{\Psi}(z, t) = \hat{\Psi} \left( z - \int_0^t v_g(\tau) \, d\tau, 0 \right).
\]

This corresponds to a polariton that moves in \( z \) direction with a velocity \( v_g = c \cos^2 \theta \) while its shape is unchanged: \( \hat{\Psi}(z, t) = \hat{\Psi}(z - v_g t, 0) \).

2.3.2 Storage of light

Finally, we have arrived at the tool that we need to create our interface. As we have seen, the character of the polariton (light or atomic) depends on the control field, as does the propagation velocity. Consequently, by slowly varying the control field intensity \( \Omega_c \) we can control the speed of propagation through the medium, and, more importantly, the properties of the light field part of the polariton are slowly transferred to the atomic part.

One last comment should be made here. This transfer is only valid because a dark state polariton is an eigenstate of the interaction Hamiltonian with zero eigenvalue. In this case, if the system of atoms and light described in the previous section is in a dark state and we change the control parameter \( \Omega_c \) slowly, the system will remain in a dark state.

This enables us to implement a memory for pulses of light following the next steps.
Storage of light

(see Fig. 2.4):

• First the control field $\Omega_C$ is turned on, then a pulse of signal light with a bandwidth close to that of the EIT transparency window enters the medium. The medium adiabatically follows into a dark state corresponding to the input quantum state of the light pulse. This dark state polariton travels through the medium with a velocity given by $\Omega_C$ (a).

• While the pulse is within the medium, we adiabatically reduce the control field strength to zero and so the polariton comes to a complete stop. Here all the information of the system remains as a collective atomic excitation (b).

• After some delay the control field is switched on again and the polariton finally emerges as an optical pulse and leaves the medium thanks to the EIT conditions created by the control field (c).
2.3.3 Experimental EIT storage to date

The theory we reviewed briefly in the last section has served as a basis for several experimental breakthroughs in the last ten years. Together with the experiments of the Hau group, the group of Lukin at Harvard University were the first to demonstrate the storage of light by sending pulses of light resonant to $^{87}$Rb atoms [9, 23]. For these particular experiments, the two ground states chosen were Zeeman degenerate from the $F = 2$ to $F' = 1$ transition of the D1 line. After this, several groups reproduced these results [24] and several extensions also followed, some of them using the hyperfine splitting of Rubidium as ground states [25, 26]. The importance of this last result will become evident in the next chapters.

In the last years, the effort has turned to improving several points in the storage procedure: Gorshkov et al. have studied the transfer efficiency of information to the atomic media by changing the shape of the retrieval pulses [27], Javan et al. [19] and Novikova et al. [28] have studied the decoherence processes involved in the atomic phase of the storage, together with a study about the quality of the cells and buffer gas amount and its relation to the storage times [29, 30].

At the same time, several groups also explored the potential of EIT-based memory using different physical systems. Turukhin et al. [31] at MIT demonstrated the possibility of using the storage of light in ion doped crystals and the work of Longdell et al. [32] expanded the possibilities even more by demonstrating enormous storage times in these systems. Another interesting experiment has been the demonstration of EIT in photonic band gap fibers by Ghosh et al. [33] and Rubidium filled hollow-core fibers by Light et al. [34].

Another major breakthrough has been the implementation of EIT and storage using cold atoms [23]. Nowadays, a number of groups around the world have the capability to perform EIT experiments using cold atoms trapped in magneto-optical traps. The new
properties this last technique provides increased the chances of actually extrapolating our room temperature EIT techniques to a region more useful for processing of quantum information.

2.4 EIT with non-classical light

2.4.1 Experimental work: towards quantum repeaters

As a result of all the experimental success using EIT to store classical information, the next question to ask is whether it is possible to repeat these experiments in the quantum regime.

A major motivation in this field has been the promise of the implementation of quantum communication, for instance, the transmission of individual photons over long distances while preserving their quantum state. A proposed solution to this challenge is the implementation of quantum repeaters [35] which enable polynomial, rather than exponential, decrease in the bit transfer rate as distance increases. The implementation of quantum repeaters requires quantum memory for individual photons. From here on, we refer to quantum memory as the storage of the quantum state of an electromagnetic mode in an atomic coherence with a successful preservation of this state in the retrieved light.

This realization would involve a merge of two of the fields in physics which have had the greatest development over the last decade: quantum optics and atomic physics.

Since the quantum optics community has made advances in creating and manipulating states of light that can only be described via quantum mechanics, the question to address has become whether potential exists to mix these results with our atom-light techniques, such as EIT-based storage. The first experiment where EIT was observed in the quantum regime was achieved by the Kozuma group in 2004 [10] using squeezed light as the signal
field. Since then, a number of groups have researched the implementation of EIT using different quantum states of light.

The relevant question to answer here is how to implement sources of light that have quantum properties in the electric field, while, at the same time, being resonant to atomic transitions. Moreover, they have to fulfill a more stringent requirement, that is, their bandwidth has to be small in order to interact with atoms in the EIT regime.

There are two main solutions to this problem. The first, designed by Duan, Lukin, Cirac and Zoller [36], is the use of atoms to produce single photons. The last two requirements are automatically fulfilled because the light is produced within the bandwidth of the atoms. To prove that the produced light has quantum properties is somewhat more challenging. The second requires a more standard technique using spontaneous parametric down conversion and will be studied in a subsequent chapter.

2.4.2 Duan-Lukin-Cirac-Zoller (DLCZ) protocol

In this section I will briefly explain the idea of single photon generation using EIT. As usual, we consider an ensemble of three level atoms. We first pump all the atoms to $|B\rangle$. Later we send an off-resonant pulse (known as the writing pulse) resonant to the $|B\rangle$ to $|A\rangle$ transition. This results in the spontaneous Raman scattering of so-called Stokes photons with a frequency close to the $|C\rangle$ to $|A\rangle$ resonance [36]. If we condition on detecting a single Stokes photon using single photon detectors (SPDC) we ensure that only one atom in the ensemble has made the transition. Since it is unclear which of the atoms was addressed, we project the atomic ensemble into a collective atomic coherence (or spin wave) with a single excitation, where all atoms have an equal probability of being in state $|C\rangle$.

After the preparation, we can retrieve the atomic coherence onto a single photon using EIT. A retrieve pulse is applied in resonance to the $|C\rangle$ to $|A\rangle$ transition, converting the
atomic coherence into an anti-Stokes photon resonant to the $|B\rangle$ to $|A\rangle$ transition. The retrieve laser performs the function of the control field, therefore achieving EIT for the emerging single photon. Fig. 2.5 shows a schematic of the procedure.

The immediate advantage of this technique is control of the bandwidth of the anti-Stokes photon via the retrieve laser. The timing of emission can, in principle, be controlled with the moment we apply the retrieve laser.

Once we prove the quantum nature of the anti-Stokes photons, we can consider building a quantum memory for these photons by putting another atomic ensemble in the path of these photons and using our EIT methods to retrieve the created atomic coherence.

Storage of single photons through the use of this protocol was achieved independently by Eisaman et al. and Chaneliere et al. [11, 12]. These two experiments were the first demonstrations of storage of non-classical light using EIT. Nevertheless, both techniques relied on measuring the statistics between the Stokes and anti-Stokes photons to prove the quantum nature of the retrieve state. Therefore, full reconstruction of this state was not performed.

Using this technique, we can think about generating the cornerstone of quantum communications: entanglement of atomic ensembles. According to the DLCZ proposal one can illuminate both ensembles by synchronized classical writing pulses (see Fig. 2.6),
and the generated Stokes photons can interfere at a 50/50 beam splitter, with the outputs detected by single photon detectors. In this configuration a single click detector implies that one spin wave is created in one of the two ensembles, but we cannot distinguish which of the two ensembles emitted the photon which, therefore creates entanglement.

This technique to manipulate atomic ensembles has applications for long distance quantum communication. In order to accomplish this, we rely on the entanglement swapping process [36]. The entanglement swapping is achieved by interfering two photons coming from different entangled pairs, transferring the entanglement to the pair of photons that were not interfered. In this probabilistic scheme, the quantum mem-
ory is essential for polynomial scaling of the required time with distance, compared to exponential scaling for the case of direct entanglement generation.

2.4.3 Duan Lukin Cirac Zoeller protocol: Experiments up to date

Of all the attempts to implement quantum information, quantum repeaters and quantum networking technology is one of the areas that has seen the most success. Much of this progress has been achieved over the last three years, and further developments are continually occurring.

Apart from the aforementioned experiments by Lukin and Kuzmich, the Kimble group in Caltech was the first to publish results on measurement induced entanglement of two different atomic ensembles [37]. The Schmiedmayer group followed with a similar experiment where the phase of the optical path for both Stokes photons was locked to achieve maximum entanglement [38]. Work on the decoherence of this atomic entanglement has recently been published [39, 40, 41]. Even more, pioneer work on entanglement distribution has been reported already [42].

Progress has been made in this direction of research, fueled mainly by the promise of long distance quantum communication. In the following chapters I will describe an alternative technique to generate light with quantum properties and extend the presented results in this section by combining several quantum optics techniques to overcome the problem of characterizing the quantum state of the signal field after the storage procedure.
In the previous section, we provided a brief introductory description of EIT theory along with its importance in the quantum optics community. We now turn our attention to the experimental results involving EIT systems produced in our laboratory. For the purposes of this thesis, I follow a historical approach to give insight on how and why the experiments were developed leading to the final goal of realizable quantum memory.

For brevity, most of the technical details are omitted in the following chapters. For a complete description of some the elements commented here, the reader is referred to the Appendices and the thesis of my colleague Juergen Appel [17]. Therefore, the following chapters will be directly focused on the experimental results obtained. Specifically in this chapter, we will discuss our early experiments in generating EIT, characterization of atomic resonances, and finally a little glimpse of the coherent control that can be achieved in these systems. All of the aforementioned work has already been published [1, 2, 3].

3.1 How to create EIT resonances in Rubidium

The first key issue we must consider when communicating with atoms is the light sources required, not every coherent source will suit our purposes. There are technical restrictions that exist such as wavelength stability and bandwidth that must be overcome in order to correctly address atomic transitions.

Our first project was the construction of laser sources needed for the pertinent transitions. Rubidium is fortunate in this respect since atomic wavelengths of common use are achievable using laser diodes widely employed in industry. For example, wavelengths
of 780 nm and 795 nm can be obtained from laser diodes commonly used as readers in CD drives.

The requisite of a narrow bandwidth can be solved through use of these diodes in an external cavity arrangement, e.g. in a Littrow configuration. Our group has been successful in building several of these diode lasers, the details of which are presented in Appendix A.

Another (more expensive) method of accessing atomic resonances is using Titanium-Sapphire tunable lasers. These systems available commercially are characterized by low bandwidths on the order of tens of kHz. Thus they are well suited for our purposes. With sources tuned to the right wavelength, the experimental setup to see EIT is not too complicated.

Furthermore, to see EIT we need to provide an atomic ensemble. As mentioned, we can use atomic vapor or cold atoms with similar results. Throughout this thesis, we will focus on the first case. We employ isotopically pure Rubidium 87 due to its well known and clean transitions that are readily combined into lambda and double lambda configurations. To achieve atomic densities where EIT can be seen, we use rubidium cells heated to near 65 °C.

A crucial factor in designing the apparatus is shielding of the cell from the Earth’s magnetic field. This external field splits the the degeneracy of the atomic transitions and prevents EIT from happening. Details relevant to the Rubidium cells, the magnetic shielding necessary and the oven design are located in Appendix C.

The cells used in the experiments were produced by Triad Technologies Inc. and feature anti-reflection coatings in the windows surfaces. For reason that will become evident during our review of the experiments, they are also filled with neon as buffer gas.

We now have the tool necessary to generate Λ-type systems composed of two ground levels |B⟩, |C⟩ and one excited state |A⟩. In Rubidium, there exist two such configurations
In one configuration we need only to address a single hyperfine transition, and the ground states will consist of different magnetic sub-levels of the lower hyperfine level. In this configuration only one laser source is needed, since linearly polarized light in combination with a quarter wave plate before the cell is sufficient to obtain both circular polarizations.

Each polarization separately corresponds to our signal and control fields, with the ratio of their Rabi frequencies given by the original linear polarization. Separation of both polarizations can be achieved later via a polarizing beam splitter and a second quarter wave plate right after the cell. We call this setting "Zeeman".

The "Zeeman configuration" that will be used for the reminder of the chapter is formed by control and signal fields of wavelength $\lambda = 795$ nm that couple pairs of Zeeman sublevels of the atomic ground state ($5S_{1/2}, F = 2$) via the excited state ($5P_{1/2}, F' = 1$) (See Fig. 3.1). This is the configuration used in the first experimental demonstration of light storage.

The concern however, is that to separate control and signal fields after the cell, we can...
only rely upon the polarization difference. This poses a limitation for EIT experiments with quantum light since the control field is typically $10^{12}$ times stronger than the signal, but only a level of extinction $10^6$ is attainable with normal PBS's.

In the second case, we use two separate hyperfine transitions, demanding two different laser sources with a locked phase difference to maintain atomic coherence. This strict requirement is essential for this configuration. Using the signal and control fields fixed at different frequencies and the implementation of filtering techniques (e.g. cavities or etalons), we can reach the remaining $10^6$ in the extinction factor, making it a system suitable for quantum memory experiments. We call this configuration “Hyperfine”.

In our hyperfine experiments, the signal field was obtained via an additional diode laser that was phase locked at 6.8 GHz, from the Ti:Sapphire laser to ensure two-photon resonance with the $(|B\rangle = |5S_{1/2}, F = 1\rangle, |A\rangle = |5P_{1/2}, F = 2\rangle, |C\rangle = |5S_{1/2}, F = 2\rangle)$ transitions (See Fig. 3.2).
3.2 Characterization of the atomic coherence decay for the storage of light

The main goal of the project is to demonstrate quantum memory. Thinking along the lines of building the seminal blocks of quantum repeaters and long distance quantum communication, we began by analyzing the storage process from a classical perspective. We were also interested in the tools that could in principle be used to have control of the EIT lines and the storage procedure itself.

As we know, light storage relies on the transfer of the quantum state of light onto a coherent superposition of atomic ground levels. Understanding the limitations of the memory lifetime requires figuring out the processes that modify this superposition during the storage time.

The atoms that are created in the superposition will experience movement and collisions (they are in a room temperature cell) with other atoms. These effects will make the atoms lose the created coherence at a rate given by the physical process involved, we call these rates decoherence. In principle we can reduce these collisions by adding a buffer gas, but realistically this coherence can not be preserved forever. In this part of our experiments, we measured these decoherence effects and developed explanations regarding the physics involved.

Here, we used three different measurement techniques, each dedicated to gaining more insight into the physical processes observed. The first experiment involved storage of light where we related the decoherence time to the decay rate of the restored pulse energy against the storage time. For the second experiment we created a coherent superposition of the atomic ground levels by applying two-photon resonant Raman fields, and then non-adiabatically moved one of the fields out of resonance. This causes the ground state coherence to oscillate between dark and bright states in and out of phase with respect to
the light fields. If we then monitor the intensity of this beam after propagation through rubidium, we can see an oscillatory absorption signal (Hanle oscillations). We related the decay time for these oscillations to the power of the control field. When the control field is zero we have a limiting case that we can relate to the decoherence rate. Lastly, we measured the dependence of the EIT resonance width against the power of the control field and related the limiting case of zero pump field to the decoherence process. The results of the three experimental procedures proved to be very similar. Fortunately for us, these measurements had important implications in justifying our later experiments.

For the rest of this chapter, we will make use of the nomenclature developed in the previous chapter. For clarity I repeat some definitions: our levels are coupled by a pair of electromagnetic fields: a probe field with Rabi frequency $\Omega_b$ and detuning $\delta$ and a stronger driving field with Rabi frequency $\Omega_c$ and detuning $\Delta$. $\Gamma_b$ and $\Gamma_c$ are rates of spontaneous emission into the respective states $|B\rangle$ and $|C\rangle$ whereas $\gamma_{bc}$ describes the coherence decay rate between these states, the quantity of interest in this chapter. Fig. 2.1 shows the atomic structure with the relevant decay channels.

3.2.1 Storage of light

In our first experiment, we performed storage of light and measured the energy of the restored pulses as a function of the storage time. Our first approach in characterizing the decoherence processes was to relate the decoherence time of the ground state to the decay rate of the restored pulse’s energy. This crude measurement does not give us insight into the physical processes involved but sets an upper limit for the decoherence rate.

As mentioned in the previous chapter, the transfer of light states to stationary atomic excitations can be explained in terms of so-called dark-state polaritons [22], which are particle-like entities into which light pulses convert when propagating through an EIT medium. The dark-state polariton consists of both atomic and optical components. The
optical component, as well as the propagation velocity, vanishes when the EIT pump field is adiabatically turned off. The quantum state carried by the original light pulse is transferred into a coherent superposition of the atomic ground states (the atomic part of the polariton).

When the pump field is turned back on, the polariton resumes its propagation and the light pulse leaves the atomic sample in its original quantum state. Assuming that dark-state polaritons decohere at a rate $\gamma_{bc}$, the intensity of the restored pulse (which is proportional to the square of the polariton amplitude) decays at a rate $e^{-2\gamma_{bc} \tau}$, where $\tau$ is the storage time. This behavior has been observed by several groups [25, 28, 43].

**Experimental set up**

For the experiments related to this project, we used atomic $^{87}$Rubidium vapor at temperatures of 60–100 °C in a 5 cm long cell with neon as a buffer gas at a pressure of 1 Torr. The cell was located inside a temperature controlled oven, surrounded by three coaxial cylinders of $\mu$-metal that attenuate external fields to below 1 $\mu$G. (See Appendix C)

For the Zeeman experiments, the pump and probe fields were composed of orthogonal circular polarizations derived from a linearly polarized laser beam. The light fields coupled pairs of Zeeman sublevels of ground state $5^2S_{1/2}, F = 2$ Rb atoms via the excited state $5^2P_{1/2}, F = 2$ (Fig. 3.1). The light source for both fields was a Coherent MBR-110 Ti:Saphire laser with narrow spectral width ($\sim$ 40kHz) and high long-term frequency stability.

At first the beam was split into pump and probe beams by a polarizing beam splitter (PBS). Each component was then sent to an acousto-optical modulator (AOM) in a double-pass configuration for independent control of their frequencies. Each AOM was driven via a 80 MHz signal. More information on the AOM driving system can be found in Appendix B.
Figure 3.3: Schematic of the experimental setup for the measurement of decoherence rates. The same setup is slightly modified to perform experiments in both the Zeeman and Hyperfine configuration. For the Zeeman measurements only the Ti:Sapphire laser is used and quarter wave plates are placed in front and after the interaction zone. For the hyperfine experiment, an additional diode laser is added and the quarter plates are removed.

After the EIT interaction, the beams were recombined in a PBS to form an interferometer (see Fig. 3.3). A $\lambda/4$ plate (QWP) converted the linear polarization of the beams to circular before they entered the rubidium vapor sample. The measured diameter of the beams just before the cell was $\sim 10$ mm for the pump and $\sim 2$mm for the probe. Upon exiting the cell, the linear polarization was recovered with a $\lambda/4$ plate and the probe field was detected. A scheme of the experiment is shown in Fig. 3.3.

For the hyperfine experiments we slightly modified the Zeeman setup (see Fig. 3.3,
dotted part). The Ti:Sapphire laser, resonant with the $5^2S_{1/2}, F = 2$ to $5^2P_{1/2}, F = 2$ hyperfine transition of the Rb D1 line, was only used as the probe. The control field was obtained from a diode laser resonant to the $5^2S_{1/2}, F = 1$ to $5^2P_{1/2}, F = 2$ transition (Fig 3.2). The diode laser was phase locked at 6.8 GHz to the Ti:Sapphire to ensure a two-photon resonance with the rubidium ground state hyperfine splitting. The phase lock was implemented by observing interference of the two lasers via a 12 GHz photodiode and providing feedback to the diode laser’s injection current. More information on the phase lock design can be found in the thesis of my colleague Juergen Appel [17].

The polarization of both beams was linear for the light-atom interaction. The probe field was again detected after filtering out the pump with a Glan-Thompson prism. The diameters of the beams were similar to those used for the Zeeman experiments.

Using these configurations, we can use the technique of Philipps and co-workers [9] to store pulses of light by timing the production of signal pulses to the switching of the control field. After retrieving the stored light pulses and measuring their energy as a function of the storage time, we observed exponential behavior with decay rates $2\gamma_{bc}$ of $\sim$8000 s$^{-1}$ (1.3 kHz) for the Zeeman configuration (Fig. 3.4) and $\sim$12000 s$^{-1}$ (1.9 kHz) for the hyperfine configuration (Fig. 3.5).

These results motivated us to design a new experimental setup to measure the decoherence of the ground state for comparison to the storage results. By obtaining similar results we could conclude that this decoherence is the main responsible for information lost during the storage procedure. Different results would imply that other effects not considered play a major role in the storage of light.

3.2.2 Transient coherence oscillations

To design an experiment specifically for measuring the decoherence of the ground state, we decided to change the conditions of this coherence and monitor its evolution. We created
Figure 3.4: Exponential fit to the decrease of the retrieved pulse energy as a function of the storage time for the Zeeman configuration (decoherence rate fitted: 8000 s\(^{-1}\)).

Figure 3.5: Hyperfine configuration (decoherence rate fitted: 12000 s\(^{-1}\)). The inset in (b) shows the behavior of the restored pulses against the storage time for the hyperfine experiment. The leftmost is the original pulse. Experiments were performed at temperatures \(\sim 90 \, ^\circ\text{C}\).
a coherent superposition of the atomic ground levels by applying two-photon resonant Raman fields, and then non-adiabatically detuned the frequency of one of the fields out of resonance by $\Delta_R$. Technically speaking, this was achieved by abruptly changing the driving frequency of one of the AOM's.

The ground state coherence then starts to oscillate between dark and bright states, and we can observe modulation of the probe field absorption. This process has been theoretically investigated by Park et al. [45] and this modulation can be proven to be of the form:

$$\alpha = A e^{-\gamma t} + B \cos(\Delta_R t + \phi)e^{-\frac{\gamma_{TC}}{\gamma_{TC}} t},$$

(3.1)

where $\alpha$ is the absorption index, the first term in the sum accounts for spontaneous decay at a rate $\gamma$ into the atomic levels outside the closed $\Lambda$-scheme and the second term represents the transient coherence oscillations, which decay at a rate

$$\gamma_{TC} = \gamma_c + \frac{\Omega^2}{\gamma},$$

(3.2)

where $\Omega_c = \Omega_b = \Omega$.

Extrapolation to the case $\Omega \to 0$ thus gives a value for $\gamma_c$. Similar oscillations were measured by Mair et al. [46] as well as in Valente et al. [47], but in that case Raman detunings were created by applying an external magnetic field.

**Experiments**

For this experiment we first created equal-amplitude atomic coherence between the ground states by using pump and probe fields of equal intensity. This is a slightly different condition compared to the EIT scheme, but since we are only interested in measuring the decay rate of the coherence it can as well be used.
One of the fields was rapidly shifted in frequency by $\Delta R$. This detuning had to exceed the width of the EIT window, but still be within the oscilloscope bandwidth such that the oscillations could be registered. In the Zeeman configuration, we implemented a driving frequency change of $\Delta R \sim 30$ kHz with one of the acousto-optical modulators.

In the hyperfine setup, we rapidly changed the reference frequency of the phase lock resulting in a shift of the frequency difference between the two lasers of $\Delta R \sim 2$ MHz. This initiated the oscillation of the ground state coherence with respect to the laser field, which in turn caused periodic modulation of the absorption (see Figs. 3.6 and 3.7, insets). The transmitted probe intensity was recorded and fit with Eq. (3.1), from which the decay parameter $R_{TC}$ was obtained.

According to Eq. (3.2), the decay rates depend quadratically on the Rabi frequency of the pump field and therefore linearly upon its power. In both experimental configurations, such linear dependence was indeed observed, except for very low pump powers in the Zeeman configuration, where the measured decay rates did not change. From our fits, we estimated $\gamma_{bc} \sim 6000 \pm 1000$ s$^{-1}$ in the Zeeman configuration and $\gamma_{bc} \sim 12000 \pm 1000$ s$^{-1}$ in the hyperfine.

