Decoherence and decoherence suppression in quantum memories with collective atomic excitations

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Kurzfassung


Weiterhin wurden im Rahmen einer Kooperation mit der Arbeitsgruppe von M. Drewsen an der Universität in Aarhus (Dänemark) numerische Rechnungen zur Speicherung von Ein-Photon-Wellenpaketen unter Berücksichtigung realistischer experimenteller Verhältnis- se durchgeführt.


gezeigt werden, dass bei geeigneter Wahl der Zustände des Unterraumes, wie sie z.B. durch die kollektiven Speicherzustände realisiert werden, Übergänge innerhalb des Unterraumes invers mit der Anzahl der Atome skalieren und somit bei grosser Teilchenzahl vernachlässigbar sind.

Abstract

In the thesis "Decoherence and decoherence suppression in quantum memories with collective atomic excitations" a scalable procedure for quantum information processing with collective atomic excitations based on electromagnetically induced transparency (EIT) has been developed. This procedure is based on the method developed by M. Fleischhauer and M.D. Lukin [1–3] utilizing an adiabatic transfer of a photonic wave packet into symmetric collective states of an $N$ atom system.

The transfer process between states in the Hilbertspace of an $N$ atom system and of a quantized light field as proposed in [1,3] is based on Stimulated Raman Adiabatic Passage (STIRAP) [4] which is a generalization of the well known process for single atoms in high Q-cavities. In a system with a single atom the small absorption cross section leads to the experimental challenging strong coupling condition. This restrictive requirement can be avoided in an atomic ensemble by the collective interaction of light with the atoms.

In the first part of the thesis the requirements of a storage system with atomic ensembles are discussed in detail. For this purpose quantitative corrections of the usually used adiabatic approximation have been calculated. The basis of those calculations is a quasi particle picture of the transfer process which uses so called dark and bright state polaritons. An effective equation of motion for the dark state polariton operators has been derived which contains, besides the already known parts from the adiabatic limit, further time dependent correction terms, which are discussed in detail. The analytical derived conditions have been compared with numerical simulations in which the corresponding Maxwell-Bloch equations have been solved numerically without any specific assumptions. Furthermore in cooperation with the experimental group of M. Drewsen at the university of Aarhus (Denmark) numerical simulations for the storage process of one photon wave packets under realistic experimental assumptions have been carried out.

In context with the practical use of the proposed transfer process for quantum information storage of photons in systems with inhomogeneous broadened Raman transitions, which appear for example in atomic vapors with Doppler broadening or in doped crystals, the influence of the two photon detuning on the storage process has been analyzed. Using a
problem adapted perturbative expansion in the two photon detuning it was possible to derive an effective equation of motion for the dark state polariton operators which have been solved analytically. The conditions for a successful and reversible transfer process of the photonic excitations are discussed.

Since the robustness of a quantum memory against decoherence plays a crucial role for its experimental realization, the sensitivity of the proposed quantum memory with collective atomic excitations with respect to different decoherence effects is subject of the second part of the thesis. Hereby the existence of equivalence classes of storage states has been shown. Equivalence classes of storage states are classes of collective atomic states which are equivalent to a single stored state of the electromagnetic field. Decoherence processes which lead to transitions within such an equivalence class do not effect the fidelity of the quantum memory. In conclusion it has been shown that a quantum memory based on collective atomic excitations is not more sensitive to decoherence than single atom systems.

The compensation of the enhanced error probability in quantum memories with collective excitations is based on the fact that the adiabatic read out process is only sensitive to dark state polaritons and therefore additional excitations in bright state polaritons do not play any role. Those additional excitations are produced by decoherence during the storage process. A non adiabatic read out process should lead to a wrong result. However by modelling a non adiabatic read out process it has been shown that even such a process does not lead to significant higher error probability.

In the third part of the thesis it has been shown that by providing an accurate additional interaction it is possible to generate quasi decoherence free subspaces of dimension $d \geq 2$ for the collective storage states even if individual and independent reservoir interactions are assumed. The existence of a sufficient large energy gap between a single non degenerated ground state and all other states leads to an exponential suppression of the decay out of this state since a transition in an energetically higher state requires sufficient energy. But the existence of such singular quasi decoherence free states is not of interest for the purpose of quantum information storage. The generation of an energy gap between a subspace and all other states suppresses the decay out of this subspace but does not suppress the transitions caused by the environmental interaction within this subspace. However it was possible to show that by a suitable choice of the storage states in a collective atomic ensemble transitions within the subspace spanned by the storage state scale as the inverse of the number of atoms in the atomic ensemble and therefore can be neglected for a large number of atoms.

In the last part of the thesis the suppression of decoherence caused by collisions in atomic vapors is discussed for a specific system with collective atomic excitations. Hereby the collisions between the storage atoms as well as the collisions of those with buffer gas
atoms have been included. By introducing an additional high frequency interaction, as used for example in NMR systems to control the spin dynamics, it has been shown that the dephasing processes within magnetic Zeeman levels caused by the collisions can be suppressed. Nevertheless spin flips caused by the collisions can not be suppressed for the collective storage states. For this reason in addition to the periodic interaction the energy gap discussed in the third part of the thesis has been introduced. This combined system allows the suppression of spin flips as well as dephasing processes caused by atomic collisions.
Chapter 1

Introduction

Since the discovery of the quantum nature of light in 1900 by M. Planck quantum mechanics has proven to be a very powerful description of nature. There is so far no experimental observation contradicting the basic principles of quantum mechanics. However some of the predictions of quantum mechanics are far from intuitive. The entanglement of quantum states, first introduced by E. Schrödinger in 1935 [5], is one of those predictions which already in the early days of quantum mechanics was subject of much discussion.

Nowadays entanglement is one of the basic ingredients of quantum computation. Quantum computation has attracted much attention in recent years because of its ability to perform certain classes of computations exponentially faster than any classical computer [6–8]. Besides the necessity to perform logical operations using quantum logic gates quantum computation also requires the possibility to transmit and store quantum information. Photons are very robust and readily available transmitters of quantum information, in fact quantum cryptographic devices based on photons are already commercially available [9]. However the storage of the photonic quantum information in quantum bits (qubits) has proven to be quite challenging especially with respect to its scalability to a large number of qubits. While substantial progress has been made in this direction on small systems [10,11], scalability to large system with many qubits remains a major challenge. For example in the case of nuclear magnetic resonance (NMR) [12–14] the number of qubits is limited by the number of spins in a molecule. Similarly for quantum systems based on ion traps the number of ions that can be coupled by a phonon mode [15] or through photon exchange using cavity QED [16,17] is limited. However in recent days several proposals for scalable quantum information memories and processors have been put forward. Among those the most promising ones are based on atomic lattices or arrays of micro traps [18–20] or use linear optical elements and detection in connection with sources of entangled photons [21]. Much success has been achieved in using supercon-
ducting Josephson junctions for quantum information storage and processing [22, 23]. Another possible approach is to store and process the quantum information in collective atomic ensembles. M. Fleischhauer et al. have proposed a technique for a controlled transfer of the quantum state of a photon wave packet to and from a collective atomic spin excitation [1–3] using electromagnetically induced transparency (EIT) [24] and Raman adiabatic passage [4, 25]. In contrast to previous approaches involving single atoms, the technique developed by M. Fleischhauer et al. does not require the strong coupling regime corresponding to a high-Q micro cavity. The key mechanism hereby is the use of an optically dense many atom system in which single photons couple to collective excitations. Therefore the corresponding coupling strength exceeds that of an individual atom by the square root of the number of atoms in the atomic ensemble. Since this fact alleviates most of the stringent requirements of single-atom cavity QED, it could become the basis for a fast and reliable quantum network. Recent experiments [26–28] have already demonstrated the dynamic group velocity reduction and the adiabatic following in the so called dark-state polaritons [3]. The central goal of the present work is to analyze and extend the potentials of a scalable system of quantum information processing based on photonic networks with atomic ensembles.

In chapter 2 the basic concepts of the proposed storage scheme are discussed. First of all the idea of electromagnetically induced transparency (EIT) [24, 29], a technique which eliminates absorption of light in an otherwise optically thick medium is reviewed. Furthermore the idea of using slow light pulses to store quantum information and its limitations are described. Some of those limitations can be overcome by using explicitly time dependent electromagnetically induced transparency. This mechanism first proposed by M. Fleischhauer and M.D. Lukin [1] allows to store and release light pulses while its quantum state is preserved. Its description and the concept of dark and bright state polaritons is also subject of this chapter.

Essential for a successful storage procedure as discussed in chapter 2 is an adiabatic transfer process. Therefore the limitations and restrictions of this storage scheme arising from non adiabatic processes [3, 30] are subject of chapter 3. Furthermore the influence of a finite two photon detuning on the storage process is analyzed [31], which is of particular practical importance in gas experiments with different pump and probe frequencies [26,27] or different propagation directions of the fields since two photon Doppler shifts are no longer negligible in these cases. An estimate of the two photon linewidth of light storage is furthermore interesting for applications in rare earth doped solid state materials with inhomogeneously broadened two-photon transitions [28].

In chapter 4 the problem how reliable the proposed scheme is in view of a quantum information memory is analyzed. This question is very important with respect to various
fields in quantum information processing like network quantum computing [32], secure long distance quantum communication [33, 34] and quantum teleportation [35, 36]. In atomic ensembles the quantum information is stored in collective many particle states which are highly entangled states and are known to be very sensitive to decoherence processes. Therefore in chapter 4 the influence of various decoherence mechanisms on the fidelity of a quantum memory are analyzed. A basis of dark and bright state operators is introduced and it is shown that each quantum state of the radiation field stored in the atomic ensemble corresponds to a whole class of many particle states. Due to the existence of these equivalence classes quantum memories based on collective many particle states do not show an enhanced sensitivity to decoherence as compared to a quantum memory with a single atom.

In order to perform quantum computation in a quantum network a quantum memory is an essential part but additional techniques for the implementation of logical gates are required. These quantum gate operations can for example be implemented by using a scheme based on nonlinear interactions of all atoms as proposed by M. Lukin et al. [32]. However such a quantum logic gate is much more sensitive to decoherence then the quantum memory discussed in chapter 4, i.e. a single spin flip would lead to an incorrect result of the quantum gate operation. Therefore the elimination of decoherence effects is a necessary condition for a successful implementation of a quantum network based on photons and collective atomic excitations and is addressed in chapter 5. One possibility to suppress decoherence is the concept of decoherence free subspaces. Since this concept usually requires an experimentally hard to fulfill permutation symmetry in the system environment interaction an approach based on quasi decoherence free subspace is investigated. It is shown that by choosing appropriate collective storage states and providing for an additional nonlinear interaction of all atoms, it is possible to construct a quasi decoherence free subspace for the storage state even if all atoms of the atomic ensemble couple to individual and independent reservoirs which lead to individual atomic spin flips. Furthermore this idea is combined with the open-loop control well known from the field of nuclear magnetic resonance to suppress collisional decoherence in a storage scheme using atomic vapors.
Chapter 2

Motivation and overview

One of the major problems in quantum information processing is the actual storage of quantum information. In the case of many atom systems this corresponds to the transfer of quantum states of light into metastable states of matter. How this transfer can be carried out using electromagnetically induced transparency is subject of this chapter. First of all the idea of electromagnetically induced transparency [24, 29], a technique which eliminates absorption of light in an otherwise optically thick medium is discussed in section 2.1. The electromagnetically induced transparency is accompanied by a reduction of the group velocity of the propagating light pulse [37]. This leads to the idea of using ultra slow light pulses to store quantum information in a medium. A quantum description of slow light and its limitations for quantum information storage is discussed in section 2.2. Some of these limitations can be overcome by using explicitly time dependent electromagnetically induced transparency as first proposed by M. Fleischhauer and M.D. Lukin [1]. This scheme allows to store and release light pulses, while preserving their quantum state. This mechanism and the concept of dark and bright state polaritons is discussed in sections 2.3 and 2.4.

2.1 Electromagnetically induced transparency

Electromagnetically Induced Transparency (EIT) is an effect of reduction or even cancellation of light absorption by an atomic resonance. The interaction between light and atoms depends strongly on the frequency of the light. In the resonant case, i.e. if the frequency of the light matches a particular atomic transition, the light propagation is accompanied by strong absorption and dispersion. Under certain conditions however it is possible for atomic coherence to cancel the absorption,
Electromagnetically induced transparency

Figure 2.1: Three-level atomic system for electromagnetically induced transparency, with ground state $|b\rangle$, excited state $|a\rangle$ and metastable state $|c\rangle$. $\Omega$ and $E$ denote the drive and probe field respectively.

making a resonant opaque medium transparent by means of quantum interference.

K.-J. Boller, A. Imamâglu and S.E. Harris [38] reported the first experimental observation how an otherwise optically thick medium can be rendered transparent to a probe field by applying an additional electromagnetical drive field. The observed transparency is a special case of the formalism and examples given by P. Lambropoulos [39, 40], J.H. Eberly [41, 42], G.S. Agarwal [43, 44], P.L. Knight [45] and their co-workers. Since the first observation of EIT by K.-J. Boller et al. [38] the research of EIT has attracted great attention. A major driving force behind this interest is the potential application of EIT in nonlinear optics [46–48], sub-recoil laser cooling [49–52], ultra-slow group velocities [47,53–55] and light storage [1–3,26–28]. In the following section a model to illustrate the effect of EIT and the transparency condition will be discussed. Recent overviews concerning the EIT technique are given for example by S.E. Harris [24], M.O. Scully and M.S. Zubairy [29] or M. Fleischhauer, A. Imamoglu and J.P. Marangos [56].

2.1.1 Model

Consider a system consisting of $\Lambda$-type 3-level atoms as indicated in figure 2.1. A sufficiently strong coherent drive field with complex amplitude $E_0e^{-i\nu dt}$ and constant Rabi frequency $\Omega = \wp_{ac} E_0/\hbar$ couples the excited state $|a\rangle$ to a metastable state $|c\rangle$. $\wp_{ac}$ is the dipole moment of the $|a\rangle \rightarrow |c\rangle$ transition and $\nu_d$ is the drive field frequency. The levels $|a\rangle$ and $|b\rangle$ are coupled by a probe field of amplitude $E$ and frequency $\nu$ whose dispersion and absorption will be calculated in the following. The combined effect of these two fields is the stimulation of the atoms into a coherent superposition of the states $|b\rangle$ and $|c\rangle$, i.e. the atoms occupy both states with a well defined phase relation. In such a case the two possible pathways of interaction of light with the atoms can interfere destructively which
2.1. Electromagnetically induced transparency

leads to a vanishing absorption of light, as will be shown in detail in section 2.1.2. The interaction Hamiltonian $\hat{H}_{\text{int}}$ for the atom and the two fields in rotating-wave and dipole approximation is given by [29]

$$\hat{H}_{\text{int}} = \varrho_{ab} E e^{-i\nu t} |a\rangle\langle b| - \hbar \Omega e^{-i\nu dt} |a\rangle\langle c| + H.c., \quad (2.1)$$

where $\varrho_{ab}$ is the dipole moment of the $|a\rangle \rightarrow |b\rangle$ transition. Within the rotating wave approximation the interaction between the atoms and the fields can be described by density-matrix equations. The equations of motion for the density matrix elements $\varrho_{ab}$ and $\varrho_{cb}$ are given by [29]

$$\dot{\varrho}_{ab} = - (\gamma_{ab} - i\delta) \varrho_{ab} - \frac{i\varphi}{\hbar} E e^{-i\nu t} (\varrho_{aa} - \varrho_{bb}) + i \Omega e^{-i\nu dt} \varrho_{cb}, \quad (2.2)$$

$$\dot{\varrho}_{cb} = - (\gamma_{cb} + i\omega_{cb}) - \frac{i\varphi}{\hbar} E e^{-i\nu t} \varrho_{ca} + i \Omega^* e^{i\nu dt} \varrho_{ab}, \quad (2.3)$$

where $\gamma_{ab}$ and $\gamma_{cb}$ denote transversal decay rates, $\omega_{ab}$ and $\omega_{cb}$ are the atomic resonances. Assuming that the Rabi frequency of the probe field $E$ is much smaller than the Rabi frequency $\Omega$ of the strong coherent drive field the coupled atomic equations (2.2) and (2.3) can be treated perturbatively in $E$. The coherent field coupling the states $|a\rangle$ and $|c\rangle$ is however large and must be treated exactly, i.e. keeping all orders of $\Omega$. Initially the atoms are assumed to be in the ground state $|b\rangle$ which means that only $\varrho_{bb}^{(0)} = 1$ differs from zero. Substituting this into the density matrix equations and introducing slowly varying variables according to

$$\varrho_{ab} = \tilde{\varrho}_{ab} e^{-i\nu t} \quad \text{and} \quad \varrho_{cb} = \tilde{\varrho}_{cb} e^{i(\nu_d - \nu)t}, \quad (2.4)$$

the following coupled set of equations is obtained

$$\dot{\tilde{\varrho}}_{ab} = -(\gamma_{ab} - i\delta) \tilde{\varrho}_{ab} + \frac{i\varphi}{\hbar} E + i \Omega \tilde{\varrho}_{cb}, \quad (2.5)$$

$$\dot{\tilde{\varrho}}_{cb} = -(\gamma_{cb} - i\omega_{cb}) - \frac{i\varphi}{\hbar} E \tilde{\varrho}_{ca} + i \Omega^* \tilde{\varrho}_{ab}. \quad (2.6)$$

Here $\delta = \nu - \omega_{ab}$ is the detuning between the atomic transition frequency $\omega_{ab}$ and the frequency $\nu$ of the probe field. In the case that the carrier frequency $\nu_d$ coincides with the atomic resonance $\omega_{ac}$ this set of equations can be solved exactly which yields for $\varrho_{ab}$ the solution [29]

$$\varrho_{ab}(t) = \frac{i\varphi E e^{-i\nu t}(\gamma_{cb} - i\delta)}{\hbar(\gamma_{ab} - i\delta)(\gamma_{cb} - i\delta) + |\Omega|^2}. \quad (2.7)$$

2.1.2 Transparency condition

The polarization $P$ in the medium is given by $P = \varrho \rho_{ab} e^{i\nu t}$ which together with the relation $P = \epsilon_0 \chi E$ results in the following expression for the real and imaginary part of
2.1. Electromagnetically induced transparency

![Figure 2.2: Real (solid line) and imaginary (dashed line) parts of the linear susceptibility (in arbitrary units) as a function of normalized detuning $\delta/\gamma_{ab}$ for $\Omega = 1/\gamma_{ab}$ and $\gamma_{cb} = 10^{-4}\gamma_{ab}$.](image)

Figure 2.2: Real (solid line) and imaginary (dashed line) parts of the linear susceptibility (in arbitrary units) as a function of normalized detuning $\delta/\gamma_{ab}$ for $\Omega = 1/\gamma_{ab}$ and $\gamma_{cb} = 10^{-4}\gamma_{ab}$.

The complex susceptibility $\chi = \chi' + i\chi''$

\[
\chi'(\delta) = \frac{N_a\psi^2 \delta[(|\Omega|^2 + \gamma_{ab}\gamma_{cb} - \delta^2) - \gamma_{cb}(\gamma_{cb} + \gamma_{ab})]}{\epsilon_0\hbar (|\Omega|^2 + \gamma_{ab}\gamma_{cb} - \delta^2)^2 + \delta^2(\gamma_{ab} + \gamma_{cb})^2},
\]

(2.8)

\[
\chi''(\delta) = \frac{N_a\psi^2 \delta^2(\gamma_{ab} + \gamma_{cb}) + \gamma_{cb}(|\Omega|^2 + \gamma_{ab}\gamma_{cb} - \delta^2)}{\epsilon_0\hbar (|\Omega|^2 + \gamma_{ab}\gamma_{cb} - \delta^2)^2 + \delta^2(\gamma_{ab} + \gamma_{cb})^2}.
\]

(2.9)

Here $N_a$ is the atom number density. In figure 2.2 the real and imaginary part of the complex susceptibility $\chi$ are shown as a function of the detuning $\delta$ from resonance. $\chi'$ and $\chi''$ represent the dispersion and loss per unit wavelength respectively and are related to the complex refractive index $n = n' + in''$

\[
n^2(\delta) = 1 + \chi(\delta).
\]

(2.10)

The real and imaginary part of the complex refractive index, $n'$ and $n''$ represent the refractive index and the absorption coefficient of the medium. Combining (2.8), (2.9) and (2.10) this yields

\[
n' = \left\{\frac{1[(1 + \chi')^2 + \chi''^2]^{1/2} + (1 + \chi')}{2}\right\}^{1/2},
\]

(2.11)

\[
n'' = \left\{\frac{1[(1 + \chi')^2 + \chi''^2]^{1/2} - (1 + \chi')}{2}\right\}^{1/2} \text{sgn}(\chi'').
\]

(2.12)
2.1. Electromagnetically induced transparency

Since the imaginary part $\chi''$ of the susceptibility represents the loss per unit wavelength the intensity transmission coefficient $T$ is given by

$$T(\delta) = \exp\{-\chi''(\delta)kL\},$$

(2.13)

where $L$ is the length of the medium.

The medium becomes transparent if the intensity transmission coefficient $T$ is equal to one. For zero detuning, i.e. $\delta = 0$, the real part $\chi'$ in (2.8) of the complex susceptibility is equal to zero and the imaginary part $\chi''$ in (2.9) is proportional to $\gamma_{cb}$. This means that ideal transparency is obtained in the limit of vanishing relaxation of the lower level spin coherence ($\gamma_{cb} = 0$). In this case the two possible pathways of interaction of light with the atoms interfere destructively and there is no absorption at atomic resonance (see figure 2.3). For a small detuning $\delta$ the interference is not perfect and the medium becomes absorbing. Hence the transparency peak that appears in the transmission spectrum is typically very narrow. As the interference becomes more robust by using a stronger coupling field, it is possible to increase the tolerance to frequency mismatch by using stronger coupling fields.

In an ideal EIT medium ($\gamma_{cb} = 0$) the atoms are decoupled from the light fields and at resonance ($\delta = 0$) the susceptibility $\chi$ vanishes and the refractive index $n'$ (2.11) is equal to unity. This means that the velocity of a phase front is equal to that in vacuum. But at the same time the refractive index $n'$ shows a large normal dispersion (see figure 2.2 (b)). Associated with the linear dispersion is a reduction of the group velocity $v_g$. The possibility of reducing the group velocity and manipulating the refractive properties in an EIT medium was first pointed out by S.E. Harris et al. [37] and will be discussed in more detail in the following section.

**Figure 2.3:** (a) Spectrum of transmission and (b) refractive index corresponding to EIT. Numerical parameters as given in figure 2.2.
2.2 Quantum description of slow light propagation

The narrow transparency resonance is accompanied by a very steep variation of the refractive index with frequency. As a result the envelope of a wave packet inside the medium propagates with a changed group velocity which can be much smaller than the speed of light in vacuum. This effect was first discovered by S.E. Harris et al. [37]. Many experimental demonstrations by A. Kasapi et al. [53], M. Xiao et al. [47] and M.D. Lukin et al. [57] followed. In an remarkable experiment by L.V. Hau et al. [55] group velocities down to 17 m/s were achieved in an ultra-cold gas of atoms. Experimental work using hot atomic vapors were done by D. Budker et al. [58] and M.M. Kash et al. [59]. In the following the mechanism of the slow down process and its limitations for the purpose of quantum information storage devices are discussed.

2.2.1 Model

Consider a quasi-one-dimensional\(^1\) system consisting of Λ-type 3-level atoms with two metastable lower states as shown in figure 2.4 (a). A quantized electromagnetic field \(\hat{E}\) couples resonantly the transition between the ground state \(|b\rangle\) and the excited state \(|a\rangle\). The time dependent electric field operator for modes propagating in \(z\) direction and one polarization is given by

\[
\hat{E}(z, t) = \sum_k \frac{\hbar \nu_k}{2\epsilon_0 V} \hat{a}_k(t) e^{ikz} + H.a.,
\]

\(^1\)In this context quasi-one-dimensional means that only the propagation direction \(z\) of the electromagnetic wave is used as spatial variable.
2.2. Quantum description of slow light propagation

where $\hat{a}_k$ denotes the creation operator and $\nu_k$ the frequency of the $k$th field-mode. $\nu$ is the carrier frequency of the optical field. The quantization volume $V = AL$ is the product of the cross section $A$ and the length $L$ of the interaction region along the propagation direction of the quantized field. The upper state $|a\rangle$ is furthermore coupled to the metastable state $|c\rangle$ via a coherent control field with Rabi frequency $\Omega$. The interaction between light and atoms is governed by the Hamiltonian $\hat{H}_{int}$ which in rotating-wave and dipole approximation is given by [3,30]

$$\hat{H}_{int} = -\varphi \sum_{j=1}^{N} \hat{\sigma}_{ab}^j(t) \hat{E}^+(z_j, t) - \hbar \sum_{j=1}^{N} \hat{\sigma}_{ac}^j(t) \Omega(z_j, t) + H.a. \quad (2.15)$$

Here $\varphi$ is the dipole matrix element between the states $|a\rangle$ and $|b\rangle$, $z_j$ denotes the position of the $j$th atom, $N$ is the number of atoms in the quantization volume and

$$\hat{\sigma}_{\alpha\beta}^j \equiv |\alpha_j\rangle \langle \beta_j|, \quad \alpha, \beta \in \{a, b, c\} \quad (2.16)$$

defines the atomic flip operators. $\hat{E}^+(z_j, t)$ is the positive frequency part of the electric field in (2.14). In analogy to [3] it is assumed that the carrier frequencies $\nu$ and $\nu_d$ of the quantum and control fields coincide with the atomic resonances $\omega_{ab}$ and $\omega_{ac}$. Slowly varying variables are introduced according to

$$\hat{E}^+(z_j, t) = \sqrt{\hbar \nu 2\epsilon_0 V} \hat{E}(z_j, t) \exp\left\{i \frac{\nu}{c} (z_j - ct)\right\}, \quad (2.17)$$

$$\hat{\sigma}_{\alpha\beta}^j(t) = \hat{\sigma}_{\alpha\beta}^j(t) \exp\left\{-i \frac{\omega_{\alpha\beta}}{c} (z_j - ct)\right\}, \quad \alpha, \beta \in \{a, b, c\}, \quad (2.18)$$

$$\Omega(z_j, t) = \hat{\Omega}(z_j, t) \exp\left\{i (k_d z_j - \nu_d t)\right\}, \quad (2.19)$$

where $k_d = \vec{k}_d \cdot \vec{e}_z = \nu_d \cos \vartheta / c$ defines the projection of the wave vector $\vec{k}_d$ of the control field on the propagation axis $\vec{e}_z$ of the quantum field (see figure 2.4 (b)).