We believe this to be the first experiment where Hanle-like oscillations of the ground state were used to determine the decoherence rate. The observed linear dependence of the decay rates on the pump power (see Figs. 3.6 and 3.7) confirmed the theoretical predictions made by Park et al. [45]. Note also that this experiment worked for a variety of detunings ($\Delta R \sim 30$ kHz – 2 MHz).

As can readily be seen, the decay times from these experiments are very close to our first measurements of light storage time. This provided strong evidence supporting our starting hypothesis. Thus further motivating us to design another experimental progression such that we could extract more information on the physics behind this decoherence.
Figure 3.6: Transient coherence oscillations of the ground state, behavior of the decay rate against pump power for the Zeeman experiment @ 90 °C. Inlet: Transmitted probe intensity vs. time showing the modulation of absorption @ 30 KHz.

Figure 3.7: Transient coherence oscillations of the ground state, behavior of the decay rate against pump power for the hyperfine experiment @ 85 °C. Inlet: Transmitted probe intensity vs. time showing the modulation of absorption @ 2 MHz.
3.2.3 Width of the EIT resonances

Having proved the decoherence of the ground state to be our main contributor to information lost in storage experiments, we next turned our attention into gaining more insight about the actual processes responsible for it.

Decoherence in EIT is a consequence of several mechanisms, such as flight-through broadening, population exchange, atom-atom and atom-wall collisions, etc., but it was still not clear which if any was the most significant.

It turned out that the Scully group at Texas A & M has already explored this problem and proposed to answer it by measuring the width of the EIT resonance as a function of the control field intensity. By assuming different physical mechanisms, the predicted results for such an experiment will change. In their theoretical treatment [19, 44], the authors assume the population exchange between the ground levels $|B\rangle$ and $|C\rangle$ to be the main source of decoherence.

With population exchange (or population shuffling) we understand the following effect: atoms getting into the EIT interaction zone being in a random ground state which affects the atomic distribution (diagonal elements of the density matrix) and hence the coherence. It can also be viewed as the created spin wave losing coherence due to random interactions with new atoms. Physically, it can happen if the pumping preparation is not optimum or if we have inelastic collisions between the rubidium atoms. To include these effects in the theory presented in chapter 2, we have to introduce decay in the diagonal terms in the density matrix which also has an effect on the coherence terms.

The key consequence of this assumption is the prediction of a non-linear dependence of the width of the EIT resonance against weak control field powers. This theory was widely accepted in the EIT community as the main standard for decoherence.

Interestingly, most of the experiments performed in atomic vapors [48, 49, 50, 51] showed this dependence to be linear, with an exception published by Ye et al. citeYe,
using non-conventional conditions, without buffer gas and relatively small beam diameters.

With these motivational reasons, we measured the width of the EIT resonance on the D1 transition in rubidium vapor in a variety of settings. In agreement with previous measurements, our results showed a consistent linear behavior with the y-axis intercepts (as in the other experiments, the extrapolation to zero control field) on the order of a few kHz.

From these results, we proposed an alternative theory to the one of Ref. [19] based on a different mechanism to explain the linear effect. We assumed pure dephasing (i.e. only decay of the off-diagonal density matrix elements) as the dominant decoherence mechanism.

One could in principle imagine our atoms prepared in a coherence moving away (physically) from the interaction zone while simultaneously interacting with other atoms, but not losing their coherence, rather only changing the phase of it. This effect arises from elastic collisions or atoms moving in and out of the interaction region.
This treatment of decoherence does predict linear dependencies and hence, yields much better fits to our data. From the fits, we also obtained the ground state decoherence rates on the scale of a few kHz, which are consistent with our other measurements.

Experiments

The experiments were performed in atomic $^{87}$Rb vapor at temperatures of 60–100 °C, which correspond to the optically thick regime. Relative frequencies of both fields were precisely controlled by acousto-optical modulators. The measured diameter of the control beam just before the cell was $\sim 10$ mm. We used linear polarizations for the pump and signal.

To measure the full width half maximum (FWHM) of the EIT resonance, we swept the signal field frequency to obtain a full shot of the EIT resonance. A typical scan is depicted in the inset of Fig. 3.9 and approximates a Lorentzian distribution. The pump power was varied from 100 $\mu W$ to 1.2 mW ($\Omega_c \sim 1-3$ MHz) while the signal power was kept constant at about 20 $\mu W$ ($\Omega_b \sim 500$ kHz).

Fig. 3.9 shows the results of the Zeeman measurement. The behavior is linear, temperature independent, and shows a $y$-intercept of 3 kHz. Similar measurements were repeated in a 10 Torr Ne buffer gas cell (not depicted), showing a similar behavior and a slightly different $y$-intercept.

Experimental results for the hyperfine configurations are presented in Fig. 3.10. Similarly, a linear behavior was observed but the slope and the $y$-intercept do depend on temperature. Additional measurements were done with a 0.1 Torr cell (not depicted), showing a comparable trend.

Figures 3.9 and 3.10 also display the best fit obtained using the theory of Javan et al. [19, 44]. These are not in agreement with the experimentally observed data and yield unrealistically low values for the decoherence rates: 110 Hz and 117 Hz, respectively. We
Figure 3.9: Measured width of the Zeeman configuration EIT resonances as a function of the control laser power for different temperatures together with linear fits and a fit to the theory of Javan et al.\cite{19, 44}, assuming a decoherence rate of $\gamma_{bc} = 110$ Hz. The inset shows an example of the measured EIT resonance.
concluded that the population exchange cannot be the dominant decoherence mechanism in our cells. If this were the case, we should have been able to observe narrower EIT lines for low control field powers.

Since our results were in clear disagreement with the existent theory, we proceed to re-formulate the theory by considering a different mechanism for the decoherence of the ground state. Specifically, we proposed the ground state decoherence to be dominated by dephasing, i.e. decay of the off-diagonal density matrix element $\rho_{bc}$ at a rate $\gamma_{bc}$.

Recalling the notation we defined at the beginning of the chapter, we can work with the density matrix element that defines the susceptibility of the media. The responsible matrix element (see chapter 2 Eq. 2.3) should be integrated over all atomic velocities in order to average the susceptibility of the medium over the different atom velocities.
in a thermal cloud (see Eq. 2.5-2.7). Performing the integration (over the two photon detuning), we find the average susceptibility for the $\Omega_b$ field (Eq. 2.8).

The EIT linewidth is orders of magnitude smaller than the Doppler width $2W_d$, hence it is possible to neglect several terms in the denominator.

The absorption coefficient $\alpha$ can then be expressed as:

$$\alpha(\delta_2) = \frac{\omega}{c} \text{Im}(\chi_b) = \alpha_{\text{max}} - \frac{\alpha_{\text{max}} - \alpha_{\text{min}}}{1 + \left(\frac{\delta_2}{\text{FWHM}}\right)^2}$$  \hspace{1cm} (3.3)

with

$$\alpha_{\text{max}} = 2\frac{\omega \rho}{c} \sqrt{\pi} \ln 2 \frac{1}{2W_d + \Gamma}; \hspace{1cm} (3.4)$$
$$\alpha_{\text{min}} = 2\frac{\omega \rho}{c} \sqrt{\pi} \ln 2 \frac{1}{2W_d + \Gamma + 2\frac{|\Omega_c|^2}{\gamma_b}}; \hspace{1cm} (3.5)$$
$$\text{FWHM} = 2\gamma_b + \frac{4|\Omega_c|^2}{2W_d + \Gamma}. \hspace{1cm} (3.6)$$

The important result of this theory is that now, since $|\Omega_c|^2$ is proportional to the beam intensity, the EIT linewidth scales linearly with control power and intersects the $y$-axis at a minimum of $2\gamma_b$. Clearly, this results provides a more accurate explanation of the experimental values.

Evident from Figs. 3.9 and 3.10, the linear dependence of Eq. 3.6 provides an excellent fit to our experimental data. The $y$-axis intercept is twice the ground state decoherence rate, which we measured by two different methods. In our setup, storage times of 100-250 $\mu$s were observed, and are in reasonable agreement with the measured intercepts.

At this point, we could gain an insight about the physics involved in our measure-
ments. Decoherence in atomic vapor cells is known to be dominated by atoms moving away from the interaction zone (flight-through mechanism). The atoms arrive into the interaction area in an arbitrary ground state, so we would expect a signature of population exchange decoherence, surprisingly none is present.

In order to explain this, we attributed the lack of population exchange decoherence to be an effect of the geometry of the laser beams, which in our case was close to Gaussian. Before entering the interaction area, an atom initially in a random ground state, propagates through the "wings" of the Gaussian profile.

In this region, the signal field is negligible because its diameter is smaller, but the pump field is already sufficiently strong to pump the atom out of $|C\rangle$. When entering the central part of the interaction region, most of the atoms will be in state $|B\rangle$, albeit with a random phase.

This idea gave us another insight into the dephasing process. Due to the fact that we followed a first order approximation in $\Omega_b$ to obtain Eq. 3.6, assuming population decay only from $|C\rangle$ to $|B\rangle$ in our density matrix formalism will result in exactly the same result. We can argue then, that the physical reason for dephasing is this particular decay.

The temperature dependence of the slope in the hyperfine configuration can be explained by absorption of the control laser upon entering the saturated rubidium vapor. We quantified this effect by integrating the susceptibility equation over the length of the cell, assuming that the pump absorption is governed by Beer's law. The temperature dependence of the slope derived by this procedure (Fig. 3.10, second inset) was compared to the measured slopes. The best fit was obtained for the 12 mm beam diameter, which agrees well with our measured diameter of 10 mm. In the Zeeman configuration, the pump absorption was negligible at all temperatures, which explains a constant slope.

The central message here is the experimentally observed behavior of the EIT linewidth
as a function of the control power, exhibited consistently linear behavior in different A
configurations and temperatures. Furthermore, this was in agreement with the explanation assuming that the main ground state decoherence mechanism is dephasing.

3.2.4 Comparison of the methods

The experimental results for the three methods exhibited order-of-magnitude agreement
with each other and with related results already published [9, 25, 28]. Some discrepancy among the results of the three experiments can be attributed to simplifications in
theoretical models used to obtain Eqs. (3.1) and (3.6).

The observed discrepancy is not surprising for the measurements of the EIT linewidth
in the hyperfine case, where the pump absorption prevented us from obtaining a correct intercept of the linewidth curve. In the Zeeman case, the observed intercept may not be
a true measure of $\gamma_{bc}$ either, for the following reason:

The population exchange theory [19],[44] predicts that at high pump intensity, the
dependence of the EIT linewidth on the pump power approaches the linear expression,

$$\text{FWHM} \rightarrow 4 \gamma_{pe} \frac{W_d}{\Gamma} + \frac{2|\Omega_0|^2}{W_d},$$

(3.7)

with $\gamma_{pe}$ being the population exchange rate. This asymptotic dependence has a slope
similar to that in Eq. 3.6, but its intercept for $\gamma_{pe} \sim \gamma_{bc}$, is higher by a factor of
$\sim 180$. Therefore, although our experimental result provides a clear evidence in favor
of dephasing as the dominant mechanism of the ground state decoherence, even a tiny
amount of population exchange may have a significant effect on the observed $y$-intercept,
without influencing the general linear shape of the experimental curve. By comparing
the observed intercept with the values of $\gamma_{bc}$ obtained in our two previous measurements
(where minor population exchange cannot be expected to have a major effect on the
decay rate), we can estimate $\gamma_{pe}$ to be on the scale of 50–100 Hz.

Similarly, the theory of the transient coherence decay [45] has been derived for a
closed three-level system, assuming dephasing to be the main ground state decoherence
mechanism. The role of population exchange and additional decay channels in the be-
havior of the decay constant as a function of the pump power was a subject of further
experimental investigation (See subsection 3.2.6).

3.2.5 Overall Conclusions

We have presented experimental measurements of the decoherence of the ground level
($5S_{1/2}$) rubidium atoms in a vapor cell with a buffer gas. We determined the decay
rates of coherences between the Zeeman states of the same hyperfine sublevel, as well as
between different hyperfine sublevels. The measurements were made using three different
techniques. All methods showed decoherence rates of the same order of magnitude $\sim 10^4$
s$^{-1}$; discrepancies can be attributed to various simplifications in the theoretical treatment.

We proved that creating Hanle-like oscillations in a $\Lambda$ configuration and then mea-
suring their behavior against pump power give us a good measurement of decoherence.
By showing a linear dependence of the decay rates against pump power we verified the
theoretical predictions of Park et al. [45].

We also have experimentally observed the behavior of the width of an EIT resonance
as a function of the pump power in different $\Lambda$ configurations and temperatures. Consis-
tently linear behavior was observed and explained theoretically with an assumption that
the main ground state decoherence mechanism is dephasing.

We believe that the procedures shown in this chapter can be applied to estimate
the decoherence in a medium with a $\Lambda$ energy level configuration, and thus provide an
estimate of the maximum achievable quantum optical storage time.
I will mention these results in subsequent chapters, because they became also important as the basis for a theoretical formulation of the interaction of squeezed light with an EIT media.

3.2.6 EIT with added decoherence mechanism

Before exploring the storage of light in the quantum regime, it was decided to perform an additional experiment in which we could create an artificial population exchange ratio in order to test the conditions in which the population exchange mechanism could be valid. We implemented this mechanism via a counter-propagating repumper field at the control transition detuned by 160 MHz.

We tried two different configurations: a wide re-pumper beam (wider than the control and signal fields) and a hollow repumper beam with no overlap with the EIT beams. The experimental setup is presented in Fig. 3.11.

When the experiments were performed using a cell with buffer gas, the behavior with a hollow repumper beam and without repumper was identical. From this, we concluded...
that the decoherence outside the EIT beams had little influence on the EIT linewidth.

With full repumper, the minimum EIT linewidth was much larger that in the normal case: over 200 kHz (without repumper, it is in the order of 2-5 kHz). We attributed this phenomenon to the AC Stark shift due to the repumper field since atoms moving with different velocities experience different shifts.

Exchanging the cell with one without buffer gas and using a hollow repumper beam the behavior was nonlinear at low pump powers. This shows that population exchange plays a significant role in the decoherence.

The behavior with a hollow repumper beam and without repumper is different (as it was not the case with a cell with buffer gas). This is because because either wall collisions do not result in full decoherence or the repumper field brings more atoms in $|C\rangle$ than pure decoherence does. Results for these experiments are plotted in Figs. 3.12 and 3.13.

These last small tests showed that it is possible to reproduce experimental conditions where population exchange can play a major role in the decoherence but also proved that such conditions are not the general case regarding how storage of light experiments are performed. Thus, validating our conclusions of last section that population exchange definitively does not play a significant role in the decoherence in the experiments we are interested in.

This line of investigation allowed us to choose further directions for our experiment: we decided to keep advancing in our domain of coherent control processes using EIT while at the same time started to build the preliminaries of our non-classical source. The experimental results of the first path have been already discussed in the Thesis of my colleague Juergen Appel [17] so I will mention them here briefly. The results of the second path will be discussed in full detail in the next chapter of the thesis.
Figure 3.12: Behavior of EIT resonance against control power for different experimental conditions to explore decoherence mechanisms. In this case a 1 Torr buffer gas cell was used.

Figure 3.13: Behavior of EIT resonance against control power for different experimental conditions to explore decoherence mechanisms. In this case the Ribidium cell does not contain additional buffer gas.
3.3 Frequency conversion and routing of quantum information in atomic media

Based on our gained understanding of our system, we decided to tackle a more complicated problem in order to gain more insight into the adiabatic control of atomic systems. By performing this experiment we gathered information that will later prove essential for addressing quantum experiments.

As mentioned in the introduction, one of the three objectives of creating an interface between light and atoms is to prove that useful protocols can be built using our EIT procedures (e.g. nonlinear optics and quantum information processing). For this particular case we decided to tackle the problem of routing information using atomic samples as the router. We wished to develop an interface, taking advantage of the properties induced in atomic media by EIT, capable of converting an input state into several correlated output states (sharing the properties of the input).

This idea is very attractive since an essential element of a quantum optical communication network is a tool for transferring quantum information between optical modes (possibly of different frequencies). This is important not only for routing quantum information, but also for memory-based quantum repeaters [9, 36].

This was not the first attempt to tackle this problem: experiments on frequency conversion of quantum states of light have been performed using nonlinear optical effects in crystals [53, 54] and periodically-poled waveguides [55]. Conservation of quantum information in such a transformation was already demonstrated in Ref [56].

Approaches similar to our experiment have already been put in place using double-lambda systems [57, 58], which involved storage of light by means of electromagnetically induced transparency (EIT) [8] and its subsequent retrieval on another optical transition. This approach is subject to decoherence mechanism we analyzed in the previous section.
and can only be used reliably in a brief window of time after the storage.

We developed our approach on the basis of a theoretical proposal that uses double- and multi-\(\Lambda\) configurations that contain two or more excited levels, which are excited by several control fields (see Fig. 3.14). In this case, if the control fields couple to the same ground state, and the signal fields to different excited states, one could naively expect all of the signals fields to experience EIT. Interestingly enough, this is not the case. In fact only a linear combination of the signal modes will experience EIT while the others will be absorbed [60].

Using this configuration, one can show that by an adiabatic change of the control fields, a transfer of quantum optical states between different signal modes or their linear combinations can be implemented. This procedure resembles stimulated Raman adiabatic passage (STIRAP) [59], but applies to optical rather than atomic states. Our method is related to the method mentioned before [57, 58], but in our case the information is transferred during the propagation, thus avoiding the losses associated with the storage of light.

As has been the case so far in this thesis, I will only make a small overview of the theory in order to understand the results included. For a more detailed discussion, see [60].

It is possible to show that there is a linear superposition of signal states that sees EIT in this system:

\[
\hat{b}_Q = \frac{1}{\Omega_{\perp}} \sum_{q=1}^{Q} \frac{\Omega_q^*}{g_q^*} \hat{a}_q,
\]

(3.8)

where \(g_q\) describes the vacuum Rabi frequency of the signal transition. \(\Omega_q\) is the slowly varying Rabi frequency of the corresponding control field and \(\Omega_{\perp} = \sqrt{\sum_{q=1}^{Q} |\Omega_q|^2}.\)

Signal pulses in the \(\hat{b}_Q\) mode interact with the atoms of a multi-\(\Lambda\)-medium in a fashion
Figure 3.14: a) Multi $\Lambda$-system: $Q$ excited states $|A_q\rangle$ are each coupled by a classical control field $\Omega_q$ with detuning $\Delta$ to the ground state $|C\rangle$ and by a quantized field $\hat{a}_q$ with detuning $\delta$ to another ground state $|B\rangle$.

completely analogous to pulses propagating through the well understood standard EIT system.

3.3.1 Raman Adiabatic Transfer of Optical States

Based on this formalism we can define a protocol for transfer of quantum information between optical modes (Raman adiabatic transfer of optical states, RATOS).

If the intensities of the control fields are changed slowly, the eigenstates follow the new conditions adiabatically. If we use a double lambda system, this allows for transfer of quantum information from an optical mode $\hat{a}_1$ to another mode $\hat{a}_2$:

- First only one strong control field $\Omega_1$ is switched on. The medium then exhibits electromagnetically induced transparency for the $\hat{b}_Q = \hat{a}_1$ mode.

- An incoming quantum pulse in the $\hat{a}_1$ mode can enter the EIT medium without absorption.
• Once the pulse is completely inside the medium, the control field $\Omega_1$ is replaced by another field $\Omega_2$ adiabatically.

• A pulse with a different frequency but in the same optical quantum state as the original pulse exits the medium in mode $\tilde{a}_2$.

If the first control field is left on, the optical state that has entered the cell in mode $\tilde{a}_1$ will leave it in a superposition $\tilde{b}$ (superposition of $\tilde{a}_1$ and $\tilde{a}_2$). This allows the implementation of beam splitting for optical modes of different frequency, with the final intensities of the two control pulses determining the outcome of the process. The beam splitting ratio is given by

$$\frac{\langle \tilde{a}_2 \rangle}{\langle \tilde{a}_1 \rangle} = \frac{g_1 \Omega_2}{g_2 \Omega_1}.$$  

(3.9)

Alternatively, if the pump field is instead switched off adiabatically while mode $\tilde{b}$ is propagating through the cell, the initial quantum state of mode $\tilde{a}_1$ will be fully transferred to mode $\tilde{a}_2$, thus completing the RATOS protocol.

3.3.2 Experimental Setup

The experiment was performed in $^{87}$Rb vapor at 60 °C in a 5 cm long cell filled with 5 Torr of neon as a buffer gas. The cell is mounted within a magnetically shielded oven. The double-$\Lambda$ system is realized using the hyperfine levels of the 795 nm D1 line, with the $5S_{1/2}$ hyperfine levels as the two ground states, which are coupled via three laser fields to the $5P_{1/2}$ levels (Fig. 3.15).

The optical setup is shown in Fig. 3.16. The signal field was provided by a Coherent MBR-110 Ti:Sapphire laser with a narrow spectral width (~40 kHz). The beam was frequency shifted by 160 MHz using an acousto-optical modulator (AOM) in a double-pass configuration, which allowed switching of the signal field. The pump ($\Omega_1$) and retrieve ($\Omega_2$) fields were obtained from external-cavity diode lasers sequentially phase-
Figure 3.15: The transitions used in the experiment: the natural double lambda system in the D1 line of Rubidium.
locked to each other and the Ti:Sapphire laser. Both phase-lock circuits were programmed so that the frequency difference among the three fields entering the cell corresponded to the hyperfine splitting frequencies of the ground and excited levels of the rubidium D1 transition: 6835 MHz and 817 MHz, respectively. The bandwidth of the beat signal in each phase lock circuit did not exceed 10 Hz. The switching required for each diode laser beams was controlled by an AOM driven at 80 MHz.

Before entering the rubidium cell, the beams were combined with the signal beam on a polarizing beam splitter, so the linear polarization of the control beams is orthogonal to that of the signal and RATOS beams. The spatial modes of the input optical fields were carefully matched to each other.

After passing through the cell the two signal fields were separated from the control beams using a polarizing beam splitter, and subjected to heterodyne detection on a fast photodiode. The role of the local oscillator was played by the unmodulated field of the
Figure 3.17: Temporal profiles of the fields. The two control pulses $\Omega_1$ and $\Omega_2$ are switched, respectively, off and on on when the signal pulse enters the cell completely, giving rise to a Ratos pulse ($\hat{a}_0$). Also displayed is the signal pulse $\hat{a}_1$ in the absence of the cell (scaled down) and the slowed down signal pulse in the presence of a constant pump field (regular EIT).

Ti:Sapphire laser. The beat signal at 160 MHz (signal beam) or 657 MHz (RATOS field) was measured using a spectrum analyzer in the zero span mode, with a temporal resolution of 200 ns.

Figure 3.17 shows typical pulse forms for the two control fields, the incoming signal field, and the RATOS pulse. Also shown is the slowed down signal field, observed in normal EIT conditions (constant pump field, no retrieve field). As evident from the figure, RATOS is indeed capable of transferring light from mode $\hat{a}_1$ into $\hat{a}_2$, with an efficiency of approximately 20% with respect to the slowed pulse for this particular measurement. This benchmark can be increased to over 70% by improving the mode-matching between the fields and hence the achievable transparency.
3.3.3 Optical Beam Splitting

In order to realize an optically controlled beam splitter, the pump field $\Omega_1$ could be kept on continuously, while $\Omega_2$ was turned on when the signal pulse had fully entered the rubidium cell. In this case the quantum state of the signal mode $\hat{a}_1$ was transferred into a superposition $\hat{b}$ of the modes $\hat{a}_i$. In the experiment the power of the pump field was kept constant at 4 mW, and the signal fields $\hat{a}_1$ and $\hat{a}_2$ were measured for different powers of the retrieval field.

The results of the optical beam splitting experiment are shown in Fig. 3.18. The three waveforms of the output signal and RATOS fields at different retrieval field powers illustrate the dynamics of multimode dark-state polaritons in the EIT medium. Even though the pump field remains the same in all three plots, the group velocity of the signal pulse (which couples to the pump field through an excited level) increases with the intensity of the retrieval field. The waveforms in Fig. 3.18 also show that the energy ratio of the RATOS and output signal fields increases with the retrieval intensity.
3.3.4 Discussion

In this project we demonstrated the possibility for adiabatic frequency conversion and routing of optical information carried by light between two EIT modes in a multi-Λ energy level configuration. At the same time we gained valuable expertise in atomic coherent control and developed tools that later will be required in the more advance experiments.

These results opened the possibility for a new branch of experiments that could be implemented in the future. For example, the same procedure can be easily implemented using the signal field resonant to the rubidium D1 transition, while the D2 line is used to create the RATOS field. The demonstrated technique is also of particular interest in solid state systems [61], where the level structure allows access to nearly arbitrary frequencies for the created RATOS pulse. Research in this area is currently ongoing by several groups [62].

3.4 Small summary

In this chapter we have presented an experimental study of decoherence of the ground energy levels of $^{87}$Rb atoms in vapor cells. We measured the decoherence of the ground state using three different methods: measuring the decay constant of the storage of light in atomic vapor, the decay rates of transient coherence oscillations of the ground state, and the width of the electromagnetically induced transparency resonances. The measurements showed decoherence rates on the scale of $10^4$ $s^{-1}$.

The dependence of the EIT linewidth on the power of the pump field was investigated, at various temperatures, for the ground states of the lambda-system associated with different hyperfine levels of the atomic $5S_{1/2}$ state as well as magnetic sublevels of the same hyperfine level. Strictly linear behavior was observed in all cases. A theoretical analysis of our results showed that dephasing in the ground state is the main source of
decoherence, with population exchange playing a minor role.

We also showed a quantum communication protocol that enables frequency conversion and routing of quantum optical information. The protocol is based on electromagnetically-induced transparency in systems with multiple excited levels: transfer and/or distribution of optical states between different signal modes was implemented by adiabatically changing the control fields.
Chapter 4

Source of light with non-classical properties

4.1 Useful tools from quantum optics

At this point, we had spent almost two years building our atomic experiments and also gained expertise in the control and manipulation of EIT schemes. It was now time to turn our attention to a more subtle problem: Since we want to study light with non-classical properties interacting with atomic systems under EIT conditions, it was necessary to create a light source with such characteristics.

Since we are interested in the storage of squeezed light, we cannot rely on the atomic-based generation of non-classical light (DLCZ) that we reviewed in chapter 2. As we will develop in this chapter, our interest is the full reconstruction of the quantum state of light after the storage procedure. Because of this, we use another process to generate non-classical light that is more suited to this ultimate purpose: parametric down conversion.

Fortunately, over the last few years our group has developed a strong background in the generation and characterization of non-classical states of light using this physical process.

I begin this chapter with a brief introduction to quantum optics, presenting the main results that are the basis of our non-classical source.

4.1.1 The Quantized Light Field and the Quadratures

To understand the relevant effects for the generation of non-classical light states one has to deal with a quantized radiation field. The quantization of the light field is carried out in many text books and will not be presented here. A nice derivation can be found in
Following this approach, the operators for the electric and magnetic fields in free space are found to be

\[
\hat{E}(x,t) = i \int d^3 k \sum_{\sigma} a_{k\sigma} \sqrt{\frac{\hbar \omega_k}{2(2\pi)^3 \epsilon_0 \omega_k}} e^{i k x - i \omega_k t} + H.C.
\]

\[
\hat{B}(x,t) = i \int d^3 k \sum_{\sigma} a_{k\sigma} \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega_k}} (k \times \epsilon_{k\sigma}) e^{i k x - i \omega_k t} + H.C.
\]

where the sum over \( \sigma \) runs over the two independent polarizations and \( a_{k\sigma} \) is the annihilation operator (satisfying the bosonic commutation relation: \( [a_{k\sigma}, a_{k'\sigma'}] = \delta_{kk'} \delta_{\sigma\sigma'} \)) for the mode with wave vector \( k \) and polarization \( \sigma \). Using the classical expression for the energy density we can calculate the Hamiltonian for the electromagnetic field.

\[
\hat{H} = \frac{\epsilon_0}{2} \int d^3 x \left( \hat{E}^2(x,t) + c^2 \hat{B}^2(x,t) \right) = \int d^3 k \sum_{\sigma} \hbar \omega_k \left( a_{k\sigma}^\dagger a_{k\sigma} + \frac{1}{2} \right)
\]

As we can readily see, the Hamiltonian is just a sum over simple harmonic oscillators for the different modes. We can consider only a single-mode electric field. We can then simplify and write the field as:

\[
\hat{E}(t) = \frac{1}{2} \mathcal{E} \left( a e^{-i \omega t} + a^\dagger e^{i \omega t} \right) = \hat{E}^+(t) + \hat{E}^-(t),
\]

where \( \hat{E}^{(\pm)}(t) \) represent the positive and negative frequency parts. In analogy to the harmonic oscillator, it is intuitive to introduce the two quadrature operators \( \hat{P} \) and \( \hat{Q} \).
\[\hat{Q} = \frac{1}{2}(a + a\dag)\]
\[\hat{P} = \frac{1}{2i}(a - a\dag)\]
with \([\hat{Q}, \hat{P}] = \frac{i}{2}\), \((4.5)\)

which are essentially dimensionless position and momentum operators. In terms of the quadratures the single mode electric field operator can be written as

\[\hat{E}(t) = E \left( \hat{Q} \cos(\omega t) + \hat{P} \sin(\omega t) \right)\]
\((4.6)\)

The operators \(\hat{Q}\) and \(\hat{P}\) are now seen to be the amplitude of the two field components, that have a phase difference of \(\pi/2\). The amplitude of the part of the field that is in phase with a chosen reference, is given by \(\hat{Q}\). The amplitude of the part that is out of phase by \(\pi/2\) is given by \(\hat{P}\).

We can think about \(\hat{Q}\) and \(\hat{P}\) as conjugate variables, a well known fact from the theory of the harmonic oscillator. The uncertainty relation for the two quadratures is:

\[\Delta \hat{Q} \Delta \hat{P} \geq \frac{1}{4}\]
\((4.7)\)

4.1.2 Coherent States

Since we are dealing with lasers sources, it is worthwhile to take a look at the description that quantum mechanics has for this fields. Coherent states represent a quantum mechanical description of a classical field.

To generate a coherent state we could choose a classical polarized medium such as a bulk crystal, in this case the polarization \(P\) of the medium is not quantized and can be represented in the form \(P = Pe^{i\omega t} + P^*e^{-i\omega t}\).

Since we are now interested in the quantum state of light generated by this polarization, we look at the interaction of the polarization with a quantized radiation field that
oscillates with the same frequency $\omega$. The Rotating Wave Approximation-Hamiltonian in this situation is given by:

$$\hat{H}_{\text{int}} = Pe^{i\omega t} \hat{E}(+) + P^* e^{-i\omega t} \hat{E}(-), = \beta a + \beta^* a^\dagger$$

(4.8)

where we have used Eq. 4.3 and defined $\beta = 1/2 PE$. To obtain the state of light generated in the interaction we have to solve the Schrödinger equation in the interaction picture:

$$i\hbar \partial_\tau |\psi_I\rangle = \hat{H}_{\text{int}} |\psi_I\rangle$$

(4.9)

Since the interaction Hamiltonian is time-independent we can perform the integration to obtain:

$$|\psi_I(\tau)\rangle = e^{-i\hat{H}_{\text{int}} \tau} |\psi_I(0)\rangle = e^{\alpha a^\dagger - \alpha^* a} |\psi_I(0)\rangle := \hat{D}(\alpha) |\psi_I(0)\rangle,$$

(4.10)

where $\tau$ is the interaction time and $\alpha = -i/\hbar \beta \tau$. The introduced operator $\hat{D}(\alpha)$ is called displacement operator.

To see what quantum state of light the classical polarization generates, we choose the vacuum state $|0\rangle$ as our initial state:

$$|\alpha\rangle := \hat{D}(\alpha) |0\rangle = e^{-|\alpha|^2/2} \sum_{n=0}^\infty \frac{\alpha^n}{\sqrt{n!}} |n\rangle$$

(4.11)

The generated displaced vacuum state is called a coherent state $|\alpha\rangle$. An alternative definition could be obtained considering that coherent states are eigenstates of the annihilation operator $a$.
Using this property one can readily calculate the variances for the two quadratures \( \hat{Q} \) and \( \hat{P} \).

\[
(\Delta Q)^2(\alpha) = \frac{1}{4} \left( \alpha^2 + \alpha^*^2 + 2|\alpha|^2 + 1 \right) - \left( \frac{1}{2} [\alpha + \alpha^*] \right)^2 = \frac{1}{4} 
\]  
(4.13)

\[
(\Delta P)^2(\alpha) = -\frac{1}{4} \left( \alpha^2 + \alpha^*^2 - 2|\alpha|^2 - 1 \right) - \left( \frac{1}{2\hbar} [\alpha - \alpha^*] \right)^2 = \frac{1}{4}.
\]  
(4.14)

According to the Heisenberg uncertainty principle, the last result implies that coherent states are minimum uncertainty states with symmetric fluctuations in both quadratures. Another important feature is that the uncertainties do not depend on the strength \( \alpha \) of the coherent states and are independent of time.

In quantum optics, states of radiation field are often represented in phase space. The notion phase space is in analogy to that of classical mechanics. In terms of the harmonic oscillator the two quadratures correspond just to position and momentum operator, and they span the phase space well known from classical mechanics.

The quantum phase space is spanned by the two quadratures \( \langle \hat{Q} \rangle \) and \( \langle \hat{P} \rangle \). Calculating the two expectation values for a coherent state, one obtains that \( \langle \hat{Q} \rangle \) and \( \langle \hat{P} \rangle \) are given by the real and imaginary part of the amplitude \( \alpha \), respectively. The uncertainties in the two quadratures correspond to fluctuations in the amplitude and phase of the electric field.

Two more points should be made that are useful for our description: First, coherent states can be characterized by a complex amplitude \( \alpha \), where the two quadratures are given by the real and imaginary part respectively. This means the classical amplitude \( \mathcal{E} \)
translates into the amplitude $\alpha$. Secondly, coherent states are not a point in phase space, the amplitude and phase of the electric field are not exactly defined. This means that while performing measurements there are fluctuations, detected as noise on the measured signal due to the nature of quantum mechanics.

4.1.3 Relevant Light-Matter-Interaction

After describing the fields generated by a laser, we now must briefly address a result that is the foundation of the sources that we will explore in brief. The relevant process we use to generate the states of light we are interested in are normally light-matter-interactions.

When an electromagnetic field propagates through a medium, it induces an oscillating polarization. As we can see from Maxwell’s equations, this polarization generates a new electric field. If the polarization is oscillating at a different frequency to that of the incident electric field, the medium will emit radiation at this other frequency. This process is known as frequency conversion.

We can express the polarization of the medium in terms of the incident electric field as follows

$$P_i = \chi^0 + \chi_{ij}^1 E_j + \chi_{ijk}^2 E_j E_k + \chi_{ijkl}^3 E_j E_k E_l + O(|E|^4)$$  \hspace{1cm} (4.15)

For low light intensities only the first two terms have to be taken into account. This is the regime of linear optics. With the invention of the laser, high intensity phenomena became relevant and many medium were found to show nonlinear behavior.
4.2 Squeezing

4.2.1 Parametric down conversion

After describing these tools we are ready to review the main process we mentioned at the beginning of the chapter. Nowadays, spontaneous parametric down conversion (SPDC) is one of the corner-stones of quantum optics.

First I start with a small physical description: a non linear crystal can split incoming photons into pairs of photons of lower energy whose combined energy and momentum is equal to the energy and momentum of the original photon. The process is spontaneous in the same sense as the atomic spontaneous emission, because it is stimulated by random vacuum fluctuations. Consequently, the photon pairs are created at random times. A very interesting situation can be noted here, if one photon of the pair is detected, we know the partner has to be present. In this sense, by detecting this photon, we are creating a field that contains only one photon. This process has been studied for many years now and is still one of the fundamental tools to create optical fields that present quantum properties.

The SPDC process may be viewed as a coherent three photon process where a crystal is illuminated by a pump beam. This pump beam is intense enough to drive the oscillations of the electrons in the crystal into the non-linear regime. This interaction results in the annihilation of a pump photon and the creation of two down-converted photons. The photon pairs that are created are entangled in space-time, or in wave number and frequency.

Conservation of energy and momentum (i.e. \( \omega_1 + \omega_2 = \omega_p \) and \( k_1 + k_2 = k_p \)), requires a phase matching condition for the three frequency interaction. Due to this condition, the polarization of the primary wave could be different from the polarization of the generated waves. For the case in which the created photons have the same polarization we have
Type I phase-matching. In addition, by choosing the orientation of the optical axis of the crystal, we can generate emergent photons that are collinear or non-collinear. Therefore, different types of crystal configurations will allow the generation of states with different quantum properties.

After our physical description, we can address the theory of SPDC in a bit more detail. As mentioned in section 4.1.3, in order to generate light with quantum properties a nonlinear interaction of the polarization and the incident light field is needed. To describe the process observed in our experiment we take the first nonlinear term in Eq. 4.15 and take an incident field that contains two different frequencies $\omega_1$ and $\omega_2$. We obtain for the nonlinear part of the polarization:

$$\tilde{\mathcal{P}}^{(nl)} \sim \chi^{(2)} \tilde{E} \tilde{E} = \chi^2 \left[ \left( \hat{c}^{(+)}_1 e^{-i\omega_1 t} + \hat{c}^{(-)}_1 e^{i\omega_1 t} \right) + \left( \hat{c}^{(+)}_2 e^{-i\omega_2 t} + \hat{c}^{(-)}_2 e^{i\omega_2 t} \right) \right]^2$$

$$\tilde{\mathcal{P}}^{(nl)} \sim \chi^2 \left[ \left( \hat{c}^{(+)}_1 \hat{c}^{(+)}_2 e^{-i(\omega_1+\omega_2)t} + \hat{c}^{(-)}_1 \hat{c}^{(-)}_2 e^{i(\omega_1+\omega_2)t} \right) + \left( \hat{c}^{(+)}_1 \hat{c}^{(-)}_2 e^{-i(\omega_1-\omega_2)t} + \hat{c}^{(-)}_1 \hat{c}^{(+)}_2 e^{i(\omega_1-\omega_2)t} \right) \right],$$

where we have disregarded the terms where the fields are not mixed. Under these assumptions, the nonlinear part of the polarization oscillates with the sum and difference of the frequencies contained in the incident electric field.

For the SPDC process, we are only interested in the process that generates the...
difference-frequency. Thus we will only consider the part of the polarization oscillating with the difference-frequency.

\[
\hat{P} \sim \frac{\chi(2) \hat{E}_1^{(+)\dagger} \hat{E}_2^{(-)\dagger} e^{-i(\omega_1-\omega_2)t}}{\hat{\rho}^{(+)\dagger}} + \frac{\chi(2) \hat{E}_1^{(-)} \hat{E}_2^{(+)\dagger} e^{i(\omega_1-\omega_2)t}}{\hat{\rho}^{(-)}}
\]  

(4.17)

In the rotating wave approximation (RWA), the interaction Hamiltonian is then given by:

\[
\hat{H}_{\text{int}} = \hat{P}^{(+)} \hat{E}_3^{(-)} + \hat{P}^{(-)} \hat{E}_3^{(+)} = \epsilon_1 \hat{E}_2^{(-)} \hat{E}_3^{(-)} + \epsilon^* \hat{E}_1^{(-)} \hat{E}_2^{(+)\dagger} \hat{E}_3^{(+)} = \gamma \hat{a} \hat{b} + \gamma^* \hat{a}^\dagger \hat{b} \hat{c}
\]  

(4.18)

where the operators \(\hat{a}, \hat{b}, \hat{c}\) are the annihilation operators for the three different modes. In a photon picture the first term of the Hamiltonian describes the process of generating a photon in modes \(b\) and \(c\) by the annihilation of a photon in mode \(a\). This process describes SPDC. The second term describes the inverse process, i.e. the generation of a photon in mode \(a\) by the expense of a photon in mode \(b\) and \(c\). As one can see in both cases energy is conserved. Also important is that momentum is conserved, i.e. the wavevectors of the two generated photons have to sum up to the wavevector of the third one.

Since we have a reliable tool to generate states with non-classical fluctuations of the field, now we can turn our attention to describe a viable measuring method that would allow us to reconstruct their properties and unambiguously prove their quantum nature.

4.2.2 Brief introduction to Homodyne tomography

In this subsection, I will describe another key tool of quantum optics. Since we have to measure fluctuations in the field that are very small, a sensitive measuring technique has
Figure 4.2: Homodyne detection. Left: a strong coherent state interferes with the weak quantum state $|\psi\rangle$ on a 50% transparent beam splitter. The difference of the intensities in both beam splitter outputs then forms the homodyne output signal. Right: an alternative configuration using polarizing beam splitters instead of one symmetric non-polarizing splitter. The waveplate allows to tune the intensity of both outputs to exactly 50%.

As mentioned in chapter 2, several experiments related to ours use photon counting as a tool to describe the quantum nature of the generated states. This method however, lacks phase sensitivity and cannot be used to generate a complete description of the state. One way to overcome these obstacles is to use homodyne detection: a strong coherent beam (the so called "local oscillator, LO") is sent to a symmetric (50%) non-polarizing beam splitter where it interferes with a quantum state $|\Psi\rangle$ that is to be measured. The spatial and polarization modes of the local oscillator and the signal beam have to be matched with great care.

Both output ports of the beam splitter are then each sent to photodetectors and the resulting photo-currents are subtracted. The difference current $\Delta I$ is the output signal. It can be shown to be proportional to the field fluctuations of the incoming quantum field [75]. By using the properties of the beam splitter we eliminate the intensity fluctuations of the local oscillator in order to measure only the fluctuations of the field of interest.

Theoretically proposed in [63] and first experimentally implemented in the early 1990s
[64], Optical Homodyne Tomography (OHT) has become a standard tool of the quantum technology of light, in particular in quantum information applications.

The LO is a strong classical field characterized by the parameter $\beta$, and it should be in exactly the same temporal and spatial mode as the mode to be analyzed, i.e. for cw homodyne detection the LO has to have the same frequency, beam size and divergence as the incoming mode.

The annihilation operator of the incoming and local oscillator modes are denoted $\hat{a}$ and $\hat{b}$, respectively. The two output modes reaching the detector are described by the annihilation operators $\hat{c}$ and $\hat{d}$. They are connected to the input modes as follows:

$$\hat{c} = \frac{1}{\sqrt{2}}(\hat{a} + \hat{b}) \quad \text{and} \quad \hat{d} = \frac{1}{\sqrt{2}}(-\hat{a} + \hat{b}). \quad (4.19)$$

The photocurrents of the two detectors are proportional to the intensities of the incoming fields, which are essentially given by photon numbers $n_c$ and $n_d$. Therefore the difference of the two photocurrents is determined by the operator:

$$\hat{n}_{cd} = \hat{n}_c - \hat{n}_d = \hat{a}^\dagger \hat{b} + \hat{b}^\dagger \hat{a}. \quad (4.20)$$

Considering that the input state $|\psi\rangle$ at the 50-50 beam-splitter is a product state of the state in mode $a$ and the large coherent state in mode $b$. The two modes $a$ and $b$ are independent, i.e. $\langle \hat{a} \hat{b} \rangle = \langle \hat{a} \rangle \langle \hat{b} \rangle$. The measured signal is then given by:

$$i \sim \hat{n}_{cd} = 2|\beta|\hat{q}(\theta) \quad \text{with} \quad \hat{q}(\theta) = \frac{1}{2} \left( \hat{a} e^{-i\theta} + \hat{a}^\dagger e^{i\theta} \right), \quad (4.21)$$

where $\hat{q}(\theta)$ is a rotated quadrature where the rotation angle is determined by the phase $\theta$ of the LO. The signal $i$ is proportional to the quadrature of the squeezed light and scales with the amplitude of the LO. The variance of the signal is found to be:
Remarkably, if we assume the LO to be powerful enough to be able to neglect its quantum fluctuations, the variance $\Delta i$ does not contain any noise of the LO. The quadrature is chosen by scanning the phase $\theta$ of the LO. In this way all quadratures can be investigated.

Two of the main homodyne setups are shown in Fig. 4.2.

The setup (a) uses a 50-50 beam-splitter (BS) to superimpose the local oscillator field and the quantum light. The main problem in this setup is that a slight change in the angle of incident changes the splitting ratio of the beam splitter, making the detector hard to balance.

In setup (b) the 50-50 beam-splitter is replaced by two polarizing beam splitters (PBS) and three half-wave-plates (HWP). The first two HWPs are used to rotate the polarizations of the signal light and the local oscillator in such a way, that the LO is completely transmitted through the PBS and the signal is totally reflected at the PBS. With the third HWP the polarizations of both fields are rotated by $45^\circ$. Due to this rotation, the two fields split at the second PBS into two parts as in a 50-50 BS. The splitting ratio now depends on the angle of the third HWP and therefore the balancing of the homodyne detector is easier to achieve.

4.2.3 Squeezed light

After reviewing homodyne detection, we can finally describe the properties of the quantum state we will use in our experiments. First, I will mention a very interesting idea that follows readily from our measurement setup. For example, if the input to our homodyne detector is vacuum (in this context meaning no photons), what are we actually measuring? Here we have to remember a very important axiom in quantum optics: the

\[ (\Delta i)^2 \sim (\eta_{vd})^2 = 4|\beta|^2 (\Delta q(\theta))^2. \]  \hspace{1cm} (4.22)
number of photons and the electric field are not simultaneous observables, meaning that
the more accuracy with which we know the number of photons involved in an experiment,
the less we know about the electric field.

For example, the number of photons is clearly defined for the vacuum, therefore, even
though there are no particles of light, the field of the vacuum itself still has noise. In
other words, it exhibits fluctuations. The noise we can measured while doing homodyne
tomography of the vacuum is called the shot noise.

In principle, we could think that this an intrinsic limit of nature: light that is turned
off should generate the least amount of noise. Fortunately, it is not the case. If we
combine the ideas of spontaneous parametric down conversion and vacuum fluctuations,
we could produce states of light in which the field fluctuations are less that the vacuum.
These states are called squeezed and I will comment more on them in the next subsections.
Given its characteristics, squeezed light can be used to improve the sensitivity of several
experiments like spectroscopy and interferometry [65]. Furthermore, it can be used as a
tool for quantum information [66].

At this point we have to remember our main objective for the project: store quantum
fluctuations of the field in an atomic coherence and retrieve it. Several groups have
investigated the generation of squeezed light since the late 80's with great success [67]. As
we mentioned before, squeezed light also allows us to measure the quantum fluctuations
of the phase. Considering all this, we decided to produce squeezed light and convert
the fluctuations reduction of the field with respect to vacuum in our essential figure to
demonstrate EIT based quantum memory.

In the next subsections I will briefly review the theory of squeezed light states.
4.2.4 A General Definition of Squeezing

In quantum mechanics the knowledge we can get about a physical observable and the accuracy of this measurement, is limited by the knowledge we have about the canonical conjugate variable or in general, by any knowledge we have about the state of the system. If we consider two Hermitian operators $A$ and $B$ which satisfy the commutation relation:

$$[A, B] = iC.$$  \tag{4.23}

According to the Heisenberg uncertainty relation, the product of the uncertainties in determining the expectation values of two variables $A$ and $B$ is given by:

$$\Delta A \Delta B \geq \frac{1}{2}|\langle C \rangle|.$$  \tag{4.24}

A state of the system is called a squeezed state if the uncertainty in one of the observables ($A$) satisfies the relation:

$$(\Delta A)^2 < \frac{1}{2}|\langle C \rangle|.$$  \tag{4.25}

If, in addition to this condition, the variances satisfy the minimum-uncertainty relation:

$$\Delta A \Delta B = \frac{1}{2}|\langle C \rangle|,$$  \tag{4.26}

then the state is called an ideal squeezed state [14].

In a squeezed state, therefore, the quantum fluctuations in one variable are reduced below their value in a symmetric minimum-uncertainty state ($(\Delta A)^2 = (\Delta B)^2 = |\langle C \rangle|/2$) at the expense of the corresponding increased fluctuations in the conjugate variable such
that the uncertainty relation is not violated.

\[ \omega_3 = \omega_1/2 \]

Figure 4.3: Schematic representation of *degenerate spontaneous parametric down-conversion*. The strong classical field is converted into a field with half the frequency due to the interaction with vacuum that catalyzes the conversion process due to the fluctuations in the electric field.