If the slowly varying quantum field does not change in a length interval $\Delta z$ which contains $N_z \gg 1$ atoms, a continuum approximation is justified. In this case it is possible to introduce continuum atomic variables

$$\hat{\sigma}_{\alpha\beta} = \frac{1}{N_z} \sum_{z_j \in \left(\frac{\Delta z}{2}, \frac{\Delta z}{2}\right)} \hat{\sigma}_{\alpha\beta}^j(t), \quad \alpha, \beta \in \{a, b, c\} \quad (2.20)$$

and make the replacement $\sum_{j=1}^{N} \rightarrow (N/L) \int dz$. This yields the continuous form [1,3] of the interaction Hamiltonian in (2.15)

$$\hat{H}_{int} = -\hbar \frac{N}{L} \int dz \left[ g \hat{\sigma}_{ab}(z, t) \hat{E}(z, t) + \hat{\sigma}_{ac}(z, t) \hat{\Omega}(z, t) e^{i\Delta k z} + H.a. \right]. \quad (2.21)$$
2.2. Quantum description of slow light propagation

Here $g = \sqrt{\nu/2\hbar v_0 V}$ is the quantum field Rabi frequency and $\Delta k = k_d - \omega_{ac}/c = \omega_{ac}(\cos \theta - 1)/c$.

The evolution of the Heisenberg operator corresponding to the optical field can be described in a slowly varying amplitude approximation by the propagation equation

$$
\left( \frac{\partial}{\partial t} + \frac{\partial}{\partial z} \right) \hat{E}(z,t) = igN\tilde{\sigma}_{ba}(z,t). \tag{2.22}
$$

The atomic evolution is governed by a set of Heisenberg-Langevin equations

\begin{align*}
\dot{\tilde{\sigma}}_{aa} &= -\gamma_a \tilde{\sigma}_{aa} - ig[\hat{E}\dagger \tilde{\sigma}_{ba} - H.a.] - i[\tilde{\Omega}^{\ast} \tilde{\sigma}_{ca} e^{-i\Delta k z} - H.a.] + F_a, \tag{2.23} \\
\dot{\tilde{\sigma}}_{bb} &= \gamma_{a\rightarrow b} \tilde{\sigma}_{aa} + ig[\hat{E}\dagger \tilde{\sigma}_{ba} - H.a.] + F_b, \tag{2.24} \\
\dot{\tilde{\sigma}}_{cc} &= \gamma_{a\rightarrow c} \tilde{\sigma}_{aa} + i[\tilde{\Omega}^{\ast} \tilde{\sigma}_{ca} e^{-i\Delta k z} - H.a.] + F_c, \tag{2.25} \\
\dot{\tilde{\sigma}}_{ba} &= -\gamma_{ba} \tilde{\sigma}_{ba} + ig\hat{E}[\tilde{\sigma}_{bb} - \tilde{\sigma}_{aa}] + i\tilde{\Omega}\tilde{\sigma}_{bc} e^{i\Delta k z} + F_{ba}, \tag{2.26} \\
\dot{\tilde{\sigma}}_{ca} &= -\gamma_{ca} \tilde{\sigma}_{ca} + i\tilde{\Omega}[\tilde{\sigma}_{cc} - \tilde{\sigma}_{aa}] e^{i\Delta k z} + ig\hat{E}\dagger \tilde{\sigma}_{bc} + F_{ca}, \tag{2.27} \\
\dot{\tilde{\sigma}}_{bc} &= -ig\hat{E}\dagger \tilde{\sigma}_{ca} + i\tilde{\Omega}^{\ast} \tilde{\sigma}_{ba} e^{-i\Delta k z}. \tag{2.28}
\end{align*}

$\gamma_a = \gamma_{a\rightarrow b} + \gamma_{a\rightarrow c}$, where $\gamma_{a\rightarrow b}$ and $\gamma_{a\rightarrow c}$ denote longitudinal decay rates and $\gamma_{a\beta}$ denotes the transversal decay rate. Dissipative population-exchange processes due to, e.g. spin-flip collisions and dephasing of the transition between the lower states will be disregarded in the following discussion but will be discussed in detail in chapter 4. This assumption can be justified in the case that the interaction time is sufficiently short compared to the characteristic times of these processes. $F_a$ and $F_{a\beta}$ are $\delta$-correlated Langevin noise operators whose explicit form is not of interest here.

2.2.2 Low intensity and adiabatic approximation

The propagation problem can be solved by assuming that the Rabi frequency $g$ of the quantum field is much smaller than the Rabi frequency $\Omega$ of the classical control field. Furthermore the number density of photons in the input pulse has to be much smaller than the number density of atoms. In this case the atomic equations can be treated perturbatively in $\hat{E}$. If we assume that the atoms are prepared initially in the ground state $|b\rangle$, in zeroth order perturbation theory only $\tilde{\sigma}_{bb} = 1$ is different from zero. In first order (2.26) and (2.28) have the following form

\begin{align*}
\tilde{\sigma}_{bc} &= -\frac{g}{\Omega} \hat{E} e^{-i\Delta k z} - \frac{i}{\Omega} \left[ \left( \frac{\partial}{\partial t} + \gamma_{ba} \right) \tilde{\sigma}_{ba} - F_{ba} \right] e^{-i\Delta k z}, \tag{2.29} \\
\tilde{\sigma}_{ba} &= -\frac{i}{\Omega^{\ast}} \frac{\partial}{\partial t} \tilde{\sigma}_{bc} e^{-i\Delta k z}. \tag{2.30}
\end{align*}
By substituting (2.30) in (2.29) and (2.22) the interaction of the probe pulse with the medium can be described by the electric field $\hat{E}$ and the collective spin variable $\tilde{\sigma}_{bc}$

$$\frac{\partial}{\partial t} + c \frac{\partial}{\partial z} \right) \hat{E} = \frac{gN}{\Omega^*} e^{i\Delta k z} \frac{\partial}{\partial t} \tilde{\sigma}_{bc},$$

$$\tilde{\sigma}_{bc} = -\frac{g}{\Omega} \hat{E} e^{-i\Delta k z} + i \left[ \gamma_{ba} + \frac{1}{T} \frac{\partial}{\partial \tilde{t}} \right] \frac{i}{\Omega^*} \frac{\partial}{\partial \tilde{t}} \tilde{\sigma}_{bc} + F_{bc} e^{-i\Delta k z}. \tag{2.32}$$

The propagation equations simplify considerably if for $\tilde{\Omega}(z, t)$ a sufficiently slow change with time is assumed, i.e. if adiabatic conditions apply [60–63]. Normalizing the time to a characteristic scale $T$ via $\tilde{t} = t/T$ and expanding the right hand side of (2.32) in powers of $1/T$ yields

$$\tilde{\sigma}_{bc} = -\frac{g}{\Omega} \hat{E} e^{-i\Delta k z} + i \frac{i}{\Omega} \left[ \gamma_{ba} + \frac{1}{T} \frac{\partial}{\partial \tilde{t}} \right] \frac{i}{\Omega^*} \frac{\partial}{\partial \tilde{t}} \tilde{\sigma}_{bc} + \frac{1}{\sqrt{T}} F_{bc} e^{-i\Delta k z}, \tag{2.33}$$

where $\langle F_{a}(t) F_{b}(t') \rangle \propto \delta(t - t')$ has been used. In the lowest non vanishing order one obtains

$$\tilde{\sigma}_{bc}(z, t) = -\frac{g}{\Omega(z, t)} \hat{E}(z, t)e^{-i\Delta k z}. \tag{2.34}$$

Substituting this into the field equation (2.31) yields the propagation of the quantum light pulse in the perturbative and adiabatic limit

$$\left( \frac{\partial}{\partial \tilde{t}} + c \frac{\partial}{\partial \tilde{z}} \right) \hat{E}(z, t) = -\frac{g^2 N}{\Omega^* (z, t)} \frac{\partial}{\partial \tilde{t}} \hat{E}(z, t). \tag{2.35}$$

### 2.2.3 Electromagnetically induced transparency and slow light

One of the most important phenomena associated with pulse propagation in the described systems is called electromagnetically induced transparency and has been discussed in section 2.1. The complex susceptibility for the discussed system, which is an ideal EIT medium, is given by

$$\chi(\delta) = \frac{g^2 N}{kc} \frac{\delta}{|\Omega|^2 - \delta^2 - i\gamma_{ba}\delta} \approx \frac{g^2 N}{|\Omega|^2} \left[ \frac{\delta}{|\Omega|^2} + i\gamma_{ba} \left( \frac{\delta}{|\Omega|^2} \right)^2 + O(\delta^3) \right], \tag{2.36}$$

where $\delta = \nu - \omega_{ab}$ is the probe detuning. In figure 2.5 the imaginary and real part of the susceptibility $\chi = \chi' + i\chi''$ are shown as a function of the detuning from resonance. At resonance the complex susceptibility $\chi$ vanishes and the refractive index $n' \approx 1 + \frac{1}{2} \chi'$ is equal to unity. This means that the phase velocity of the probe pulse is equal to that in vacuum. But at the same time the real part $\chi'$ shows a large normal dispersion. Associated with this dispersion is a drastic reduction of the group velocity [37].
2.2. Quantum description of slow light propagation

Figure 2.5: Typical susceptibility spectrum for the probe field $\hat{E}$ as a function of normalized detuning for resonant and constant drive field.

If $\tilde{\Omega}(z,t) = \tilde{\Omega}(z)$ is constant in time, this effect can be seen directly in the propagation equation (2.35) of the quantum light pulse

\[
\left( \frac{\partial}{\partial t} + v_g(z) \frac{\partial}{\partial z} \right) \hat{E}(z,t) = 0. \tag{2.37}
\]

The group velocity $v_g$ of the quantum field is given by

\[
v_g(z) = \frac{c}{1 + n_g(z)} \quad \text{with} \quad n_g(z) = \nu \frac{d\chi}{d\nu}|_{\delta=0} = \frac{g^2 N}{|\tilde{\Omega}(z)|^2}. \tag{2.38}
\]

$n_g$ is the so called group index. Since the medium is non absorbing high densities can be used and therefore rather small group velocities can be achieved.

The solution of the wave equation (2.37)

\[
\hat{E}(z,t) = \hat{E} \left( 0, t - \int_0^z dz' \frac{1}{v_g(z')} \right) \tag{2.39}
\]
describes a pulse propagation with a spatial varying group velocity $v_g$ and an invariant temporal pulse shape. Here $\hat{E}(0,t')$ denotes the field entering the interaction region at $z = 0$. The slow down of the quantum probe field is a lossless linear process and hence all properties of the quantum pulse are conserved. Its total number of photons is reduced by the ratio of the group velocity $v_g$ to the vacuum speed of light $c$ since the time integrated
2.2. Quantum description of slow light propagation

Figure 2.6: Schematic (see [64,65]) of the spatial compression of a light pulse when it enters an EIT medium. Photons are converted into flipped atomic spins. The slow photonic and the spin wave propagate together inside the medium.

number of photons crossing a plane perpendicular to the propagation direction stays constant. Thus photons must be temporarily stored in the combined system of atoms and control field. Furthermore the spectrum of the pulse

\[
S(z, \omega) = \int_{-\infty}^{\infty} dt' \langle \hat{E}^\dagger(z, t) \hat{E}(z, t) \rangle e^{-i\omega t'} = S(0, \omega) \tag{2.40}
\]

and therefore the spectral width

\[
\Delta \omega_p(z) = \Delta \omega_p(0) \tag{2.41}
\]

remains unchanged.

The spatial shape is not conserved and depends on the spatial profile of \(v_g\). This leads to a compression of the spatial pulse profile. If the group velocity is statically reduced to a value \(v_g\), the spatial pulse length \(\Delta l\) is modified to

\[
\Delta l = \frac{v_g}{c} \Delta l_0, \tag{2.42}
\]

where \(\Delta l_0\) is the free space value.

In figure 2.6 the dynamics of light propagating inside an EIT medium is illustrated in analogy to [64,65]. Initially the pulse is outside the medium and all atoms are in their ground state \(|b\rangle\) which is illustrated as spin down configuration in figure 2.6. Upon entering the medium the front edge of the pulse is rapidly decelerated. The part of the pulse which is outside the medium still propagates with vacuum speed of light \(c\). Thus by entering
the EIT medium the spatial extent of the pulse is compressed by the ratio $c/v_g$ whereas its amplitude remains unchanged. The energy of the light pulse inside the medium is therefore smaller than outside. The rest of the photons are expended to establish the coherence between the states $|b\rangle$ and $|c\rangle$ which can be illustrated by flipping the atomic spins. The excess energy of the spin flip process is carried away by the control field. The wave of the flipped spins propagates together with the light pulse inside the medium. They form a combined excitation of photons and spins called a dark state polariton [1] which is well known in condensed matter physics (see for example [66]) and will be discussed in detail in section 2.3. The group velocity of the polariton is proportional to the magnitude of its photonic component. As the pulse exits the medium again the atoms return to the ground state and therefore the spatial extent of the pulse increases again. By propagating through the EIT medium the pulse is thus delayed by $(1/v_g - 1/c)L_m$ where $L_m$ is the length of the medium.

### 2.2.4 Slow light and delay time limitations

The achievable delay time $\tau_d$ in an EIT medium with a very small group velocity is limited by the finite lifetime of the low-frequency spin coherence which has been neglected in the preceding section. If $\gamma_{cb}$ denotes the dephasing rate of the $b-c$ transition, it is required that $\tau_d \leq \gamma_{cb}^{-1}$.

However a much stronger condition arises from the requirements of the adiabatic approximation. As discussed in section 2.1.2 the EIT medium is only non absorbing within a very small frequency window around the two-photon resonance. The adiabatic approximation requires that the slow down procedure takes place within this frequency window. If this is not the case, absorption and higher order dispersion need to be taken into account.

The transparency window is defined by the intensity transmission coefficient $T(\delta, z)$ of the medium (2.13). Together with (2.36) this yields

$$T(\delta, z) = \exp\{-\chi'' k z\} \approx \exp\left\{-\frac{\delta^2}{\Delta \omega^2_{tr}}\right\},$$

(2.43)

where the transparency width is given by

$$\Delta \omega_{tr} = \left[\frac{c \tilde{\Omega}^2}{\gamma_{ba} l n_g}\right]^{1/2} = \frac{|\tilde{\Omega}|^2}{\gamma_{ba} \sqrt{\alpha}}.$$

(2.44)

Here $l$ is the propagation length in the medium and $\alpha \equiv \frac{3}{8\pi^2} \phi \lambda^3 k l$ is the opacity of the medium in the absence of EIT. The width of the transparency window $\Delta \omega_{tr}$ decreases with increasing group index $n_g$. According to (2.38) the transparency window gets smaller.
with decreasing group velocity $v_g$. This can be directly related to the pulse delay time $\tau_d = n_g l / c$

$$\Delta \omega_{tr} = \frac{\sqrt{\alpha}}{\tau_d}. \quad (2.45)$$

Hence large delay times $\tau_d$ imply a narrow transparency window which on the other hand requires a long pulse time. Thus during the slow down process of a pulse as the group velocity $v_g$ becomes smaller the transparency window narrows. In the case of a time independent control field the spectral width of the pulse stays constant during the slow down process. As a consequence the transparency window becomes smaller than the spectral width $\Delta \omega_p$ of the pulse. This situation is illustrated in figure 2.7 in which the transmission spectrum of the EIT medium and the frequency spectrum of the quantum pulse are shown as a function of the normalized group velocity. As soon as the transparency window becomes to small, the adiabatic condition is violated and the pulse is absorbed. Therefore for a successful slow down it is required that the spectral width of the pulse stays much smaller than the spectral transparency window

$$\Delta \omega_p \ll \Delta \omega_{tr}. \quad (2.46)$$

This condition leads to an upper limit for the ratio of achievable delay time to the initial pulse length of the photon wave packet. Together with the requirement due to the finite lifetime of the low frequency spin coherence this yields

$$\frac{\tau_d}{\tau_p} \ll \sqrt{\alpha} \quad \text{and} \quad \tau_d \ll \frac{1}{\gamma_{cb}}, \quad (2.47)$$

where $c \tau_d = c / \Delta \omega_p$ is the pulse length. The ratio $\tau_d / \tau_p$ is the figure of merit for any memory device. In practice the achievable opacity $\alpha$ of atomic vapor systems is limited to values below $10^4$ which results in an upper limit for the ratio of time delay to pulse length of the order of 100. Thus EIT media with ultra small group velocities achieved by a spatially dependent control field are only of limited use as temporary storage devices. As will be shown in the following section this limitation can be overcome by introducing a time dependent control field.
2.2. Quantum description of slow light propagation

Figure 2.7: (a) Transmission spectrum of EIT in units of $\delta/(g\sqrt{N})$ as a function of the group velocity $v_g(z)$ and (b) constant pulse spectrum for an space dependent drive field $\tilde{\Omega}(z)$. When the transmission spectrum becomes narrower than the pulse spectrum, strong absorption sets in.
2.3 Dark state polaritons

In the preceding section the propagation of a quantum field in an EIT medium with a spatially varying control field has been discussed. The essential limitation of EIT for a temporary memory and light stopping is the inverse proportionality between the spectral transmission width and the pulse delay time (see equation (2.45)). Thus below a certain value of \( v_g \) the transmission window becomes narrower than the pulse spectrum and the pulse is absorbed unless its spectrum narrows as well. Changing the spectrum of a pulse in a linear medium requires that the medium properties are changing in time. If a hypothetical medium with time dependent group velocity is assumed, the corresponding wave equation could according to (2.37) look like

\[
\left( \frac{\partial}{\partial t} + v_g(t) \frac{\partial}{\partial z} \right) \hat{E}(z, t) = 0. \tag{2.48}
\]

The solution of this hypothetical equation

\[
\hat{E}(z, t) = \hat{E} \left( z - \int_{t_0}^{t} dt' v_g(t'), 0 \right) \tag{2.49}
\]

describes a spatially invariant pulse propagation. The temporal profile however is not conserved and the spectrum of the probe field changes during propagation. Assuming that \( v_g \) changes slowly compared to the field amplitude, the spectral width changes according to

\[
\Delta \omega_p(t) \approx \Delta \omega_p(0) \frac{v_g(t)}{v_g(t_0)}. \tag{2.50}
\]

The spectrum of the probe pulse narrows in the same way as the transparency width while the group velocity is reduced and therefore the bandwidth limitations of EIT can be overcome. One way to make the group velocity time dependent is to allow a time dependent control field \( \Omega \).

By using a time dependent control field M. Fleischhauer and M.D. Lukin were the first to show that the limitations of EIT can be overcome and light pulses can be decelerated and trapped in which case their shape and quantum state are mapped onto meta stable collective states of matter [2, 3]. First experiments done by C. Liu et al. [26] and D.F. Phillips et al. [27] have verified the basic concept and showed that weak laser pulses can be trapped and released after some storage time. The longest storage time achieved so far is 1.5 ms [26]. In an experiment done by A. Mair et al. [67] the phase coherence of the trapping process was demonstrated.

In the following the propagation of a probe pulse in an EIT medium with an explicitly time dependent control field will be discussed.
2.3. Dark state polaritons

2.3.1 Definition of dark- and bright-state polaritons

In order to describe the propagation of the weak probe pulse in an explicitly time dependent EIT medium a spatially homogenous and real valued control field $\tilde{\Omega} = \tilde{\Omega}(t) = \tilde{\Omega}(t)^*$ which is a function of time is considered. The propagation problem (2.31) and (2.32) can be solved in a quasi particle picture. The quasi particles are called dark- and bright-state polaritons\(^2\) and are two new quantum fields. They are defined by a rotation in the space of the electric field $\hat{E}$ and the atomic spin coherence $\tilde{\sigma}_{bc}$. The dark polariton operator $\hat{\Psi}$ is defined by

$$\hat{\Psi}(z,t) = \cos \theta(t) \hat{E}(z,t) - \sin \theta(t) \sqrt{N} \tilde{\sigma}_{bc}(z,t) e^{i \Delta kz} \quad (2.51)$$

and the bright polariton operator is given by

$$\hat{\Phi}(z,t) = \sin \theta(t) \hat{E}(z,t) + \cos \theta(t) \sqrt{N} \tilde{\sigma}_{bc}(z,t) e^{i \Delta kz}. \quad (2.52)$$

The mixing angle $\theta$ is defined in the following way

$$\tan^2 \theta(t) = \frac{g^2 N}{\Omega(t)} = n_g(t). \quad (2.53)$$

$\hat{\Psi}$ and $\hat{\Phi}$ are superpositions of the electromagnetic field operator $\hat{E}$ and the collective atomic operator $\tilde{\sigma}_{bc}$ whose admixture can be controlled by changing the strength of the external control field which leads to a varying mixing angle $\theta$. In analogy to the plane wave decomposition of the electric field operator $\hat{E}$ the dark- and bright-state polariton operators can be decomposed in the same way

$$\hat{\Psi}(z,t) = \sum_k \hat{\Psi}_k(t) e^{i k z} \quad (2.54)$$

$$\hat{\Phi}(z,t) = \sum_k \hat{\Phi}_k(t) e^{i k z}. \quad (2.55)$$

The mode operators $\hat{\Psi}_k$ and $\hat{\Phi}_k$ obey the following commutation relations

$$\left[ \hat{\Psi}_k, \hat{\Psi}^\dagger_{k'} \right] = \delta_{k,k'} \left( \cos^2 \theta + \sin^2 \theta \frac{1}{N} \sum_{j=1}^{N} (\hat{\sigma}_{bb}^j - \hat{\sigma}_{cc}^j) \right), \quad (2.56)$$

$$\left[ \hat{\Phi}_k, \hat{\Phi}^\dagger_{k'} \right] = \delta_{k,k'} \left( \sin^2 \theta + \cos^2 \theta \frac{1}{N} \sum_{j=1}^{N} (\hat{\sigma}_{bb}^j - \hat{\sigma}_{cc}^j) \right), \quad (2.57)$$

$$\left[ \hat{\Psi}_k, \hat{\Phi}^\dagger_{k'} \right] = \delta_{k,k'} \sin \theta \cos \theta \left( 1 - \frac{1}{N} \sum_{j=1}^{N} (\hat{\sigma}_{bb}^j - \hat{\sigma}_{cc}^j) \right). \quad (2.58)$$

\(^2\)A review on dark states and coherent population trapping is given by E. Arimondo [68].
Hence the number density of the photons is assumed to be much smaller than the density of the atoms, the approximation $\hat{\sigma}_{bb}^{j} \approx 1$ and $\hat{\sigma}_{cc}^{j} \approx 0$ is justified. In this limit the new fields $\hat{\Psi}_{k}$ and $\hat{\Phi}_{k}$ fulfill bosonic commutation relations

$$\left[\hat{\Psi}_{k}, \hat{\Psi}_{k}^\dagger\right] \approx \left[\hat{\Phi}_{k}, \hat{\Phi}_{k}^\dagger\right] \approx \delta_{k,k}\quad\text{and}\quad\left[\hat{\Psi}_{k}, \hat{\Phi}_{k}^\dagger\right] \approx 0.$$  

(2.59)

For this reason it is possible to identify them with bosonic quasi particles, the polaritons. Furthermore all states $|n_{k}\rangle$ created by $\hat{\Psi}_{k}^\dagger$ do not contain the excited atomic state $|a\rangle$ and are thus immune against spontaneous emission

$$|n_{k}\rangle = \frac{1}{\sqrt{n_{k}}} (\hat{\Psi}_{k}^\dagger)^{n} |0\rangle |b_{1}, b_{2}, \ldots, b_{N}\rangle, \quad n \in \mathbb{N}.$$  

(2.60)

Here $|0\rangle$ denotes the vacuum field and $|b_{1}, b_{2}, \ldots, b_{N}\rangle$ denotes the state in which all atoms are in the atomic ground state $|b\rangle$. Furthermore the states $|n_{k}\rangle$ are eigenstates of the interaction Hamiltonian $H_{int}$ (2.21) with zero eigenvalue

$$H_{int}|n_{k}\rangle = 0.$$  

(2.61)

For these reasons the quasi particle $\hat{\Psi}$ is called-dark state polariton. The elementary excitations created by $\hat{\Phi}_{k}$ can be identified with the bright states in a three level system (see e.g. [68,69]). For this reason these quasiparticles are called bright-state polaritons.