At this point we have to make a connection with the SPDC process to introduce how squeezed states are generated. For this we have to focus our attention to the particular case of SPDC where one photon with frequency \( \omega_1 \) is split into two photons with frequency \( \omega_1/2 \). This process is called *degenerate down-conversion*. The interaction Hamiltonian (Eq. 4.18) then simplifies to:

\[ \hat{H}_{\text{int}} = \zeta \hat{b}^2 + \zeta^* \hat{b}^2 \]  

(4.27)

Solving the Schroedinger equation in the interaction picture and proceeding similarly to section 4.1.2 we can obtain:

\[ |\psi_I(\tau)\rangle = e^{-\frac{i}{\hbar} \hat{H}_{\text{int}} \tau} |\psi_I(0)\rangle = e^{i(\xi^2 - \xi^* \phi)} |\psi_I(0)\rangle =: \hat{S}(\xi) |\psi_I(0)\rangle, \]  

(4.28)

where \( \hat{S}(\xi) \) is called the *squeezing operator* and \( \xi = \rho e^{i\phi} \) is referred to as the *squeezing parameter*. The absolute value \( r \) of the squeezing parameter will determine the size of squeezing. It essentially depends on the nonlinearity of the medium and the interaction time, which is mainly given by the length of the medium.

If the electromagnetic field is initially prepared in the vacuum state, i.e. there is no incoming field with frequency \( \omega_2 \), it evolves under the action of the squeezing operator
into the state:

\[ |\xi\rangle = \hat{S}(\xi) |0\rangle \quad \text{with} \quad \langle \xi | \hat{H}_{\text{field}} |\xi\rangle = \frac{1}{2} + \sinh^2(r), \quad (4.29) \]

which is called a *squeezed vacuum state*. As we can see, the squeezing process adds energy to the state. The first term in Eq. 4.29 is the energy of the vacuum and the later term quantifies the added fluctuation energy due to the squeezing process. A *squeezed coherent state* is obtained by the squeezing operator acting on a coherent state:

\[ |\alpha, \xi\rangle = \hat{S}(\xi) \hat{D}(\alpha) |0\rangle \quad \text{with} \quad \langle \alpha, \xi | \hat{H}_{\text{field}} |\alpha, \xi\rangle = |\alpha| + \frac{1}{2} + \sinh^2(r), \quad (4.30) \]

After looking at how squeezed states can be generated, I will now review the uncertainties in the two quadratures for squeezed states since it is there where we can find the quantum signature of these states. We can do it for squeezed coherent states, since they include squeezed vacuum. It is useful to do the calculation with quadratures \( \hat{q} \) and \( \hat{p} \) that are rotated with respect to the original quadratures \( \hat{Q} \) and \( \hat{P} \) by an angle \( \phi/2 \),

\[ \hat{q}\hat{p} = \begin{pmatrix} \cos \frac{\phi}{2} & \sin \frac{\phi}{2} \\ -\sin \frac{\phi}{2} & \cos \frac{\phi}{2} \end{pmatrix} \hat{Q}\hat{P}, \quad (4.31) \]

where \( \phi \) is the phase of the squeeze parameter. As we have seen while discussing homodyne detection, the quadratures simply corresponds to a different choice of reference phase.

Using Eq. 4.21 as well as the following transformation properties for the displacement
and squeeze operator:

\[ \hat{D}(\alpha) \hat{a} \hat{D}(\alpha) = \hat{a} + \alpha \quad \text{and} \quad \hat{S}(\xi) \hat{a} \hat{S}(\xi) = \hat{a} \cosh r - \hat{a}^\dagger e^{i\phi} \sinh r, \]  \hspace{1cm} (4.32)

and their Hermitian conjugate, the calculation of the quadrature fluctuations can be done to find:

\[ \langle \Delta q^2 \rangle_{(\alpha, \xi)} = \langle q^2 \rangle_{(\alpha, \xi)} - \left( \langle \hat{q} \rangle_{(\alpha, \xi)} \right)^2 \]

\[ \langle \Delta q^2 \rangle_{(\alpha, \xi)} = \frac{1}{4} \langle \hat{q}^2 e^{i\phi} + \hat{q}^2 e^{-i\phi} + 2\hat{q}^\dagger \hat{q} + 1 \rangle_{(\alpha, \xi)} - \frac{1}{4} \left( \langle \hat{q} e^{-\phi/2} \hat{q}^\dagger e^{i\phi/2} \rangle_{(\alpha, \xi)} \right)^2 = \frac{1}{4} e^{-2r}. \]  \hspace{1cm} (4.33)

Similarly, we find for \( \Delta p^2 \):

\[ \langle \Delta p^2 \rangle_{(\alpha, \xi)} = \frac{1}{4} e^{2r}. \]  \hspace{1cm} (4.34)

and thus the product for the two uncertainties is given by:

\[ \Delta q \Delta p = \frac{1}{4}. \]  \hspace{1cm} (4.35)

We can see that squeezed coherent states are minimum uncertainty states and thus they are ideal squeezed states. The error circle of the initial coherent state is squeezed into an error ellipse (See Fig. 4.4).

The principle axis of the ellipse lie along \( q \) and \( p \) that are rotate by an angle \( \phi/2 \) with respect to \( Q \) and \( P \). This means that the phase \( \phi \) of the squeezing parameter, which is essentially given by the phase of the classical field \( E_1 \) with respect to the reference, gives the "direction" of squeezing. As we can see, the degree of squeezing is determined by the
absolute value of the squeeze parameter $r$.

One can generalize the single mode squeezed state to a multi-mode squeezed state. In order to do that we have to consider a strong classical field $E_1$ but at this time the photons with frequency $\omega_1$ will be split into two different frequencies. As we have already reviewed, during the conversion process, energy must be conserved. Thus, the frequencies of the two generated photons are symmetrically placed around the frequency $\nu = \omega_1/2$ of the mode in the degenerate case. The interaction Hamiltonian then takes the form

$$\hat{H}_{\text{int}} = \beta \hat{b}_{\nu+\nu'}^\dagger \hat{b}_{\nu-\nu'}^\dagger + \beta^* \hat{b}_{\nu+\nu'} \hat{b}_{\nu-\nu'} \quad \Rightarrow \quad \hat{S}_2(\xi) = e^{\xi \hat{b}_{\nu+\nu'}^\dagger \hat{b}_{\nu-\nu'}^\dagger + \xi^* \hat{b}_{\nu+\nu'} \hat{b}_{\nu-\nu'}}$$

(4.36)

where $\hat{b}_{\nu\pm\nu'}$ are the annihilation operators for the two sidebands $\nu \pm \nu'$.

Proceeding similarly to the previous cases we can find that the created states are ideally squeezed two-mode states [14].

Two-mode squeezed states are also often referred to as Einstein-Podolsky-Rosen entangled light because of their quantum correlations.

To understand the importance of multi-mode squeezing we have to look at our ho-
modyne detection method. As mentioned above, in the conversion process energy and momentum have to be conserved. In the experiment this condition is not only fulfilled for the degenerate down conversion but as well for pairs of sidebands. Meaning that we actually create two-mode squeezed states for each sideband. In our detection scheme we interfere the generated photons with a beam of frequency $\nu$. Therefore the interference signal beats with the frequencies $\pm \nu'$. Because experimentally we are not able to distinguish between positive and negative frequencies (only frequency differences), using homodyne detection allow us to detect two correlated sidebands at the same time, thus measuring the properties of the two-mode state for each sideband.

Actually, in the performed experiments we are only detecting multi-mode squeezing. This is due to the fact that in the degenerate case, where the frequency of the LO and the squeezed light are identical, a DC photo-current is generated in the two photo-diodes. Since the electronics in the homodyne detector have $1/f$-noise, the fluctuations of the DC photo-current are covered in noise and are thus not measurable

We could detect the degree of squeezing at $\nu' = 0$ by frequency shifting the LO by $\Delta \nu$. Then the squeezed light generated in degenerate down conversion produces a photocurrent with frequency $\Delta \nu$, due to the interference with the LO, and is observed in the squeezing spectrum at $\Delta \nu$.

After reviewing the properties of squeezed light, its production and detection, it is time to turn our attention on how to interact it with atomic ensembles.

Two main challenges have to be overcome in order to interact the squeezed light with the atoms. First, squeezed light resonant to rubidium atoms had not yet been produced prior to commencement of this thesis work. Therefore, we have to adapt the existent developments to our 795 nm wavelength. Additionally, the bandwidth of the squeezed light has to be in the same order than the EIT line-width. As we have seen in the previous chapter, this width is in the order of less that 1 MHz. Fortunately enough, the problem to
reduce the bandwidth has been solved previously using optical parametric amplifiers (OPA). We present these devices in the next section.

4.3 Optical parametric amplifiers

The general idea of the OPA is rather simple, we use the normal SPDC process in a non-linear crystal to produce correlated pairs of photons. In this case a Type I collinear down conversion crystal will help our purposes, since we need to make the two photons completely indistinguishable. Then we put an optical cavity surrounding the crystal and force the emerging photons to travel resonantly in the cavity. By doing this we increase the probability that the quantum field of the photons interferes with the fluctuations of the vacuum. When this interference is destructive, the squeezing increases. The idea is simple, but as we will see in this chapter, it is technically very challenging.

Since we need light tuned to the rubidium transitions, we used the same source we used as signal for the EIT experiments described in the last chapter. The light from the Ti:Sapphire laser has to be frequency doubled (we reviewed the second harmonic generation process briefly in the last section but a better and more complete review can be found in [14]). These blue energetic photons will be the pump of our SPDC process, and the bandwidth of the final state will be given by the bandwidth of the cavity. The normal election for the cavity is a bow-tie configuration (this choice will become clear when we analyze the technical details) with out-coupling through one of the flat mirrors.

4.3.1 Small introduction to the theory of OPA's

Before reviewing the implementation of our OPA, we quickly highlight some of the most important results of the theory of OPA's.

The physical observables that we are actually interested in are the electric field amplitudes at sideband frequencies $\Delta \omega$. These are accessible to measurements by homodyne
detection, where the output of the OPO is mixed on a symmetric beam splitter with a strong coherent beam. A photocurrent is generated that is proportional to the intensity difference of the two output ports of the beam splitter. The measured RF power of this current then is proportional to a superposition of the two sideband quadrature operators:

$$\hat{q}_\phi(\omega) = \frac{1}{\sqrt{2}} \left[ \hat{a}(\omega)e^{i\theta} + \hat{a}^\dagger(-\omega)e^{-i\theta} \right].$$ (4.37)

In order to obtain a theoretical expression for the quadrature noise of the squeezed vacuum, we express the noise magnitude as:

$$V(\omega) = \langle \hat{q}_\phi(\omega)\hat{q}_\phi(-\omega) \rangle$$ (4.38)

This noise is calculated in [67]:

$$V^\pm(\omega) = 1 \pm \eta \frac{4\sqrt{P/P_{th}}}{\omega/\gamma^2 + (1 \mp \sqrt{P/P_{th}})^2},$$ (4.39)

where $\omega$ is the sideband frequency, $\eta$ is the overall quantum efficiency, $P/P_{th}$ is the ratio of the OPA pump and threshold powers, and $\gamma$ is the cavity bandwidth.

The maximum squeezing level can be increased by reducing the reflectivity of the OPA front mirror. However, this is at the expense of greatly raising the OPA threshold and is only feasible with a very powerful pump laser.

As we have seen before at the beginning of the chapter, the noise reduction in the $(+)$ quadrature comes with an increase of the noise in the $(-)$ quadrature. While the first one is bounded by the losses, the latter one can get arbitrarily high near threshold.
4.4 Elements to create the pumping beam

In order to create the 397.5 nm radiation required to pump the OPA, several stages of conversion are needed. The main source for this purpose is a Verdi V10 diode-pumped solid state Nd:Yag laser capable of producing 10 W of CW output power at 532 nm.

This laser pumps a Coherent MBR-110 continuous wave Ti:Sapphire laser. The MBR-110 laser is tunable from 750 to 900 nm, and has a linewidth of less than 40 kHz when stabilized to its reference cavity. When tuned to emit radiation at 795 nm, up to 1.7 W of power can be obtained. As we have seen in chapter 3, this laser is our main source of radiation resonant to rubidium atoms.

1 W of the output of the Ti:Sapphire laser is directed to a resonant frequency doubler FD-SF-07 custom made by TekhnoScan for our purposes. The cavity is a conventional bow-tie design (as analyzed in Appendix E) with a BBO crystal between the two curved mirrors. The fundamental radiation is kept resonant to the cavity using the Hansch-Couillaud locking scheme [68]. This technique is applicable to a cavity containing a birefringent material, which gives a polarization dependent phase shift. The error signal is obtained using a polarization analyzer consisting of a polarization beam splitter, a quarter wave plate and two photo-detectors. The obtained error signal is fed to a piezo mounted on the back of one the plane mirrors in order to keep the length of the cavity stable. Under this condition, when the fundamental radiation power is tuned to 795 nm the doubler outputs up to 200 mW of radiation at 397.5 nm. A schematic of this part of the experiment is presented in Fig. 4.5.

4.5 Implementation of the OPA

Since many groups have reported squeezing at several different wavelengths, we had the hope that implementing the OPA would be an easy task. In actuality, this proved to be
Figure 4.5: Schematics of the squeezed light source.

quite challenging, since we had to develop some of the components from the beginning.

The first critical element is the non-linear crystal. Only a few companies have the capability to produce the crystals with characteristics suitable for our purposes. We interacted with one of them (Raicol Inc.) to develop crystals with the required characteristics. More on this development can be found in A4.

For the first implementation of the OPA we used a periodically poled KTiOPO$_4$ (PPKTP) crystal with a 3.175 $\mu$m poling period. The crystal was 1 mm x 2 mm in cross-section and $L_c = 5$ mm long. It was cut for propagation along the crystallographic $X$-Axis and its end faces were polished and anti-reflection coated for both (397.5 nm) and (795 nm) light (measured loss of $L/2 = 0.5\%$ per surface).

The cavity itself followed the following considerations (see Fig. 4.6 for clarity):

- Mirrors with curvature radi of 100 mm ($M_1,M_2$) were placed approximately one focal length apart from the crystal. This provides for a small beam waist inside the crystal. This waist will prove later to be very influential in the squeezing that could be obtained.
Two flat mirrors (M3,M4) were placed closely behind and beside the curved mirrors so that the reflection angle is just large enough for the light path not to be obstructed by the crystal mount. The distance between the two mirrors is $2l_2 = 200$ mm and their lateral displacement is $h = 20$ mm with respect to the crystal axis.

One of the flat mirrors (M4) is the output coupler and has a reflectivity of 93.1(2)%. All other mirrors are high reflectors with reflectivities > 99.9% for light around 795 nm and low reflectance (< 3%) for 397.5 nm light. The back faces of all mirrors were anti-reflection coated for both wavelengths.

The other flat mirror (M3) is mounted on a piezoelectric actuator to allow for a stable locking of the cavity length.

The design of the cavity was done using the ABCD matrix formalism for gaussian beams to determine the region where the cavity will work stably and also to quantify the astigmatism (See Appendix E). We then could predict the beam waist inside the crystal.
dependent on the distance between the two curved mirrors. By adjusting this distance, focus sizes in the range of 15-40 μm could be set. We measured a distance of 124(1) mm which agrees well with the predicted stability range and delivers an intermediate focus size. By aligning a red seed beam inside the cavity, we could measure its properties, for example a free spectral range of 464 MHz which agrees very well with our measurements of 460.0(4) MHz.

4.5.1 Alignment

For the first step of the alignment procedure we set up the cavity as a frequency doubler using the output coupler as the input of red light and optimized the crystals position and its temperature to obtain maximum SHG and a clean TEM$_{00}$ mode.

The red reflection off the output coupler M4 was then used to stabilize the resonator length by means of the Hansch-Couillaud technique [68]. (See Fig. 4.7 for a detailed setup).

Once this is achieved, the next step was to align the blue pump light, so that it provides parametric gain in the crystal for photons resonant with the cavity’s TEM$_{00}$ mode. Our main objective was to maximize the mode-matching between the blue pump and the blue light generated in the OPA cavity with the idea that this will maximize the squeezing. This simple step took us several months of careful lens placement and alignment. An alternative method would have been the use of a mode matching cavity [65].

Once the pump beam was aligned, we proceeded to set up an auxiliary laser beam to stabilize the cavity length. An attenuated beam from a dedicated diode laser was mode matched to the OPO cavity mode entering through the output coupler in the direction counter-propagating to that of the blue pump and the parametric radiation.

A λ/2 waveplate was used to match the polarization to the OPA mode. The reflection
Figure 4.7: Schematics for the first part of the procedure to align the OPA. A red beam is injected into the cavity to produce blue radiation using the non-linear properties of the crystal. The cavity is stabilized to obtain a stable output. Inset: Hansch-Couilloud locking signal and the cavity modes of the cavity. Lower curve: Transmission of the OPO cavity at the fundamental wavelength (795 nm); middle: corresponding locking signal obtained with the difference-detector; upper curve: Emission of second harmonic (397.5 nm) in the OPO cavity. The dotted lines depict the reversal points of the piezo actuator.
off the output coupler was collected and detected by a fast photodetector of bandwidth 50 MHz. See Fig. 4.8 for details.

The frequency of the diode laser was electronically phase locked to the Titanium-Sapphire laser at a frequency offset of three free spectral ranges of the OPA cavity (1.38 GHz). The laser diode injection current was also modulated with a weak 30 mV, 20 MHz signal via a bias-tee producing weak phase-modulation sidebands on the laser’s output. This enabled us to obtain a Pound-Drever-Hall (PDH) error signal [69].

We sent the seed beam mentioned in section 4.5 into the system to see the output of the cavity while locked. Stabilizing the cavity length with the PDH-lock provided a steady beam of light exactly in the TEM$_{00}$ spatial mode. This light was counter propagating to the PDH locking beam and was detected once it separated from the PDH beam. We used this beam to align the rest of the experiment and to mode match the local oscillator.

### 4.6 Detection of squeezed light

Once we mode matched the local oscillator to the seed beam coming from the cavity, we sent the squeezed light to the Homodyne detection setup.

In our early experiments, the circuit for homodyne detection used two Hamamatsu S3883 photodiodes of 94% quantum efficiency in a “back-to-back” configuration, akin to that employed in Refs. [70, 71], followed by a OPA847 operational preamplifier. The detector exhibited excellent linearity and an up to 12-17 dB signal-to-noise ratio over a 100 MHz bandwidth, with local oscillator powers up to 10 mW. At a 1 MHz sideband the highest signal-to-noise ratio of the detector exceeded 17 dB. We analyzed the detector output in the frequency domain using an Agilent ESA spectrum analyzer.

The maximum level of squeezing that we could obtain with the original setup was 3dB. This squeezing level was achieved with a high pump power of 260 mW.
Figure 4.8: Schematics for the second part of the procedure to align the OPA. A red beam which frequency is locked 3 Free Spectral Ranges apart from the main Ti:Sa laser and modulated with a 20 MHz signal is injected into the cavity in the counter-propagating direction with respect to the SPDC generated in the crystal. The cavity is stabilized using the Pound-Drever-Hall technique to obtain a stable output of seed beam/squeezed light. Inset: Pound-Drever-Hall locking signal and the cavity modes of the linear cavity. Lower curve: Transmission of the OPO cavity at the fundamental wavelength (795 nm); upper curve: corresponding locking signal obtained with the fast-detector.
Figure 4.9: Quadrature noise spectrum of the squeezed vacuum generated by the OPA, obtained by a simultaneous scan of the local oscillator phase and the detection frequency. The green line is the shot noise spectrum (plus electronic noise).

Figure 4.9 shows the electronic spectrum of squeezed vacuum field generated by the OPA. Up to 3 dB of squeezing is observed within the cavity linewidth. Almost simultaneously, sources with similar characteristics were reported by the Lam group in Canberra [72] and the Kozuma group in Tokyo [73]. This indeed opened a race to achieve EIT interaction with the squeezed light.

4.7 Electronic Noise in Optical Homodyne Tomography

Once the OPA was operational, we worked in a side project, where the properties of the source were useful to prove new results in homodyne tomography theory.

Optical homodyne tomography (OHT) is prone to a variety of inefficiencies, the main one being the optical losses (OLs). This inefficiency is due to absorption in beam paths, non-unitary efficiency of the photodiodes, imperfect balancing of the detector, and, partially, imperfect mode matching between the signal and the local oscillator modes [74]. Also, the presence of non-mode-matched signal light appears as added noise in the de-
tector, raising the apparent noise floor [74]. All OLs can be modeled by an absorber in the signal beam path. The effect of an OL in quantum state reconstruction is well understood, and there are ways of both its quantitative evaluation and compensation for its effects in the quantum state reconstruction procedure [75, 76].

An important additional inefficiency source is the electronic noise (EN) of the homodyne detector. Its effect is the addition of a random value to each quadrature measurement, which causes errors in quantum state reconstruction. Unlike the optical inefficiency, the EN has not yet been widely studied, and is usually neglected whenever efficiency analyses are made [77, 78, 79, 80, 81, 82, 83].

The purpose of this project was to analyze the role of the electronic noise in OHT. We showed its effect to be equivalent to that of optical losses. The key to this equivalence is the detector calibration procedure, which involves measuring the quadrature noise of the vacuum state. Applying this procedure to a vacuum state contaminated with EN results in rescaling of the field quadratures, analogous to that taking place due to optical absorption.

We confirmed these findings using the OPA squeezing experiment, in which we vary the local oscillator power to achieve different signal-to-noise ratios of the homodyne detector.

Normally, a non-unitary optical efficiency $\eta$ has to be taken in account to consider the optical loses when performing reconstruction. Moreover, in the absence of the electronic noise, the electric signal $V$ generated by the homodyne detector is proportional to the quadrature sample $X$: $V = \alpha X$. If we include now the electronic noise in our analysis using $T$, the mean square noise magnitude, it is possible to prove that we may treat the electronic noise as an additional optical attenuator with a transmission equal to $\eta_{eq}$ [4]. Accordingly, the known numerical procedures for correcting for optical losses can be readily applied to the electronic noise.
\[ \eta_{eq} = \frac{\alpha^2}{\alpha'^2} = \frac{\alpha^2}{\alpha^2 + T}. \] (4.40)

For experimental applications, it is more convenient to express the equivalent efficiency in terms of the signal-to-noise ratio of the detector. By the latter we understand the ratio between the observed mean square noise of the vacuum state (in the presence of the EN) and the mean square electronic noise,

\[ S = \frac{\alpha^2 + T}{T}. \] (4.41)

Re-expressing the equivalent efficiency in terms of \( S \) yields

\[ \eta_{eq} = \frac{S - 1}{S}. \] (4.42)

As evidenced by this equation, reasonably low electronic noise does not constitute a significant efficiency loss. For example, the original time-domain detector of Smithey and co-workers [77, 64], featuring a 6 dB signal-to-noise ratio (i.e. the rms quadrature noise being only twice as high as the rms EN) has an equivalent quantum efficiency of 75%. A similar equivalent efficiency is featured by a high-speed detector by Zavatta et al. [71]. A 14 dB time-domain detector of Hansen and co-workers [70] has an equivalent efficiency of 96%, and a typical frequency-domain detector with a signal-to-noise ratio of 20 dB has an efficiency of 99%.

To test these predictions, we stabilized the pump level of the OPA at 140 mW and set the spectrum analyzer to run in the zero span mode at a detection frequency of 1 MHz. The signal-to-noise ratio of the detector was varied by changing the local oscillator...
intensity. At each intensity value, we recorded three spectrum analyzer traces: electronic noise (measured by blocking both photodiodes of the homodyne detector), shot noise (measured by blocking the squeezed vacuum signal) and the phase-dependent squeezed vacuum quadrature. The highest \( \langle Q_+^2 \rangle \) and lowest \( \langle Q_-^2 \rangle \) quadrature noise levels of the squeezing measurement were then determined and normalized to the shot noise level. The uncertainty of \( \langle Q_+^2 \rangle \) was estimated as 0.2 dB. The results of these measurements are shown in Fig. 4.10.

To determine the quantum efficiency, we used the fact that for the pure squeezed state, the product of the mean square quadrature uncertainties is the minimum allowed by the uncertainty principle, i.e. \( \langle Q_{\text{pure,}+}^2 \rangle \langle Q_{\text{pure},-}^2 \rangle = 1/4 \) [4], where the normalization factor corresponds to the shot noise level. After undergoing an optical loss, the squeezed state remains Gaussian, but loses its purity. Using the beam splitter model of absorption, we found that upon propagating through an attenuator of transmission \( \eta \), the quadrature noise reduces by a factor of \( \eta \), but gains excess vacuum noise of intensity \( 1 - \eta \):

\[
\begin{align*}
\langle Q_+^2 \rangle &= \langle \eta Q_{\text{pure,}+}^2 \rangle + (1 - \eta)/2; \\
\langle Q_-^2 \rangle &= \langle \eta Q_{\text{pure,}-}^2 \rangle + (1 - \eta)/2.
\end{align*}
\]

\( \text{(4.43a)} \)

Measuring \( \langle Q_+^2 \rangle \) and \( \langle Q_-^2 \rangle \) and solving Eqs. (4.43), we could find the value of \( \eta \), i.e. how much loss a pure squeezed state would have experienced to generate the state with the quadrature noise levels observed [84]:

\[
\eta = \frac{(2\langle Q_+^2 \rangle - 1)(1 - 2\langle Q_-^2 \rangle)}{2\langle Q_+^2 \rangle + 2\langle Q_-^2 \rangle - 2}.
\]

\( \text{(4.44)} \)
Figure 4.10: Experimental results. a) Observed shot noise as a function of the local oscillator power. Also displayed is a linear fit and the electronic noise level. b) Observed squeezing as a function of the local oscillator power.
The result of applying this method to our squeezed vacuum data is displayed in Fig. 4.10. To obtain a theoretical fit for these results, we expressed the overall quantum efficiency as

\[ \eta = \eta_{\text{esc}}\eta_{OL}\eta_{\text{det}}\eta_{\text{eq}}, \]

(4.45)

where \( \eta_{\text{esc}} \) is the escape efficiency of the optical cavity, \( \eta_{OL} \) is the propagation linear loss, \( \eta_{\text{det}} \) is the quantum efficiency of the homodyne detector and \( \eta_{\text{eq}} \) is the equivalent efficiency of the electronic noise. The first three factors remained constant throughout our measurement, and the fourth one can be evaluated from Eq. (4.42). A fit with \( \eta_{\text{esc}}\eta_{OL}\eta_{\text{det}} = 0.51 \) is shown in Fig. 4.11 with a solid curve.

The first three factors in Eq. (4.45) can be estimated independently as \( \eta_{\text{esc}} = T/(T + L) = 0.88 \) [85], \( \eta_{OL} = 0.95 \) and \( \eta_{\text{det}} = 0.85 \). The product of these values equals 0.71. We believe that the additional efficiency loss occurred due to blue-light-induced infrared absorption in the crystal in the presence of the pump field. In order to eliminate this loss while retaining the amount of squeezing, in the experiments I will describe in the next chapters, we used a cavity with a wider waist and a longer nonlinear crystal.

### 4.8 OPA second generation

After these results were published, we dedicated some time to improve the squeezing generated in the OPA. With that goal in mind, we switched the 5mm crystal for a 20 mm crystal with the same properties of centered wavelength and temperature of operation. Interestingly enough neither the free spectral range nor the line-width of the cavity changed significantly, remaining at 460 MHz and approx. 6 MHz respectively. We also changed the diameter of the blue pump inside the crystal to almost twice of its original value. This allowed us to obtain more additional squeezing (almost 4 dB) while at the same time using less pump power, up to 50 mW of 397.5 nm light (See Fig. 4.12).
Figure 4.11: Equivalent quantum efficiency $\eta_{eq}$ as a function of the detector's signal-to-noise ratio $S$. This ratio is expressed in decibels as $10 \log_{10} S$. Both the experimental results and the theoretical prediction (multiplied by an optical efficiency factor of 0.51) are displayed.

Figure 4.12: Improved squeezing using the 20mm crystal. The shot noise includes the electronic noise contribution which is 15 dB below.
Achieving these results, we were one step away from observing EIT in the quantum regime.

4.9 Pulse creation and reconstruction

Since we require pulses of squeezed light to be able to observe EIT based slow down and storage, the generation of lossless pulses is of prime importance. For this particular experiment, we can not use AOM's akin to the experiments reviewed in chapter 3 because of the intrinsic loses in these devices. It is also required to have short pulses with pulse lengths of approx. 1 μs so that the frequency content of such pulses is comparable to the EIT width. To that end an ultra-fast chopper wheel was constructed from a hard drive. The design of the chopper can be found in my colleague Juergen Appel's thesis [17].

The output light from the OPA is focused to a 25 μm beam size. A 50 μm slit (Thorlabs, S50R) is mounted to the rim of an aluminum disk attached to the hard drive control. The disk is then positioned such that when rotating, the slit passes through the focused laser beam. Using this method, it is possible to create pulses of about 1 μs width (See Fig. 4.13). A PBS is used to mix the squeezed light and a local oscillator (See Fig. 4.2 b). Careful mode-matching is performed to achieve 97% visibility between the local oscillator and the non-classical light. This allows the use of Homodyne Tomography to reconstruct the properties of the squeezed light pulses.

For this particular experiment, we designed and constructed a different homodyne detector, using two Hamamatsu S3883 photodiodes of 94% quantum efficiency. The detector exhibits excellent linearity and an up to 20 dB signal-to-noise ratio over a 6 MHz bandwidth, with local oscillator powers up to 10 mW. The detector has two outputs one of which is DC coupled, the other being AC coupled.

The chopper wheel is programmed to a rotation frequency of 250 Hz. For the majority
of the rotation period the squeezed light passes through the chopper wheel unobstructed. When the slit carrier passes into the beam it interrupts the light for 50 $\mu$s, generates a 700 ns light pulse and blocks the light for another 50 $\mu$s. (See Fig. 4.14)

Since this configuration uses pulses, we cannot only detect squeezing using frequency domain tomography as before. In this case we have to measure the squeezing in all the frequencies contained in the pulse bandwidth to determine the quantum state of the pulsed optical mode generated by the chopper. This procedure is known as time domain homodyne tomography [86].

For this procedure it was important to know the relative optical phase of the local oscillator beam with respect to the quantum state. In our setup, air density fluctuations
(mainly due to the spinning of the hard drive itself) lead to random drifts of the relative phase between the two beams.

During the period when the squeezed light is not blocked by the slit, the AC output of the homodyne detector was directed to a spectrum analyzer and used to detect the phase difference between the squeezed light and the local oscillator. Since the chopper wheel blocks the signal for < 2% of its rotation period, the introduction of the wheel does not change the spectrum analyzer's signal significantly compared to the CW regime.

The oscilloscope was set to its frame acquisition mode and was triggered by a photointerrupter attached to the chopper wheel. It recorded the AC output of the homodyne detector in a 10 µs window around the 700 ns pulse with a sampling rate of 100 MHz. Over 2 s, 500 pulses were recorded. The data was then transferred to a computer in 0.5 s, and the procedure was repeated 100 times. (See, Fig 4.15)

For each of the 100 acquisition runs a waveform generator synchronously generated a triangular signal. This signal was applied to a piezo which controlled the local oscillator phase, so that it was scanned over > 2π every 2.5 s acquisition sequence.

During each sequence, the spectrum analyzer recorded the power of the noise in the DC output of the homodyne detector.

Over a time of 250 s we gathered a total of 50000 pulses consisting of 1002 samples, along with 100 spectrum-analyzer traces of the average homodyne noise power. At the same time, we were able to measure the squeezed light pulses and additional vacuum state measurements.

We then associated a local oscillator phase, determined by the spectrum analyzer power level at the specific time of a given pulse, to each of the 50000 pulses.

To analyze the squeezing, we convoluted the noise detected with a weighting vector corresponding to a classical pulse from the chopper wheel (See Fig. 4.16). This ensured that only the noise corresponding to a pulse of squeezed light is included in the analysis.
Figure 4.15: Experimental set-up to retrieve pulsed squeezing.

Figure 4.16: Spatio-temporal mode used to analyze the pulsed squeezing.
Figure 4.17: Quadratures for the pulsed squeezing.

Figure 4.18: Noise measurements of the pulsed squeezing after the Time Domain Homodyne Tomography.

Figure 4.17 shows the reconstruction of the pulsed squeezed noise coming from the OPA after comparison with the measured vacuum noise. The reconstruction is in good agreement to what was observed with a spectrum analyzer using frequency domain detection (See Fig. 4.18).

We then proceeded to perform a Maximum Likelihood reconstruction [87] of the data using the quadrature values to obtain the density matrix and the Wigner function [75] of the pulsed state (See Figs. 4.19 and 4.20).

Once we proved that we had a reliable source of pulses of squeezed light, it was time to get to the interesting experiments combining the EIT and the non classical source.
Figure 4.19: Density matrix of the pulsed squeezing.

Figure 4.20: Wigner function of the pulsed squeezing.
The next two chapters are related to this step and are the core of the results presented in this thesis.

4.10 Small summary

A novel ultra-narrow-bandwidth source of squeezed light was designed and built: an optical parametric amplifier is pumped below threshold by a frequency doubled and stabilized Ti:Sapphire laser. The produced squeezed vacuum is resonant to the 795 nm, $D_1$ transition of Rubidium atoms. The source uses especially manufactured periodically poled KTP crystals that allow for high squeezing values of up to 4 dB. 1 μs. Pulses of squeezed light were generated by a fast chopper and a full tomographic analysis of the generated states was performed.
After achieving pulses of squeezed light, the next natural step was to perform simple EIT experiments using squeezed light. The first step was to send CW squeezing (no chopper) and see if the squeezing was preserved after the atomic interaction. Successful experiments of this kind could open possibilities for more elaborated schemes.

These experiments allowed us to address specific questions regarding the noise processes that degrade the quantum properties of light while propagating in the atomic cloud. A full understanding of these processes was crucial in order to estimate the possibilities of this system when being used as a quantum memory.

For the particular case of squeezing, and as pointed out by Hsu et al. [88], noise processes inherent to the atomic interaction should be taken into account to describe the degradation of squeezing. For example, the specific influence of the decoherence of the ground state in the quadrature noise of the signal after the interaction should be addressed [89]. Also, investigating the efficiency of mapping the light squeezing to a spin squeezing in the atomic cloud could help us understand another source of degradation [90].

These processes have been investigated theoretically by several authors, for example for the transmission of CW squeezed light [91], and also in the pulsed regime in cavity and single pass configurations [91, 92, 93]. As well, a detailed exploration of the noise sources for the transmission of squeezed light together with a proposal to implement quantum information delay using squeezed light has been published [89].

However, due to the fact that narrow-band squeezed light sources have only become recently available, none of the previous publications on slowdown and storage of
continuous-variable optical states reported satisfactory comparison of theoretical models with experimental data. As we will see in this part of the thesis, the experimental details will be the key for a successful understanding of these processes.

In this chapter I present a full study of the transmission of squeezed light through an EIT medium, first in the continuous-wave and later in pulsed regime. Our theoretical model identifies all main sources of degradation of squeezing, enabling us to predict the experimental result with more than 99 % quantum-mechanical fidelity with respect to the measured values.

For the purposes of this project, we performed a series of spectral and time-domain measurements of transmission of classical fields under EIT and fit the acquired data using a standard expression for the linear susceptibility of a 3-level atomic system. Subsequently we calculated the effect of propagation through an EIT medium upon the squeezed vacuum generated by our OPA, both in the continuous-wave (CW) and pulsed cases. We then performed two experimental tests: in the continuous regime, we measured the quadrature noise spectra of the transmitted squeezed vacuum and in the pulsed regime, we performed complete quantum-state reconstruction via time-domain homodyne tomography.

Our experiment is shown schematically in Fig. 5.1. The experiments were performed in atomic $^{87}\text{Rb}$ vapor at 65 °C, using a Λ energy level configuration formed by one of the hyperfine sublevels of the $5P_{1/2}$ state and two hyperfine sublevels of the $5S_{1/2}$ state (See inset Fig.5.1). The signal field is squeezed vacuum provided by our continuous-wave optical parametric amplifier (OPA), with a squeezing of approximately 3 dB. The control field is obtained from an additional diode laser phase locked, at 6.8 GHz to the main Ti:Sapphire laser to ensure stability of the two-photon detuning. In this setting, linear polarizations for the control and signal were used.
Figure 5.1: Experimental setup for the slowdown of squeezed light experiment. The inset shows the fields in the lambda configuration with signal field being squeezed vacuum and control field $\Omega_c$. The auxiliary measurements using coherent light are performed using a red seed beam in the OPA cavity.
5.1 Transmission of squeezed light through an EIT medium

The first step in our analysis is to investigate the transmission of CW squeezed light under EIT conditions. The first experiment in this direction was performed by Akamatsu et al. [10] with broadband squeezing produced in wave-guides.

An important observation should be noted here: good transmission of squeezed light under EIT conditions can only be achieved by using broadband EIT lines to ensure the transmission of both sidebands of squeezed light from the OPA. Nevertheless, a compromise has to be achieved because if the EIT line is too wide, we would not have any delay and hence the storage procedure will not work.

In order to achieve higher Rabi frequencies for the control field, we had to experiment with focused beams. Experimental measurements were first performed using collimated beams with a diameter of approximately 1 mm, but no squeezing transmission was observed. In the actual setup we focused both beams to a size of about 130 μm which resulted in good squeezing transmission. Careful mode-matching between the control and the signal was essential for the achieved results.

Another crucial fact should be pointed out here: the existing theory of propagation of squeezed light under EIT conditions assumes a symmetric EIT line. This is the first essential difference with the actual experiment. In our measurements, the EIT lines present an asymmetry with respect to symmetric sidebands of the squeezed light. At this point the mechanism responsible for this effect it is not clear yet. However we believe that is an addition of two main factors. First, the tight focusing of the beams could play a role since this asymmetry was not present in our previous EIT experiments performed with wider beams. Second, our EIT system is not a simple lambda but rather a combination of several of them, therefore optical pumping to different Zeeman sub-levels could also produce this effect.
Due to this experimental fact, better squeezed light transmission in the CW regime is achieved by applying a small detuning from the two-photon resonance (between 300-540 kHz). Another important factor is the dependence of the transmitted squeezing on the atomic density. This explains why better results are obtained by detuning the central frequency of the squeezed light by 630 MHz from the main D1 resonance since moving the signal from resonance is equivalent to changing the atomic density. We could then think that better experimental results could be achieved at lower temperatures, but again, another compromise have to be reached, since the delay depends strongly on the temperature.

To measure the transmission of squeezed light we performed CW homodyne tomography in the frequency domain of the signal after the EIT. Figure 5.2 shows the output of the spectrum analyzer for the squeezing after the EIT interaction. It could be seen that by analyzing the signal transmission after the EIT with a zero span mode at 500 kHz, the action of the control field resulted in an increase in both the transmitted squeezing and anti-squeezing as compared to the transmission without the control field present.

By using the spectrum analyzer with a finite span in the span mode we can recover the spectrum of frequencies at which squeezing can be observed. These frequencies roughly correspond to the measured EIT line-width obtained using a coherent state as a signal. Furthermore, as another clear evidence of the EIT for the squeezed light, the bandwidth of the squeezing after atomic interaction changes with the control power (see Fig. 5.3). Similar measurements were recently reported by Arikawa et al [94] however no theory was presented explaining the results.

In order to explain our measurements, we constructed a theory based on the transmission of the multi mode squeezing generated in the OPA through the atoms under EIT conditions. In this particular case, the effect of our asymmetric EIT lines must be taken into account.
Figure 5.2: Transmission of squeezed light with and without EIT conditions at a sideband frequency of 500 kHz.

Figure 5.3: Anti-squeezing bandwidth dependence as a function of the control power.
5.2 Transmission of classical light with EIT

The transmission of coherent light through rubidium atoms in the presence of a control field was measured in order to characterize the absorption and dispersion of the medium. As mentioned in the previous chapters, these properties are governed by the linear susceptibility which is modified to consider an ensemble of Doppler broadened atoms:

\[ \chi(\Delta_p, \delta_2) \propto \frac{i\gamma_{bo} + \delta_2}{|\Omega|^2 - (i\gamma_{bo} + \delta_2)(\Delta_p + iW)}. \]  

(5.1)

In the above equation, \( \Omega \) is the control Rabi frequency, \( \Delta_p \) is the one-photon detuning of the signal field, \( \delta_2 \) is the two-photon detuning, \( \gamma_{bo} \) is the ground state dephasing rate, and \( W \) is the width of the Doppler-broadened line. The susceptibility determines the amplitude transmissivity of the atomic ensemble as \( T = e^{i\chi L} \), where \( L \) is the medium length and \( k \) is the wavenumber.

As explained in chapter 3, Equation 5.1 assumes pure dephasing (decay of the off-diagonal element of the density matrix) to be the main mechanism of the ground state decoherence. A competing decoherence process, exchange of population between the two ground states, does not significantly affect the EIT transmission spectrum, as evidenced by our earlier investigations presented in chapter 3. Interestingly enough, this distinction will become important when we have to evaluate the extra-noise added by the EIT interaction.

The parameters in the susceptibility equation were determined from the following two measurements, each performed with EIT control powers between 3 and 7 mW. First, the intensity transmission spectrum of the EIT window was determined by scanning the frequency of the control field laser while keeping the power and frequency of the signal beam constant [See Fig. 5.4]. Second, transmission of 600 ns signal pulses (FWHM) was measured in the time domain, revealing information about the dispersion of the medium.
Figure 5.4: EIT line shape together with a fit using the susceptibility for Doppler-broadened atoms.

[see Fig. 5.12]. As expected from an EIT system, significant (up to a factor of a thousand) reduction of the group velocity of light has been observed.

The results were fitted with Eq. 5.1. The data for different control powers were fitted with the same set of parameters, except the control Rabi frequency, which was proportional to the square root of the control field intensity. Although treating the rubidium atom as a three-level system is a simplification, it turns out to be sufficient to obtain good agreement, as evidenced by Fig. 5.12.

5.3 Squeezed vacuum: CW measurements

CW squeezing was observed in the frequency domain by feeding the homodyne detector output to a spectrum analyzer. In this measurement, the noise reduction at a specific electronic frequency $\omega$ is a result from quadrature entanglement between the sidebands $\Omega \pm \omega$ of the optical signal field, where $\Omega$ is the optical frequency of the local oscillator.
Squeezing in the transmitted mode is retained only if both sidebands are transmitted through the EIT transparency window. Due to the asymmetry of our EIT lines [See Fig. 5.4], squeezed light transmission is improved by detuning the signal from the two-photon resonance with the control field by 540 kHz.

Figure 5.5 shows the frequency dependence of the quadrature noise as it exits the OPA (a) and after propagating through the atomic ensemble (b) with a 5 mW control field power. At each sideband frequency, the local oscillator phase was varied and the highest and lowest noise levels were recorded.

As we have seen in chapter 4, a theoretical prediction for the OPA noise spectrum below threshold is given by [95]

\[
V^\pm(\omega) = 1 \pm \eta \frac{4\sqrt{P/P_{th}}}{(\omega/\gamma)^2 + (1 + \sqrt{P/P_{th}})^2},
\]  

(5.2)

where \( \omega \) is the sideband frequency, \( \eta \) is the overall quantum efficiency, \( P/P_{th} \) is the ratio of the OPA pump and threshold powers, and \( \gamma \) is the cavity bandwidth. This equation was fit to the observed noise spectrum by varying these parameters.

In order to obtain a theoretical expression for the quadrature noise of the squeezed vacuum after the cell, we express the noise magnitude as \( V(\omega) = \langle \hat{q}_\phi(\omega) \hat{q}_\phi(-\omega) \rangle \), where

\[
\hat{q}_\phi(\omega) = \frac{1}{\sqrt{2}} [\hat{a}(\omega)e^{i\theta} + \hat{a}^\dagger(-\omega)e^{-i\theta}]
\]  

(5.3)

is the frequency-domain quadrature operator and \( \theta \) is the local oscillator phase.

Upon propagation through a medium with amplitude transmissivity \( T(\omega) \) (which can be estimated from our asymmetric susceptibility), the field operator undergoes transfor-
mation $\hat{a}'(\omega) = T(\omega)\hat{a}(\omega) + \sqrt{1 - |T(\omega)|^2}\hat{a}(\omega)$, where $\hat{a}(\omega)$ denotes vacuum. Substituting the field operator into the above expression for $V(\omega)$, we find for the highest and lowest quantum noise:

$$V^{\pm}(\omega) = \frac{T(\omega)^2 + T(-\omega)^2}{2} (V^+(\omega) + V^-(\omega))$$

$$+ T(\omega)T(-\omega) (\pm V^+(\omega) \mp V^-(\omega))$$

$$+ \frac{1 - T(\omega)^2}{4} + \frac{1 - T(-\omega)^2}{4} + V_{\text{noise}}(\omega).$$

Interestingly, this noise depends only on the magnitude of $T(\omega)$, but not the phase shift introduced by the EIT cell. This is because the asymmetric phase shift $\phi_{\pm}$ acquired by the field at sidebands $\pm \omega$ can be compensated by adjusting the local oscillator phase by $(\phi_+ + \phi_-)/2$.

The degradation of squeezing in transmission through EIT occurs not only due to the absorption but also because of the extra noise $V_{\text{noise}}(\omega)$ generated by the atoms in the cell [88]. As investigated theoretically by Peng et al. [89] and Hetet et al. [96], this noise is a consequence of the population exchange of the rubidium ground states. Interestingly, according to the theory presented in the above references, the dephasing of the ground states does not contribute to the extra noise.

We evaluated this population exchange noise independently by performing homodyne detection of the field emerging from the EIT cell in the absence of the input squeezed vacuum. The result of this measurement, with the shot noise subtracted, is shown in the inset to Fig. 5.5.
Figure 5.5: Squeezed and antisqueezed quadrature noise from the OPA (a) and after the EIT medium (b), along with the theoretical prediction of Eqs. (5.2,5.4). Inset: atomic extra noise measured in the absence of the signal field. The control field power is 5 mW.

The theoretical prediction of [96], can be expressed as:

$$V_{\text{noise}}(\omega) = (1 - T(\omega)) N_f,$$  \hspace{1cm} (5.5)

where:

$$N_f = \frac{4\gamma_c \Omega_c^2}{2\gamma_0 \Omega_c^2 + \omega^2 (2\Gamma + \gamma_0 - 3\gamma_c)}.$$  \hspace{1cm} (5.6)

In the last equation $\Gamma$ is the atomic decay, $\gamma_0$ is the dephasing rate and $\gamma_c$ is the population exchange rate, each of which was discussed in chapter 3.
In the inset of Fig. 5.5 we also depict the theoretical fit according to the previous equations. The only additional fitting parameter here is the population exchange rate, which determines the overall magnitude of the extra noise. Strictly speaking, the theory of Ref. [96] was developed for the case of zero one-photon detuning, i.e. symmetric EIT line. Using this approximation when constructing the fit still gives good results. We estimate the population exchange to account for less than 10 % of all decoherence processes, which justifies the assumption given in chapter 3.

Applying the extra noise theory along with Eqs. (5.2, 5.4), we have constructed a theoretical prediction for the spectrum of the transmitted squeezed vacuum [Fig. 5.5(b)]. No additional fitting parameters have been used. This is indeed very interesting, because using a simplified theory it was still possible to obtain very good agreement with the actual measurements.

5.4 Slowing down pulses of squeezed light

Now that the dynamics of the CW squeezed light EIT has been understood, we turned our attention to the pulsed regime. As it was seen in the last section, the asymmetric transmissions for twin side bands of the squeezing is the main mechanism responsible for squeezing losses. In the pulsed regime, this asymmetricity becomes even more crucial since the squeezing recovered using the time domain homodyne tomography relies on an integration over all the frequencies contained in the bandwidth of the pulse. In this case, asymmetric phase shifts for each twin mode due to the unequal dispersion should be taken into account in order to estimate the overall squeezing.

In order to generate lossless pulses of squeezed light we used an ultra-fast chopper wheel constructed from a hard drive to generate short pulses with pulse lengths of approximately 600 ns. As discussed in chapter 4 this technique combined with our time
domain tomography reconstruction can be used to obtain the properties of the pulse after the slowdown procedure.

5.4.1 Squeezed vacuum: pulsed measurements

In this part of the project, we studied the propagation of pulses of squeezed vacuum through the EIT medium by means of time-domain homodyne tomography.