### 2.3.2 "Stopping" and accelerating photon wave packets under adiabatic conditions

Within the new fields the equations of motion for the electric field $\hat{\mathcal{E}}$ and the collective atomic component $\hat{\sigma}_{bc}$, (2.31) and (2.32), transform to [3]

$$\left[\frac{\partial}{\partial t} + c \cos^{2} \theta \frac{\partial}{\partial z}\right] \hat{\Psi} = -\dot{\theta} \hat{\Phi} - \sin \theta \cos \theta c \frac{\partial}{\partial z} \hat{\Phi},$$  

(2.62)

$$\hat{\Phi} = \frac{\sin \theta}{g^{2} N} \left(\frac{\partial}{\partial t} + \gamma_{ba}\right) \left[\tan \theta (\sin \theta \hat{\Psi} - \cos \theta \Phi)\right] + i \frac{\sin \theta}{g \sqrt{N}} F_{ba},$$  

(2.63)

where the mixing angle $\theta$ is a function of time. The group velocity $v_{g}$ of the slow light propagation is thus given by $v_{g}(t) = c \cos \theta(t)$. Following section 2.2.2 and introducing the adiabaticity parameter $\varepsilon \equiv (g \sqrt{NT})^{-1}$ with the characteristic time $T$ and expanding the equations of motion (2.62) and (2.63) in powers of $\varepsilon$ yields in lowest order, i.e. in the adiabatic limit

$$\hat{\Phi} \approx 0.$$  

(2.64)
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This means that the electric field $\hat{E}$ as well as the spin coherence $\tilde{\sigma}_{bc}$ are entirely determined by the dark-state polariton

$$\hat{E}(z, t) = \cos \theta(t) \hat{\Psi}(z, t) \quad \text{and} \quad \sqrt{N} \tilde{\sigma}_{bc} = -\sin \theta(t) \hat{\Psi}(z, t). \quad (2.65)$$

Furthermore all terms proportional to $\hat{\Phi}$ in (2.62) vanish in the adiabatic limit and the dark-state polariton operator $\hat{\Psi}$ obeys the following simplified equation \[1,3\]

$$\left[ \frac{\partial}{\partial t} + v_g(t) \frac{\partial}{\partial z} \right] \hat{\Psi}(z, t) = 0 \quad \text{with} \quad v_g(t) = c \cos^2 \theta(t). \quad (2.66)$$

The solution of (2.66)

$$\hat{\Psi}(z, t) = \hat{\Psi} \left( z - \int_{t_0}^{t} dt' v_g(t'), 0 \right) \quad (2.67)$$

describes a shape- and quantum-state preserving propagation with instantaneous velocity $v_g(t) = c \cos^2 \theta(t)$. For a strong external drive field, i.e. $\tilde{\Omega}^2 \gg g^2 N$, the mixing angle $\theta$ is approximately zero and the dark-state polariton has purely photonic character. In the opposite limit, $\tilde{\Omega}^2 \ll g^2 N$, the mixing angle $\theta$ is equal to $\pi/2$ and the polariton becomes spin wave like. At the same time its propagation velocity is reduced to zero

$$\hat{E}(z) \leftrightarrow \tilde{\sigma}_{bc}(z'), \quad \text{with} \quad z' = z + \int_{t_0}^{\infty} dt' v_g(t'), \quad (2.68)$$

$$\theta = 0 \leftrightarrow \theta = \frac{\pi}{2}. \quad (2.69)$$

This is the essence of the transfer technique of quantum states from photon wave packets to stationary atomic excitations. By adiabatic rotation of the mixing angle $\theta$ from 0 to $\pi/2$ the dark-state polariton is decelerated to a full stop and its character is changed from purely electromagnetic to purely atomic. Due to the linearity of (2.66) the quantum state of the dark-state polariton stays unchanged during the slow down process. This allows to accelerate the polariton again to its initial speed and to transfer the stored quantum state back to the electric field. The propagation of the dark state polariton is plotted in figure 2.8. The adiabatic rotation of the mixing angle $\theta$ is shown in figure 2.9. The essential point of the described technique is not the storage of the energy or the momentum carried by the photons but the storage of their quantum states. In the EIT medium itself almost no momentum or energy is stored. They are transferred in or from the control field in such a way that the optical pulse is coherently converted into a low energy spin wave.
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Figure 2.8: Propagation of a dark state polariton with envelope \(10^{-3} \exp\left\{-\left(z/10\right)^2\right\}\). The coherent amplitude of the polariton \(\langle \hat{\Psi} \rangle\) is plotted in (a) and the electric field amplitude \(\langle \hat{E} \rangle\) and the matter component \(|\langle \hat{\sigma}_{bc} \rangle|\) in (b) and (c) respectively. The time \(t\) is given in units of \(g\sqrt{N}\) and the position \(z\) is given in units of \(g\sqrt{N}/c\) with \(c = 1\). \(|\hat{\Psi}(z, t)|\), \(|\hat{E}(z, t)|\) and \(|\hat{\sigma}_{bc}(z, t)|\) are given in arbitrary units.

2.3.3 Simultaneous narrowing of transparency window and pulse spectral width

As discussed in section 2.2.4 the transparency window of an EIT medium decreases with the group velocity \(v_g\) which yields in the case of a time dependent control field \(\hat{\Omega}\)

\[
\Delta \omega_{tr}(t) = \frac{\cot^2 \theta(t)}{\cot^2 \theta(t_0)} \Delta \omega_{tr}(t_0),
\]

where \(\Delta \omega_{tr}\) is the spectral width of the transparency window. To avoid absorption the bandwidth of the quantum pulse must always stay within this frequency range. In the following it will be shown that for the an EIT medium with a time dependent control field the bandwidth of the quantum pulse narrows in the same way as the transparency window in analogy of the hypothetical case at the beginning of the section. According to
2.3. Dark state polaritons

Figure 2.9: Adiabatic rotation of the control field $\tilde{\Omega}(t)$. The mixing angle is rotated from 0 to $\pi/2$ and back according to $\cot \theta(t) = 100(1 - 0.5 \tanh[0.1(t - 15)] + 0.5 \tanh[0.1(t - 125)])$.

(2.65) the solution for the electric field $\hat{E}$ in the adiabatic limit is given by

$$\hat{E}(z, t) = \frac{\cos \theta(t)}{\cos \theta(t_0)} \hat{E} \left( z - c \int_{t_0}^{t} dt' \cos \theta(t'), 0 \right).$$

(2.71)

During the slow down process the spatial profile and therefore the length $\Delta l$ of the wave packet does not change

$$\Delta l = \Delta l_0,$$

(2.72)

where $\Delta l_0$ is the initial pulse length. As can be seen in equation (2.71) the amplitude of the electric field is reduced and its temporal profile gets stretched due to the reduction of the group velocity. The opposite happens for an increasing group velocity. The consequence is a changing spectrum $S(z, \omega)$ during propagation

$$S(z, \omega) = \int_{-\infty}^{+\infty} dt'e^{-i\omega t'} \langle \hat{E}^\dagger(z, t)\hat{E}(z, t - t') \rangle \approx \frac{\cos^2 \theta(t)}{\cos^2 \theta(t_0)} S \left( 0, \frac{\omega}{\cos^2 \theta(t)} \right).$$

(2.73)

Here a slow change of $\cos \theta(t)$ compared to the electric field amplitude has been assumed. This gives the spectral width $\Delta \omega_p$ of the probe field

$$\Delta \omega_p(t) \approx \frac{\cos^2 \theta(t)}{\cos^2 \theta(t_0)} \Delta \omega_p(t_0).$$

(2.74)

Combining (2.70) and (2.74) one finds that for practically relevant cases the ratio of the shrinking spectral width of the wave packet and the spectral transparency width remains
finite
\[
\frac{\Delta \omega_p(t)}{\Delta \omega_{tr}(t)} \approx \frac{\sin^2 \theta(t)}{\sin^2 \theta(t_0)} \frac{\Delta \omega_p(t_0)}{\Delta \omega_{tr}(t_0)}.
\] (2.75)

This is due to the fact that the upper value for the Rabi-frequency \( \tilde{\Omega} \) is experimentally limited and \( \sin^2 \theta(t)/\sin^2 \theta(t_0) \) stays always close to unity. Equation (2.75) shows that in the dynamic light trapping method absorption can be prevented as long as the input pulse spectrum lies within the initial transparency frequency window of the EIT medium. The simultaneous narrowing of the transparency frequency window and the pulse spectrum is illustrated in figure 2.10.

In conclusion the basic ideas of a quantum memory for light based on dark state polaritons have been discussed in the last sections. In the case of a time dependent control field a complete slow down of the light pulse and transfer of the quantum state to the atomic system can be achieved in spite of the narrowing of transparency window and thereby enables an effective storage for light pulses.
Figure 2.10: (a) Transmission spectrum of EIT in units of $\delta/(g\sqrt{N})$ as a function of the group velocity $v_g(t)$ and (b) pulse spectrum for a time dependent EIT medium. The spectral pulse width $\Delta\omega_p$ is narrow in the same way as the spectral width $\Delta\omega_{tr}$ of transparency window.
2.4 Quantum memories in resonators with $N$ atoms

An important class of schemes for quantum communication and computing [16,17,70–76] is based on an elementary process in which single excitations are transferred back and forth between atom and photon number states of the radiation field [77]. The transfer process is achieved within the framework of single atom cavity Quantum ElectroDynamics (QED) where a single atom is strongly coupled to the mode of a high Q micro cavity [78]. The proposal by M. Fleischhauer, S.F. Yelin and M.D. Lukin [2,69] suggests an alternative root to overcome the challenging conditions due to the strong coupling conditions in single cavity QED and is therefore an ideal technique for the purpose of quantum information storage. In the following the storage technique introduced in the last sections will be used to map the quantum states of single photon fields onto collective metastable states of an optically dense, coherently driven medium inside an optical resonator.

2.4.1 Intracavity EIT with quantum fields and family of dark-states

In the following the properties of intracavity EIT [79] due to the interaction of the combined cavity atomic system with one photon quantum field will be discussed. For a more detailed description see M. Fleischhauer et al. [2] and M.D. Lukin et al. [69]. The system consists of a single mode cavity which contains $N$ identical three level atoms, see figure 2.11 (a). The atomic level scheme which describes the system is equivalent to

![Figure 2.11](image_url)

**Figure 2.11:** (a) Cavity setup with $N$ atoms interacting with the cavity mode and a classical control field with Rabi frequency $\Omega$. $\gamma$ is the empty cavity decay rate. (b) Simplified cavity setup to optimize the coupling of the cavity dark state to free field modes. $R$ and $T$ are amplitude reflectivity and transmission of the input mirror. $E_{in}$, $E_{out}$ and $E_c$ denote input, output and circulating field components [2].
2.4. Quantum memories in resonators with $N$ atoms

Figure 2.12: (a) Three level atoms in Λ configuration interacting with a quantum field and a classical control field with Rabi frequency $\Omega(t)$. $g$ is the Rabi frequency of the quantum field. (b) Interaction of a single excited mode with $N$ three level atoms in the collective state basis.

de the level scheme introduced in section 2.2 and shown in figure 2.12 (a). One of the two optically allowed transitions is coupled by a cavity mode. The other allowed transition is again coupled via a coherent time dependent control field. The coherent field remains essentially unaffected by the interaction and therefore can be represented by a time dependent complex Rabi frequency $\Omega(t)$ [2]. The dynamics of the system is described by the following interaction Hamiltonian

$$\hat{H}_{int} = \hbar g\hat{a}\hat{\Sigma}_{ab} + \hbar \Omega(t)e^{-i\nu_d t}\hat{\Sigma}_{ac} + H.c.,$$  \hspace{1cm} (2.76)

where $g$ is the Rabi frequency of the quantum field and $\nu_d$ is the frequency of the control field. $\hat{\Sigma}_{\alpha\beta}$ denotes collective atomic operators which are given by [2]

$$\hat{\Sigma}_{\alpha\beta} = \sum_{j=1}^{N} \hat{\sigma}_{\alpha\beta},$$  \hspace{1cm} (2.77)

where the atomic flip operators $\hat{\sigma}_{\alpha\beta}^j$ are defined in analogy to equation (2.16). The collective operators couple only symmetric Dicke like states [80] which are defined in the
following way

\[
|b\rangle \equiv |b_1 \ldots b_N\rangle, \\
|a\rangle \equiv \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |b_1 \ldots a_j \ldots b_N\rangle, \\
|c\rangle \equiv \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |b_1 \ldots c_j \ldots b_N\rangle, \\
|aa\rangle \equiv \frac{1}{\sqrt{2N(N-1)}} \sum_{j=1}^{N} \sum_{k=1}^{N} |b_1 \ldots a_j \ldots a_k \ldots b_N\rangle, \\
|ac\rangle \equiv \frac{1}{\sqrt{N(N-1)}} \sum_{j=1}^{N} \sum_{k=1}^{N} |b_1 \ldots a_j \ldots c_k \ldots b_N\rangle, \text{ etc.}
\] (2.78)

The photonic number state inside the cavity mode is denoted by \(|n\rangle\). The interaction caused by the interaction Hamiltonian is illustrated in the collective atom basis in figure 2.12 (b).

Under conditions of two photon resonance the interaction has again a family of dark eigenstates with zero eigenvalues. The dark state involving one photon inside the cavity is given by [2,69]

\[
|D, 1\rangle = -i \frac{\Omega |b, 1\rangle - g \sqrt{N} |c, 0\rangle}{\sqrt{\Omega^2 + g^2 N}} = -i \cos \theta |b, 1\rangle + i \sin \theta |c, 0\rangle.
\] (2.79)

The time dependent mixing angle \(\theta(t)\) is again defined by \(\tan \theta(t) = g^2 N/\Omega(t)\). By changing the mixing angle from 0 to \(\pi/2\), i.e. by varying the Rabi frequency \(\Omega(t)\) of the classical control field, it is possible to change the character of the cavity dark state from purely photonic to purely atomic. This perfect dark state even exists for a single photon field if the phase of the latter is undetermined [77].

In contrast to the scheme introduced in section 2.3 the interaction of the cavity dark state with its environment has to be considered which can be done analogue to [79]. Three important mechanism of dissipation and decay have to be distinguished. As already mentioned in section 2.3 the dark state is immune against decay out of the upper levels since it contains no components of those states. But it is sensitive to the decay of the coherence between the two lower levels. This decay \(\gamma_{bc}\) sets the upper limit for the lifetime of the cavity dark state. In addition to this mechanism the effect of the finite lifetime of the cavity has to be considered. Following [79] a bare cavity decay rate \(\gamma\) leads to the
2.4. Quantum memories in resonators with $N$ atoms

effective decay rate $\gamma_D$ of the cavity dark state $|D, 1\rangle$ and is given by [2]:

$$\gamma_D = \gamma \cos^2 \theta(t). \quad (2.80)$$

This equation shows that by varying the mixing angle $\theta(t)$ it is possible to influence the coupling of the cavity dark state to its environment. For $\cos^2 \theta \ll 1$, i.e. $g^2 N \gg \Omega$, the effect of the cavity decay is substantially reduced. M. Fleischhauer et al. [1] have shown that this allows to effectively load the cavity system with an excitation resulting from an incoming photon wave packet and to reload the wave packet after some storage period.

2.4.2 Storage and reloading single photon excitation by adiabatic following

The problem of transferring a single photon state of the free field to a single photon cavity dark state and vice versa can be achieved by adiabatically rotating the mixing angle $\theta(t)$ in a specific way. Consider an effective one dimensional model with a Fabry Perot type cavity as shown in figure 2.11 (b). The $z$ axis is assumed to be parallel to the propagation direction of the in- and outgoing modes. $z = 0$ is the position of the partially transmitting input mirror of the cavity. The second mirror is assumed to be 100% reflecting.

The input and output process is modelled by introducing a continuum of free space modes with field operators $\hat{b}_k$ and $\hat{b}_k^\dagger$. They are coupled to the selected cavity mode with a coupling constant $\kappa$ which is for simplicity assumed to be equal for all relevant cavity modes. The hamiltonian $\hat{H}_{cav-free}$ which describes the interaction between the cavity modes and the free field outside the cavity is given by [2]

$$\hat{H}_{cav-free} = \hbar \kappa \sum_k \hat{a}^\dagger \hat{b}_k + H.c. \quad (2.81)$$

The input field is a general single photon state and given by [2]

$$|\Psi_{in}(t)\rangle = \sum_k \xi_k^{in}(t)|1_k\rangle \text{ with } \xi_k^{in}(t) = \xi_k^{in}(t_0)e^{-i\omega_k(t-t_0)}. \quad (2.82)$$

Here $t_0$ denotes the arrival time of the photon at the input mirror. The state $|1_k\rangle$ is defined by $|0\ldots1_k\ldots0\rangle$ and $\xi_k$ fulfils the normalization condition $\sum_k \xi_k^{in} = 1$. These fields can be described by an envelope wave function $\Phi_{in}$ defined by [2]

$$\Phi_{in}(z, t) = \sum_k \langle 0_k|\hat{b}_k e^{ikz}|\Psi_{in}(t)\rangle. \quad (2.83)$$

In the continuum limit $\xi_k(t) \rightarrow \xi(\omega_k, t)$ and $\sum_k \rightarrow \frac{L}{2\pi} \int dk$ the envelope wave function is given by

$$\Phi_{in}(z, t) = \frac{L}{2\pi c} \int d\omega_k \xi_k^{in}(\omega_k, t)e^{ikz} \quad (2.84)$$
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with the quantization length $L$. The normalization condition of the Fourier coefficients gives the normalization of the input wave function

$$\frac{1}{L} \int dz |\Phi_{in}(z, t)|^2 = 1.$$  (2.85)

The most general state $|\Psi(t)\rangle$ of the combined system of the single photon wave packet, the cavity mode and the atoms is given by [2]

$$|\Psi(t)\rangle = b(t)|b, 1, 0_k\rangle + c(t)|c, 0, 0_k\rangle + a(t)|a, 0, 0_k\rangle + \sum_k \xi_k(t)|b, 0, 1_k\rangle.$$  (2.86)

In this notation $|b, 1, 0_k\rangle$ for example stands for the collective atomic state $|b\rangle$, the cavity mode is in the single photon state $|1\rangle$ and there are no photons in the outside modes $|0_k\rangle$. It is important to note that again only symmetric collective states (2.78) are coupled by the interaction Hamiltonian. In the following calculation it is assumed that the bare frequency of the cavity $\nu_c$ coincides with the frequency $\omega_{ab}$ of the atomic $a \rightarrow b$ transition and with the carrier frequency $\omega_0$ of the input wave packet, i.e. $\nu_c = \omega_{ab} = \omega_0$. Furthermore the frequency $\nu_d$ of the classical control field is assumed to be equivalent to the frequency $\omega_{ac}$ of the atomic $a \rightarrow c$ transition, i.e. $\nu_d = \omega_{ac}$. These assumptions already imply the necessary condition of two photon resonance condition for the storage mechanism. To model the decay processes such as spontaneous emission and the finite lifetime of the dark state an open system approach is used and the decay rates $\gamma_a$ and $\gamma_c$ are introduced. The equations of motion for the slowly varying state amplitudes are obtained by a transformation into the rotating frame [2]:

$$\frac{d}{dt} a(t) = -\frac{\gamma_a}{2} a(t) - ig\sqrt{N} b(t) - i\Omega(t) c(t),$$  (2.87)

$$\frac{d}{dt} b(t) = -ig\sqrt{N} a(t) - i\kappa \sum_k \xi_k(t),$$  (2.88)

$$\frac{d}{dt} c(t) = -\frac{\gamma_c}{2} c(t) - i\Omega(t) a(t),$$  (2.89)

$$\frac{d}{dt} \xi_k(t) = -i\Delta_k \xi_k(t) - i\kappa b(t).$$  (2.90)

Here $\Delta_k = \omega_k - \nu_c$ is the detuning of the free field modes from the cavity resonance. In comparison to the one atom case the enhanced coupling of the atoms to the cavity mode is manifested in the scaling factor $\sqrt{N}$. The decay process instead is not increased since it acts on each atom individually. To solve the coupled equations of motion a basis of dark and bright states, $|D, 1\rangle$ and $|B, 1\rangle$, is introduced in analogy to the dark and bright state polariton operators in section 2.3:

$$|D, 1\rangle \equiv -i \cos \theta(t)|b, 1, 0_k\rangle + i \sin \theta(t)|c, 0, 0_k\rangle,$$  (2.91)

$$|B, 1\rangle \equiv \sin \theta(t)|b, 1, 0_k\rangle + \cos \theta(t)|c, 0, 0_k\rangle$$  (2.92)
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With the corresponding state amplitudes \( D(t) \) and \( B(t) \) the equations of motion (2.87)-(2.90) can be rewritten in the following form [2]

\[
\frac{d}{dt}a(t) = -\frac{\gamma_a}{2} a(t) - ig^2 N \csc^2 \theta(t) B(t),
\]

(2.93)

\[
\frac{d}{dt}B(t) = -i\dot{\theta}(t)D(t) - ig^2 N \csc^2 \theta(t) a(t) - i\kappa \sin \theta(t) \sum_k \xi_k(t),
\]

(2.94)

\[
\frac{d}{dt}D(t) = -i\dot{\theta}(t)B(t) + i\kappa \cos \theta(t) \sum_k \xi_k(t),
\]

(2.95)

\[
\frac{d}{dt}\xi_k(t) = -i\Delta_k \xi_k(t) - i\kappa \sin \theta(t) B(t) - \kappa \cos \theta(t) D(t).
\]

(2.96)

where the decay rate \( \gamma_c \) of the meta-stable atomic state \( c \) has to be assumed sufficiently small and therefore can be neglected during the time required for the in- and output process. But \( \gamma_c \) must be taken into account during the storage period. If the characteristic time \( T \) of the process is large compared to the radiative lifetime \( \gamma_a^{-1} \) of the upper state \( a \), i.e. \( \gamma_a T \gg 1 \), an elimination of the excited state amplitude \( a(t) \) is possible. Furthermore the bright state amplitude \( B(t) \) can be eliminated by disregarding non-adiabatic corrections, which are proportional to \( \dot{\theta}(t) = d\theta(t)/dt \) in the equations (2.94) and (2.95). This yields the following two equations of motion:

\[
\frac{d}{dt}D(t) = \kappa \cos \theta(t) \sum_k \xi_k(t),
\]

(2.97)

\[
\frac{d}{dt}\xi_k(t) = -i\Delta_k \xi_k(t) - \kappa \cos \theta(t) D(t).
\]

(2.98)

From this equations it follows that under adiabatic conditions the total probability of finding the system in a free field single photon state or in the cavity dark state is conserved and therefore only an exchange of probability between the free field states and the cavity dark state is possible. Formal integration of equation (2.98) leads to [2]

\[
\xi(\omega_k, t) = \xi^{in}(\omega_k, t) e^{-i\Delta_k(t-t_0)} - \kappa \int_{t_0}^{t} d\tau \cos \theta(\tau) D(\tau) e^{-i\Delta_k(t-\tau)}.
\]

(2.99)

Substituting this solution into the equation (2.97) yields

\[
\frac{d}{dt}D(t) = \frac{\kappa L}{2\pi c} \cos \theta(t) \int d\omega_k \xi^{in}(\omega_k, t_0) e^{-i\Delta_k(t-t_0)}

- \kappa^2 \cos \theta(t) \int_{t_0}^{t} d\tau \cos \theta(\tau) D(\tau) \frac{L}{2\pi c} \int_{t_0}^{t} d\omega_k e^{-i\Delta_k(t-\tau)}.
\]

(2.100)

The first term in equation (2.100) can be identified with the input photon wave function at \( z = 0 \). Invoking a Markov approximation \( \int d\omega_k e^{-i\Delta_k(t-\tau)} \rightarrow 2\pi \delta(t - \tau) \) and assuming
2.4. Quantum memories in resonators with $N$ atoms

that no photons arrive to the cavity before $t_0$, i.e. $\Phi_{in}(0,t)=0$ for all $t \leq t_0$, leads to the solution [2]

$$D(t) = \sqrt{\gamma c \frac{\kappa}{L}} \int_{t_0}^{t} d\tau \cos \theta(\tau) \Phi_{in}(0,\tau) \exp \left\{ -\frac{\gamma}{2} \int_{\tau}^{t} d\tau' \cos^2 \theta(\tau') \right\},$$

(2.101)

where the empty cavity decay rate $\gamma$ is defined by $\gamma = \kappa^2 L/c$. Substituting this into equation (2.99) leads to the following input-output relation

$$\Phi_{out}(0,t) = \Phi_{in}(0,t) - \gamma \cos \theta(t) \int_{t_0}^{t} d\tau \cos \theta(\tau) \Phi_{in}(0,\tau) \exp \left\{ -\frac{\gamma}{2} \int_{\tau}^{t} d\tau' \cos^2 \theta(\tau') \right\}.$$  

(2.102)

The equations (2.101) and (2.102) show that the atoms simply cause a change of the cavity decay rate according to $\gamma \rightarrow \gamma \cos^2 \theta$. By using techniques of adiabatic transfer [4,60,81] it is in analogy to section 2.3.2 possible to capture and to subsequently release a single photon state. Equation (2.97) shows that the coupling of the cavity dark state and the free field modes can be controlled via the time dependent Rabi frequency $\Omega(t)$ of the classical control field. If $\Omega$ is large, i.e. $\cos \theta$ is large respectively, the coupling between the cavity dark state and the free field is strong and the photons of the free field modes can leak in and out the cavity. This process is effectively stopped if $\Omega$ is small. Therefore by first accumulating the free field in a cavity mode and then adiabatically switching off the classical control field an initial free space wave packet can be stored in a long lived atomic dark state. The latter can be released by reversing the process, i.e. by adiabatically increasing the Rabi frequency $\Omega$ of the control field again. Even if the previous discussion is restricted to the storage of a single photon wave packet M.D. Lukin et al. [69] have also discussed the generalization to multiphoton states.