The output of the homodyne detector was multiplied by the temporal mode function $W(t)$ (see Fig. 5.6) and integrated over time, producing a single sample of the field quadrature. The weight function was equal to the square root of the slowed down classical pulse intensity [See Fig. 5.6].

In this manner, 50,000 quadrature samples were obtained. The local oscillator phase values associated with each quadrature were determined using the method described in chapter 4. (Fig. 5.7 shows the reconstruction compared to the values measured with the spectrum analyzer.)

These data were used to reconstruct the density matrix and the Wigner function of
5.4.2 Slow down and squeezing dependence on the atomic density

After reconstructing squeezing in slowed down pulses, we made a brief experiment to investigate the conditions in which we could see maximum slowdown while still recovering fluctuations below shot noise. As indicated in [20] the group velocity reduction depends strongly on the atomic density. We repeated our measurements for different temperatures of the cell. As we can see in Fig. 5.11, we could obtain very good squeezing with little delay for low temperatures (40 °C) or very good delay (almost one third of the pulse) but almost no noise reduction for higher temperatures (65 °C).
Figure 5.9: Density matrix of a slowed down pulse of squeezed light.

Figure 5.10: Wigner function of a slowed down pulse of squeezed light.
Figure 5.11: Homodyne reconstruction of a slowdown pulse of squeezed light for different temperatures of the rubidium cell. Five points were taken for temperatures ranging from 40-65 °C. As it can be seen, the atomic density influences greatly the amount of squeezing that can be retrieved.

In a sense, this measurement indicates that the population exchange rate responsible for the extra noise is temperature dependent. Partially similar results have been already reported by D. Akamatsu et al. [97] albeit full reconstruction of the quantum state was not performed.

5.5 Theory for the pulsed measurements

In order to understand the results of the last two sections, we had to extend our theory from the CW case to the pulsed regime. First, from the fit of the absorption profile, we could estimate the dispersion properties for each on the squeezing sidebands and therefore, the phase difference induced for the sideband pairs. Once our susceptibility was reconstructed completely, we sent pulses of coherent light through the atomic media and measured the properties of its slowdown. In order to demonstrate that the reconstructed dispersion correctly described our system, we transformed a classical pulse of width 1us generated with our chopper using the estimated susceptibility and compared it with our
Figure 5.12: Slowdown of a classical pulse together with the estimation provided by our susceptibility

experimental measurements of the slowdown of pulses of coherent light. As can be seen, the theory resembles closely the properties of the slow down pulse, and the obtained delay for different control powers faithfully follows the measured values (see Figs. 5.12 and 5.13).

For the case of pulses of squeezed light, we followed a procedure similar to the CW case, by transforming the twin side band quadratures with an asymmetric susceptibility.

The experimental data on classical transmission of the EIT line, specifically the parameters of the OPA and of the atomic extra noise spectra, are sufficient to make theoretical predictions for the transmission of squeezed vacuum in the pulsed regime. Suppose the output of the OPA cavity is expressed by a time-dependent annihilation operator \( \hat{a}(t) \). After the chopper this operator will change into \( \hat{a}'(t) = \tau(t) \hat{a}(t) + \rho(t) \hat{v}(t) \) with \( \tau(t) \) representing the time-dependent transmission of the chopper, \( \hat{v}(t) \) the vacuum field operator, and \( \rho(t) = \sqrt{1 - \tau(t)^2} \).

In order to account for the propagation through the cell, we switch to the frequency
Figure 5.13: Delay obtained for different control powers together with the theoretical estimation domain:

\[
\bar{\alpha}'(\omega) = \bar{\tau}(\omega) * \bar{\alpha}(\omega) + \bar{\rho}(\omega) * \bar{\theta}(\omega), \tag{5.7}
\]

where \( \bar{\tau}(\omega) \) and \( \bar{\rho}(\omega) \) are, respectively, the Fourier images of the \( \tau(t) \) and \( \rho(t) \), and the asterisk denotes convolution. Absorption in the rubidium cell is then described by

\[
\bar{\alpha}''(\omega) = T(\omega) \bar{\alpha}'(\omega) + \sqrt{1 - T(\omega)^2 \bar{\theta}(\omega)}, \tag{5.8}
\]

where \( \bar{\theta}(\omega) \) is a vacuum contribution.
The measured (time-integrated) quadrature is given by

\[ \hat{Q}_\phi = \frac{1}{\sqrt{2}} \int_{-\infty}^{+\infty} W(t) \left[ \hat{a}''(t)e^{i\phi} + \hat{a}'''(t)e^{-i\phi} \right] dt \]

\( = \int_{-\infty}^{+\infty} \tilde{W}(-\omega) \hat{q}_\phi''(\omega) d\omega, \)

where the time-dependent quadrature operator is defined by Eq. 5.3. Knowing the transmission function \( T(\omega) \) from the classical measurement, we can express the variance of \( \hat{Q}_\phi \) after the rubidium atoms as

\[ V_\phi'' = \frac{1}{2} \int_{-\infty}^{+\infty} (W(\omega)T^*(\omega)) \ast \tilde{\tau}(\omega)^2 \]

\[ (V^+(\omega) + V^-(\omega) - 1) \, d\omega + \frac{1}{2} + V_{\text{noise}} \]

\[ + \frac{1}{2} \int_{-\infty}^{+\infty} F(\omega) (V^+(\omega) - V^-(\omega)) \cos(2\theta + \phi(\omega)) d\omega \]

with \( F(\omega) = \left[ [W^*(\omega)T(\omega)] \ast \tilde{\tau}(\omega) \right] \left[ [W(\omega)T(-\omega)] \ast \tilde{\tau}(\omega) \right] = F(\omega)e^{i\phi(\omega)}. \) The pulsed atomic extra noise is obtained from the measured frequency-domain extra noise by integrating

\[ V_{\text{noise}} = \int_{-\infty}^{+\infty} |W(\omega)|^2 V_{\text{noise}}(\omega) d\omega. \]

No fitting parameters are required for this calculation.

An important conclusion can be stated here, unlike the CW case, the phase shift imposed by the EIT line does lead to additional degradation of squeezing. Because the EIT line is asymmetric, \( T(-\omega) \neq T^*(\omega) \) and \( \phi(\omega) \) is not a constant, leading to mixing of
Figure 5.14: Maximum and minimum quadrature noise of pulsed squeezed vacuum transmitted through the EIT cell (theory and experiment).

Figure 5.14 shows the result of the calculation compared to the experimentally obtained values for several control powers. For all control powers, over 99% fidelity between the theoretically predicted and experimentally observed states was reached.

At this point, we can discuss the results of our pulsed squeezing experiment. I would like to address two points that had to be taken into account in order to obtain reasonable agreement with the measured values. First, akin to the measurement performed for the CW case, time domain homodyne tomography of the vacuum plus the control field, gives a measurement of the extra noise induced by the control. In this case, an integrated version over all the frequencies included in the bandwidth of the pulse. This is to be expected from the theory of the CW case. However, another point can be made here: since both lasers coupling the lambda system are phase locked in order to maintain a constant two-photon detuning, one could expect this phase lock noise to be translated into atomic noise. It is surprising that the effect of this noise is minimal since the reconstruction
is already good without considering it. Further experiments in this direction should be performed in order to understand this more clearly.

Another important interpretation can be stated here. As proved by our theoretical fits of the measured values, the mechanisms for losses in the squeezing can be readily identified: asymmetric EIT lines and extra-noise induced by the population exchange in the ground states.

The propagation of squeezing can be seen as a simultaneous coupling between the quantum fluctuations of the signal field and those of the atomic ground state coherence. In a sense, the atomic coherence gets squeezed and, for this particular experiment, almost by the same amount as the squeezed light propagating in the media after the transmission losses and the extra noise.

In conclusion, in this chapter we have performed a thorough theoretical and experimental investigation of squeezed light propagation through an EIT medium in both the CW and pulsed regime. Starting with a theoretical expression for the susceptibility of the EIT medium, we determined the degradation of squeezing in each spectral component of the squeezed vacuum. For the pulsed case, we calculated and measured the quadrature noise after propagation. We identified the main mechanisms leading to the degradation of squeezing: absorption in the EIT medium, asymmetry of the EIT line and extra-noise induced by the control field. Surprisingly, a very simple 3-level model is sufficient to fully explain our experimental results. This results opened the possibility to try an even more daring experiment: the storage of the squeezing. We dedicate the next chapter to this experiment.
5.6 Small summary

In this chapter we have analyzed the transmission of continuous-wave and pulsed squeezed vacuum through rubidium vapor under the conditions of electromagnetically induced transparency.

We have also developed a full theoretical treatment for a squeezed state of light propagating through temporal (pulsing) and spectral (EIT propagation) filters and detected using time and frequency domain homodyne tomography.

A model based on a three-level atom allowed us to evaluate linear losses and extra noise that degrade the nonclassical properties of the squeezed vacuum and eventually predict the quantum states of the transmitted light with a high precision.
Chapter 6

Quantum memory for squeezed light

We are finally arriving at the highlight of this thesis. After proving (and actually understanding) the transmission of squeezed light under EIT conditions, we proceed to the logical extension, storage. One interesting point to note is, as we have seen, that transferring the quantum fluctuations of the field will imply that, somehow, the atomic coherence will carry squeezing as well. At this point, we can think about these fluctuations as very fragile entities, and ask if it is possible to preserve them in a noisy environment such as thermal vapor.

Another point to bear in mind is that we are putting together, for the first time, three of the main tools recently developed by the quantum optics community: generation of narrow band squeezed light tuned to atomic transitions, EIT based light storage, and time domain homodyne detection and reconstruction [87]. In a sense, this experiment is the first proof of quantum memory for optical states which presents full reconstruction and comparison of the input and retrieved states.

By doing this experiment, we could also demonstrate the compatibility of the quantum repeater technology with the continuous-variable domain of quantum optics.

At the end of this project, we were able to show that the light in the retrieved mode retains quadrature squeezing, albeit degraded by absorption and atomic decoherence. In addition, we demonstrated that the optical phase of the retrieved squeezed vacuum faithfully reproduces that of the input.

In order to characterize the retrieved state, we used pulsed, time-domain homodyne tomography [64, 75, 98]. This method, allowing complete reconstruction of the quantum state of a specific spatiotemporal mode, has been applied to a variety of optical states
[78, 79, 99], but not yet to the characterization of quantum memories. By using homodyne tomography, we have determined the density matrices of the input and output states, evaluated the memory fidelity and found it to exceed a classical benchmark.

6.1 Improving the storage of light

After realizing the conditions in which better squeezed light transmission can be achieved (e.g. broad EIT lines, small beam sizes) we had to find a configuration where good storage efficiency could be achieved. To this end we used a configuration akin to the one presented in chapter 3 where the signal field was the seed beam in the mode of the OPA cavity (see Fig. 6.1). In this case, if we use a smaller beam size, the atoms will leave the interaction zone very quickly, so we had to find a compromise in the size that would allow broad enough EIT lines and low decoherence rates. Our first approach was to find conditions of the control field where the storage efficiency was as high as possible. To put things in perspective, the best efficiencies experimentally achieved to date are on the order of 40 per cent of the energy of the incoming field. To achieve this value it is necessary to use feedback techniques [29], that cannot be applied to the quantum case. After some work, we were able to achieve efficiencies of 25 per cent after storage times of 1 us (See Fig. 6.3).

In order to maximize the storage efficiency, maximum power of the control field had to be used. After the AOM alignment, about 10 mW could be put into the experiment. The first series of experiments were done in this configuration.

6.2 Experimental setup

For clarity, in this section I will review the experimental setup, including some aspects explained in the previous chapters.
The master laser, a Coherent MBR-110 Ti: Sapphire with a narrow spectral width (~40 kHz) and high long-term stability is tuned to the $^{87}\text{Rb} |5S_{1/2}, F = 1\rangle - |5S_{1/2}, F = 1\rangle$ transition at 795 nm. Its second harmonic, produced by means of a TekhnoScan frequency doubling cavity, pumps our squeezed light source, a degenerate optical parametric amplifier (OPA), with a power of 50 mW.

The bowtie cavity of the OPA consists of four mirrors, among which two are flat and two are concave with a curvature radii of 100 mm. The cavity is singly resonant for the generated 795 nm wavelength. We use one of the flat cavity mirrors, with a reflectivity of 93%, as the output coupler. Other cavity mirrors have over 99.9% reflectivity. The cavity exhibits a 460 MHz free spectral range with a resonance linewidth of approximately 6 MHz. The nonlinear element, a 20 mm long periodically poled KTiOPO$_4$ crystal, is placed at the beam waist between the curved mirrors.

The cavity is locked on resonance by means of an additional diode laser phase locked to the master oscillator at an offset of 3 OPA free spectral ranges (1.3 GHz). The cavity lock is implemented using the Pound-Drever-Hall method, with a beam modulated at 20 Hz phase lock.
MHz and counterpropagating relative to the generated non-classical mode. When the signal output of the OPA is measured with a homodyne detector, it exhibits, on average, a 3 dB noise reduction with respect to the shot noise in the squeezed quadrature and 6 dB excess noise in the antisqueezed quadrature on the 500 kHz sideband.

Our goal is to observe storage of squeezed light, so the OPA output has to be chopped into microsecond pulses. Because optical losses degrade squeezing, we avoid using electro- or acousto-optical modulators, and employ a fast mechanical chopper custom-made from a standard computer hard disk (see chapter 4). A 50 μm slit (of cylindrical shape with a diameter of 9 mm) is mounted to the rim of the disk. The disk is then positioned so that when rotating, the slit passes through the OPA output beam focused to a 25 μm size. The disk control is programmed to a rotation frequency of 250 Hz. For most of the rotation period the squeezed vacuum passes through the chopper wheel unobstructed, which allows us to get information about the phase of the squeezed vacuum. When the slit material enters the beam it interrupts the light for about 70 μs, generates a 600 ns (FWHM) squeezed vacuum pulse and blocks the light for another 70 μs.

Originally a 10 mW EIT control field is provided by a separate diode laser phase locked to the master oscillator. The frequency difference between the signal and control fields was set to optimize, on one hand, the classical light storage efficiency, and on the other hand, transmission of squeezed light through the cell under EIT conditions. As we have seen in chapter 5, we found these requirements to be best fulfilled when the frequencies of both fields are red detuned from the center of the Doppler-broadened atomic line by \( \Delta_{1-\text{photon}} = 530 \) MHz. In addition, the transmission of squeezing is improved when the fields are two-photon detuned from the 6834.683 MHz hyperfine splitting resonance by about \( \Delta_{2-\text{photon}} = +0.4 \) MHz. This is because the EIT resonance line is asymmetric, so such detuning permits better accommodation of the entire bandwidth of pulsed squeezed vacuum into the transparency window.
The experiments were performed in atomic $^{87}$Rb vapor at 65 °C, using a Λ energy level configuration formed by one of the hyperfine sublevels of the $5P_{1/2}$ state and two hyperfine sublevels of the $5S_{1/2}$ state (Fig. 6.2, inset). The rubidium vapor cell used for storage has 5 Torr of neon buffer gas and is contained in a magnetically shielded oven (See Appendix C). The control and signal fields were orthogonally linearly polarized, which allows their combination and separation at the oven entrance and exit. The spatial modes of the control and signal fields were carefully matched to each other.

The beam width of the fields inside the cell was 130 μm. This geometry was chosen as a compromise between two factors. On one hand, a narrower beam increases the flight-through decoherence [1] of the atomic ground levels, which reduces the memory lifetime. On the other hand, decoherence of the atomic dark state leads to Raman scattering of the control field into the signal mode. In quadrature measurements of the control field, this emission manifests itself as low-frequency noise [88] which degrades squeezing in transmission through the EIT cell. This noise appeared to increase with the beam width.

The control field was on for most of the 4 ms experimental cycle. It was turned off, by means of an acousto-optical modulator, at time $t = 1.95 \mu s$ when the majority of the signal pulse has entered the cell, and turned back on after the 1 μs storage period. After an additional 4 μs, when the stored signal has been fully retrieved, the control field was turned off again briefly to obtain a clean sample of the vacuum state noise. The intensity waveform associated with the storage and retrieval of a classical laser pulse is given by the solid curve in Fig. 6.3.

Upon separation from the control field, the signal was subjected to homodyne detection using a local oscillator (LO) derived from the master oscillator. The LO had constant intensity, but its phase was varied, with a period of 2.5 s, by means of a piezoelectric transducer. The homodyne detector uses two Hamamatsu S3883 photodiodes of 94% quantum efficiency and offers an over 6 MHz bandwidth. The signal from the ho-
Figure 6.2: Experimental setup for the first set of experiments of squeezing storage. The inset shows the atomic level configuration.

modyne detector, corresponding to the quantum quadrature noise of the detected field, was fed to a digital oscilloscope and a spectrum analyzer for simultaneous time- and frequency-domain measurements (See chapter 4 for details).

6.3 Storage of squeezing results I

The last step of our procedure was to perform storage of the pulses of squeezed light.

Figure 6.3 shows a typical storage experiment where approximately 20% of the original pulse is recovered while still having a squeezed light transmission of about 1 dB in the CW regime. The power of the control field was 10 mW.

It is essential to notice that the presence of the control field caused a shift in the vacuum level measured by the homodyne detector (see Fig. 6.3). This can be explained by considering the Raman scattering with the same frequency as the local oscillator created due to population exchange decoherence. This agrees with the prediction by Tsu et al
[88], but poses the question - what is the true level which needs to be overcome in order to demonstrate squeezing after the storage procedure? We designed our acquisition to be able to measure both vacuum levels in the same experimental run. After comparison, we could claim this extra noise results in an offset of the vacuum level by an amount of 0.1-0.3 dB depending on the strength of the control field.

The dot-dashed curve in Fig. 6.3 shows the pointwise variance of 50,000 oscilloscope traces registering the homodyne detector output during the storage/retrieval procedure. When the control field is off and the signal pulse has terminated (2.4 $\mu$s < $t$ < 2.9 $\mu$s), the signal is in the vacuum state, so the detector outputs shot noise. Prior to the beginning of the signal pulse ($t$ < 1.4 $\mu$s), and upon completion of the storage procedure ($t$ >3.5 $\mu$s), when the control field is on, the shot noise is elevated by about 0.1 dB due to the Raman scattering mentioned above. When the front of the squeezed vacuum pulse is transmitted through the cell (1.4 $\mu$s < $t$ <2 $\mu$s), the phase-averaged quadrature noise is significantly higher than the shot noise level. When the control field is turned back on after the storage period (3 $\mu$s < $t$ <3.5 $\mu$s), the noise level increases again due to the retrieval of the stored squeezed state.

6.3.1 Quantum state reconstruction

We characterized the states of the electromagnetic field modes corresponding to the input and retrieved pulses as we did in chapters 4 and 5. We defined the temporal amplitude shape $f(t)$ of these modes by the square root of the associated classical intensity waveforms (the mode shape of the retrieved state is shown by the dotted line in Fig. 6.3). We used a continuous local oscillator, and post-processed the homodyne photocurrent by multiplying it by $f(t)$ (see Fig. 6.3) and subsequently integrating over time [86]. In this fashion, we processed the photocurrent from 50,000 retrieved pulses and obtained a set of values that are proportional to the field quadrature noise samples of the retrieved state.
Figure 6.3: Time-dependent intensity of the classical input pulse without storage (blue dashed curve), and with storage (yellow solid curve). The $y$-axis units are arbitrary, but the intensity ratio between the fields is preserved. The green dot-dashed line shows time-domain pointwise variance of quantum quadrature noise as measured by the homodyne detector, normalized to the shot-noise level. The red dotted line is the temporal mode $f(t)$ in which the retrieved state is reconstructed.
The proportionality coefficient was determined from the variance of 50,000 quadrature measurements of the vacuum state acquired in the same temporal mode.

Quantum state reconstruction required knowledge of the local oscillator phase values associated with each quadrature sample. This was the purpose of the frequency-domain registration of the homodyne detector output discussed in chapter 4. The spectrum analyzer acquired data continuously in the zero-span mode at a 500 kHz sideband with a resolution bandwidth of 30 kHz and a sweep time of 2.5 seconds. Because this acquisition was relatively slow, and because the chopper blocks the signal for less than 2% of its rotation period, the chopper had very little effect on the acquired signal, which corresponded to the phase-dependent quadrature noise of the squeezed vacuum generated by the OPA. The local oscillator phase was determined by this signal.

Figure 6.4 shows the phase dependent quadrature noise reconstructed with our time domain technique, together with the Maximum Likelihood reconstruction for two separate analyses using the vacuum level considering the extra noise and the vacuum level without the presence of the control field.

As can be seen in both reconstructions, noise reduction below the shot noise level was obtained. In the case of vacuum plus extra-noise, squeezing of 0.4 dB was achieved, whereas in the case of just vacuum, a maximum squeezing of 0.05 dB was observed.

At this point quantifying the error in our measurements is of utmost importance, especially in the case of the squeezing level compared to the vacuum without control field. In principle we can assign a statistical error from the Maximum Likelihood reconstruction. We estimate this error to be in the order of 0.05 dB. This already proves that we retrieved squeezing in the case of vacuum with extra-noise. To prove the same for the second case we analyze the variance of the data where minimum squeezing can be found (only 3000 points instead of the original 50000) and we found a variance of 0.479 +/- .013 compared to the normalized vacuum variance of .5. This second analysis proves that we
Figure 6.4: Binned quadrature noise of the retrieved squeezed state after storage and maximum likelihood reconstruction of the same state for: vacuum plus extra noise (upper plots) and only vacuum (lower plots). 0.05 dB of squeezing were found in these preliminary experiments when considering only vacuum.
also recovered squeezing compared to the vacuum with no control field present, albeit very little.

6.4 Storage of squeezing results II

Once we have exhausted the possibilities of our set-up, we tried different configurations in order to improve the retrieved squeezing. We tried configurations in which the storage efficiency was not that high, but since the control field was also less powerful, less extra noise was generated. This, added to a different configuration for the detunings that allowed for better squeezed light transmission, finally allowed us to measure squeezing after retrieving that was beyond the error limit of our reconstruction.

Having changed to a 2.5 mW EIT control field our original results were improved. We found the best results could be obtained when the frequencies of both fields are red detuned from the center of the Doppler-broadened atomic line by $\Delta_{1,\text{photon}} = 630 \text{ MHz}$. In addition, the transmission of squeezing is improved when the fields are two-photon detuned from the 6834.68 MHz hyperfine splitting resonance by about $\Delta_{2,\text{photon}} = +0.54 \text{ MHz}$. We repeated the procedure mentioned in the last section using the new field configurations.

We applied the iterative likelihood-maximization algorithm [87] to reconstruct the input and retrieved states from 50,000 quadrature-phase pairs obtained in this manner [Fig. 6.5 (a)]. This procedure yielded the density matrices in the Fock basis [Fig. 6.5 (b)] from which the Wigner function of the state in question was calculated [Fig. 6.5 (c)] as well as the phase-dependent behavior of the quadrature noise [Fig. 6.5 (d)].

Both the input and retrieved states were, to a large accuracy, squeezed thermal states, and both exhibited the minimum quadrature noise at the level below the standard quantum limit. The squeezing for the input state was 1.23 dB and for the retrieved state it was
Figure 6.5: Quantum state of the input (left column) and retrieved (right column) states. 50,000 samples of phase-dependent quadrature noise (a), maximum-likelihood reconstruction of the density matrices (absolute values, b), Wigner functions (c) and quadrature noise variances (d) are displayed. In (d), the solid lines are calculated from the density matrices while the points with error bars show variances of binned quadrature values, as discussed in the text.
0.28 dB. The corresponding antisqueezing levels amounted to 3.74 dB and 1.77 dB, respectively. The uncertainty of these values was estimated as described in Ref. [87] as 0.05 dB. We could see that the time-domain measurement of the input state already showed significant degradation of squeezing as compared to the frequency domain (See chapter 4). This happened because (1) time-domain reconstruction includes low-frequency sidebands, where the squeezing is reduced and additional technical noise is present and (2) in the shoulders of the pulse, the slit edges clip the beam. The squeezing in the retrieved state was further reduced compared to the input. Again, two main factors are at play here. First, the storage efficiency can be estimated from the classical data to be about 15%. The second degrading factor is the Raman scattering of the control field.

We also verified the presence of squeezing in the retrieved mode by direct calculation of the phase-dependent quadrature variance. To this end, we binned the acquired quadrature values into 16 groups of 3,125 points according to their phases and evaluated the mean square variance of each group [Fig. 6.5 (d), right column]. Two groups exhibited variance below the standard quantum limit, with the difference about twice as high as the margin of error. The latter was evaluated as the statistical error of estimating the width $\sigma$ of a Gaussian distribution from $N$ samples, and equals $\sigma\sqrt{2/N}$ [100].

We performed two additional tests in order to verify our interpretation of this experiment as a demonstration of quantum memory for squeezed light. First, instead of storing the squeezed state, we stored vacuum and analyzed the quantum state in the retrieved optical mode. As expected, we detected a state with almost phase-independent quadrature noise, elevated by 0.1 dB with respect to the vacuum due to Raman scattering of the control field. Second, we stored the squeezed state, but did not turn the control field back on for retrieval. We acquired and analyzed the optical state in the same spatiotemporal mode as before and found, as expected, that, without the control field, this state is almost exactly vacuum.
6.4.1 How to check the performance of a quantum memory?

Since quantum memory with complete characterization was not achieved before, criteria judging the quality of the memory performance did not yet exist. Our first approach to quantify the performance of our memory setup was by means of the fidelity, which, for the input and output quantum states $\hat{\rho}_{in}$ and $\hat{\rho}_{retr}$ is defined as:

$$F = \text{Tr}[(\hat{\rho}_{in}^{1/2} \hat{\rho}_{retr} \hat{\rho}_{in}^{1/2})^{1/2}]^2.$$  (6.1)

Comparing the input and retrieved states, we determine $F = 0.97$. This value is significantly higher than the "classical" fidelity of 0.73, defined by adding two units of the vacuum noise to the input state $|101\rangle$. The measured fidelity is also higher than 0.82, which would be obtained in a "memory" procedure where the input state is replaced by the vacuum. This indeed looks very promising, but a direct comparison of the incoming and retrieved squeezing would tell us differently. I will elaborate more on this point at the end of the next section.

6.4.2 Important remarks

Once these experiments were repeated several times to confirm our findings, it was time to make some important comments.

In this particular set of experiments, we have demonstrated a proof of principle experiment of the storage and retrieval of pulses of squeezed light using EIT and for the first time, we achieved a characterization of the quantum state after the storage procedure.

We have characterized our state considering two different vacuum levels. First considering the extra noise inherently caused by the experiment and later against what can be considered the real vacuum level. In both cases squeezing has been demonstrated, therefore proving the nonclassicality of the retrieved states.
While this work was being prepared for publication, we became aware of the work [102], where storage and retrieval of squeezed vacuum in a cold atom ensemble was been demonstrated, albeit without tomographic reconstruction.

6.4.3 Phase evolution

As an additional measurement that permits us to make a little bit of sense of the presented results, we used our setup to study the evolution of the atomic coherence during the storage time with respect to that of the laser fields.

Because the signal and control field are not in exact two-photon resonance with respect to the atomic hyperfine splitting, the atomic and optical systems evolve with different frequencies during the storage time. As a result, the retrieved state's phase, measured with respect to the local oscillator, is different from the input by

\[ \phi = 2\pi \Delta_{\text{2-photon}} t_{\text{storage}}, \]  

so we expected a linear behavior in the retrieved phase if either the two-photon detuning or the storage time were changed. We verified this dependence, reconstructing the retrieved state at different values of these parameters. This reconstruction yielded phase values modulo \( \pi \), so in order to fit the theoretical prediction (6.2) we added multiples of \( \pi \) to each point. The result of this procedure is shown in Fig. 6.6 and exhibits good agreement with the theory.

These additional results allowed us to have more certainty about the phase we were using to reconstruct the retrieved states, since we could in principle reproduce its evolution.
Figure 6.6: Phase dependence of the reconstructed squeezed state with respect to the local oscillator as a function of the storage time with a constant $\Delta_{\text{2-photon}} = 384$ kHz (a) and as a function of the two-photon detuning with a constant $\tau_{\text{storage}} = 1$ µs (b). The solid lines correspond to Eq. (6.2)

6.5 Storage of squeezing results III

After sending the previous results for publication, one of the referees suggested that we perform extra measurements that showed in a clear way the reduction in the variance of the noise. For this particular experiment several changes were made to the original set-up, since further investigations on the behavior of the memory were already ongoing. In the original experiment the beams were focused inside the rubidium cell and the classical memory time was short, we then switched to a different configuration were the classical memory time was larger but it was still possible to retrieve squeezing. With this configuration, more control power is needed in order to achieve the same Rabi frequencies. The control field power for this experiment was 5 mW. This time we acquired 100,000 quadrature points to achieve better statistics.

In Fig. 6.7 is shown the analog to Fig. 6.3 for the new experimental configuration.

We reconstructed the state in the same way as we did before.

We applied the iterative likelihood-maximization algorithm to reconstruct the input and retrieved states from 100,000 quadrature-phase pairs obtained in this manner [Fig. 6.8 (a)]. This procedure yielded the density matrices in the Fock basis [Fig. 6.8 (b)] from
Figure 6.7: Our best storage results: Time-dependent intensity of the classical input pulse without storage (a), and with storage (b). The y-axis units are arbitrary, but the intensity ratio between the fields is preserved. Line (c) shows time-domain pointwise variances of the phase-averaged homodyne detector photocurrent, in the units of SNL. Curve (d) is the temporal mode $f(t)$ in which the retrieved state is reconstructed.
which the Wigner function of the state in question was calculated [Fig. 6.8 (c)] as well as the phase-dependent behavior of the quadrature noise [Fig. 6.8 (d)].

Both the input and retrieved states still looked like squeezed thermal states, and both exhibited the minimum quadrature noise at the level below the standard quantum limit. The squeezing for the input state was 1.86 dB (corresponding to 0.65 SNL on a linear scale) and for the retrieved state it was 0.21 dB (0.95 SNL). The corresponding anti-squeezing levels amounted to 5.38 dB (3.45 SNL) and 1.32 dB (1.36 SNL), respectively. The uncertainty of these values was estimated for this new experiment to be 0.04 dB.

We again verified the presence of squeezing in the retrieved mode by direct calculation of the phase-dependent quadrature variance. To this end, we partitioned the acquired quadrature set into 5 bins of \( N = 20,000 \) points according to phases and evaluated the mean square variance within each bin [Fig. 6.11 (d)]. One of the bins exhibited a variance of \((0.187 \pm 0.043) \) dB below the SNL. The margin of error is evaluated here as was done in the previous section.

As before, we performed two additional tests in order to test our interpretation of this experiment as a demonstration of quantum memory for squeezed light.

We quantified the performance of our memory setup again in terms of fidelity. This time we determined \( F = 0.89 \) compared to the “classical” fidelity of 0.74. This obtained fidelity value is high due to a large vacuum component in both \( \hat{\rho}_{in} \) and \( \hat{\rho}_{retr} \).

As discussed before, an alternative figure of merit for our interface is needed that gives us a better idea about the performance of the memory. This could be done by evaluating the degrees of nonclassicality of both states in terms of their entanglement potentials [103], equaling 0.309 for \( \hat{\rho}_{in} \) and 0.036 for \( \hat{\rho}_{retr} \). We see that only a small fraction of the input state’s nonclassicality is transferred to the retrieved pulse. In other words, the quantum memory works but not that well.

The results of this experimental run allowed us to reduce the error in the estimation
Figure 6.8: Quantum state of the input (left column) and retrieved (right column) states for the experiment with better statistics. Raw samples of phase-dependent quadrature noise (a), maximum-likelihood reconstruction of the density matrices in the Fock basis (absolute values, b), Wigner functions (c) and quadrature noise variances (d) are displayed. In (d), the solid lines are calculated from the density matrices while the points with error bars show variances of binned quadrature values, as discussed in the text.
of the variances of the binned quadrature values (particularly important to better show noise reduction in the squeezing region). As can be seen by comparing Fig. 6.5 (d) and 6.8 (d), the statistical error was reduced by a factor of 2. This however comes with the price of reconstructing slightly less squeezing.

6.6 Small summary

In this chapter we presented experiments in which we produced a 600 ns pulse of 1.86 dB squeezed vacuum at 795 nm in an optical parametric amplifier and stored it in a rubidium vapor cell for 1 us using electromagnetically induced transparency. The recovered pulse, analyzed using time-domain homodyne tomography, showed up to $0.21 \pm 0.04$ dB of squeezing. We identified the factors leading to the degradation of squeezing and also investigated the phase evolution of the atomic coherence during the storage interval.

6.7 Conclusions

After presenting all the results in the last chapters, it is time to put everything that was achieved in perspective. We have been able to produce results which have had a noticeable impact in the quantum optics community. First, we published a detailed experimental analysis of decoherence processes for the storage of light [1, 2]. In this work we proved dephasing to be the dominant decoherence mechanism in this type of experiments, therefore improving the general idea conceived by Lee and Javan [19, 44]. We have also reported our results on adiabatic transfer of optical frequencies using multi-lambda schemes [3]. This work represented a step further with respect to the original work of Zibrov [57] and also defined a path of research that can lead to routing of quantum information (See chapter 3).

We also implemented a source of squeezed vacuum tuned to Rubidium transitions [4].
The difficulty of this work is shown by the fact that only two more sources with the same characteristics have been reported [72, 73] (See chapter 4).

Moreover, we have interacted our light source with rubidium atoms in a thermal vapor under EIT conditions. These experiments represent the major results of this thesis, since we were able to observed both slowdown [5] and storage of pulses of squeezed light [6] (see chapters 5 and 6).

These results have only been matched recently by the work of Kozuma [?]. In addition, after the EIT interaction we performed a full reconstruction of the retrieved state by using time domain homodyne tomography. This is what make our results unique; we have been the only group so far to put together three separated techniques at the same time: non-classical light generation, EIT storage, and full state reconstruction using homodyne detection, therefore constructing a universal toolbox for quantum optical memory.

Regarding the quantum memory, in chapter 6 we have reviewed that is possible to preserve the quantum character of the light (in this particular case the noise reduction present in the squeezed light). At this point, it is fair to ask the question of what criteria must be met to justify our claim of an operational quantum memory. Since answering this question requires an analysis of the maximum average fidelity achievable when the states are stored by a classical channel in order to create a benchmark which has to be surpassed by the quantum memory in order to outperform any classical strategy (akin to the analysis presented in [104] for coherent states), it falls outside of the scope of this thesis. Nevertheless, I will refer to a recently published work [105] addressing a quantum benchmark for storage of squeezed light. In this work, the authors establish a benchmark of 81.5 % fidelity in order to outperform any classical strategy. Moreover, the authors consider the work in this thesis and assign a fidelity value of 89 %, neatly surpassing their benchmark.

The last point to address is the importance of these experiments regarding a possible
quantum process tomography (QPT). If such a process is ever to be performed on a quantum memory, the results presented in this thesis will be necessary in order to corroborate the results of such experiment.

After this small discussion, in the last chapter I will describe the present status of the experiment and discuss the possible paths it could go in future years.
Chapter 7

Outlook

After finally achieving the main goal of the project (which in turn comes all the way from 2003 when the group was still in Germany) it is time to look into further developments. In this chapter, I will address the present state of the project and future perspectives for its evolution.

7.1 Present state of the experiment

After proving the storage of squeezed light, we followed the logic approach and started analyzing the behavior of the quantum memory against the storage time. As we mentioned before, quantum memories where experimental data could be reliably compared with theory only became available in the last year. Given this situation, there is a lack of literature describing their behavior. To address this, we followed the theory by Hetet et al. [96] that was also used to construct the theory of the propagation of squeezed light under EIT conditions. In this paper, even though the analysis of the memory includes atomic noise sources, one of the main conclusions that the authors state is that the exponential memory decay only depends on the decoherence rate, but not on the state stored.

This is the same result of the dark state polariton theory by Lukin and Fleischhauer [22] despite the fact that this analysis does not include noise sources. In other words, we should expect a decay of the quantum memory similar to the one of a memory where classical pulses are employed. This is a not a surprising result: once the atomic coherence is squeezed, the main decoherence mechanisms that affect the classical storage are still
Figure 7.1: Life time of the quantum memory for squeezed light. The results are in a linear scale to be able to fit an exponential decay.

the main influence in the experiment.

From a naive experimentalist perspective, one should think that more effects should probably be taken in account to have a complete picture of the evolution of the squeezing during the atomic-storage phase.

We have to say here as well that performing this experiment was not easy. As mentioned before, there are too many parts of the experiment that have to work at the same time in order to produce trustworthy results. For these measurements, we had to take enough experimental data to make an accurate estimation for each storage time value. Even though the experiments are reproducible, not all runs provided useful data. We have made two complete runs of this characterization and both gave the same results. Figure 7.1. presents the results of this experiment for storages times between 700 ns and 1.7 μs.

As we can see, the memory lifetime is only 1.3 μs before the squeezing effectively disappears. From this result immediately follows the question: how do these results com-
Figure 7.2: Classical pulses to evaluate the lifetime of the classical memory. The classical memory lifetime can be clearly seen larger than the squeezed light memory.

pare to the same experiment using classical pulses? According to theory, the decoherence mechanisms should be the same for both experiments.

7.1.1 Comparing quantum and classical memories

To answer this question, we proceed to analyze the behavior of the memory under the same physical conditions for classical pulses of light with a procedure akin to the one used in chapter 3. The results are plotted in Fig. 7.2.

Our measurements show a classical memory lifetime in the order of 50 μs, i.e. more than one order of magnitude more than the quantum counterpart. This discrepancy of course raised questions about which different mechanism are occurring that cause the losing of quantum information.

Our first approach to explain the discrepancy was the effect of the two photon detuning. As we briefly discussed in chapter 6, the presence of a two photon detuning might have an influence on the storage and retrieval since it makes the atomic system evolve differently when compared to the laser system. As mentioned in chapter 6 the storage of
squeezing experiments were performed with a two photon detuning of 500 kHz. In this stage, several experimental runs were made with different two photon detunings but still the classical memory lifetime did not change (See Fig. 7.3).

7.1.2 Memory for coherent states

In order to answer this question, our approach was to repeat the experiment with a different kind of optical states: in this case we checked the memory for highly attenuated coherent states (See chapter 4). This is an experiment similar to the one performed to obtain the classical pulses used to evaluate the noise in the pulsed squeezing experiments (See chapters 4 and 5). For this particular case the classical beam in the cavity mode is highly attenuated to reach an average photon number close to one. In order to fully reconstruct the state, we performed time domain homodyne tomography as in the previous
Figure 7.4: Quadrature noise of a reconstructed coherent state.

case. For this experiment, the phase for each quadrature measurement is determined using a second oscilloscope instead of the spectrum analyzer that we used for the squeezing experiments. Figure 7.4 shows the reconstructions of a pulsed coherent state. Reconstructing the $\alpha$ parameter of the coherent state using maximum likelihood reconstruction allow us to estimate the mean number of photons. This number is the complete analogy to the energy of the pulse we measured in chapter 3. By monitoring how the average photon number changes when we change the storage time we can estimate the behavior of the memory.

Our idea was to investigated a case that is close to the quantum regime but still classical in nature, and see if the measurement of the life time is more related to the squeezed light memory or the complete classical regime. These experiments are still ongoing and at the moment we are improving the reconstruction of the pulsed coherent states.
These results might give us answers about the mechanisms that have not been identified in order to better understand quantum memories. Currently, we are researching such mechanisms while trying to develop a theory that explains this behavior. Other experiments with results relevant to our experiment have also been recently performed by the Pinard group in France [106] reporting values similar to that of the classical memory lifetime.

7.1.3 Near future experiments

Given that there is still research to be done in order to understand the behavior of the quantum memory, the next series of experiments will be aimed at investigating several aspects that could influence the outcome of the memory. Since we are able to explain what happens to the pulses of squeezed light when they propagate under EIT conditions, we have to focus our attention on two aspects: First, what happens to the squeezing when we perform the mapping to the atomic coherence together with the reverse mapping to light again? Also we have to investigate in more detail how the phase of the spin squeezing evolves and how this changes the phase of the retrieved quantum state with respect to the phase of the original squeezed pulse.

The first problem has been recently addressed theoretically by Hetet et al. [96]. Here the authors assume the creation of an atomic coherence dependant on a transfer function quantifying the losses due to the finite EIT bandwidth, and the finite length of the cell.

This result is of prime importance in the overall conclusion of the theory presented in [96] since it allows a complete efficiency for the light to atoms mapping. It assumes a symmetric susceptibility, which as we have seen in chapter 5, is not the case for the actual experiments. Perhaps the inclusion of this asymmetry allow us to better explain the results of the squeezed light memory.

Another possible experiment related to the ongoing storage of attenuated coherent
states will be the analysis of the behavior of the memory lifetime regarding the photon number of the input coherent state. This could be a key element to explain the short memory lifetime in the case of squeezing.

7.2 Outlook of the project

7.2.1 Future projects

After a successful conclusion to our analysis of the quantum memory for squeezed light, we plan to modify our OPA to set up the next generation of quantum memory experiments: the EIT interaction of entangled modes. This experimental realization is important for quantum communications since it will mean the possibility to operate in one of the entangled modes (slowdown, storage) without losing the entanglement with the other mode. In this thesis, we have sent both entangled modes into the EIT interaction region, but separated interaction has not yet been extensively investigated.

Recently, there have been preliminary attempts to achieve these goals. Broadbent et al. showed the preservation of energy entanglement in a slow light medium by sending broadband bi-photons through different paths, one of them containing a Rubidium cell [107], and Hetet et al. showed the preservation of entanglement after one of the modes was sent through a EIT delay medium [108]. For this particular experiment, the entanglement was created by splitting the output of a squeezed light source.

In order to create our entangled source, we will take advantage of our OPA design. Since we know squeezing can be viewed as Einstein-Podolsky-Rosen entanglement between sidebands $\omega_{\text{pump}}/2 \pm \Delta \omega$, we can cleverly choose the length of the OPA cavity to generate entangled sidebands well separated in frequency.

For the particular configuration of our OPA for the squeezed light experiment, the cavity is presently locked to the main frequency $\omega_0$ of the Ti:Sapphire laser. If we now
lock it detuned from this frequency by a FSR/2 = 230 MHz, twin side-bands at \( \omega_\pm = \omega_0 \pm 230 \text{ MHz} \) will possess quadrature phase entanglement as suggested by Reid [109] (See. Fig. 7.5).

Since the signal and idler modes emerging from the OPA are separated in frequency by 1 FSR, we need to design a filter cavity that will allow for spatial separation of the modes.

The length of this filtering cavity has to be stabilized to maintain resonance with the \( \omega_+ \) mode and total reflection for the \( \omega_- \) mode. This will require the implementation of a Pound-Drever-Hall locking scheme.

After the filtering cavity the signal and idler modes will propagate in different directions and each will be subjected to homodyne detection. If the two modes can be efficiently separated, using a double homodyne detection experiment we can prove that the entanglement is preserved by measuring the sum and difference of the photocurrents.
of each detector and then using a Fourier analysis to look for correlations (See Fig. 7.6 for schematics of the proposed setup).

If we can prove that entanglement is preserved after separation, this will be the basis for future experiments including the study of entanglement after the EIT delay of one of the modes, as well as the storage of one of the entangled modes.

7.3 Outlook of the quantum memory field

Over the last two decades, many discoveries in have been made in quantum optics. Several groups embarked on a journey to realize how these new findings can find practical applications. These new quantum discoveries have indeed opened the possibility for real-
izations like quantum computing, quantum communication and quantum cryptography.

Over the last 6 years, while doing research in quantum devices using semiconductors and later working in novel ways to interact non-classical light with atomic ensembles, I have developed a clear idea of which fundamental problems could be addressed in the near future by the quantum memory community. In other words, I will give my two cents on future experiments I would like to see done by myself or some other friendly group.

I can divide these directions in five main categories, some I believe are the result from weighting my laboratory experience and some are result of observing the directions in which the field has evolved.

1. Interaction of quantum light with atoms using electromagnetically induced transparency

   a) Generation of non-classical light tuned to atomic transitions using optical parametric amplifiers (OPA’s)

   It has been proven than we can effectively produce squeezed states of light using OPA’s with a degree of squeezing higher than 9dB at certain wavelengths [110]. Nevertheless, once we try to design our sources with a wavelength tuned to atomic transitions, our ability to reach squeezing decreases dramatically [72, 73, 4].

   I believe it is still necessary to tackle the fundamental problems creating these differences. Are they just technical problems, or is there a fundamental limitation on the the non-linearity that can be achieved at those wavelengths? Can we find new materials with higher non-linearities?

   b) EIT interaction: storage and slowdown
As a part of my PhD work, I have been studying the slowdown and storage of squeezed light using EIT [6]. Since these are state of the art experiments, new questions are arising as old ones are answered. Particulary interesting problems relate to the nature of the decoherence mechanisms during the storage process since, in principle, they seem to be different than what we would normally expect for a classical channel [39, 40]. Can we achieve better storage times just by improving our techniques or is it a more fundamental matter of the fragility of the stored quantum state that can not be overcome?

c) Storage of entanglement

Another direction is the creation of continuous-variable entanglement using OPA’s (as discussed in chapter 7). This will allow for possible tests of preservation of entanglement after one or both entangled channels have been subjected to either slowdown or storage in atomic media. This will require simultaneous homodyne tomography in both arms of the experiment, something which has not yet been achieved.

2. Coherent control and adiabatic interactions in atomic media using non-classical light

Having non-classical light tunable to atomic transitions opens the possibility to realize coherent transfer of quantum states between atomic levels [59] or optical modes [3]. This transfer would have a variety of applications in quantum information and communication. How well can we implement such transfer and what are the main technical challenges?

3. Quantum dot single photon sources and coherent control in semicon-
ductor structures

This is a very interesting field where lots of fundamental research needs to be done, particularly in the use of quantum dots as single photon sources using the radiative decay of the biexcitons states, while also performing full state characterization using homodyne tomography. This has been a subject that I have followed closely as a consequence of the pioneering work from Yamamoto and Williams [111, 112]. Is it possible to prove creation of entanglement in these sources?

Along these lines, another topic which I find to be of considerable interest is the optical properties of a set of several quantum dots embedded in an optical waveguide. Can we expect properties related to those of an atomic ensemble?

4. EIT and storage of light using solid state materials

Since the pioneering work of Manson [32], the possibility of using solid state materials for EIT and memory for light with storage times greater that seconds has caught the community’s attention. Are there materials useful for practical quantum information processing? Can we create multi-lambda configurations in these materials? Is it possible to reach storage efficiencies comparable to that of atomic ensembles? Can these material be used to perform interaction with non-classical light?

5. Quantum repeaters and quantum networking

Despite this success, important questions still remain unanswered, such as our capability to produce robust enough experiments to actually implement long distance communications. Can entanglement distillation techniques be applied to this architecture? Is
it possible to achieve better lifetimes of the atomic entanglement? Can we extend these techniques to the regime of quantum gases?

6. Controlled reversible inhomogeneous broadening

This technique, based on controlling the artificial broadening of an absorption line using external fields, has received lots of attention recently due to its prospects to store and recall arbitrary light states [113]. Nevertheless, fundamental questions still need to be addressed regarding which solid state materials are suitable for the technique, as well as if can it be applied experimentally to quantum states of light.
Appendix A

External cavity laser diodes

Almost all the sources of light for our EIT experiments are quantum well laser diodes emitting with a wavelength close to 795 nm. As mentioned in chapter 3, one of our first tasks was to design and build all the elements for an external cavity diode laser system based on the design proposed by Ricci et al. [114] and Haensch et al. [115].

We construct an external cavity for the laser diode via a diffraction grating. This configuration is normally referred as Littrow.

With the feedback obtained from this configuration it is possible to drive the laser diode to emit in a narrow band. Moreover, if we place the grating in a holder that allow us to change its position via a piezo electric transducer, we can change the frequency for which the diode receives feedback. This allows us to scan the frequency of the emitted field by changing the length of the piezo. In principle, we can scan the frequency of the laser by 10 GHz without mode-hops.

To achieve these features, the diode has to be mounted in a set of mechanical parts that facilitate the alignment of the grating via precision screws. A schematic of the laser parts is presented in Fig. A.1.