In order to achieve a maximum transfer of free field photons into cavity photons the outgoing field components should be minimized. Differentiating the input-output relation (2.102) and assuming $\Phi_{out} = \frac{d}{dt} \Phi_{out}(t) = 0$ yields

$$-\frac{d}{dt} \ln \cos \theta(t) + \frac{d}{dt} \ln \Phi_{in}(t) = \frac{\gamma}{2} \cos^2 \theta(t)$$

(2.103)

which corresponds to a dynamical impedance matching [82]. The output field $\Phi_{out}$ remains zero if the Rabi frequency $\Omega(t)$ of the control field is chosen such that $\cos \theta(t)$ fulfills equation (2.103) with the asymptotic condition $\cos \theta(t) \rightarrow 0$ for $t \rightarrow \infty$. The first term on the left hand side of equation (2.103) describes internal losses due to coherent Raman adiabatic passage and the second term appears due to the time dependence of the input field $\Phi_{in}$. The right hand side of this equation can be interpreted as an effective cavity decay rate which is reduced due to intra-cavity EIT [79]. In analogy to the classical impedance
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Figure 2.13: Input and output wave function for an hyperbolic secant input wave packet $\Phi_{in} = \sqrt{2/(\gamma T)} \text{sech}(2t/T)$, $\gamma T = 4$ and optimized $\cos \theta(t)$. To release the photon wave $\cos \theta(t)$ is time reversed at $t \approx 15T$.

Matching equation (2.103) reflects the condition for complete destructive interference of the directly reflected and the circulating components which leads to a vanishing outgoing wave. Solving equation (2.103) leads to [69]

$$
\cos^2 \theta(t) = \frac{\Phi_{in}^2(t)}{\gamma \int_{-\infty}^{t} d\tau \Phi_{in}^2(\tau)},
$$

(2.104)

which corresponds to $D(t \to +\infty) \to 1$. Hence by suitable variation of the classical driving field any single photon pulse can be trapped ideally if its pulse length is longer than the bare cavity decay time. For further discussions concerning the impedance matching in the case of a time dependent input field see for example [2,69]. To release the photon wave packet the mixing angle $\theta(t)$ is time reversed after the storage period. The input and output wave functions for an hyperbolic secant input wave packet using the impedance matching condition 2.104 optimized $\cos \theta(t)$ are shown in figure 2.13.

Before proceeding the conditions for the adiabatic elimination of the bright state amplitude has to be derived. This can be done by substituting the formal integral (2.98) in the equations of motion (2.87)-(2.89) and performing the Markov limit. This leads to the following adiabatic conditions [2,69]

$$
\Omega^2(t) + g^2 N \gg \max \left[ \gamma \gamma_a, \frac{\gamma_a}{T}, \sqrt{\frac{\gamma}{T} \gamma_a} \right].
$$

(2.105)
The condition $\Omega^2(t) + g^2N \gg \gamma\gamma_a$ is the most stringent one since the characteristic input pulse length and therefore the characteristic time $T$ have to be larger or equal to the bare cavity decay time $\gamma^{-1}$. For a time dependent control field the condition $g^2N \gg \gamma\gamma_a$ is sufficient to guarantee adiabatic following. But in contrast to the one atom case the proposed scheme does not require a strong coupling regime due to the large factor $N$ in condition (2.105).

In the discussion above the finite lifetime $\gamma_c^{-1}$ of the meta-stable state has been neglected. This is justified during the loading process since $\gamma_c$ is assumed to be sufficient small. But it needs to be taken into account during the storage interval. For a time $t_1$, such that $\Phi_{in}(0, t) = 0$ for all $t > t_1$ and $\cos \theta(t_1) = 0$, the output pulse $\Phi_{out}(0, t)$ can be obtained from the input-output relation (2.102):

$$\Phi_{out}(0, t) = -\sqrt{\frac{L}{c}} D(t_1) \cos \theta(t) \exp \left\{ -\frac{\gamma}{2} \int_{t_1}^{t} d\tau \cos^2 \theta(\tau) \right\}. \quad (2.106)$$

If the dark state decay is again neglected during the unloading process the amplitude of the output wave function only depends on the dark state amplitude at the releasing time $t_1$. The ultimative fidelity of the storage process is since determined by the decay of the collective dark state during the storage period. With the decay rate $\gamma_c$ of the meta-stable state the dark state amplitude $D(t_1)$ after the storage period $t_1 - t_0$ is given by

$$D(t_1) = D(t_0) \exp \left\{ -\frac{\gamma_c}{2} (t_1 - t_0) \right\}. \quad (2.107)$$

Since the decay only affects those atoms which are in the meta-stable state and each atom has only a probability of $1/N$ to be in this state the decay of the collective dark state is identical to the single atom case even if the coupling strength to the cavity mode is enhanced by a factor $\sqrt{N}$.

It is essential that in the scheme proposed by M. Fleischhauer et al. [2,69] all the transfer operations can be achieved without invoking the strong coupling regime of single atom cavity QED. The involved dark state consists of a large number of multi level atoms interacting with a single cavity mode. By adiabatic rotation of these cavity dark state quantum impedance matching can be achieved for a single photon input wave packet. In this case the quantum state of the radiation field can be stored with nearly 100% efficiency to a metastable state of the atoms. By reversing the adiabatic rotation the stored state can be transformed back into a well defined output wave packet and can therefore be used as quantum memory. For a more detailed discussion see references [2,69].
Chapter 3

Adiabatic and non adiabatic effects during the propagation of dark state polaritons

In the previous chapter the technique for a controlled transfer of the quantum state of a photon wave packet to and from a collective atomic spin excitation using EIT [1, 2] has been introduced. Essential for a high fidelity of the transfer process is an explicitly time dependent control field which varies adiabatically.

In section 3.1 the limitations and restrictions of the transfer process arising from non adiabatic processes will be discussed [3, 30]. Adiabatic following leads to a parallel narrowing of the spectral width of the probe pulse and the transparency window of the EIT medium provided the carrier frequency of the probe pulse and the time dependent control field are in precise two photon resonance. Therefore the question arises how a finite two photon detuning affects this storage scheme. This question is of particular practical importance in gas experiments with different pump and probe frequencies [26, 27] or different propagation directions of the fields since two photon Doppler shifts are no longer negligible in these cases. An estimate of the two photon linewidth of light storage is furthermore interesting for applications in rare earth doped solid state materials with inhomogeneously broadened two-photon transitions [28]. Therefore in section 3.2 the influence of a finite two photon detuning on the storage process will be analyzed. In section 3.3 numerical calculations for realistic experimental conditions are presented. These calculations have been carried out in collaboration with the experimental group of M. Drewsen at the university of Aarhus (Denmark). The experimental realization is currently being developed by A.V. Mortensen [83] in the same group.
3.1 Adiabaticity condition and non adiabatic corrections

As discussed in chapter 2 in the case of a time dependent control field the spectrum of the polariton and thus of the probe pulse narrows in the same way as the EIT transparency window. Therefore the adiabatic slow down of the dark state polariton overcomes the bandwidth limitation of EIT and the light pulse can be stopped and stored inside the EIT medium. In order to prove the validity of the simple analytic solution (2.67) the assumption of an adiabatic slow down process has to be justified. In the following the adiabaticity requirements are investigated [3,30].

In section 3.1.1 the model of section 2.2.1 is revisited in view of these requirements and in section 3.1.2 the adiabaticity parameter \( \varepsilon \) is introduced. In section 3.1.3 the requirements resulting from the adiabaticity condition are derived and discussed.

3.1.1 Model

To describe the non adiabatic effects the quasi one dimensional system of \( \Lambda \)-type 3-level atoms with two metastable lower states which was introduced in section 2.2.1 (see figure 2.4) is considered. As already discussed in section 2.2.1 the Hamiltonian describing the interaction of the atoms with the quantum field \( \hat{E} \) and the classical control field with Rabi frequency \( \Omega \) is given in the continuous form by

\[
\hat{H}_{int} = -\hbar \frac{N}{L} \int dz \left[ g \tilde{\sigma}_{ab}(z, t) \hat{E}(z, t) + \tilde{\sigma}_{ac}(z, t) \tilde{\Omega}(z, t) e^{i\Delta k z} + H.a. \right].
\] (3.1)

Here \( g = \varphi \sqrt{\nu/2\hbar \omega_0 V} \) is the atom-field coupling constant and \( \Delta k \) is given by \( \Delta k = k_d - \omega_{ac}/c = \omega_{ac}(\cos \vartheta - 1)/c \). Furthermore, we assume that \( \Omega \) the is only a function of time. This can be realized either by perpendicular incidence of the control field or, in the case of copropagating fields, if the group velocity of the probe pulse is at all times much less than that of the coupling field. In the latter case retardation effects of the control field can be disregarded.

The evolution of the Heisenberg operator \( \hat{E} \) corresponding to the optical field is described in a slowly varying amplitude approximation by the propagation equation [1]

\[
\left( \frac{\partial}{\partial t} + \frac{\partial}{\partial z} \right) \hat{E}(z, t) = igN \tilde{\sigma}_{ba}(z, t).
\] (3.2)
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The atomic evolution is governed by the set of Heisenberg-Langevin equations [3]

\[
\dot{\sigma}_{aa} = -\gamma_a \sigma_{aa} - ig[\hat{E}_\dagger \sigma_{ba} - H.a.] - i[\hat{\Omega}^* \sigma_{ca} e^{-i\Delta k_z} - H.a.] + F_a, \tag{3.3}
\]

\[
\dot{\sigma}_{bb} = \gamma_a \rightarrow b \sigma_{aa} + ig[\hat{E}_\dagger \sigma_{ba} - H.a.] + F_b, \tag{3.4}
\]

\[
\dot{\sigma}_{cc} = \gamma_a \rightarrow c \sigma_{aa} + [\hat{\Omega}^* \sigma_{ca} e^{-i\Delta k_z} + F_c], \tag{3.5}
\]

\[
\dot{\sigma}_{ba} = -\gamma_{ba} \sigma_{ba} + ig\hat{E}[\sigma_{bb} - \sigma_{aa}] + i\hat{\Omega} \sigma_{bc} e^{i\Delta k_z} + F_{ba}, \tag{3.6}
\]

\[
\dot{\sigma}_{ca} = -\gamma_{ca} \sigma_{ca} + i\hat{\Omega}[\sigma_{cc} - \sigma_{aa}] e^{i\Delta k_z} + ig\hat{E} \sigma_{bc} + F_{ca}, \tag{3.7}
\]

\[
\dot{\sigma}_{bc} = -ig\hat{E} \sigma_{cb}^\dagger + i\hat{\Omega}^* \sigma_{ba} e^{-i\Delta k_z}. \tag{3.8}
\]

\[\gamma_a = \gamma_a \rightarrow b + \gamma_a \rightarrow c,\] where \(\gamma_a \rightarrow b\) and \(\gamma_a \rightarrow c\) denote longitudinal decay rates and \(\gamma_{a\beta}\) denotes the transversal decay rate.

To get a first sense of the validity of the simple analytic solution (2.67) the density matrix equations which correspond to equations (3.3)-(3.8) are solved numerically. For this reason a MATLAB programm has been developed which solves the initial boundary value problems for the given partial differential equations. The main part of this program (see appendix A) is based on the MATLAB function pdepe.m [84] which solves initial boundary value problems for small systems of parabolic and elliptic partial differential equations in one space and one time variable. The comparison of the analytic results in section 2.3.2 (see figure 2.8) with the numerical results (see figure 3.1) reveals an excellent agreement of both results.

To analyze the exact conditions for the adiabatic transfer process analytically the first and second order corrections to the adiabatic solution [1,3]

\[\hat{\Psi}(z, t) = \hat{\Psi} \left( z - \int_{t_0}^{t} d\tau v_g(\tau), t_0 \right) \text{ with } v_g(t) = c \cos^2 \theta(t) \tag{3.9}\]

will be taken into account in the following.

### 3.1.2 The adiabaticity parameter \(\varepsilon\)

In section 2.3 the dark and the bright polariton operators, \(\hat{\Psi}\) and \(\hat{\Phi}\), were defined as

\[
\hat{\Psi}(z, t) = \cos \theta(t) \hat{E}(z, t) - \sin \theta(t) \sqrt{N} \hat{\sigma}_{bc}(z, t) e^{i\Delta k z} \quad \text{and} \quad \hat{\Phi}(z, t) = \sin \theta(t) \hat{E}(z, t) + \cos \theta(t) \sqrt{N} \hat{\sigma}_{bc}(z, t) e^{i\Delta k z}. \tag{3.10}\]

The time dependent mixing angle \(\theta\) is defined by \(\tan^2 \theta(t) = \frac{\epsilon^2 N}{\Omega(t)}\). Within the dark and bright state polariton fields the equations of motion for the electric field \(\hat{E}\) and the
3.1. Adiabaticity condition and non adiabatic corrections

Figure 3.1: Propagation of a dark state polariton with envelope $10^{-3}\exp\left\{-\left(z/10\right)^2\right\}$. The mixing angle is rotated from 0 to $\pi/2$ and back according to $\cot \theta(t) = 100\{1 - 0.5 \tanh[0.1(t - 15)] + 0.5 \tanh[0.1(t - 125)]\}$. The coherent amplitude of the polariton $\langle \hat{\Psi} \rangle$ is plotted in (a) and the electric field amplitude $\langle \hat{E} \rangle$ and the matter component $|\langle \tilde{\sigma}_{bc} \rangle|$ in (b) and (c) respectively.

The time $t$ is given in units of $g\sqrt{N}$ and the position $z$ is given in units of $g\sqrt{N}/c$ with $c = 1$. $|\hat{\Psi}(z,t)|$, $|\hat{E}(z,t)|$ and $|\tilde{\sigma}_{bc}(z,t)|$ are given in arbitrary units.

The adiabaticity parameter $\varepsilon$ can be introduced by [3,30]:

$$\varepsilon = \frac{1}{g\sqrt{NT}}.$$  \hspace{1cm} (3.14)
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parameter $\varepsilon$ it is useful to introduce normalized time and space variables $\tilde{t}$ and $\tilde{z}$ according to

$$t = \tilde{t}T \quad \text{and} \quad z = \lambda \tilde{z}, \quad (3.15)$$

where $\lambda = cT$ is the corresponding characteristic length scale. With the normalized variables the equations (3.12) and (3.13) read [3, 30]

$$\left[ \frac{\partial}{\partial \tilde{t}} + \cos^2 \theta \frac{\partial}{\partial \tilde{z}} \right] \tilde{\Psi} = \left[ \left( \frac{\partial \theta}{\partial \tilde{t}} \right) + \sin \theta \cos \theta \frac{\partial}{\partial \tilde{z}} \right] \tilde{\Phi}, \quad (3.16)$$

$$\dot{\tilde{\Phi}} = \varepsilon^2 \sin \theta \left( \frac{\partial}{\partial \tilde{t}} + \gamma_{ba} T \right) \left[ \tan \theta \frac{\partial}{\partial \tilde{t}} \left( \sin \theta \tilde{\Psi} - \cos \theta \tilde{\Phi} \right) \right]. \quad (3.17)$$

For the derivation of the expectation values of $\tilde{\Psi}$ and $\tilde{\Phi}$ it is allowed to neglect the quantum Langevin force $F_{ba}$ in equation (3.13).

3.1.3 Adiabatic condition and condition due to the initial pulse spectrum

To calculate the first and second order corrections to the analytic solution (3.9) the dark and bright polariton operators are rewritten in a polynomial expansion in the adiabaticity parameter $\varepsilon$:

$$\tilde{\Psi} = \tilde{\Psi}^{(0)} + \varepsilon \tilde{\Psi}^{(1)} + \varepsilon^2 \tilde{\Psi}^{(2)} + \ldots, \quad (3.18)$$

$$\tilde{\Phi} = \tilde{\Phi}^{(0)} + \varepsilon \tilde{\Phi}^{(1)} + \varepsilon^2 \tilde{\Phi}^{(2)} + \ldots, \quad (3.19)$$

where the index $(n), n \in \mathbb{N}_0$, denotes the order of the expansion. By assuming that the characteristic time $T$ of the rotation process is larger or of the same order as the decay time $\gamma_{ba}^{-1}$ of the atomic $a \rightarrow b$ transition, i.e. $\gamma_{ba} T \gtrsim 1$, equation (3.17) implies that the zeroth and first order of the evolution of $\tilde{\Phi}$ are equal to zero, i.e. $\tilde{\Phi}^{(0)} = 0$ and $\tilde{\Phi}^{(1)} = 0$. This fact leads to the following effective equation for $\tilde{\Phi}^{(2)}$

$$\tilde{\Phi}^{(2)} = \left( \dot{\theta} T \gamma_{ba} \sin^2 \theta + \dot{\theta}^2 \sin \theta \cos \theta + \ddot{\theta} \sin^2 \theta \right) \tilde{\Psi}^{(0)}$$

$$+ \left( T \gamma_{ba} \sin^2 \theta \tan \theta + \dot{\theta} (\tan^2 \theta + 2 \sin^2 \theta) \right) \frac{\partial}{\partial \tilde{t}} \tilde{\Psi}^{(0)}$$

$$+ \sin^2 \theta \tan \theta \frac{\partial}{\partial \tilde{t}} \tilde{\Psi}^{(0)}. \quad (3.20)$$

With $\tilde{\Phi}^{(0)} = 0$ and $\tilde{\Phi}^{(1)} = 0$ for $\tilde{\Psi}^{(0)}$ equation (3.16) simplifies to

$$\left( \frac{\partial}{\partial \tilde{t}} + \cos^2 \theta \frac{\partial}{\partial \tilde{z}} \right) \tilde{\Psi}^{(0)} = 0, \quad (3.21)$$
which is the linearized wave equation (2.66) for $\hat{\Psi}$ in the adiabatic limit. This equation allows to transform the time derivative of $\hat{\Psi}^{(0)}$ into a spatial derivative and therefore transforms equation (3.20) into

$$
\hat{\Phi}^{(2)} = \sin \theta \left( \dot{\theta} T_{\gamma} b \sin \theta + \dot{\theta}^2 \cos \theta + \ddot{\theta} \sin \theta \right) \hat{\Psi}^{(0)} - \sin^2 \theta \left( T_{\gamma} b \sin \theta \cos \theta + \dot{\theta} \left( 1 + 2 \cos^2 \theta - 2 \sin^2 \theta \right) \right) \frac{\partial}{\partial z} \hat{\Psi}^{(0)} + \sin^3 \theta \cos^3 \theta \frac{\partial^2}{\partial z^2} \hat{\Psi}^{(0)}.
$$

(3.22)

Combining this equation with equation (3.16) leads to an effective equation of motion for the dark state polariton operator which is correct up to the second order perturbation theory in the adiabaticity parameter $\varepsilon$.

$$
\left( \frac{\partial}{\partial t} + \cos^2 \theta \frac{\partial}{\partial z} \right) \left( \hat{\Psi}^{(0)} + \varepsilon \hat{\Psi}^{(1)} + \varepsilon^2 \hat{\Psi}^{(2)} \right) + \varepsilon^2 \left[ f_0(t) + f_1(t) \frac{\partial}{\partial z} + f_2(t) \frac{\partial^2}{\partial z^2} + f_3(t) \frac{\partial^3}{\partial z^3} \right] \hat{\Psi}^{(0)} + O(\varepsilon^3) = 0,
$$

(3.23)

where the time dependent functions $f_l(t), l \in \{0, 1, 2, 3\}$ are given by

$$
f_0(t) = \dot{\theta} \sin \theta \left[ \dot{\theta} T_{\gamma} b \sin \theta + \dot{\theta}^2 \cos \theta + \ddot{\theta} \sin \theta \right],
$$

(3.24)

$$
f_1(t) = \sin^2 \theta \left[ \dot{\theta}^2 \left( 2 \sin^2 \theta - \cos^2 \theta - 1 \right) + \dot{\theta} \sin \theta \cos \theta \right],
$$

(3.25)

$$
f_2(t) = \sin^3 \theta \cos \theta \left[ \dot{\theta} \left( 2 \sin^2 \theta - \cos^2 \theta - 1 \right) - T_{\gamma} b \sin \theta \cos \theta \right],
$$

(3.26)

$$
f_3(t) = \sin^4 \theta \cos^4 \theta.
$$

(3.27)

Equation (3.23) is an inhomogeneous equation in $\hat{\Psi}^{(0)}, \hat{\Psi}^{(1)}$ and $\hat{\Psi}^{(2)}$. Since the second summand in equation (3.23) is proportional to $\varepsilon^2$ a replacement of $\hat{\Psi}^{(0)}$ by $\hat{\Psi}^{(0)} + \varepsilon \hat{\Psi}^{(1)} + \varepsilon^2 \hat{\Psi}^{(2)}$ leads to errors in third order perturbation theory in the adiabaticity parameter $\varepsilon$. Therefore the homogeneous effective equation for the dark state polariton operator $\hat{\Psi}$ up to second order perturbation theory in the adiabaticity parameter $\varepsilon$ is given in non normalized time and space parameters by [3,30]

$$
\left[ \frac{\partial}{\partial t} + c \cos^2 \theta(t) \frac{\partial}{\partial z} \right] \hat{\Psi} = -A(t)\hat{\Psi} + B(t)c \frac{\partial}{\partial z} \hat{\Psi} + C(t)c^2 \frac{\partial^2}{\partial z^2} \hat{\Psi} - D(t)c^3 \frac{\partial^3}{\partial z^3} \hat{\Psi},
$$

(3.28)
where the time dependent coefficients $A(t), B(t), C(t)$ and $D(t)$ are given by

\[
A(t) = \left( \gamma_{ba} + \frac{1}{2} \frac{\partial}{\partial t} \right) \left( \frac{\dot{\theta}^2(t) \sin^2 \theta(t)}{g^2 N} \right), \tag{3.29}
\]

\[
B(t) = \frac{\sin \theta(t)}{3g^2 N} \left( \frac{\partial^2}{\partial t^2} \sin^3 \theta(t) - 6 \ddot{\theta}(t) \sin \theta(t) \cos \theta(t) \right), \tag{3.30}
\]

\[
C(t) = \left( \gamma_{ba} + \frac{1}{2} \frac{\partial}{\partial t} \right) \frac{\sin^4 \theta(t) \cos^2 \theta(t)}{g^2 N}, \tag{3.31}
\]

\[
D(t) = \frac{\sin^4 \theta(t) \cos^4 \theta(t)}{g^2 N}. \tag{3.32}
\]

$A(t)$ describes homogeneous losses due to non adiabatic transitions followed by spontaneous emission. $B(t)$ gives rise to a correction of the polariton propagation velocity. $C(t)$ results in a pulse spreading by dissipation of high spatial frequency components and $D(t)$ leads to a deformation of the polariton. Since all coefficients depend only on time, equation (3.28) can be solved using a Fourier transformation in space $\hat{\Psi}(k,t) = \frac{1}{\sqrt{2\pi}} \int dk \hat{\Psi}(k,t) e^{-ikz}$ and integration in time which gives [3,30]

\[
\hat{\Psi}(k,t) = \hat{\Psi}(k,0) \exp \left\{ i k \int_{t_0}^t d\tau [v_g(\tau) + CB(\tau)] \right\} \times \exp \left\{ -ik^3 c^3 \int_{t_0}^t d\tau D(\tau) \right\} \times \exp \left\{ -\int_{t_0}^t d\tau [A(\tau) + k^2 c^2 C(\tau)] \right\}. \tag{3.33}
\]

The solution of equation (3.33) is illustrated in figure 3.2 where the storage and reloading process of a Gaussian light pulse obtained from the numerical calculation and the solution of equation (3.33) are compared. The last factor in equation (3.33) contains all losses due to non adiabatic corrections. In order to minimize losses caused by non adiabatic effects the exponent of this factor needs to be small compared to unity. To obtain quantitative conditions for the adiabaticity of the rotation it is assumed in the following that the dark state polariton is decelerated and stored for a time interval which goes to infinity. This results in two condition. The first one is given by

\[
\int_{t_0}^\infty A(\tau) d\tau \ll 1 \tag{3.34}
\]

\[
\Leftrightarrow \gamma_{ba} \int_{t_0}^\infty \frac{\dot{\theta}(\tau) \sin^2 \theta(\tau)}{g^2 N} \ll 1 \tag{3.35}
\]

\[
\Leftrightarrow \gamma_{ba} \int_{t_0}^\infty \frac{\dot{\theta}^2(\tau)}{g^2 N + \Omega(\tau)} \ll 1, \tag{3.36}
\]
Figure 3.2: Comparison of approximate analytical (full line) and numerical (points) results (a) electric field amplitude in arbitrary units for $t = 0, 15, 30, 45, 60$ and (b) the same for $t = 90, 105, 120, 135, 150$. The time $t$ is given in units of $g\sqrt{N}$ and the position $z$ is given in units of $g\sqrt{N}/c$. $|\hat{E}(z,t)|$ is given in arbitrary units.
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where it has been taken into account that \( \dot{\theta}(t_0) = \dot{\theta}(t \to \infty) = 0 \) and \( \sin \theta(t) = g \sqrt{N} / \sqrt{g^2 N + \tilde{\Omega}^2(t)} \). This condition is well known from adiabatic passage [85–87] and sets a limit to the rotation velocity \( \dot{\theta}(t) \) of the mixing angle and therefore to the deceleration and acceleration of the dark state polariton. With the characteristic timescale \( T \) and the normalized time \( \tilde{t} \), see equation (3.15), the condition (3.35) can be rewritten in the following form

\[
\frac{l_{\text{abs}}}{c} \int_{t_0}^{\infty} \left( \frac{\partial}{\partial \tilde{t}} \sqrt{\frac{v_g(t)}{c}} \right)^2 d\tilde{t} \ll T, \tag{3.37}
\]

where \( l_{\text{abs}} = \frac{c \gamma_{\text{ba}}}{g^2 N} \) is the absorption length in the absence of EIT. Since the integral in equation (3.37) is always smaller than \( v_g(t_0)/c \) the condition for the adiabatic rotation results in [3,30]

\[
T \gg \frac{l_{\text{abs}} v_g(t_0)}{c}. \tag{3.38}
\]

For realistic experimental parameters the quantity of the right hand side of (3.38) is very small on all relevant time scales. However if the classical control field is switched off suddenly the entire electromagnetic component of the dark state polariton is lost. This situation is illustrated in figure 3.3 where a step function has been used in the numerical calculations to simulate a sudden sharp switch off of the control field. Figure 3.3 (a) shows the propagation of the light pulse in an EIT medium where the cosine of the mixing angle is switched abrupt from 1 to 0 at \( t_g \sqrt{N} = 50 \) and back to 1 at \( t_g \sqrt{N} = 90 \). Apart from some small and rapidly decaying Rabi oscillations the light pulse is immediately absorbed and cannot be regenerated. These losses can be reduced if the cosine of the mixing angle is initially significantly small compared to unity. This situation was observed in the experiment by C. Liu et al. [26] and is demonstrated in figure 3.3 (b) where the cosine of the mixing angle is switched abrupt from 0.1 to 0 at \( t_g \sqrt{N} = 50 \) and back to 0.1 at \( t_g \sqrt{N} = 90 \). The output amplitude of the light pulse is only reduced by 10%.