Other ways of controlling the emission wavelength are the temperature and current of operation for the diode. The diode itself is embedded on a collimation tube (Thorlabs LT230260P) which is temperature controlled via a Peltier element located below the laser cavity. The mechanical system had to be built in such a way that enabled high heat conduction rate in order to use a PID control system to keep a constant temperature in the diode. Both the temperature and current are driven with a Thorlabs ITC100 TEC Controller.
Figure A.1: Schematics for the mechanical part of the external cavity laser system.
Figure A.2: Schematics of a mounted diode laser featuring several of the electronics components mentioned in the text.

We have also built electronic controllers for the piezo. The electronic signal generated by the controller drives the piezo to obtain a homogeneous scan over the band of achievable wavelengths. The electronic controller is equipped as well with several entries for feedback and modulation signals. The electronic diagram of such a controller is shown in Fig. A.3.

The diode laser system is also equipped with an additional input for high frequency modulation of the photodiode current. For this purpose, a Bias-T (50 $\Omega$ impedance) is integrated into the diode laser housing. This modulation is, for example, used for the PDH locking of the OPA cavity.

Alternatively or additionally to the Bias-T, the laser frequency can be modulated or controlled via a FET-current control. By applying a voltage to the self conducting FET transistor, mounted in parallel to the laser diode, a part of the DC current is transmitted to the diode. This feature is used for the phase-locking of these diodes. The main
difference of the modulation circuitries is the bandwidth, for example, the Bias-T has a bigger bandwidth and can be used for modulation of tens of MHz whereas the FET circuitry can only be used up to 1.5 MHz.

![Figure A.4: FET circuitry used to apply modulation to the diode laser.](image)

Another important feature of the system is that we spent time characterizing laser diodes from different companies. Normally, the 795 nm radiation can be achieved using Toptica 795 nm diodes, the problem with those is that they can be expensive. After an extensive testing we found that the Sanyo DL7140-201S and Sharp GH0781JA2C can be driven to operate single mode in the D1 transition by pushing the capabilities of our mechanical system.

At the end of this work, five laser units have been built, with the possibility of building several more due to the development of improved drawings for the mechanical
and electronic systems.

A.1 Simple spectroscopy of laser light in a rubidium cell

Once the mechanical system is mounted and the electronic controllers for temperature, current and piezo driving are calibrated, we can tune the laser wavelength to one of the transition lines of rubidium.

For this purpose, we can build a simple setup on the optical table. Using beam splitters, we can separate the laser beam into three paths to perform three simultaneous measurements. One beam is used to measure the wavelength via a Coherent wave-meter, another measures the power via a Coherent Fieldmaster. The last beam goes to a Fabry-Perot interferometer with one of its mirrors mounted on a piezoelectric crystal. The piezoelectric transducer is driven with a 26 Hz signal to investigate the modes of the incident light once the output of the cavity is focused onto a photodetector.

The objective of the procedure is to find a region of alignment of the grating where it is possible to see a single emission mode in the Fabry-Perot cavity while at the same time the wavelength of the light is near the atomic resonance. The tuning procedure is made via the precision screws of the mechanical system. Fig. A.5 shows the diagram of this simple spectroscopy experiment.

Once the alignment is completed, the laser light is directed into a rubidium cell located in an oven which is temperature controlled. At the same time, a ramp signal is applied to the piezo in the back of the grating to obtain a stable scan. After the beam passes through the cell, it is focused into a photodetector and the obtained signal is analyzed with a digital oscilloscope. If the light is correctly tuned to the Rubidium D1 line (approximately 794.979 nm) it is possible to scan over the entire rubidium absorption spectrum. Fig. A.6. shows the spectroscopy signal obtained on the oscilloscope.
Figure A.5: Setup for rubidium spectroscopy

Figure A.6: Rubidium absorption lines as a result of the simple spectroscopy experiment.
Appendix B

Acousto optical modulators

An acousto optical modulator is a crystal into which an acoustic wave is launched. It forms a sinusoidal phase grating on which an optical field can be diffracted. The zeroth diffraction order is the transmitted beam. The angle $\theta$ between the zero and first diffraction order is given by the frequency of the acoustic wave.

This acoustic wave is usually induced by applying a radio frequency on a piezo, sitting on one side of the AOM crystal. Thus, the frequency of the first diffraction order is shifted by the frequency of the acoustic wave.

An important property of the AOMs is their speed to diffract the first order when being switched on, being on the order hundreds of nanoseconds. This fact makes them suitable for fast switching of optical beams.

For all our experiments we used ISOMET 1205-C AOMs that were driven using home-made electronics. First, it was necessary to create an 80 MHz signal to provide the acoustic modulation. For that purpose, we built frequency multipliers capable of receiving a 10 MHz input to which a multiplication factor of 8 is applied. The generated 80 MHz signal has to be switched on and off; for this purpose we used Minicircuits ZYSWA-2-50DR high frequency switches driven by a Stanford Systems delay generator. The delay generator allowed us to chose the timing of the switching pulses. Before getting to the AOM, the signal has to be amplified using Minicircuits ZHL 32 A amplifiers to generate the 0 dBm signal needed by the AOM to achieve 80 per cent diffraction efficiency.
Figure B.1: The original EIT experiment described in chapter 3 featuring the acousto-optical modulators and their drivers.
Appendix C

Magnetic Shielding

The magnetic shielding was constructed by MuShield Inc. according to our requirements to reach an attenuation of transversal fields to the order of less than micro-Gauss.

To be close to ideal, a spherical geometry of the shielding would be needed. Since this cannot be the case, we use three nested cylinders of mu-metal, an alloy that has a permeability of 80 000, each one with threaded caps to allow good conductance of flux lines, and thus maximize the shielding. Each cylinder was equipped with a coil that could be placed around it, capable of conducting enough current to be able to Degauss the mu-metal.

In order to Degauss the mu-metal we have to apply a magnetic field strong enough to push the mu metal into saturation, removing any imprinted orientations of magnetic moments, followed by oscillations of the magnetic field strength around zero with exponentially decreasing amplitude. in order to achieve saturation we have to apply around 10 A of AC current to the inner coil.

Inside the innermost mu metal cylinder, we also placed another coil with the purpose of applying small magnetic fields to the experiment. The cell is kept in place by two cell holders. This part is heated using four 1/8' C1J5 Watlow Fireroads. The temperature is kept constant by controlling the current applied to the heating roads via a solid state Crydom relay controlled by a Watlow SD3C-HCAA-AARG PID controller. The heating rods are placed outside the first shielding cylinder since the AC current applied to them generates a small magnetic field that needs to be shielded.
Figure C.1: Photograph of the components of the interaction zone featuring the oven parts and the shielding cylinders.
Figure C.2: Photograph of the interaction zone featuring the shielding cylinders and the oven parts once the system is assembled.
Appendix D

Non linear crystal characterization

One of the main elements of our OPA is the non linear crystal. As we have seen in chapter 4, the properties of the crystal are crucial to define the properties of the light at the output of the cavity. In this section, we address the characterization of our crystals.

D.1 Characterization of the PPKTP-Crystals

The crystals are flux-grown periodically-poled KTiOPO$_4$ (Potassium titanyl phosphate), 5 mm long, antireflection coated for 397.5 nm and 795 nm and with a poling period of $\Lambda = 3.14 \mu$m. We characterized type I and type II crystals (the type II crystals could be used in a later experiment to generate single photons.). Here I will only present the experimental characterization for the type I crystal since it is the essential part of the OPA.

In order to understand how periodically poled crystals work, we make use of the Sellmeier equations, empirical equations modeling the wavelength dependence of the refractive index. In the case of KTP the correspondent equation is:

$$n^2_2(\lambda) = 2.3136 + \frac{1.00012}{1 - \left(\frac{238.31\text{nm}}{\lambda}\right)^2} - \left(\frac{\lambda}{7717\text{nm}}\right)^2. \quad (D.1)$$

where $\lambda$ denotes the vacuum wavelength and $n_2$ is the refractive index of the crystal in the propagation direction.

In an unpoled KTP crystal if our wavelengths $(\lambda, \lambda/2) = (795 \text{ nm}, 397.5 \text{ nm})$ would
travel together, they would experience a phase mismatch:

$$\Delta k_{\text{unpol}} = k_{\lambda/2} - 2k_\lambda = \frac{2\pi}{\lambda/2} (n_2(\lambda/2) - n_2(\lambda)) = \frac{2\pi}{3.221 \, \mu m}. \quad (D.2)$$

In order to compensate for this effect, our crystal should have a periodic change of its orientation every $\lambda_{\text{pol}} = 3.2 \, \mu m$.

This factor is critical since the second harmonic generation efficiency scales with

$$\frac{P_{\text{SHG}}}{P_{\text{pump}}} \propto \left(\frac{\sin \Delta k L_c/2}{\Delta k L_c/2}\right)^2. \quad (D.3)$$

Therefore, a small deviation $\delta\lambda_{\text{pol}}$ of the poling period will lead to a zero doubling efficiency at the atomic resonance wavelength.

To achieve the correct poling period, we had shorter sample crystals manufactured as closely as possible to the target wavelength, and a suitable poling period was approached in an iterative procedure.

For a sample crystal its phase-matched wavelength was measured by tuning the emission of a Ti:Sapphire laser to optimal single-pass SHG efficiency and determining the wavelength with a wave-meter. According to the measured deviation from the target wavelength (at a reasonable temperature), a crystal with a corrected poling period was ordered from Ricol and its wavelength was measured again. After the third iteration, phase matching at 794.960 nm could be obtained at 25 °C.

In order to obtain the parameters of the crystal, we followed the characterization procedure presented in [116]. We performed three different measurements: First, we determined the doubling efficiency $\eta$ by measuring the generated second-harmonic power.
versus the fundamental power for $\lambda = 794.960 \text{ nm}$ as the fundamental wavelength (the Rb D1 line). In the second experiment, we measured the dependence of the phase matching against the temperature at a fixed wavelength. In the third experiment, we measured the second-harmonic power as a function of the wavelength at a fixed temperature.

A schematic of the experimental setup is shown in Fig.D.1. The source for the fundamental (FUN) was our Ti:Sapphire laser. The wavelength was measured with a wavelength meter and adjusted to the desired wavelength. By rotating the half-wave plate (HWP) in front of the polarizing beam-splitter (PBS) we could vary the fundamental power. The fundamental beam was focused into the crystal using a 100 mm lens.

To maintain precise control over the crystal orientation and the temperature, a mount was designed. It consists of copper to provide the best possible heat conductivity and holds the crystal in place with four spring loaded screws for good thermal contact. The copper block was mounted on a small 18 W Peltier thermoelectric cooler and contains a PT100 (platinum, 100 $\Omega$) resistance temperature detector. The copper block was mounted on an aluminum block, placed on a Newport 5-axis kinematic translation stage (see chapter 4).
After the crystal, the fundamental is filtered out and the generated second-harmonic is focused on the second power-meter. Using the second HWP and the translation stage (TS) the second-harmonic (SH) generation is optimized.

D.1.1 Measurement of the doubling efficiency

To measure the doubling efficiency we set the Ti:Sapphire laser to $\lambda = 794.985$ nm, and varied the fundamental power from 10 to 200 mW, measuring the corresponding second-harmonic power. The experimental data is shown in Fig. D.2.

The doubling efficiency $\eta$ is defined as $\eta := \frac{P_{2\omega}}{(P_{\omega})^2}$. The data resembled linear behavior and using a linear fit for the experimental data we obtained $\eta = (0.858 \pm 0.024)\% W^{-1}$.

According to the theoretical model of Boyd and Kleinman [117] the conversion efficiency is related to the effective non-linear coefficient as follows:

$$\eta := \frac{P_{2\omega}}{(P_{\omega})^2} = \frac{16\pi^2 L}{\lambda^3 n_1 n_2 \epsilon_0 c} d_{eff}^2 h(\xi, \sigma),$$

where $\lambda$ is the wavelength of the fundamental, $L$ is the crystal length, $c$ is the speed of light in vacuum, $\epsilon_0$ the vacuum permittivity, $n_1$ and $n_2$ are the indices of refraction for the fundamental and second-harmonic, respectively, and $h(\xi, \sigma)$ is the Boyd-Kleinman focusing factor. The focusing factor depends on the focusing parameter $\xi = L/2z_0$, with $z_0$ denoting the Rayleigh-length, and $\sigma$ the normalized phase mismatch $\sigma = \Delta k z_0$. The wave-vector mismatch for quasi-phase-matching (QPM) is given by $\Delta k = k_{2\omega} - 2k_{\omega} - 2\pi m/\Lambda$, where $m$ is an integer that denotes the QPM order.

From the fit of the second-harmonic power versus temperature (see D.1.3) we obtained for our set-up $\xi = 2.9$ and $h = 1.0674$. For crystals with QPM order $m = 1$ and optimal duty cycle, the effective non-linear coefficient $d_{eff}$ is related to the non-linear coefficient
Figure D.2: Measurement of the doubling efficiency. The red dots depict the experimental data. The slope of the linear fit (blue line) gives the doubling efficiency $\eta$. 
\( d_{33} \) (see [116]) as \( d_{\text{eff}} = 2d_{33}/\pi \). Therefore, the non-linear coefficient \( d_{33} \) is given by:

\[
d_{33} = \frac{1}{8} \sqrt{\frac{\lambda^3 n_1 n_2 \varepsilon_0 c}{L h(\xi, \sigma) \eta}}
\]

and we obtained for our crystal \( d_{33} = 11.06 \text{ pm/V} \), where the indices of refraction are \( n_1 = 1.8445 \) and \( n_2 = 1.9719 \). This value is in good agreement with the tabulated value of 16.9 pm/V.

D.1.2 Phase matching against temperature change

In order to characterize the dependence of the phase matching parameter with respect to the temperature, we set the fundamental power to 100 mW and varied the temperature. An electronic feedback control kept the crystal temperature constant to within 0.01 K. While keeping the temperature stabilization active the target temperature was scanned over a 20 K range around the optimal temperature (i.e. the temperature with highest second-harmonic power for a given fundamental wavelength). This was done for the three fundamental wavelengths 794.695 nm, 794.985 nm and 795.301 nm. An example of the measured data is shown in fig.D.3.

The full-width at half-maximum phase matching range was 2.25 K. The functional dependence of the SHG generation efficiency agreed well with the predicted shape [116].

In order to compare this result with what we would expect from theory, we used the empirical relations for the temperature dependence of the refractive index:

\[
\frac{dn_z}{dT} = \left(2.1151 + \frac{2.2762 \mu m}{\lambda} - \frac{1.3332 \mu m^2}{\lambda^2} + \frac{0.3896 \mu m^3}{\lambda^3}\right) \times 10^{-5} \text{ K}^{-1} \quad (D.4)
\]
Figure D.3: The SHG phase matching curve as a function of the crystal temperature for \( \lambda_{FUN} = 794.895 \text{ nm} \)

To shift the phase matching condition by one full linewidth a temperature change of

\[
\delta T = \frac{\lambda}{L_c} \left( \frac{dn_z}{dT} \bigg|_{\lambda/2} - \frac{dn_z}{dT} \bigg|_{\lambda} + \frac{1}{2} \frac{\lambda}{\lambda_{pol}} \kappa \right)^{-1} = 7.71 \text{ K}
\]  \hspace{1cm} (D.4)

is expected to be required (using the thermal expansion coefficient \( \kappa = 6.7 \cdot 10^{-5} \text{ K}^{-1} \)). The measured value is in good agreement since the simplified theory does not consider
the length of the crystal.

D.1.3 Phase matching against wavelength change

To determine the dependence of the phase-matching on the wavelength, we fixed the crystal temperature and varied the fundamental wavelength while keeping the fundamental power constant at 100 mW. The obtained curves look similar to the one shown in Fig. D4.

![Phase-matching dependence against the fundamental wavelength.](image)

To understand our measurements, we used the Sellmeier equation to predict the phase matching bandwidth (spectral distances of the two first zeros of the plane wave doubling efficiency)

$$2\delta \lambda = -\frac{\lambda}{L_c} \left( \left( \frac{1}{2} \frac{dn_Z}{d\lambda} \right)_{\lambda/2} - \frac{dn_Z}{d\lambda} \right) - \frac{1}{2\lambda_{pol}} \right)^{-1} = 0.25 \text{nm} \quad (D.4)$$

As it can be seen in Fig. D4, this number resembles reasonably well the measured
D.1.4 Additional measurements

Two more measurements can be made to complete the characterization of the crystals. First, we can estimate the dependence of the refractive index of the crystal on the temperature, and second, we can determine the relation between the optimal wavelength for SHG and the optimal temperature.

To estimate the dependence of the refractive index on the temperature, we have to consider the normalized phase matching parameter \( \sigma \) for first order QPM [116]:

\[
\sigma = \frac{L}{2\xi} \left( \frac{4\pi}{\lambda} \Delta n + \frac{2\pi}{\Lambda} \right)
\]

Normally, it is assumed that the difference of the refractive indices depends linearly on the temperature and the wavelength in the investigated region. Therefore we assumed the following expression for \( \Delta n \):

\[
\Delta n(T, \lambda) = \Delta n_0 + B_T(T - T_0) + B_\lambda(\lambda - \lambda_0).
\]

The parameter \( B_T \) in this equation is determined by fitting the function

\[
P_{2\omega}(T, \lambda, A, \Delta n_0, B_T, B_\lambda, \xi) = A h(\xi, \sigma) = \frac{A}{4\xi} \left| \int_{-\xi}^{\xi} e^{i\sigma(T, \lambda, \Delta n_0, B_T, B_\lambda, \xi)\xi} \frac{1}{1 + i\tau} \right|^2
\]

to the experimental data of the phase-matching tuning measurements (see Fig. B.3), where the fitting parameter \( A \) depends on the fundamental power \( P_\omega \). Making a least square fit through the experimental data we obtain: \( A = 97.6 \ \mu W, \ \xi = 2.9, \ B_T = 31.22 \cdot 10^{-6} K^{-1} \). This value is in good agreement with the tabulated value of \( 16 \cdot 10^{-6} K^{-1} \).

The phase matching tuning measurements also gave us the second quantity: the
temperature versus optimal wavelength ratio (see Fig.D.5).

We find a linear behavior for the investigated region. By making a linear fit through the experimental data we measure a slope of 0.044 nm/K. This value agrees with the reported values in [116].
In this section we review the design of the geometry of the OPA cavity using the matrix propagation procedure.

The first characteristic to be considered in our design was the stability of the cavity. For a given set of mirrors not all distances between the mirrors form a stable resonator. For many configurations the beam is just reflected outside of the cavity after a few round-trips.

The stability analysis is based on a bow-tie configuration with a crystal in one of the two foci (See fig.E.1).

The cavity consists of two curved mirrors with curvature $R_1$ and two plane mirrors. I will assume gaussian beams propagating inside the cavity. These beams can be characterized by a complex beam parameter $q$ given by

\[
\frac{1}{q(z)} := \frac{1}{R(z)} - i \frac{\lambda}{\pi w(z)^2},
\]
where $\lambda$ is the wavelength of the light, $R(z)$ describes the curvature of the beam and $w(z)$ gives the beam waist. As the beam propagates inside the cavity the beam parameter changes due to the reflection at for example, the curved mirrors.

If we analyze the transformation of the beam parameter due to any optical element we find that the transformation can always be written as

$$q_f = \frac{Aq_i + B}{Cq_i + D},$$

where $q_i$ gives the initial and $q_f$ the transformed beam parameter. This is a so-called Moebius-transformation. These transformations can be represented by 2x2 matrices and the concatenation of several of them is just given by the multiplication of the corresponding matrices. Thus, we can readily calculate the transformation of the beam parameter inside the cavity, given the matrices for each optical element inside. These matrices are the same as for the transformation of rays in classical paraxial optics. Here are two examples:

$$M := \begin{pmatrix} A & B \\ C & D \end{pmatrix}, \quad M_d := \begin{pmatrix} 1 & d \\ 0 & 1 \end{pmatrix}, \quad M_R := \begin{pmatrix} 1 & 0 \\ \frac{-2}{R} & 1 \end{pmatrix}$$

$M_d$ gives the transformation for propagation along the distance $d$ in a uniform medium and $M_R$ describes the change at a curved mirror.

To analyze the stability of the cavity we have to calculate the transformation matrix $M_{rt}$ for one round trip inside the cavity. To do so we start in the middle of the crystal.
with $n_c$ the refractive index of the crystal and $U = d_2 + \sqrt{(d_1 + d_2)^2 + 4h^2}$ the distance the beam propagates from one curved mirror to the other via the upper arm. The first and last matrix describe the propagation through a medium with refractive index $n$ and length $d$.

\[
M_{rt} = \begin{pmatrix}
1 & \frac{L_0}{2n_c} \\
0 & 1
\end{pmatrix}
\begin{pmatrix}
1 & \frac{d_1 - L_0}{2} \\
0 & 1
\end{pmatrix}
\begin{pmatrix}
1 & 0 \\
-\frac{2}{R} & 1
\end{pmatrix}
\begin{pmatrix}
1 & \frac{L_0}{2n_c} \\
0 & 1
\end{pmatrix}
\begin{pmatrix}
1 & 0 \\
0 & 1
\end{pmatrix}
\begin{pmatrix}
1 & 0 \\
0 & 1
\end{pmatrix}.
\]

To simplify the matrix $M_{rt}$ we can introduce the parameters

$$d_{\text{eff}} = d_1 + L_0\left(\frac{1}{n_c} - 1\right)$$

$$\gamma_1 = 1 - \frac{d_{\text{eff}}}{R}$$

$$\gamma_2 = 1 - \frac{U}{R}$$

with parameters $\gamma_1$ and $\gamma_2$ defined in an equivalent way as the $g$-parameters for a linear cavity. Using the new parameters we can write $M_{rt}$ as

\[
\begin{pmatrix}
2\gamma_1\gamma_2 - 1 & R\gamma_1(1 - \gamma_1\gamma_2) \\
-\frac{4\gamma_2}{R} & 2\gamma_1\gamma_2 - 1
\end{pmatrix}
\]

The cavity will be stable if after one round trip the beam parameter is the same again.
i.e. the condition is \( q_f = q_i = q \). We can solve for \( q^{-1} \) and get

\[
\frac{1}{q} = \frac{D - A}{2B} \pm \frac{1}{B} \sqrt{(\frac{A + D}{2})^2 - 1}
\]

\[
\frac{1}{q} = \frac{1}{R} - \frac{i \lambda}{\pi w^2(z)}
\]

Since \( A, B, C, D \in \mathbb{R} \) the first term of the first equation has to be real. Thus to get a physical solution (real beam waists and beam curvatures), the second term has to be imaginary. This is fulfilled for \( |(A + D)/2| \leq 1 \). In terms of the stability parameters \( \gamma \) the confinement or stability condition is

\[
0 \leq \gamma_1 \gamma_2 \leq 1.
\]

This expression is equivalent to the one found for linear cavities.

We can then find the beam waists inside the crystal to be:

\[
w^2_1 = \frac{\lambda R}{2\pi} \left[ \frac{\gamma_1}{\gamma_2} (1 - \gamma_1 \gamma_2) \right]^{\frac{1}{2}}
\]

The dependence of the two beam waists \( w_1 \) and \( w_2 \) on the distance \( d_1 \) between the two curved mirrors for our experimental setup is shown in Fig. E.3.

Another factor to consider in the design is the astigmatism, which is due to the fact that the curvature of the mirror in the propagation plane is reduced by a factor of \( \cos(\beta)^{-1} \) and the curvature in the perpendicular plane enlarged by a factor of \( \cos(\beta) \), where \( \beta \) is the angle between the direction of propagation and the mirror. This means the position
Figure E.2: Beam waist $w_1$ as a function of the distance $d_1$ for the parameters of our OPA setup. The red curve shows the behavior neglecting astigmatism. The green and blue curves correspond to beam waists perpendicular and in the plane of the cavity of the focus for the two planes does not coincide anymore and the beam gains ellipticity.

\[ R_\parallel = R \cos(\beta)^{-1} \quad R_\perp = R \cos(\beta) \]

This fact has to be taken into account for the modeling of bow-tie resonators. As one can see in Fig. E.3 this becomes very crucial near the stability limit since at that point the astigmatism increases very rapidly.
Figure E.3: Beam waist $w_2$ as a function of the distance $d_1$ for the parameters of our OPA setup. The red curve shows the behavior neglecting astigmatism. The green and blue curves correspond to beam waists perpendicular and in the plane of the cavity.
Appendix F

Full experimental setup
Figure F.1: Detailed experimental setup for the EIT with squeezed light project.
Bibliography