The second condition which results from equation (3.33) is given by

\[
k^2 c^2 \int_{t_0}^{\infty} C(\tau) d\tau \ll 1 \tag{3.39}
\]

\[
\Leftrightarrow k^2 c^2 \gamma_{\text{ba}} \int_{t_0}^{\infty} \frac{\sin^4 \theta(\tau) \cos^2 \theta(\tau)}{g^2 N} \ll 1. \tag{3.40}
\]

Here it has been used that \( \sin \theta(t_0) \approx 0 \) and \( \cos \theta(t \to \infty) = 0 \). Condition (3.40) can be brought into a transparent form by replacing \( \sin^2 \theta \) with unity and using the relation
3.1. Adiabaticity condition and non adiabatic corrections

Figure 3.3: Propagation of the light pulse in an EIT medium with sudden switching of the mixing angle $\theta(t)$ (a) $\cos^2 \theta(t)$ is switched from 1 to 0 at $t = 50$ and back at $t = 90$ (b) $\cos^2 \theta(t)$ is switched from 0.1 to 0 at $t = 50$ and back at $t = 90$, in this case only 10% of the electromagnetic component of the polariton gets lost. The time $t$ is given in units of $g\sqrt{N}$ and the position $z$ is given in units of $g\sqrt{N}/c$. $|\tilde{\Psi}(z,t)|$, $|\tilde{\mathcal{E}}(z,t)|$ and $|\tilde{\sigma}_{bc}(z,t)|$ are given in arbitrary units.
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\[ v_g(t) = c \cos^2 \theta(t) \] which results in \([3,30]\)

\[
\gamma_{ba} k^2 c \int_{t_0}^{\infty} \frac{v_g(\tau)}{g^2 N} d\tau \ll 1 \quad (3.41)
\]

\[
\Leftrightarrow \gamma_{ba} k^2 c l \ll 1, \quad (3.42)
\]

where \( l = \int_{t_0}^{\infty} v_g(\tau) d\tau \) is the propagation depth of the polariton inside the medium. Condition (3.42) has to be fulfilled for all relevant spatial Fourier frequencies \( k \) which are determined by the inverse of the initial pulse length \( L_p \), i.e. \( k \sim L_p^{-1} \). This leads to \([3,30]\)

\[
l \ll \frac{g^2 N}{\gamma_{ba} c} L_p^2 \Leftrightarrow l \ll \sqrt{\alpha L_p}, \quad (3.43)
\]

where \( \alpha = \frac{g^2 N}{\gamma_{ba} c} \) is again the opacity of the medium without EIT.

The initial spatial pulse length \( L_p \) in the medium is related to the initial group velocity \( v_g(t_0) \) and the initial pulse bandwidth \( \Delta \omega_p(t_0) \) via \( L_p \sim v_g(t_0)/\Delta \omega_p(t_0) \). The transparency width of the medium is given by equation (2.45): \( \Delta t_r(t_0) = \sqrt{\alpha/\tau_d} \), where \( \tau_d \) is the delay time of the pulse in the EIT medium. With this relations the condition (3.43) can be rewritten \([3,30]\)

\[
\frac{\Delta \omega_p(t_0)}{1 + n_g(t_0)} \ll \Delta t_r(t_0) \quad (3.44)
\]

\[
\Leftrightarrow \Delta \omega_p(t_0) \ll \Delta t_r(t_0), \quad (3.45)
\]

where it has been used that the delay time \( \tau_d \) is given by \( \tau_d = n_g(t) l/c \) and the group index \( n_g(t) = \tan \theta(t) \) is defined by equation (2.53). Condition (3.45) states that the initial pulse spectral width \( \Delta \omega_p(t_0) \) has to be much smaller than the initial width \( \Delta t_r(t_0) \) of the transparency window. This is easily satisfied if the probe pulse is not too short which practically prevents light storage of pulses shorter than a few nanoseconds.

### 3.2 Two photon linewidth of light stopping via electromagnetically induced transparency

Essential for the storage process in an atomic dense medium is an explicitly time dependent control field which varies adiabatically as discussed in the previous section. As the group velocity of the polariton approaches zero the spectral width of the transparency window in the EIT medium also narrows to zero. Adiabatic following leads to a narrowing of the spectral width of the probe pulse parallel to the narrowing of the transparency.
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window. Therefore there are no losses due to absorption effects during the slow down process. This requires however that the carrier frequency of the probe pulse and the classical control field are in precise two photon resonance. This is a highly idealized situation, therefore in order to account for more realistic experimental conditions the influence of the two photon detuning on the storage process will be discussed in the following.

In section 3.2.1 the model of section 3.1.1 is further generalized in view of the requirements for two photon detuning. Based on this model a new parameter is introduced section 3.2.2 to handle a finite two photon detuning and the changed polariton dynamics under the influence of the finite two photon detuning are discussed.

3.2.1 Model for two photon detuning

The EIT medium only renders transparent in the case of a precise two photon resonance of the involved fields. For a non vanishing two photon detuning the pulse spectrum will move outside the transparency region at some small but finite value of the group velocity. This situation is illustrated in figure 3.4. Therefore the question arises if any or what values of two photon detuning are tolerable to maintain a sufficiently high fidelity of the storage process due to its use as quantum memory.

To describe the effects of a finite two photon detuning the quasi one dimensional system of Λ-type 3-level atoms with two metastable lower states which was introduced in section 2.2.1 (see figure 2.4) is considered. Within the rotating wave approximation the interaction between the atoms and the fields can be described by the set of Heisenberg-Langevin equations given by [31]

\[
\begin{align*}
\dot{\sigma}_{aa} &= -\gamma_a \sigma_{aa} - ig[\hat{E}^\dagger \sigma_{ba} - H.a.] - i[\tilde{\Omega}^* \sigma_{ca} e^{-i\Delta k z} - H.a.], \\
\dot{\sigma}_{bb} &= \gamma_{a\rightarrow b} \sigma_{aa} + ig[\hat{E}^\dagger \sigma_{ba} - H.a.], \\
\dot{\sigma}_{cc} &= \gamma_{a\rightarrow c} \sigma_{aa} + i[\tilde{\Omega}^* \sigma_{ca} e^{-i\Delta k z} - H.a.], \\
\dot{\sigma}_{ba} &= -(\gamma_{ba} + i\delta_{ab}) \sigma_{ba} + ig\hat{E}[\sigma_{bb} - \sigma_{aa}] + i\tilde{\Omega}\sigma_{bc} e^{i\Delta k z}, \\
\dot{\sigma}_{ca} &= -(\gamma_{ca} + i\delta_{ac}) \sigma_{ca} + i\tilde{\Omega}[\sigma_{cc} - \sigma_{aa}] e^{i\Delta k z} + ig\hat{E}\sigma_{bc}^\dagger + F_{ca}, \\
\dot{\sigma}_{bc} &= -i(\delta_{ab} - \delta_{ac}) - ig\hat{E}\sigma_{ca}^\dagger + i\tilde{\Omega}\sigma_{ba} e^{-i\Delta k z}.
\end{align*}
\]

\[\gamma_a = \gamma_{a\rightarrow b} + \gamma_{a\rightarrow c}, \text{ where } \gamma_{a\rightarrow b} \text{ and } \gamma_{a\rightarrow c} \text{ denote longitudinal decay rates and } \gamma_{a\bar{b}} \text{ denotes the transversal decay rate. The fact that the carrier frequency of the probe pulse is not resonant to the frequency of the classical control field is manifested in the additional detunings } \delta_{ab} \text{ and } \delta_{ac} \text{ in equations (3.49)-(3.51). Since we are only interested in the expectation values of the atomic and the electric field operator, the Quantum Langevin forces are already neglected. To simplify the following analytic discussion it is useful to consider}\]
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Figure 3.4: (a) Transmission spectrum $1 - T(\omega)$ of EIT in units of $\delta/(g\sqrt{N})$ as a function of the group velocity $v_g(t)$ and (b) pulse spectrum $S(\omega)$ for a time dependent EIT medium. The spectral pulse width $\Delta\omega_p$ narrows in the same way as the spectral width $\Delta\omega_r$ of the transparency window. But due to the finite two photon detuning the two spectra are shifted against each other and absorption of the pulse sets in at a finite group velocity $v_g(t)$.
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a resonant probe field, i.e. \( \delta_{ab} = 0 \), while keeping \( \delta_{ac} \neq 0 \). For a simpler nomenclature we define \( \delta = \delta - ac \). The evolution of the probe pulse is described in the slowly varying amplitude approximation by the propagation equation (see section 2.2.1):

\[
\left( \frac{\partial}{\partial t} + \frac{\partial}{\partial z} \right) \hat{E}(z,t) = igN\tilde{\delta}_{ba}(z,t).
\] (3.52)

This equation and the equations for the atomic variables can again be transformed into equations for the dark and bright state polariton operators \( \hat{\Psi} \) and \( \hat{\Phi} \) which yields

\[
\left[ \frac{\partial}{\partial t} + c \cos^2 \theta \frac{\partial}{\partial z} - i\delta \sin^2 \theta \right] \hat{\Psi} = -\left[ \dot{\theta} + \sin \theta \cos \theta \left( \frac{c}{\partial t} + i\delta \right) \right] \hat{\Phi},
\] (3.53)

\[
\hat{\Phi} = \frac{\sin \theta}{g^2N} \left( \frac{\partial}{\partial t} + \gamma \right) \left[ \tan \theta \left( \frac{\partial}{\partial t} - i\delta \right) \right] (\sin \theta \hat{\Psi} - \cos \theta \hat{\Phi}),
\] (3.54)

with \( \gamma \equiv \gamma_{ba} \).

### 3.2.2 Polariton dynamics for finite two photon detuning

In order to find approximative analytic solutions of equations (3.53) and (3.54) the characteristic timescale \( T \) during which the adiabatic rotation of the storage process is fulfilled is introduced in analogy to section 3.1.2. The normalized time and space variables, \( \tilde{t} \) and \( \tilde{z} \), are defined by equation (3.15). It is now possible to identify two expansion parameters \( \varepsilon_1 \) and \( \varepsilon_2 \). The first expansion parameter \( \varepsilon_1 = (g\sqrt{NT})^{-1} \) is the adiabaticity parameter defined by equation (3.14). The second expansion parameter \( \varepsilon_2 \) characterizes the magnitude of the two photon detuning and is given by [31]

\[
\varepsilon_2 = \frac{\delta}{g\sqrt{N}}.
\] (3.55)

Since the non adiabatic effects during the propagation of the dark state polariton has already been discussed in section 3.1.3 it is assumed that the characteristic time \( T \) is much larger than the decay time, i.e. \( \gamma T \gg 1 \). With the normalized time and space variables and the assumption \( \gamma T \gg 1 \) equations (3.53) reads [31]

\[
\left[ \varepsilon_1 \left( \frac{\partial}{\partial \tilde{t}} + \cos^2 \theta \frac{\partial}{\partial \tilde{z}} \right) - i\varepsilon_2 \sin^2 \theta \right] \hat{\Psi} = -\left[ \dot{\theta} + \sin \theta \cos \theta \left( \frac{c}{\partial \tilde{t}} + i\delta \right) \right] \hat{\Phi},
\] (3.56)

Equation (3.54) transforms to

\[
\hat{\Phi} = \frac{\gamma T \sin^2 \theta}{\cos \theta} \left[ \varepsilon_1 \frac{\partial}{\partial \tilde{t}} - i\varepsilon_1 \varepsilon_2 \right] (\sin \theta \hat{\Psi} - \cos \theta \hat{\Phi}).
\] (3.57)
3.2. Two photon linewidth of light stopping via electromagnetically induced transparency

In order to calculate the first and second order corrections to the analytic solution (3.9) the dark and bright polariton operators are rewritten in a polynomial expansion in the perturbation parameters $\varepsilon_1$ and $\varepsilon_2$:

\[
\hat{\Psi} = \hat{\Psi}^{(00)} + \varepsilon_1 \hat{\Psi}^{(10)} + \varepsilon_2 \hat{\Psi}^{(01)} + \varepsilon_1^2 \hat{\Psi}^{(20)} + \varepsilon_2^2 \hat{\Psi}^{(02)} + \varepsilon_1\varepsilon_2 \hat{\Psi}^{(11)} + \ldots, \\
\hat{\Phi} = \hat{\Phi}^{(00)} + \varepsilon_1 \hat{\Phi}^{(10)} + \varepsilon_2 \hat{\Phi}^{(01)} + \varepsilon_1^2 \hat{\Phi}^{(20)} + \varepsilon_2^2 \hat{\Phi}^{(02)} + \varepsilon_1\varepsilon_2 \hat{\Phi}^{(11)} + \ldots, 
\]

where the index $(n, m)$, $n, m \in \mathbb{N}_0$, denotes the order of the expansion due to the perturbative parameters $\varepsilon_1$ and $\varepsilon_2$. Since it is assumed that $\varepsilon_1$ and $\varepsilon_2$ have the same order of magnitude they can be treated equally and therefore the sum $m + n$ gives the total order of the perturbative expansion.

In the adiabatic limit and for small detuning the equations (3.56) and (3.57) simplify for $\hat{\Psi}^{(00)}$ and $\hat{\Phi}^{(00)}$ to

\[
\left( \frac{\partial}{\partial t} + c\cos^2\theta \frac{\partial}{\partial z} - i\delta \sin^2\theta \right) \hat{\Psi}^{(00)}(z, t) = 0, \\
\hat{\Phi}^{(00)} = 0. 
\]

In this limit the bright state polariton is not excited. The dark state polariton propagates with a form stable envelope with group velocity $v_g(t) = c\cos^2\theta(t)$. The small two photon detuning simply causes a time dependent additional phase factor. The solution of equation (3.60) is given by [31]

\[
\hat{\Psi}^{(00)}(z, t) = \hat{\Psi}^{(00)} \left( z - c \int_{t_0}^t \cos^2\theta(\tau)d\tau, t_0 \right) \exp \left\{ i\delta \int_{t_0}^t \sin^2\theta(\tau)d\tau \right\}. 
\]

Equation (3.57) furthermore implies that not only the zeroth order $\hat{\Phi}^{(00)}$ of the bright state polariton is equal to zero but also the first order $\hat{\Phi}^{(10)}$ and $\hat{\Phi}^{(01)}$ vanishes. This leads to the following effective equation for the second order of perturbation for the bright state polariton

\[
\varepsilon_1^2 \hat{\Phi}^{(20)} + \varepsilon_2^2 \hat{\Phi}^{(02)} + \varepsilon_1\varepsilon_2 \hat{\Phi}^{(11)} = \frac{\gamma T \sin^2\theta}{\cos\theta} \left[ \varepsilon_1^2 \frac{\partial}{\partial t} - i\varepsilon_1\varepsilon_2 \right] \sin\theta \hat{\Psi}^{(00)}. 
\]

The equation (3.60) for the zeroth order of the dark state polariton allows to replace the time derivative $\frac{\partial}{\partial t} \hat{\Psi}^{(00)}$ by $(i\delta \sin^2\theta - c\cos^2\theta \frac{\partial}{\partial z}) \hat{\Psi}^{(00)}$. A longer calculation gives the following excitation of the bright state polariton up to second order due to non adiabatic couplings and the non vanishing two photon detuning

\[
\hat{\Phi}(z, t) = \frac{\gamma \sin^2\theta}{g^2N} \left[ \hat{\theta} - i\delta \sin\theta \cos\theta - c\sin\theta \cos\theta \frac{\partial}{\partial z} \right] \hat{\Psi}(z, t), 
\]

\[
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\]
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where it has been used that a replacement of \( \hat{\Psi}^{(00)} \) by \( \hat{\Psi}^{(00)} + \varepsilon_1 \hat{\Psi}^{(10)} + \ldots + \varepsilon_1 \varepsilon_2 \hat{\Psi}^{(11)} \) just causes errors in the third order of the perturbative treatment. Substituting this result into the right hand side of equation (3.56) gives an effective equation of motion for the dark state polariton operator \( \hat{\Psi} \) up to second order perturbation theory in \( \varepsilon_1 \) and \( \varepsilon_2 \) [31]

\[
\left( \frac{\partial}{\partial t} + c \cos^2 \theta(t) \frac{\partial}{\partial z} - i \delta \sin^2 \theta(t) \right) \hat{\Psi} = -[A_0(t) + \delta^2 A_1(t)] \hat{\Psi} - i \delta B_0(t) c \frac{\partial}{\partial z} \hat{\Psi} + C_0(t)c^2 \frac{\partial^2}{\partial z^2} \hat{\Psi},
\]

(3.65)

where the time dependent coefficients \( A_0(t), A_1(t), B_0(t) \) and \( C_0(t) \) are given by

\[
A_0(t) = \frac{\gamma}{g^2 N} \hat{\theta}^2(t) \sin^2 \theta(t), \quad (3.66)
\]

\[
A_1(t) = \frac{\gamma}{g^2 N} \sin^4 \theta(t) \cos^2 \theta(t), \quad (3.67)
\]

\[
B_0(t) = -\frac{2\gamma}{g^2 N} \sin^4 \theta(t) \cos^2 \theta(t), \quad (3.68)
\]

\[
C_0(t) = \frac{\gamma}{g^2 N} \sin^4 \theta(t) \cos^2 \theta(t). \quad (3.69)
\]

Since all coefficients depend only on time, equation (3.65) can be solved by a Fourier transformation in space \( \hat{\Psi}(z,t) = \frac{1}{\sqrt{2\pi}} \int dk \hat{\Psi}(k,t)e^{-ikz} \) and integration in time which gives [31]

\[
\hat{\Psi}(k,t) = \hat{\Psi}(k,t_0) \exp \left\{ i \int_{t_0}^t [ck \cos^2 \theta(\tau) + \delta \sin^2 \theta(\tau)]d\tau \right\} \times \exp \left\{ -\int_{t_0}^t [A_0(\tau) - c^2 k^2 C_0(\tau)]d\tau \right\} \times \exp \left\{ -\int_{t_0}^t [\delta^2 A_1(\tau) - c k \delta B_0(\tau)] \right\}. \quad (3.70)
\]

The first factor leads to the changed group velocity \( v_g(t) = c \cos^2 \theta(t) \) and the additional phase factor which has been discussed due to the solution (3.62) in the adiabatic limit and for small detunings. The factor which includes \( A_0(t) \) causes dissipative losses due to non adiabatic couplings and leads to the restriction (3.38) of the speed of the adiabatic rotation and was discussed in section 3.1.3. \( C_0(t) \) leads to dissipative losses of the high frequency components of the dark state polariton and restricts the spectral width of the light pulse (see equation (3.45).

The factor which contains \( A_1(t) \) and \( B_0(t) \) limit the two photon linewidth of the storage process. The term which contains \( A_1(t) \) is a frequency independent dissipation term. The term which contains \( B_0(t) \) accounts for deviations from this value depending on the \( k \).
space Fourier frequency of the dark state polariton. The frequency independent losses
due to a finite two photon detuning are thus given by

$$\exp \left\{ -\frac{\gamma \delta^2}{g^2N} \int_{t_0}^{t} \cos^2 \theta(\tau) \sin^4 \theta(\tau) d\tau \right\}. \quad (3.71)$$

To minimize these losses the exponent of this factor needs to be small compared to unity,
which results for an infinite storage period in [31]:

$$-\frac{\gamma \delta^2}{g^2N} \int_{t_0}^{\infty} \cos^2 \theta(\tau) \sin^4 \theta(\tau) d\tau \ll 1. \quad (3.72)$$

Equation (3.72) shows that there are only contributions to the losses for times when
neither $\sin \theta(t)$ nor $\cos \theta(t)$ are zero. $\sin \theta(t) = 0$ corresponds to the limit of an infinite Rabi
frequency $\tilde{\Omega}(t)$ of the control field. In this case the Autler Townes splitting of the upper
level of the $\Lambda$ level atomic scheme caused by the control field suppresses any absorption
despite the finite two photon detuning. If $\cos \theta(t) = \pi/2$ the dark state polariton is entirely
matter like and thus two photon detuning is of no relevance. The characteristic time for
rotating the mixing angle $\theta(t)$ from 0 to $\pi/2$, i.e. the time of transferring the dark state
polariton from pure electromagnetic to pure matter excitations, can be therefore defined
by [31]

$$T \equiv \int_{t_0}^{\infty} \cos^2 \theta(\tau) \sin^4 \theta(\tau) d\tau. \quad (3.73)$$

Together with equation (3.72) this leads to the following condition for the two photon
detuning $\delta$ [31]:

$$\delta \ll \delta_{2ph} = \frac{g\sqrt{N}}{\sqrt{\gamma T}}, \quad (3.74)$$

where $\delta_{2ph}$ is the two photon linewidth of the storage process. It is important to note
that $\delta_{2ph}$ is proportional to the collective Rabi frequency $g\sqrt{N}$. Therefore in an optically
thick medium rather large two photon detuning can be tolerated. This fact is illustrated
in figure 3.5 where the normalized integrated intensity of the dark state polariton after
storage and release is plotted as a function of $\varepsilon_2 = \frac{\delta}{g\sqrt{N}}$. 

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Figure 3.5: (a) Absolute value of the dark state polariton amplitude at $t = 0$ (input) and after storage and release at $t = 150$ (output) for finite two photon detuning $\delta = 0.2g\sqrt{N}$ and $\delta = 0.5g\sqrt{N}$, $\gamma T = 1$. Shown are the analytical expressions (full line) obtained from equation (3.62) and a numerical result (dotted line) obtained by solving the full Maxwell Bloch equations corresponding to equations (3.46)-(3.51). The mixing angle $\theta(t)$ is rotated according to $\cot\theta(t) = 100 - 50\tanh[0.1(t-15)] + 50\tanh[0.1(t-125)]$. (b) Integrated output intensity of the dark state polariton as a function of $\delta/(g\sqrt{N})$ normalized to its value at $\delta = 0$ obtained from the numerical solution of the Maxwell Bloch equations (dots). The full line shows the analytic approximation (3.71). The time $t$ is given in units of $g\sqrt{N}$ and the position $z$ is given in units of $g\sqrt{N}/c$. $|\Psi(z, t)|$ is given in arbitrary units.
3.3 Light storage in ion Coulomb crystals

Experimental realizations of light pulse storage in optically thick media were realized by using ultra cold atomic ensembles [26] or an hot atomic gas [27]. Another possible implementation are large ion Coulomb crystals as an atomic ensemble. These ion crystals can be realized in Penning or Paul traps by laser-cooling the trapped ions to temperatures in the millikelvin range. [88–92]. The cloud of ions then condenses and forms a quasicrystalline spatial structure [93]. The open configuration of a linear Paul trap [94] provides easy access for lasers, making it possible to use the storage scheme of section 2.4 to store the quantum information of photons in ion Coulomb crystals. For this case numerical simulations were done in collaboration with M. Drewsen and A.V. Mortensen [83] at the department of physics and astronomy at the University of Aarhus (Denmark). The calculations use realistic experimental parameters and served the experimental group in Aarhus to build up a new experimental setup. In section 3.3.1 the calculations of the storage and release process of quantum states of single photon fields will be presented. They go beyond the adiabatic limit discussed in section 2.4 and show the dependence of the fidelity of the storage process on the cavity decay rate.

3.3.1 Numerical simulations of photon storage in non ideal cavity systems

The technique proposed in [2] and reviewed in section 2.4 uses an optically dense many atom system inside an optical resonator to avoid the requirements of a strong coupling condition of single atom cavity. Photons couple to collective excitations associated with a large number of atoms which enhances the coupling by the square root of the number of atoms.

In analogy to section 2.4.1 a single mode cavity filled with identical Λ-type atoms is considered (see figure 2.12). One of the two optically allowed transitions is coupled by the cavity mode, the other one is coupled by a time dependent coherent classical control field. As shown in [2] the control field remains essentially unaffected by the interaction and can therefore be presented by a time dependent Rabi frequency $\Omega(t)$. In the case of two photon resonance this control field induces transparency for the cavity field. The associated linear dispersion substantially reduces the group velocity of the latter. The velocity reduction is associated with the formation of a quasi particle with electromagnetic and atomic components.

By changing the Rabi frequency $\Omega(t)$ of the classical control field one can change the coupling of the quasi particle to the free space modes. In section 2.4.2 it has been shown...
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that in the adiabatic limit it is possible to load the cavity system with any excitation resulting from an incoming photon wave packet, provided the shape of the wave packet is known, and to subsequently release it back into a desired photon wave packet after some storage time $t_s$.

To discuss the limits of the transfer process, an effective one dimensional model with a Fabry Perot type cavity (see figure 2.11) is considered. The propagation of the incoming and outgoing modes are assumed to be parallel to the $z$-axis. $z = 0$ denotes the position of the partially transmitting input mirror of the cavity. The other mirror of the cavity is assumed to be 100% reflecting. In the following the single photon state of the input field is described by the envelope function $\Phi_{\text{in}}(z, t)$ in equation (2.84) with the normalization condition $\int dz |\Phi_{\text{in}}(z, t)|^2 = 1$, where $L$ is the quantization length. During interaction of the single photon wave packet with the combined system of cavity modes and atoms, the total state vector can be written in the form

$$|\Psi(t)\rangle = b(t)|b, 1, 0_k\rangle + c(t)|c, 0, 0_k\rangle + a(t)|a, 0, 0_k\rangle + \sum_k \xi_k(t)|b, 0, 1_k\rangle,$$

where for example $|b, 1, 0_k\rangle$ denotes the state corresponding to the atomic system in the collective state $|b\rangle$, the cavity mode in the single photon state and no photons in the outside modes. The equations of motion for the amplitude functions are given in equations (2.87)-(2.90). Under the condition of perfect two photon resonance one finds for the envelope of the outgoing wave packet

$$\Phi_{\text{out}}(z = 0, t) = \Phi_{\text{in}}(z = 0, t) - i\gamma b(t),$$

where $\kappa$ is the coupling constant of the free space modes and the selected cavity mode and $\gamma$ is the empty cavity decay rate. Here the system of equations (2.87)-(2.90) has been solved analytically in the adiabatic limit $g^2N \gg \gamma\gamma_a$, where $\gamma_a$ is the decay rate of the excited state population. In order to avoid reflection from the input mirror during the loading process, in the adiabatic limit the mixing angle needs to be changed according to the impedance matching condition (2.104).

In the following the behavior of the system if the adiabatic condition $g^2N \gg \gamma\gamma_a$ is not or only barely fulfilled will be discussed. The resulting equations of motion

$$\frac{d}{dt}a(t) = -\frac{\gamma_a}{2}a(t) - ig\sqrt{N}b(t) - i\Omega(t)c(t), \quad (3.77)$$

$$\frac{d}{dt}b(t) = -ig\sqrt{N}a(t) - i\kappa \Phi_{\text{in}}(z = 0, t) - \frac{\gamma}{2} b(t), \quad (3.78)$$

$$\frac{d}{dt}c(t) = -i\Omega(t)a(t), \quad (3.79)$$
3.3. Light storage in ion Coulomb crystals

Figure 3.6: Input and Output wave functions for a hyperbolic secant input wave packet at space position $z = 0$ in the adiabatic limit and optimized $\cos \theta(t)$. At $t \approx 15$ the mixing angle $\theta(t)$ is time reversed to release the photon wave packet. The time $t$ is given in units of $g\sqrt{N}$ and the position $z$ is given in units of $g\sqrt{N}/c$. $|\Phi_{in}|$ and $|\Phi_{out}|$ are given in arbitrary units.

are solved numerically. In the simulations a normalized hyperbolic secant input pulse and the corresponding optimized mixing angle from equation (2.104) are used. In order to release the stored photon into free field photons after some storage time $t_s$, one can simply reverse the rotation of the mixing angle $\theta(t)$. The equations of motion (3.77)-(3.79) are solved for a fixed value of $\gamma_a$ but with a varying cavity decay rate $\gamma$.

As a measure for the quality of the photon storage process the fidelity

$$F = \frac{\int d\tau |\Phi_{out}(z = 0, \tau)\Phi_{in}(z = 0, \tau - t_s)|^2}{\int d\tau |\Phi_{in}(z = 0, \tau)|^2}$$

(3.80)

is calculated in dependence on $\frac{\gamma_a}{g\sqrt{N}}$. In the adiabatic limit the fidelity is very close to unity and the shape of the outgoing wave packet is again a hyperbolic secant as can be seen in figure 3.6. The dependence of the outgoing wave packet on the change of the empty cavity decay rate $\gamma$ or the decay rate $\gamma_a$ of the excited state population and thereby violating the adiabaticity condition is shown in figure 3.7. Since the impedance matching condition (2.104) is only strictly valid for the adiabatic limit, by violating this condition a part of the incoming wave packet is reflected.

The numerical results discussed in the following are calculated for $\gamma_a = 0.1g\sqrt{N}$ whereas $\gamma$ has been varied between $0.1g\sqrt{N}$ and $100g\sqrt{N}$. As can be seen in figure 3.8 the fidelity
Figure 3.7: (a) Output wave functions for a hyperbolic secant input wave packet at space position $z = 0$ for $\gamma/(g\sqrt{N}) = 0.1, 10, 50, 100$ (lines a-d) and $\gamma_a/(g\sqrt{N}) = 0.1$. At $t \approx 15$ the mixing angle $\theta(t)$ is time reversed to release the photon wave packet. The time $t$ is given in units of $g\sqrt{N}$ and the position $z$ is given in units of $g\sqrt{N}/c$. $|\Phi_{\text{out}}|$ is given in arbitrary units.

(b) Output wave functions for a hyperbolic secant input wave packet at space position $z = 0$ for $\gamma_a/(g\sqrt{N}) = 0.1, 10, 50, 100$ (lines a-d) and $\gamma/(g\sqrt{N}) = 0.1$. At $t \approx 15$ the mixing angle $\theta(t)$ is time reversed to release the photon wave packet. The time $t$ is given in units of $g\sqrt{N}$ and the position $z$ is given in units of $g\sqrt{N}/c$. $|\Phi_{\text{out}}|$ is given in arbitrary units.
3.3. Light storage in ion Coulomb crystals

Figure 3.8: Logarithm fidelity $\ln(F)$ of the storage process for the storage period $t_s = 15$, logarithm fidelity $\ln(F_{\text{eff}})$ of the storage process for the effective storage period $t_{s,\text{eff}}$ which maximizes the overlap integral (3.80) and logarithm of the norm $\ln(|n|)$ of the released wave packet without components reflected during the loading process. The empty cavity decay rate $\gamma$ and the decay rate $\gamma_a$ of the excited state population are given in units of $g\sqrt{N}/c$. $\gamma_a = 0.1$.

of the system exhibits an approximately exponential decay with increasing cavity decay rate. The calculation has been carried out for a storage time $t_s = 15T$, by comparing the output wave with the input wave shifted by $t_s$. This slightly underestimates the fidelity of the system since non adiabatic effects also change the effective storage time. This can be corrected by replacing $t_s$ by an effective storage time $t_{s,\text{eff}}$ which maximizes the overlap integral in equation (3.80). As can be seen in figure 3.8 the fidelity of the system is in this case very close to the norm of the released wave packet (without components reflected during the loading process), which represents an upper limit of the fidelity. The effective storage time $t_{s,\text{eff}}$ increases with increasing cavity decay rate due to the change of the shape of the released wave packet. The non adiabatic losses are mainly caused by increased reflections of the input wave packet at $t = 0$. It is interesting to note, that the storage fidelity may be further enhanced in this case by detecting these reflected photons and post select storage events without reflection.
Chapter 4

Decoherence in single mode quantum memories with collective ensembles

One of the essential ingredients for quantum information processing with photons as information carrier is a reliable quantum memory which allows to store and reload the quantum state of the photons after a certain storage period. Such a memory is essential for various fields of quantum information processing like network quantum computing [32], for secure long distance quantum communication [33, 34] and for quantum teleportation [35, 36]. The conceptually simplest storage system for photonic qubits are individual atoms where coherent transfer techniques have been developed that allow a controlled and reversible transfer of quantum information from light to an individual atom [16,17]. However cavity QED settings require the experimentally challenging strong coupling regime to achieve reasonable fidelities for the state transfer. On the other hand the scheme discussed in sections 2.3 and 2.4 which uses the enhanced coupling of an atomic ensemble to light has less challenging requirements and a coherent and reversible transfer technique for photons [1–3] has been proposed and in part experimentally implemented [26,27].

In atomic ensembles the quantum information is stored in collective many particle states which are highly entangled states and are known to be very sensitive to decoherence processes. In fact one might naively expect that the lifetime of quantum correlations drastically decreases with the number of atoms involved in the storage state. In this case a quantum memory with collective atomic ensembles would be practically useless for quantum information storage. Therefore in this chapter the influence of various decoherence mechanisms on the fidelity of a quantum memory will be analyzed. In section 4.1 a basis of dark and bright state polariton operators for the considered Hilbert space is introduced. In the following section 4.2 it is shown that each quantum state of the radiation field stored in the atomic ensemble corresponds to a whole class of many particle
4.1 Dark and bright state polaritons

The most essential properties of a quantum memory based on collective excitations can be understood by looking at the case of a single mode of the radiation field as realized for example in a single mode cavity and has been discussed in detail in sections 2.3 and 3.3. In analogy to them now an ensemble of $N$ three level atoms with internal states $|a\rangle$, $|b\rangle$ and $|c\rangle$ are considered, as illustrated in figure 4.1. One of the two optically allowed transitions is coupled by a cavity mode. The other allowed transition is coupled via a classical coherent control field with time dependent Rabi frequency $\Omega(t)$. The dynamics of the system are described by a non Hermitian Hamiltonian

$$\hat{H} = \hbar \omega \hat{a}^\dagger \hat{a} + \hbar (\omega_a - i\gamma) \sum_{j=1}^{N} \hat{\sigma}_a^j + \hbar \omega_c \sum_{j=1}^{N} \hat{\sigma}_c^j$$

$$+ \hbar g \sum_{j=1}^{N} (\hat{a} \hat{\sigma}_a^j + \hat{a}^\dagger \hat{\sigma}_a^j \hat{a}^\dagger) + \hbar \sum_{j=1}^{N} (\Omega(t)e^{-i\nu t} \hat{\sigma}_a^j + \Omega(t)e^{i\nu t} \hat{\sigma}_a^j^\dagger),$$

(4.1)

**Figure 4.1:** Three level atoms in Λ configuration interacting with a quantum field and a classical control field with Rabi frequency $\Omega(t)$. $g$ is the coupling constant between the quantum field and the atoms.
where the atomic flip operators $\hat{\sigma}_{\alpha\beta}^j$ are defined in analogy to equation (2.16) and the vacuum Rabi frequency $g$ of the considered cavity mode is assumed to be equal for all atoms. Furthermore the energy of state $|\alpha\rangle$ is chosen to be equal to zero, i.e. $E_b = \hbar \omega_b = 0$. The complex Hamiltonian emerges from a Lindblad Liouville operator that includes the decay from the excited state to other internal states of the atoms after projection onto the subspace spanned by the states $|\alpha\rangle$, $|\beta\rangle$ and $|\gamma\rangle$. Therefore the imaginary part of the Hamiltonian (4.1) takes into account losses from the excited state $|\alpha\rangle$ for example via spontaneous emission. The model does not include relaxation from the excited state $|\alpha\rangle$ back into the lower states $|\beta\rangle$ and $|\gamma\rangle$. Spontaneous emission into the resonator mode accompanied by a transition from state $|\alpha\rangle$ into state $|\beta\rangle$ is however automatically included in the model.

If all atoms are initially prepared in state $|\beta\rangle$ the interaction given in equation (4.1) only couples totally symmetric Dicke like states [80] which were introduced in section 2.4.1 and are given by

\begin{align}
|\beta\rangle_N &= |b_1 \ldots b_N\rangle, \quad (4.2) \\
|\alpha^1\rangle_N &= \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |b_1 \ldots a_j \ldots b_N\rangle, \quad (4.3) \\
|\gamma^1\rangle_N &= \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |b_1 \ldots c_j \ldots b_N\rangle, \quad (4.4) \\
|\beta^2\rangle_N &= \frac{1}{\sqrt{2N(N-1)}} \sum_{j=1}^{N} \sum_{k=1}^{N} |b_1 \ldots a_j \ldots a_k \ldots b_N\rangle, \quad (4.5) \\
|\gamma^2\rangle_N &= \frac{1}{\sqrt{2N(N-1)}} \sum_{j=1}^{N} \sum_{k=1}^{N} |b_1 \ldots c_j \ldots c_k \ldots b_N\rangle, \quad (4.6) \\
|\alpha^1\gamma^1\rangle_N &= \frac{1}{\sqrt{2(N-1)}} \sum_{j=1}^{N} \sum_{k=1}^{N} |b_1 \ldots a_j \ldots c_k \ldots b_N\rangle, \quad \text{etc. (4.7)}
\end{align}

The couplings within the atomic sub system corresponding to a single and a double excitation are shown in figure 4.2. The set of collective states can be divided into groups with a specific excitation number $n$ and an atom number $N$ which will become important in the discussion of equivalence classes of storage states in section 4.2.

Since the influence of a finite two photon detuning on the transfer process itself has been discussed in section 3.3 two photon resonance and for simplicity single photon resonance
4.1. Dark and bright state polaritons

Figure 4.2: Interaction of a single and double excited mode with $N$ three level atoms in Λ configuration in the collective state basis.

are assumed in the following. Under these conditions the interaction of the $N$ atom system with the quantized radiation mode $|n\rangle$ has a family of dark eigenstates with zero eigenvalue

$$|D, n\rangle_N = \sum_{k=0}^{n} \xi_{nk} [-\sin \theta(t)]^k [\cos \theta(t)]^{n-k} |c_k, n - k\rangle_N,$$

with $\xi_{nk} = \sqrt{\frac{n!}{k!(n-k)!}}$ and $\tan \theta(t) = \frac{g\sqrt{N}}{\Omega(t)}$. (4.8)

Adiabatic rotation of the mixing angle $\theta(t)$ from 0 to $\pi/2$ leads to a complete and reversible transfer of all photonic states to a collective atomic excitation if the maximum number of photons $n$ is less than the number of atoms $N$. One notices that there are no transitions between dark states with different excitation number. Due to the symmetry of the interaction Hamiltonian this even holds if non adiabatic corrections are taken into account.

If the initial quantum state of the single mode light field is in a mixed state described by a density matrix $\hat{\rho}_f = \sum_{m,n} \rho_{mn} |m\rangle\langle n|$ the adiabatic transfer process generates a quantum state of collective excitations given by

$$\sum_{m,n} \rho_{mn} |m\rangle\langle n| \otimes |b\rangle_{NN}\langle b| = \sum_{m,n} \rho_{mn} |D, m\rangle_{NN}\langle D, n|.$$ (4.9)

As discussed in section 2.3 the dark states of the $N$ atom system can be identified as quasi particle excitations of the dark state polariton operator $\hat{\Psi}$ in the Hilbert space of the atoms and the cavity mode

$$|D, n\rangle_N = \frac{1}{\sqrt{n!}} \left(\hat{\Psi}^\dagger\right)^n |b, 0\rangle_N,$$ (4.10)
where $|b\rangle$ is the total ground state of the $N$ atom system and $|0\rangle$ is the vacuum state of the cavity mode. The dark state polariton operator is as defined in section 2.3.1 given by

$$\hat{\Psi} = \cos \theta(t) \hat{a} - \sin \theta(t) \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{\sigma}_{bc}^j. \quad (4.11)$$

It is a mixture of the resonator mode and the collective spin corresponding to the $|b\rangle \leftrightarrow |c\rangle$ transition. Associated with the dark state polariton operator is a bright state polariton operator given by

$$\hat{\Phi}_0 = \sin \theta(t) \hat{a} + \cos \theta(t) \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{\sigma}_{bc}^j. \quad (4.12)$$

To obtain a complete set of operators in the space of the cavity mode and the $N$ atoms in internal states $|b\rangle$ and $|c\rangle$ the operators $\hat{\Phi}_l$, $l \in \{1, 2, \ldots, N-1\}$, are introduced in the following and will be referred to as bright state polariton operators. They are defined by

$$\hat{\Phi}_l = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{\sigma}_{bc}^j \exp \left\{ 2\pi i \frac{l_j}{N} \right\} \text{ for } l \in \{1, 2, \ldots, N-1\}. \quad (4.13)$$

In the limit of a small number of atomic excitations the polariton operators obey approximately bosonic commutation relations:

$$[\hat{\Psi}, \hat{\Psi}^\dagger] \approx 1 \text{ and } [\hat{\Phi}_j, \hat{\Phi}_k^\dagger] \approx \delta_{jk} \text{ with } j, k \in \{0, 1, 2, \ldots, N-1\}, \quad (4.14)$$

where $\hat{\sigma}_{cc}^j \approx 0$ and $\hat{\sigma}_{bb}^j \approx 1$ has been used.

From equations (4.4) and (4.6) it can be seen that the collective storage states $|c^n\rangle_N$ are maximally entangled $N$ particle states which are known to be very sensitive to decoherence. If for example in the storage state $|c^1\rangle_N$ the first atom undergoes a transition from level $b$ to level $c$ the resulting state is almost orthogonal to the original one

$$\frac{1}{\sqrt{N}} (|c_1, b_2, \ldots, b_N\rangle + |b_1, c_2, \ldots, b_N\rangle + \ldots + |b_1, b_2, \ldots, c_N\rangle) \rightarrow \frac{1}{\sqrt{N}} (|c_1, b_2, \ldots, b_N\rangle + |c_1, c_2, \ldots, b_N\rangle + \ldots + |c_1, b_2, \ldots, c_N\rangle). \quad (4.15)$$

This situation can be identified with a spin flip between the two internal states $|b\rangle$ and $|c\rangle$ of the atoms, used for the storage and is illustrated for four atoms in figure 4.3.

To get a first sense of the influence of such an effect on the quantum memory with collective atomic excitations the total probability $P_{error}$ to end up in an orthogonal state is estimated for the $N$ atoms. If $p$ denotes the probability that one atom undergoes a transition from $b_j$ to $c_j$ caused by environmental interactions, one would naively expect that the total probability $P_{error}$ scales like

$$P_{error} = 1 - (1 - p)^N \approx pN. \quad (4.16)$$
4.2. Equivalence classes

Figure 4.3: Demonstration of a spin flip operation $\hat{\sigma}_1^c$ in the storage state $|c^1\rangle_N$ for $N = 4$. Spin up stands for an atom in state $|c\rangle$ and spin down has to be identified with an atom in state $|c\rangle$.

This would mean that the collective quantum memory has an $N$ times enhanced sensitivity to decoherence compared to single atom memories. It will be shown that this conclusion is incorrect which is because of the existence of so called equivalence classes, as will be explained in the following sections.

4.2 Equivalence classes

From equations (4.11) and (4.12) the cavity mode field operator $\hat{a}$ can be expressed by the dark and bright state polariton operator, $\hat{\Psi}$ and $\hat{\Phi}_0$:

$$\hat{a} = \cos \theta(t) \hat{\Psi} + \sin \theta(t) \hat{\Phi}_0.$$  (4.17)

This equation shows that for the resonator mode which is relevant for the outgoing wave packet after the storage and reloading process only excitations caused by the dark state polariton operator $\hat{\Psi}$ and the bright state polariton operator $\hat{\Phi}_0$ are important. All other excitations caused by the bright state polariton operators $\hat{\Phi}_l$, $l \in \{1, 2, \ldots, N - 1\}$ do not contribute to the outgoing wave packet. Furthermore after storage of photon states in the atomic system the electromagnetic excitations are regenerated by rotating $\theta(t)$ back from $\pi/2$ to 0, thus only excitations caused by the dark state polariton operator $\hat{\Psi}$ are relevant. From this one can draw a very important conclusion for the storage state: After the adiabatic rotation of the mixing angle $\theta(t)$ from 0 to $\pi/2$ to store the quantum information of the incoming light pulse only the reduced density operator

$$\hat{\rho} = \text{tr}_{\hat{\Phi}_0, \hat{\Phi}_1, \ldots, \hat{\Phi}_{N-1}} \{W\}$$  (4.18)
4.2. Equivalence classes

is relevant. Here $W$ denotes the total density operator of the combined atom cavity system after the storage process. For this reason all states of the system which have the same number of dark state polariton excitations but an arbitrary number of excitations caused by any bright state polariton operator are equivalent from the point of view of quantum information storage and are called equivalence classes:

$$|D, n\rangle_N = \left\{ \left( \hat{\Phi}^\dagger_i \right)^k \left( \hat{\Phi}^\dagger_j \right)^l \ldots \left( \hat{\Psi}^\dagger \right)^n |b, 0\rangle_N \right\}.$$  (4.19)

These equivalence classes can be illustrated by identifying the $N$ atoms with the relevant internal states $|b\rangle$ and $|c\rangle$ with $N$ spin-$\frac{1}{2}$-particles [80]. The obvious basis for a collection of two level atoms is provided by the collective states $|\alpha_1, \ldots, \alpha_N\rangle$ where for the $j$th atom $\alpha_j$ can take the values $b$ or $c$ to denote the two states $|b\rangle$ and $|c\rangle$ of the atom. All together there are $2^N$ such states. The Dicke states are simultaneous eigenstates of the total spin operator $\hat{J}^2$ constructed from the sum of $N$ spin-$\frac{1}{2}$ operators and of the inversion operator $\hat{J}_z$. The totally symmetric Dicke like states are the class of states with the maximal angular momentum $J = N/2$. Different number of excitations in the dark states correspond to different magnetic quantum numbers, $m = -J, -J + 1, \ldots, J$. The ground state $|b, 0\rangle$ for example corresponds to the state with the quantum numbers $(J = \frac{n}{2}, m = -\frac{n}{2})$ and the dark state with one photonic excitation to the state with the quantum numbers $(J = \frac{n}{2}, m = -\frac{n}{2} + 1)$. All other states, i.e. the non symmetric bright states, correspond to states with a smaller total angular momentum. The magnetic quantum number corresponds to the number of stored photons. The interaction created by the Hamiltonian (4.1) just couples states which belong to the subclass with the same total angular momentum and is therefore $\hat{J}^2$ conserving. Interaction of the storage system with its environment leads to excitations of non symmetric states. The equivalence classes of the quantum memory with collective atomic excitations are given by all states which, by reloading the memory, lead to the same output. In figure 4.4 those classes are illustrated for the dark state with one stored photonic excitation. Any unwanted interactions with the environment which lead only to transitions within these equivalence classes and which do not destroy the relative phase between them do not change the storage state and therefore do not affect the fidelity of the quantum memory.

Another important point is the fact that any perturbation which only acts onto states created by the bright state polariton operators does not destroy superpositions of storage states. This is due to the fact that $[\hat{\Phi}_l, \hat{\Psi}] = 0$ for $l \in \{1, 2, \ldots, N - 1\}$:

$$\hat{\Phi}_l^\dagger \sum_n \alpha_n |D, n\rangle_N = \hat{\Phi}_l^\dagger \sum_n \frac{\alpha_n}{\sqrt{n!}} \left( \hat{\Psi}^\dagger \right)^n |b, 0\rangle_N = \sum_n \frac{\alpha_n}{\sqrt{n!}} \left( \hat{\Psi}^\dagger \right)^n \left\{ \hat{\Phi}_l^\dagger |b, 0\rangle_N \right\},$$  (4.20)

1An excellent overview concerning Dicke states is for example given by H.J. Carmichael [95].
4.2. Equivalence classes

Figure 4.4: Equivalence classes of the quantum memory with collective atomic excitations in the basis of Dicke states for the dark state $|D, n = 1\rangle$.

where $\alpha_n$ is an arbitrary complex number.

Furthermore all dark states with the same number of excitations $n$ but with a different number of atoms, $N$ and $N'$, with $N, N' \gg n$, are equivalent

$$|D, n\rangle_{N} = |D, n\rangle_{N'}, \text{ with } N, N' \gg n. \quad (4.21)$$

This reflects the fact that in the readout process all dark state polariton operators corresponding to different number of atoms have the same asymptotic mapping $\hat{\Psi} \rightarrow \hat{a}$ if the mixing angle $\theta(t)$ goes to zero, i.e. $\theta \rightarrow 0$. This fact is important for the case of atom losses during the storage period and will be discussed in detail in section 4.3.3.

To understand what happens to additional excitations into bright states during the reloading process, the complex Hamiltonian (4.1) has to be expressed in terms of the dark and bright state polariton operators after adiabatically eliminating the excited state $|a\rangle$. Separating the oscillatory factor $e^{-i\omega t}$ by a canonical transformation and assuming two photon resonance, i.e. $\omega = \omega_c + \nu$, the transformed Hamiltonian is given by

$$\hat{H} = \hbar \omega \left( \hat{\Psi}^\dagger \hat{\Psi} + \sum_{l=1}^{N-1} \hat{\Phi}_l^\dagger \hat{\Phi}_l \right) - i\hbar \frac{\Omega^2(t)}{\gamma} \sum_{l=1}^{N-1} \hat{\Phi}_l^\dagger \hat{\Phi}_l$$

$$= \hbar \omega \left( \hat{\Psi}^\dagger \hat{\Psi} + \sum_{l=1}^{N-1} \hat{\Phi}_l^\dagger \hat{\Phi}_l \right) - i\hbar \frac{g^2 N}{\gamma} \cot^2 \theta(t) \sum_{l=1}^{N-1} \hat{\Phi}_l^\dagger \hat{\Phi}_l. \quad (4.22)$$
This Hamiltonian shows that the dynamics do not couple different polariton excitations. Furthermore all bright state polariton excitations caused by the operators $\hat{\Phi}_l$, $l \in \{1, 2, \ldots, N-1\}$, decay by optical pumping, i.e. by excitation to the excited state and successive spontaneous emission if $\theta(t) \neq \frac{\pi}{2}$. On the other hand the dark state polariton excitations caused by $\hat{\Psi}$ as well as the bright state polariton excitations caused by $\hat{\Phi}_0$ are immune to spontaneous emission. The Hamiltonian (4.22) shows that under adiabatic conditions, $g\sqrt{NT} \ll 1$, all excitations of the dark state polariton $\left(\hat{\Psi}^\dagger\right)^n |b, 0\rangle_N$ are conserved while all bright state polariton excitations decay via optical pumping into the excited state and subsequent spontaneous emission. If non adiabatic corrections are taken into account dark and bright state polariton excitations are coupled with a rate proportional to $\frac{d}{dt}\theta(t)$.

### 4.3 Random spin flips, dephasing and atom losses

The existence of equivalence classes of a quantum memory with collective atomic excitations, as introduced in the previous section has the interesting consequence that such a quantum memory does not show a larger sensitivity to decoherence than a quantum memory with a single atom. This fact will be discussed in detail in this section for three different decoherence mechanisms. In section 4.3.1 the random spin flip which was already mentioned in the previous section is discussed. In the following in section 4.3.2 the dephasing process of individual atoms is modelled and discussed. The loss of atoms, which presents the most dramatic event that can occur during the storage period is analyzed in section 4.3.3. In all these cases the fidelity of the quantum memory with collective atomic excitations scales like the fidelity of the quantum memory with one atom, if one takes advantage of the existence of the equivalence classes.

#### 4.3.1 Random spin flips

As shown in section 2.3.2 under adiabatic conditions the excited level $|a\rangle$ of the three level system is not involved in the readout process. However spin flips between the two internal states $|b\rangle$ and $|c\rangle$ are another possible source of information loss during the storage process and are therefore considered in the following.

To illustrate this the case a superposition of Fock states of the radiation field $\sum_n \alpha_n|n\rangle$ stored in the superposition of symmetric dark states $\sum_n \alpha_n|D, 1\rangle_N$ is considered. A random spin flip of atom $j$ from state $|b\rangle$ to state $|c\rangle$ can be described by the following
4.3. Random spin flips, dephasing and atom losses

operation

\[ |D, n\rangle_N \rightarrow \hat{\sigma}_{cb}^j |D, n\rangle_N. \]  

where \( \hat{\sigma}_{cb}^j \) is the atomic flip operator for the \( j \)-th atom and leads to a transition from state \( |b\rangle \) to state \( |c\rangle \) in the \( j \)-th atom if this atom is in state \( |b\rangle \). \( \hat{\sigma}_{cb}^j \) can be expressed with the dark and bright state polariton operators (4.11)-(4.13). Assuming that the spin flip takes place during the storage period, i.e. \( \theta(t) = \pi/2 \), the spin flip operator \( \hat{\sigma}_{cb}^j \) can be expressed by using the following transformation:

\[ \hat{\Phi}_l^\dagger = \frac{1}{\sqrt{N}} \sum_{j=1}^N \hat{\sigma}_{cb}^j \exp \left\{ -2\pi i \frac{l j}{N} \right\}, \]  

with \( l \in \{1, 2, \ldots, N - 1\} \) \( \Leftrightarrow \)

\[ \sum_{l=1}^{N-1} \hat{\Phi}_l^\dagger \exp \left\{ 2\pi i \frac{m l}{N} \right\} = \frac{1}{\sqrt{N}} \sum_{j=1}^N \hat{\sigma}_{cb}^j \sum_{l=1}^{N-1} \exp \left\{ -2\pi i \frac{l(j-m)}{N} \right\}, \]

with \( m \in \{1, 2, \ldots, N - 1\} \) \( \Leftrightarrow \)

\[ \sum_{l=1}^{N-1} \hat{\Phi}_l^\dagger \exp \left\{ 2\pi i \frac{m l}{N} \right\} + \frac{1}{\sqrt{N}} \sum_{j=1}^N \hat{\sigma}_{cb}^j \]

\[ = \frac{1}{\sqrt{N}} \sum_{j=1}^N \hat{\sigma}_{cb}^j \sum_{k=0}^{N-1} \exp \left\{ -2\pi i \frac{k(j-m)}{N} \right\}. \]  

Using \( \hat{\Psi}^\dagger = \frac{1}{\sqrt{N}} \sum_{j=1}^N \hat{\sigma}_{cb}^j \) for \( \theta(t) = \pi/2 \) and \( \sum_{l=0}^{N-1} \exp \left\{ -2\pi \frac{k(l-m)}{N} \right\} = N \delta_{jm} \) simplifies equation (4.27) to

\[ \hat{\sigma}_{cb}^j = \frac{1}{\sqrt{N}} \left( \sum_{l=1}^{N-1} \hat{\Phi}_l^\dagger \exp \left\{ -2\pi i \frac{l j}{N} \right\} - \hat{\Psi}^\dagger \right). \]  

With expression (4.28) for the spin flip operator \( \hat{\sigma}_{cb}^j \) the stored superposition of dark states \( \sum_n \alpha_n |D, 1\rangle_N \) is transformed to

\[ \sum_n \alpha_n |D, 1\rangle_N \rightarrow \left[ \sum_{l=1}^{N-1} \frac{\exp \left\{ -2\pi i \frac{l j}{N} \right\}}{\sqrt{N}} \hat{\Phi}_l^\dagger + \frac{1}{\sqrt{N}} \hat{\Psi}^\dagger \right] \sum_n \alpha_n |D, n\rangle_N \]  

Only the second summand on the right hand side of equation (4.29) belongs to a state that is not in the same equivalence class as the original state. Thus the probability of leaving the equivalence class of the stored state is only \( 1/N \). This factor exactly compensates the factor \( N \) of the total probability (4.16) of a spin flip error to occur in any of the atoms. Therefore an ensemble of \( N \) atoms is not more sensitive to spin flips compared to the single atom case if one takes advantage of the presence of the equivalence classes to store the quantum information.
4.3. Random spin flips, dephasing and atom losses

4.3.2 Dephasing

Another source of decoherence arises from dephasing interactions of the quantum memory and its environment. In an atomic vapor for example those dephasing effects can be caused by elastic collisions between the atoms. Therefore the question arises how these dephasing processes influence the storage state and how sensitive an arbitrary stored quantum state is to this decoherence process. In the following dephasing is modelled by including a random phase in each of the atomic operators $\hat{\sigma}^j_{bc}$ defining the dark state polariton operator $\hat{\Psi}$ in equation 2.51. If $p$ is the probability that the $j$th atom undergoes an arbitrary dephasing process the atomic operator $\hat{\sigma}^j_{bc}$ of atom $j$ is transformed according to

$$\hat{\sigma}^j_{bc} \rightarrow \hat{\sigma}^j_{bc} = p\hat{\sigma}^j_{bc}e^{i\varphi_j} + (1-p)\hat{\sigma}^j_{bc}. \quad (4.30)$$

The first summand of the right hand side of (4.30) accounts for the fact that dephasing in the $j$th atom takes place with the probability $p$, the second summand states that with a probability $1-p$ no dephasing affects this atom. The individual phase $\varphi_j$, $j \in \{1, 2, \ldots, N\}$, of each atom can vary from 0 to $2\pi$. They are independent random variables with zero mean value and by classical averaging the product of two of those variables is just given by the product of their individual averages:

$$\overline{e^{i\varphi_j}e^{-i\varphi_k}} = \frac{1}{2\pi} \int_0^{2\pi} \int_0^{2\pi} e^{i\varphi_j}e^{-i\varphi_k} d\varphi_j d\varphi_k = \delta_{jk}. \quad (4.32)$$

With the modified atomic operator (4.30) it is possible to calculate the storage state after the dephasing process. The storage state is defined by excitations caused by the dark state polariton operator $\hat{\Psi}$ which is defined by:

$$\hat{\Psi} = \cos \theta \hat{a} - \sin \theta \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{\sigma}^j_{bc}. \quad (4.33)$$

Due to the modified atomic operator $\hat{\sigma}^j_{bc}$ the dark state polariton operator is also modified

$$\hat{\Psi} \rightarrow \hat{\Psi} = (1-p)\hat{\Psi} - p \left[ \cos \theta \hat{a} - \sin \theta \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{\sigma}^j_{bc}e^{i\varphi_j} \right]. \quad (4.34)$$

With this modified dark state polariton operator $\hat{\Psi}$ the storage state after the dephasing process can be calculated. For simplicity first a stored Fock state $|D, n\rangle_N$ is considered. The modified stored Fock state $|\tilde{D}, n\rangle_N$ is given by

$$|D, n\rangle_N \rightarrow |\tilde{D}, n\rangle_N = \frac{1}{\sqrt{n!}} \left(\hat{\Psi}^\dagger\right)^n |b, 0\rangle_N \quad (4.35)$$
4.3. Random spin flips, dephasing and atom losses

By using the definition of the modified dark state polariton operator (4.34) the stored state $|\tilde{D}, n\rangle_N$ can be expressed by

$$
|\tilde{D}, n\rangle_N = \sum_{k=0}^{n} \left[ \frac{n!}{(n-k)!k!} \right]^{1/2} p^k (1-p)^k \frac{1}{\sqrt{k!}} \left( \cos \theta \hat{a}^\dagger - \sin \theta \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{a}_b^\dagger e^{i\varphi_j} \right)^k \\
\times \frac{1}{\sqrt{(n-k)!}} \langle \tilde{\Psi} \rangle^{-n-k} |b, 0\rangle_N
$$

is defined by:

$$
\tilde{\rho}_{\text{Fock}} = |\tilde{D}, n\rangle_N \langle \tilde{D}, n|.
$$

The explicit form of $\tilde{\rho}_{\text{Fock}}$ is given in appendix B. The modified stored Fock state $|\tilde{D}, n\rangle_N$ can be described by its density matrix

$$
f_{\text{Fock}} = \text{tr}[| \tilde{D}, n\rangle_N \langle \tilde{D}, n|] = \text{tr}[|D, n\rangle_N \langle D, n|].
$$

A longer calculation (see appendix B) leads to

$$
f_{\text{Fock}} = \sum_{k_1}^{n} \sum_{k_2}^{n} \min(k_1,k_2) \left[ \frac{n!}{(n-k_1)!(n-k_2)!} \right] \frac{n!}{(N-l)!} \frac{(N-l)!}{l! N!} \frac{\sin^2 \theta}{\cos^2 \theta} \frac{1}{k_1-l+k_2-l}.
$$
4.3. Random spin flips, dephasing and atom losses

Since $l \leq k_{1,2} \leq n \ll N$ it yields $\frac{(N-l)!}{l!N!} \approx \frac{1}{N}$ which allows an expansion of the fidelity (4.40) for a stored Fock state in orders of $1/N$:

$$f_{Fock} = (1 - p \sin^2 \theta)^{2n} + \mathcal{O}(1/N).$$

(4.41)

If one further assumes that the single atom probability $p$ for dephasing is small and that the dephasing takes place during the storage period, i.e. $\theta = \pi/2$, equation (4.41) can be written in the following form

$$f_{Fock} = (1 - p)^{2n} + \mathcal{O}(1/N) = 1 - 2np + \mathcal{O}(p^2) + \mathcal{O}(1/N).$$

(4.42)

Due to the modelling of the dephasing process in each of the atomic operators $\hat{\sigma}_{bc}$ as given in equation (4.30) the fidelity decreases with increasing number of photons in the Fock state.

By performing a similar calculation it is also possible to calculate the fidelity of the storage media for different classes of initial quantum states after dephasing of individual atoms has taken place. If the initial quantum state of the single mode light field in a mixed state is described by the density matrix $\hat{\rho} = \sum_{m_1=0}^{n} \sum_{m_2=0}^{n} \rho_{m_1 m_2} |m_1\rangle \langle m_2|$ the fidelity $f$ is given by

$$f = \sum_{m_1=0}^{n} \sum_{m_2=0}^{n} |\rho_{m_1 m_2}|^2 (1 - p \sin^2 \theta)^{m_1 + m_2} + \mathcal{O}(1/N),$$

(4.43)

which leads in the case of a stored Fock state to the already discussed result (4.41).

If the initial quantum state was in a coherent state

$$|\alpha\rangle = \sum_{m=0}^{\infty} \exp \left\{-\frac{1}{2} |\alpha|^2 \right\} \frac{\alpha^m}{\sqrt{m!}}, \quad \alpha \in \mathbb{C},$$

(4.44)

the stored state is described by the following density matrix

$$\sum_{m_1=0}^{\infty} \sum_{m_2=0}^{\infty} \rho_{m_1 m_2} |D, m_1\rangle_N \langle D, n|$$

with $\rho_{m_1 m_2} = e^{-|\alpha|^2} \frac{\alpha^{m_1} \alpha^{*m_2}}{\sqrt{m_1! m_2!}}$ and $\alpha \in \mathbb{C}$. (4.45)

The fidelity for this stored state can be calculated by using equation (4.43) and carrying out the limes $n \to \infty$. Furthermore it is assumed that the dephasing takes place during the storage period. This yields:

$$f_{coh} = \exp \{-2|\alpha|^2 p\}.$$  (4.46)

The exact calculation is given in appendix B. This result shows that a stored coherent state shows an exponential decay of the fidelity and is therefore much more sensitive to dephasing effects during the storage period than a stored Fock state. This could be
4.3. Random spin flips, dephasing and atom losses

explained by the fact that the phase of a Fock state is completely undefined which makes it not so sensitive to dephasing processes than the coherent state with the well defined phase.

As last example a Schrödinger cat state which is given by

$$N \sqrt{2} (|i\alpha\rangle + |-i\alpha\rangle)$$

(4.47)

is considered as initial quantum state. Here $|i\alpha\rangle$ and $|-i\alpha\rangle$ are defined by (4.44) and $N$ is defined by:

$$N^2 \equiv \frac{1}{1 + \exp\{-2|\alpha|^2\}}.$$

(4.48)

For this stored state the fidelity is given for $\theta = \pi/2$ by

$$f_{Scs} = \exp\{-2|\alpha|^2p\} \left[\frac{1 + \exp\{-2|\alpha|^2(1 - p)\}}{1 + \exp\{-2|\alpha|^2\}}\right]^2.$$

(4.49)

The calculation of (4.49) is given in appendix B.

4.3.3 Atom loss

Another possible mechanism caused by the interaction of the storage system with its environment and which leads to decoherence is the loss of an atom during the storage period (see figure 4.5). Because during the read out process all dark state polariton operators corresponding to different number of atoms have the same asymptotic mapping all dark states with the same number of excitations $n$ but with a different number of atoms are equivalent. These equivalence classes for different atom numbers were already discussed in section 4.1. In the following how those equivalence classes for different atom numbers help to reduce decoherence effects caused by atom losses during the storage.
process will be discussed. The calculation will first be restricted to the case where the stored state is initially a Fock state of the radiation mode and then generalized to the case of a superposition of Fock states of the radiation field. The stored Fock state $|d, n\rangle_N$ can be described by its density matrix

$$\hat{\rho}_N = |D, n\rangle_N\langle D, n|.$$  \hspace{1cm} (4.50)

The atom loss leads to a new storage state and can be modelled by tracing over an arbitrary atom in the density matrix (4.50). Without loss of generality it is assumed that the $N\text{th}$ atom is lost. The density matrix $\tilde{\rho}_{N-1}$ of the storage state after one atom has been lost is therefore given by:

$$\tilde{\rho}_{N-1} = \text{tr}_{N\text{th atom}}\hat{\rho}_N.$$  \hspace{1cm} (4.51)

The exact calculation of $\tilde{\rho}_{N-1}$ is given in appendix B. The density matrix $\tilde{\rho}_{N-1}$ splits into two summands, one corresponding to the case where the lost atom was excited in state $|c\rangle$ which means that one stored photon gets lost. The other summand corresponds to the case where the lost atom was in its ground state $|b\rangle$. The expansion of both summands up to the order $1/N$ leads to:

$$\tilde{\rho}_{N-1} = |D, n\rangle_{N-1N-1}\langle D, n|$$

$$- \frac{1}{N} \sum_{k=0}^{n} \sum_{l=0}^{n} \frac{1}{4} [(k(k+1) + l(l+1)] \left[ \frac{n!}{k!(n-k)!} \frac{n!}{l!(n-l)!} \right]^{1/2}$$

$$\cdot (-\sin\theta)^{k+l}(\cos\theta)^{2n-k-l}|c^k, n-k\rangle_{N-1N-1}\langle c^l, n-l|$$

$$+ \frac{n}{N} \sin^2\theta |D, n-1\rangle_{N-1N-1}\langle D, n-1|.$$  \hspace{1cm} (4.52)

Since the dark state $|D, n\rangle_{N-1}$ is reloaded to the same radiation mode as the dark state $|D, n\rangle_N$ the fidelity of the storage process under the influence that the $j$th atom is lost can be defined by the projection of the subspace spanned by the dark state $|D, n\rangle_{N-1}$ for $N-1$ atoms:

$$f^j_{\text{Fock}} = \text{tr}|D, n\rangle_{N-1N-1}\langle D, n|\tilde{\rho}_N,$$  \hspace{1cm} (4.53)

which leads after some calculation given in appendix B to

$$f^j_{\text{Fock}} = 1 - \frac{n}{N} \sin^2\theta + O\left(\frac{1}{N^2}\right).$$  \hspace{1cm} (4.54)

With the assumption that the probability to get lost is the same for any atom and that the loss process takes place during the storage period ($\theta = \pi/2$) the fidelity of the quantum memory for a stored Fock state is given by:

$$f_{\text{Fock}} = 1 - n + O(1/N).$$  \hspace{1cm} (4.55)
4.4 Non adiabatic couplings and decoherence

Once again the fidelity decreases linear with the number of photons initially stored. By performing a similar calculation as the one shown above it is possible to calculate the fidelity of the storage media after the atom loss of an arbitrary atom has taken place for different cases of initially stored quantum states. If the initial quantum state of the single mode light field is in a mixed state described by the density matrix $\hat{\rho} = \sum_{m_1=0}^{n} \sum_{m_2=0}^{n} |m_1\rangle \langle m_2|$ the fidelity $f$ is given by

$$f = 1 - \sin^{2} \theta \sum_{l=1}^{n} \rho_{ll} + \sin^{2} \theta \sum_{l=1}^{n} \sum_{s=1}^{n} \rho_{l-1} \rho_{s-1} \sqrt{l \beta} s + \mathcal{O}(1/N). \quad (4.56)$$

Assuming that the atom loss takes place during the storage period, i.e. $\theta = \pi/2$ this yields:

$$f = 1 - \sum_{l=1}^{n} \rho_{ll} + \sum_{l=1}^{n} \sum_{s=1}^{n} \rho_{l-1} \rho_{s-1} \sqrt{l \beta} s + \mathcal{O}(1/N). \quad (4.57)$$

If the initial quantum state was in a coherent state (4.44) the fidelity for this state can be calculated by using equation (4.56) and carrying out the limes $n \to \infty$:

$$f_{\text{Coh}} = 1 + \mathcal{O}(1/N). \quad (4.58)$$

This result shows that the coherent state is very robust against atom losses.

The fidelity for a Schrödinger cat state (4.47) is given by:

$$f_{\text{Scs}} = 1 - |\alpha|^2 \tanh |\alpha|^2 + \mathcal{O}(1/N). \quad (4.59)$$

4.4 Non adiabatic couplings and decoherence

In section 4.2 it was shown that after the adiabatic rotation of the mixing angle $\theta(t)$ from 0 to $\pi/2$ to store the quantum information of the incoming light pulse only the reduced density operator (4.18) is relevant. For this reason all states of the system which have the same number of dark state polariton excitations but an arbitrary number of excitations caused by any bright state polariton operator belong to the same equivalence class and lead by an adiabatic read out process to the same result. This is caused by the fact that the bright state polariton excitations do not couple to the dark state polariton excitations during an adiabatic readout process. Due to decoherence mechanisms bright state polariton excitations are present in the system after the storage period. As discussed in section 4.2 all bright state polariton excitations caused by the operators $\hat{\Phi}_l$, $l \in \{1, 2, \ldots, N - 1\}$ decay by optical pumping during the readout process while the dark state excitations caused by $\hat{\Psi}$ as well as the bright state polariton excitations caused by $\hat{\Phi}_0$ are immune to
4.4. Non adiabatic couplings and decoherence

spontaneous emission. Now the question arises what is happening to those bright state polariton excitations caused by $\hat{\Phi}_l$, $l \in \{1, 2, \ldots, N - 1\}$, if the read out process is not adiabatic. In the following it is shown that in the case $n = 1$ only bright state polariton excitations caused by $\hat{\Phi}_0$ couple via $\frac{d}{dt} \theta(t)$ to the dark state polariton excitation. All other bright state polariton excitations do not couple to the dark state polariton excitation and therefore do not effect the storage state even if non adiabatic corrections for the readout process are taken into account.

In section 2.4 the storage and reloading of a single photon excitation by adiabatic following was discussed. Because of the form of the interaction Hamiltonian (2.76) and (2.81) the most general state $|\Psi(t)\rangle$ of the combined system of the single photon wave packet, the cavity mode and the atoms is a total symmetric state (2.86). Decoherence effects during the storage period change $|\Psi(t)\rangle$ into a non symmetric state:

$$|\Psi(t)\rangle = b(t)|b_1, \ldots, b_N, 1, 0_k\rangle + \sum_{j=1}^{N} c_j(t)|b_1, \ldots, c_j, \ldots, b_N, 0, 0_k\rangle$$

$$+ \sum_{j=1}^{N} a_j(t)|b_1, \ldots, a_j, \ldots, b_N, 0, 0_k\rangle + \sum_k \xi_k(t)|b_1, \ldots, b_N, 0, 1_k\rangle.$$  \hspace{1cm} (4.60)

The equations of motion for the slowly varying state amplitudes are obtained by a transformation into the rotating frame:

$$\frac{d}{dt} a_j(t) = -\frac{\gamma_a}{2} a_j(t) - i g b(t) - i \Omega(t) c_j(t), \hspace{1cm} (4.61)$$

$$\frac{d}{dt} b(t) = -i g \sum_{j=1}^{N} a_j(t) - i \kappa \sum_{k} \xi_k(t), \hspace{1cm} (4.62)$$

$$\frac{d}{dt} c_j(t) = -\frac{\gamma_c}{2} c_j(t) - i \Omega(t) a_j(t), \hspace{1cm} (4.63)$$

$$\frac{d}{dt} \xi_k(t) = -i \Delta_k \xi_k(t) - i \kappa b(t), \hspace{1cm} (4.64)$$

where $\gamma_a$ and $\gamma_c$ are the decay rates of the levels $a$ and $c$, $\Delta_k$ is the detuning of the free field modes from the cavity resonance and $\kappa$ is the coupling constant between the free field modes and the cavity mode. To see the non adiabatic couplings between dark and
4.4. Non adiabatic couplings and decoherence

bright states the following basis of dark and bright states is introduced:

\[ |D\rangle = -i \cos \theta |b_1, \ldots, b_N, 1, 0_k\rangle + i \sin \theta \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |b_1, \ldots, c_j, \ldots, b_N, 0, 0_k\rangle, \quad (4.65) \]

\[ |B_0\rangle = \sin \theta |b_1, \ldots, b_N\rangle + \cos \theta \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |b_1, \ldots, c_j, \ldots, b_N, 0, 0_k\rangle, \quad (4.66) \]

\[ |B_l\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \exp \left\{ -2\pi i \frac{l_j}{N} \right\} |b_1, \ldots, c_j, \ldots, b_N, 0, 0_k\rangle, \quad (4.67) \]

with \( l \in \{1, 2, \ldots, N-1\} \).

With the collective amplitude \( A(t) = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} a_j(t) \) for states with one excitation in the atomic level \( a \) the equations of motion (4.61)-(4.64) in the new basis read:

\[ \frac{d}{dt} A(t) = -\gamma_a A(t) - i\Omega_0(t) B_0(t), \quad (4.68) \]

\[ \frac{d}{dt} B_0(t) = -i\dot{\theta}(t) D(t) - i\Omega_0(t) A(t) - i\kappa \sin \theta(t) \sum_k \xi_k(t), \quad (4.69) \]

\[ \frac{d}{dt} D(t) = -i\dot{\theta}(t) B_0(t) + \kappa \cos \theta(t) \sum_k \xi_k(t), \quad (4.70) \]

\[ \frac{d}{dt} \xi_k(t) = -i\Delta_k \xi_k(t) - i\kappa \sin \theta(t) B_0(t) - \kappa \cos \theta(t) D(t), \quad (4.71) \]

where \( \Omega_0(t) \) is defined by: \( \Omega_0(t) \equiv \sqrt{g^2 N + \Omega^2(t)} \). The non adiabatic couplings are proportional to \( \frac{d}{dt} \dot{\theta}(t) = \dot{\theta}(t) \). They couple the dark state amplitude \( D(t) \) with the bright state amplitude \( B_0(t) \) in equations (4.69) and (4.70). Since all couplings to the bright state amplitudes \( B_i(t) \) vanish in the equations of motion (4.68)-(4.71) even in the non adiabatic readout process in the case of one stored photonic excitation there is no coupling between the dark state \( |D\rangle \) and the bright states \( |B_i\rangle, l \in \{1, 2, \ldots, N-1\} \).

In summary it has been shown that the existence of equivalence classes leads to drastic reduction of the decoherence sensitivity of a quantum memory based on collective many particle states. For the three considered decoherence mechanisms, spin flips, dephasing and atom losses the system does not show enhanced decoherence sensitivity as compared to the single atom case. This is even true when non adiabatic corrections are taken into account.
Chapter 5

Quasi decoherence free subspaces in quantum memories with collective atomic excitations

In the previous chapter it has been shown that the storage of quantum information in collective atomic excitations does not have an enhanced sensitivity to environmental interactions compared to the single atom approach. All storage states with the same number of dark state polariton excitations but with an arbitrary number of excitations in any other bright state polariton mode are equivalent. Therefore decoherence induced transitions between those equivalent states do not affect the fidelity of the quantum memory.

In order to perform quantum computation in a quantum network the quantum memory is an essential part but additional techniques for the implementation of logical gates are required. Since the considered quantum memory is based on delocalized stationary qubits, many proposals which require a precise control of the spatial separation of the atoms are not applicable. Instead nonlinear interactions as for example the dipole blockade scheme suggested by M.D. Lukin et al. [32] can be used in quantum memories with collective excitations. For this scheme the arguments discussed in chapter 4 with respect to the sensitivity to single atom errors do not hold. Consequently the phase gate proposed in [32] is very sensitive to decoherence. A similar problem appears for the single photon detection scheme using stored light as suggested in [96,97]. In both schemes a single atom spin flip leads to an incorrect result. Therefore the elimination of decoherence effects is a necessary condition for a successful implementation of a quantum network based on photons and collective excitations.

In section 5.1 the concept of decoherence free subspaces is introduced. Since examples of decoherence free subspaces have so far only been developed under the assumption of a
permutation symmetry in the interaction between the system and its environment, which is often not a realistic assumption, in section 5.2 the idea of a quasi decoherence free subspace in the case of an atomic ensemble is introduced. It is shown that by choosing appropriate storage states and providing for an additional nonlinear interaction of all atoms, it is possible to construct a quasi decoherence free subspace for the storage state even if all atoms of the atomic ensemble couple to individual and independent reservoirs which can lead to individual atomic spin flips. In section 5.3 this idea is combined with the open-loop control well known from the field of nuclear magnetic resonance to suppress collisional decoherence in a storage scheme implemented in atomic vapors by using a high frequency electric field.

5.1 The concept of decoherence free subspaces

One of the most important achievements in quantum information processing is the discovery of methods to overcome the problem of decoherence. These methods are quantum error correcting codes [98–101], decoherence suppression based on the quantum Zeno effect [102–105] or dynamical decoupling schemes [106–111] as well as the concept of decoherence free subspaces [112–124].

A decoherence free subspace (DFS) is a subspace of dimension $d \geq 2$ of the system’s Hilbert space which is unaffected by decoherence or in other words: If the interaction of the system with the environment is described by a Liouville operator $L$, a subspace $H_0$ of the total Hilbert space $H$ that is invariant under the action of $L$ is called decoherence free subspace. Let be $\Pi_0$ the projector on $H_0$, the condition for $H_0$ to be a DFS is given by

$$L \Pi_0 = i \lambda \Pi_0 \text{ with } \lambda \in \mathbb{R}. \quad (5.1)$$

Examples of DFSs have so far only been developed under the assumption of a permutation symmetry in the system environment coupling. A frequently used example of a DFS takes advantage of the assumption of collective decoherence [112–114, 117, 121], which is however very hard to fulfill experimentally. Collective decoherence mechanisms assume that the environment couples in an identical fashion to all qubits which implies that all qubits undergo the same error. In this case four physical qubits are sufficient to built a logical qubit which is immune against any collective error [114]. Furthermore A.D. Lidar et al. [121] have shown, that under conditions that do not relate to a spatially symmetric system environment coupling, DFSs with $d \geq 2$ may still exist under the assumption that errors affecting single qubits are absent. This concept of DFS has unfortunately only limited practical relevance since the most common type of decoherence, independent reservoir
couplings of all particles, does not have the required symmetry nor does it exclude single qubit errors. However if qubits are not stored in individual particles but rather in collective states of interacting multi particle systems as recently suggested by A.Y. Kitaev for the example of Majorana fermions [125,126], they can effectively be protected from decoherence without requiring special environment couplings. Unfortunately the generation of Majorana fermions is experimentally very difficult and there is yet no direct concept for a coupling to quantum information carrier. Nevertheless in the next section it will be shown that storage states in collective quantum memories for photons based on dark state polaritons in an ensemble of multi level atoms can show a similar robustness against decoherence as the Majorana fermions in [125,126].

5.2 Quasi decoherence free subspaces in collective atomic ensembles

Even if a DFS for an arbitrary reservoir interaction seems not to exist, for a given interaction of qubits with individual reservoirs, it is possible to generate a so called quasi decoherence free subspace (QDFS) of dimension $d = 1$ by introducing a suitable additional interaction. If this interaction generates a sufficiently large energy gap between a single non degenerated ground state and all other possible states of the system’s Hilbert space the decay out of the ground state is exponentially suppressed because for all possible transitions from the ground state to the other states energy is necessary. Unfortunately for storing a qubit a QDFS of minimum dimension $d = 2$ is required. The concept of QDFSs can however be modified for the storage states in collective quantum memories for photons based on dark state polaritons in ensembles of multi level atoms. In the following it will be shown that by choosing appropriate storage states and providing for an additional nonlinear interaction of all atoms, it is possible to construct a QDFS of dimension $d = 2$ even if all atoms of the ensemble couple to individual and independent reservoirs.

5.2.1 The storage scheme and basic idea

To exploit the idea of a QDFS for quantum memories with collective atomic excitations the storage scheme discussed in section 2.3 has to be modified. Consider a two mode quantum memory consisting of $N$ atoms with a ground state $|b\rangle$ and two other metastable states $|c_+\rangle$ and $|c_-\rangle$ (see figure 5.1). In such a system a photonic qubit
Figure 5.1: Single atom scheme for parallel storage of two modes \( \hat{a}_+ \) and \( \hat{a}_- \) with orthogonal polarizations. The atoms are initially prepared in the ground state \( |b\rangle \). \( \Omega_+(t) \) and \( \Omega_-(t) \) represents the Rabi frequencies of the two coherent control fields.

\[
|\varphi\rangle = \alpha|1_+\rangle + \beta|1_-\rangle, \text{ with } |\alpha|^2 + |\beta|^2 = 1
\] (5.2)

with a single photon in two orthogonal modes \( \pm \) can be stored. In the two parallel \( \Lambda \) transitions the orthogonal photonic modes \( \hat{a}_+ \) and \( \hat{a}_- \) couple resonantly the transition between the ground state \( |b\rangle \) and the excited state \( |a_+\rangle \) and \( |a_-\rangle \) respectively. The upper states \( |a_+\rangle \) and \( |a_-\rangle \) are furthermore coupled to the metastable states \( |c_+\rangle \) or \( |c_-\rangle \) respectively by two coherent control fields with time dependent Rabi frequencies \( \Omega_+(t) \) and \( \Omega_-(t) \). Initially the ensemble of atoms is assumed to be prepared in the ground state \( |b\rangle \). The mixing angles \( \theta_\pm(t) \) are defined by \( \tan \theta_\pm(t) = \frac{g_\pm \sqrt{N}}{\Omega_\pm(t)} \) where \( g_\pm \) are the Rabi frequencies of the quantum field described by \( \hat{a}_\pm \). Changing the mixing angles \( \theta_\pm(t) \) from 0 to \( \pi/2 \) adiabatically in time, the photonic qubit \( |\varphi\rangle \) is transferred to a collective storage state according to

\[
|\varphi\rangle = \alpha|1_+\rangle + \beta|1_-\rangle
\] (5.3)

\[
\downarrow
\]

\[
\alpha \frac{1}{\sqrt{N}} [c_{+1}, b_2, \ldots, b_N] + |b_1, c_{+2}, b_3 \ldots, b_N\rangle + \ldots + |b_1, \ldots, b_{N-1}, c_{+N}\rangle
\]

\[
+ \beta \frac{1}{\sqrt{N}} [c_{-1}, b_2, \ldots, b_N] + |b_1, c_{-2}, b_3 \ldots, b_N\rangle + \ldots + |b_1, \ldots, b_{N-1}, c_{-N}\rangle.
\]
5.2. Quasi decoherence free subspaces in collective atomic ensembles

The relevant collective states of the ensemble can be described in analogy to section 4.1 in terms of dark and bright states, $|D_{\pm}, n\rangle_N$ and $|B_{l\pm}, n\rangle_N$, which are given by

$$
|D_{\pm}, n\rangle_N = \frac{1}{\sqrt{n!}} \left(\hat{\Psi}_{\pm}^\dagger\right)^n |b_1, \ldots, b_N\rangle,
$$

$$
|B_{l\pm}, n\rangle_N = \frac{1}{\sqrt{n!}} \left(\hat{\Phi}_{l\pm}^\dagger\right)^n |b_1, \ldots, b_N\rangle \quad \text{with} \quad l \in \{1, 2, \ldots, N - 1\}.
$$

The dark and bright state polariton operators during the storage period ($\theta_{\pm} = \pi/2$) are given by

$$
\hat{\Psi}_\pm = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{\sigma}_{bc,\pm}^j,
$$

$$
\hat{\Phi}_{l\pm} = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{\sigma}_{bc,\pm}^j \exp \left\{ \frac{2\pi i lj}{N} \right\}, \quad \text{with} \quad l \in \{1, 2, \ldots, N - 1\},
$$

$$
\hat{\Psi}_{0\pm} = \hat{a}_\pm,
$$

where $\hat{\sigma}_{bc,\pm}^j = |\mu\rangle_j \langle \nu|$ is a spin flip operator of the $j$th atom. The excited states $|a_\pm\rangle$ do not occur because under conditions for an adiabatic transfer they are neither populated during nor after the storage process. The dark states $|D_{\pm}, n\rangle_N$ given by equation (5.4) are symmetric superpositions of atomic excitations. They are eigenstates of the collective angular momentum operators $\hat{J}_{\pm}^2$ with maximum eigenvalue $J_{\pm} = N/2$. Excitations caused by all other polariton operators have smaller eigenvalues of $\hat{J}_{\pm}^2$. In the just introduced notation the stored photon $|\varphi\rangle$ corresponds to the following storage state

$$
\alpha |D_+, 1\rangle_N + \beta |D_-, 1\rangle_N.
$$

The two collective states $|D_{\pm}, 1\rangle_N$ corresponding to a stored single photon in the two modes $\hat{a}_\pm$ have a very important property: The transition probability from one state to the other upon a spin flip of any given atom scales as $1/N$. For example if the $j$th atom undergoes a spin flip $|c_+\rangle \rightarrow |c_-\rangle$ the overlap of the two storage states $|D_+, 1\rangle_N$ and $|D_-, 1\rangle_N$ after that spin flip is proportional to $1/N$. Consequently the probability that after the single atom spin flip the two resulting states cannot be distinguished is given by

$$
p = |_N\langle D_-, 1 | \hat{\sigma}_{c-,c_+}^j | D_+, 1\rangle_N|^2 = \frac{1}{N^2}.
$$

Even after taking into account that any atom can undergo a spin flip independently of all others, the total transition probability scales as $1/N$. Thus in a sufficiently large ensemble transitions between the two states $|D_+, 1\rangle_N$ and $|D_-, 1\rangle_N$ induced by single atom spin flips can practically be neglected.

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5.2. Quasi decoherence free subspaces in collective atomic ensembles

Figure 5.2: Quasi decoherence free subspace (QDFS) consisting of states $|D_+, 1\rangle_N$ and $|D_-, 1\rangle_N$. Single atom spin flip errors lead to transitions from the $|D_+, 1\rangle_N$ or $|D_-, 1\rangle_N$ to bright polariton states. For an energy gap $E_g > k_B T$ these transitions are exponentially suppressed. The transition probability between $|D_+, 1\rangle_N$ and $|D_-, 1\rangle_N$ scales as $1/N^2$.

If now a collective interaction between the atoms of the ensembles is provided which corresponds to an effective Hamiltonian $\hat{H}_g$ of the form

$$\hat{H}_g = -E_g [|D_+, 1\rangle_N \langle D_+, 1| + |D_-, 1\rangle_N \langle D_-, 1|]$$  \hfill (5.11)

the subspace $\mathcal{M} = \{|D_+, 1\rangle_N, |D_-, 1\rangle_N\}$ spanned by the states $|D_+, 1\rangle_N$ and $|D_-, 1\rangle_N$ is energetically shifted by an energy $E_g > 0$ below all other states of the Hilbertspace $\mathcal{H}$ which is spanned by all dark states with $n \geq 2$ and all bright states. This situation is illustrated in figure 5.2. If the energy shift $E_g$ is larger than the thermal energies $k_B T$ of the reservoirs which describe the environment, transitions from the storage states into all other states are exponentially suppressed. Therefore the subspace $\mathcal{M}$ should be a quasi decoherence free subspace with respect to individual spin flips. In the following this approach for a QDFS for quantum memories with collective atomic excitations will be analyzed in more detail. The starting point of the analysis is a model Hamiltonian for the interaction of the atoms with individual and uncorrelated reservoirs of finite temperature and an additional collective interaction of the type given in equation 5.11. To derive a Liouville equation the reservoir degrees of freedom are eliminated using the usual Barn and Markov approximation. The resulting Liouville equation is then the basis of the analysis of the properties and limitations of the QDFS approach.
5.2 Quasi decoherence free subspaces in collective atomic ensembles

Figure 5.3: Individual reservoir couplings of the $j$th atom which cause transition between the three internal states $|b\rangle$, $|c_+\rangle$ and $|c_-\rangle$.

5.2.2 Model of the interaction with the environment

To model the spin flips of individual atoms in the suggested scheme for QDFS in quantum memories with collective excitations an interaction Hamiltonian which models all possible spin flips between the three relevant atomic states $|b\rangle$, $|c_+\rangle$ and $|c_-\rangle$ is introduced. Since it is assumed that the spin flips happen independent from other each other, individual and independent reservoirs are introduced for each atom as indicated in figure 5.3. Since three different transition between the three relevant internal states $|b\rangle$, $|c_+\rangle$ and $|c_-\rangle$ are possible, for each atom three individual and independent reservoirs are introduced. The reservoirs are modelled by an infinite number of harmonic oscillators\(^1\). Let $\hat{A}_{kj}^\dagger$, $\hat{A}_{kj}$ be the creation and annihilation operators of a harmonic oscillator of frequency $\omega_k$ constituting the reservoir which lead to transitions between the internal states $|c_+\rangle$ and $|c_-\rangle$ of the $j$th atom. $\hat{B}_{kj}^\dagger$ and $\hat{B}_{kj}$ are the creation and annihilation operators of a harmonic oscillator of frequency $\omega_k$ constituting the reservoirs which lead to transitions between the internal states $|c_+\rangle$ and $|b\rangle$ of the $j$th atom. $\hat{C}_{kj}^\dagger$ and $\hat{C}_{kj}$ are the creation and annihilation operators of a harmonic oscillator of frequency $\omega_k$ constituting the reservoirs which lead to transitions between the internal states $|c_-\rangle$ and $|b\rangle$ respectively of the $j$th atom.

\(^1\)An excellent overview of different kinds of bathes of harmonic oscillators is given by R.R. Puri [127].
atom. The free reservoir Hamiltonian $\hat{H}_B$ for all reservoirs is therefore given by

$$\hat{H}_B = \sum_{j=1}^{N} \sum_k \hbar \omega_k [\hat{A}^\dagger_{kj} \hat{A}_{kj} + \hat{B}^\dagger_{kj} \hat{B}_{kj} + \hat{C}^\dagger_{kj} \hat{C}_{kj}].$$

(5.12)

The interaction Hamiltonian $\hat{H}_{int}$ which models the interaction between the atomic ensemble and the reservoirs reads then

$$\hat{H}_{int} = \hbar \sum_{j=1}^{N} \sum_k \left[ g_{c_i}^k \hat{A}_{kj} + g_{c_i}^{k*} \hat{A}^\dagger_{kj} \right] [\hat{\sigma}^j_{c_i} + \hat{\sigma}^{j\dagger}_{c_i}]$$

$$+ \hbar \sum_{j=1}^{N} \sum_k \left[ g_{c_i}^k \hat{B}_{kj} + g_{c_i}^{k*} \hat{B}^\dagger_{kj} \right] [\hat{\sigma}^j_{\gamma} + \hat{\sigma}^{j\dagger}_{\gamma}]$$

$$+ \hbar \sum_{j=1}^{N} \sum_k \left[ g_{c_i}^k \hat{C}_{kj} + g_{c_i}^{k*} \hat{C}^\dagger_{kj} \right] [\hat{\sigma}^j_{\gamma} + \hat{\sigma}^{j\dagger}_{\gamma}],$$

(5.13)

where the $g_{\mu\omega}$ are the system reservoir coupling constants. It should be noted that the rotating wave approximation has not been used. To simplify the notation the following symbolic operators are introduced

$$\hat{\Gamma}^j_A = \sum_k \left[ g_{c_i}^k \hat{A}_{kj} + g_{c_i}^{k*} \hat{A}^\dagger_{kj} \right] = \hat{\Gamma}^j_A,$$

(5.14)

$$\hat{\Gamma}^j_B = \sum_k \left[ g_{c_i}^k \hat{B}_{kj} + g_{c_i}^{k*} \hat{B}^\dagger_{kj} \right] = \hat{\Gamma}^j_B,$$

(5.15)

$$\hat{\Gamma}^j_C = \sum_k \left[ g_{c_i}^k \hat{C}_{kj} + g_{c_i}^{k*} \hat{C}^\dagger_{kj} \right] = \hat{\Gamma}^j_C.$$  

(5.16)

If further thermal reservoirs constituted by harmonic oscillators in the state of equilibrium at temperature $T$ are considered, the individual bath density operators are described by

$$\rho^X_{B} = \prod_{k=1}^{\infty} \left( 1 - \exp\{-\beta_k\} \right) \exp \left\{ -\beta_k \hat{X}^\dagger_{kj} \hat{X}_{kj} \right\}, \text{ with } X \in \{A, B, C\}.$$  

(5.17)

Here $\beta_k$ is defined by:

$$\beta_k = \frac{\hbar \omega_k}{k_B T},$$

(5.18)

where $k_B$ is the Boltzmann constant. For the thermal reservoirs the correlation functions which are needed in the following chapter are given by

$$\langle \hat{X}_{kj} \rangle_B = \text{tr}_B \left\{ \hat{X}_j (\omega_k) \rho_B \right\} = 0,$$

(5.19)

$$\langle \hat{X}^\dagger_{kj} \rangle_B = \text{tr}_B \left\{ \hat{X}^\dagger_j (\omega_k) \rho_B \right\} = 0,$$

(5.20)

$$\langle \hat{X}_{kj} \hat{X}_{lj} \rangle_B = \text{tr}_B \left\{ \hat{X}_j (\omega_k) \hat{X}_l (\omega_l) \rho_B \right\} = \bar{n}(\omega_k) \delta(\omega_k - \omega_l),$$

(5.21)

$$\langle \hat{X}_{kj} \hat{X}^\dagger_{lj} \rangle_B = \text{tr}_B \left\{ \hat{X}_j (\omega_k) \hat{X}^\dagger_l (\omega_l) \rho_B \right\} = [\bar{n}(\omega_k) + 1] \delta(\omega_k - \omega_l),$$

(5.22)

$$\langle \hat{X}_{kj} \hat{X}_{lj} \rangle_B = \text{tr}_B \left\{ \hat{X}_j (\omega_k) \hat{X}_l (\omega_l) \rho_B \right\} = 0 \text{ and}$$

(5.23)

$$\langle \hat{X}^\dagger_{kj} \hat{X}^\dagger_{lj} \rangle_B = \text{tr}_B \left\{ \hat{X}^\dagger_j (\omega_k) \hat{X}^\dagger_l (\omega_l) \rho_B \right\} = 0.$$  

(5.24)
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Since the individual baths are assumed to be uncorrelated, all correlations functions between different baths vanish. In equations (5.19)-(5.24) $\text{tr}_B$ is the trace over all $3N$ individual reservoirs and the density operator $\rho_B$ is the total density operator of all baths which is a direct product of all individual baths (5.17)

$$
\rho_B = \bigotimes_{j=1}^{N} \rho_{B}^{X_j}. 
$$

(5.25)

$\bar{n}(\omega_k)$ is the mean photon number for an oscillator with frequency $\omega_k$ in thermal equilibrium at temperature $T$ and is given by:

$$
\bar{n}(\omega_k) = \frac{e^{-\beta_k}}{1 - e^{-\beta_k}}. 
$$

(5.26)

In the following section it is furthermore assumed that the frequencies of the harmonic oscillators describing the individual thermal reservoirs are continuously distributed. Let $h(\omega)$ be the density of oscillators at frequency $\omega$. With this assumption it is possible to convert any sum over $k$ to an integral over $\omega$ by the following correspondence

$$
\sum_k f_k \rightarrow \int_0^{\infty} d\omega h(\omega) f(\omega), 
$$

(5.27)

where $f_k$ stands for any $k$ dependent value.

5.2.3 Derivation of the Master equation

Since the atomic ensemble interacts with its environment $E$ modelled by the individual and independent baths of harmonic oscillators (5.13) the atomic ensemble can be regarded as an open system $S$. The total system $S + E$ satisfies the usual Hamiltonian dynamics. Since one is interested in the dynamical behavior of the system $S$ alone, the master equation which describes the dynamics of the system $S$ under the influence of the interaction with its environment $E$ will be derived in the following. To derive the master equation there exists different ways. The one used in the following is based on the elimination of the environment degrees of freedom by tracing over the bath variables\(^2\). An alternative approach based on the idea of projection operators is for example presented by J. Kupsch [129] and provides a compact and general way of deriving the master equation by introducing a certain degree of abstraction.

\(^2\)An excellent overview on this topic is for example given by C.W. Gardiner and P. Zoller [128].
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The system Hamiltonian $\hat{H}_S$ is given by the free dynamics of the atomic ensemble during the storage period and therefore reads

$$\hat{H}_S = \hbar \omega \sum_{j=1}^{N} \left[ \hat{\sigma}^j_{+c+} + \hat{\sigma}^j_{-c-} \right] + \hbar \omega \left[ \hat{a}^+_+ \hat{a}^-_- + \hat{a}^+_- \hat{a}^-_+ \right] + \hbar \omega \left[ \hat{a}^+_+ \hat{a}^-_- + \hat{a}^+_- \hat{a}^-_+ \right],$$

(5.28)

where it has been assumed without loosing generality that the energy of the atomic state $|b\rangle$ is equal to zero. Since one is interested in the dynamics of the subspace $\mathcal{M} = \{|D_+, 1\rangle_N, |D_-, 1\rangle_N\}$ it is useful to rewrite the system Hamiltonian in terms of the dark and bright state polariton operators (5.6)-(5.7). Together with $\sum_{s=0}^{N-1} \exp \left\{ -2\pi i \frac{s(j-m)}{N} \right\} = N\delta_{jm}$ this yields

$$\hat{\sigma}^j_{bc\pm} = \frac{1}{\sqrt{N}} \sum_{l=1}^{N-1} \hat{\Phi}_{l\pm} \exp \left\{ -2\pi i \frac{l j}{N} \right\} \mp \frac{1}{\sqrt{N}} \hat{\Psi}_\pm,$$

(5.29)

$$\hat{\sigma}^j_{c+\pm} = \frac{1}{\sqrt{N}} \sum_{l=1}^{N-1} \sum_{m=1}^{N-1} \left[ \hat{\Phi}^\dagger_{l+} \hat{\Phi}^-_{m-} \exp \left\{ 2\pi i \frac{j(l-m)}{N} \right\} + \frac{1}{N} \hat{\Psi}^\dagger_+ \hat{\Psi}_- \right]$$

$$\mp \frac{1}{\sqrt{N}} \sum_{l=1}^{N-1} \left[ \hat{\Phi}^\dagger_{l+} \hat{\Phi}^-_{l-} \exp \left\{ 2\pi i \frac{l j}{N} \right\} + \hat{\Psi}^\dagger_+ \hat{\Phi}^-_{l-} \exp \left\{ -2\pi i \frac{l j}{N} \right\} \right].$$

(5.30)

Therefore the free system Hamiltonian (5.28) in the basis of dark and bright state polariton operators is given by

$$\hat{H}_S = E \left[ \hat{\Phi}^\dagger_{0+} \hat{\Phi}^-_{0-} + \hat{\Phi}^\dagger_{0-} \hat{\Phi}^-_{0+} \mp \sum_{l_1}^{N-1} \left( \hat{\Phi}^\dagger_{l_1+} \hat{\Phi}^-_{l_1+} \mp \hat{\Phi}^\dagger_{l_1-} \hat{\Phi}^-_{l_1-} \right) + \hat{\Psi}^\dagger_+ \hat{\Psi}_- \mp \hat{\Psi}^\dagger_- \hat{\Psi}_+ \right],$$

(5.32)

where $E = \hbar \omega$. The Hamiltonian $\hat{H}_{int}$ defined in equation (5.13) in the basis of dark and bright state polariton operators reads

$$\hat{H}_{int} = \hat{H}_A + \hat{H}_B + \hat{H}_C,$$

(5.33)

where $\hat{H}_A$, $\hat{H}_B$ and $\hat{H}_C$ are given by

$$\hat{H}_A \equiv \hbar \sum_{l=1}^{N} \sum_{m=1}^{N} \sum_{j=1}^{N-1} \left[ \hat{\Phi}^\dagger_{l+} \hat{\Phi}^-_{m-} \hat{\Gamma}^j_A \exp \left\{ 2\pi i \frac{j(l-m)}{N} \right\} + H.c. \right]$$

$$\mp \hbar \sum_{l=1}^{N} \sum_{m=1}^{N} \sum_{j=1}^{N-1} \left[ \hat{\Phi}^\dagger_{l+} \hat{\Phi}^-_{l-} \hat{\Gamma}^j_A \exp \left\{ 2\pi i \frac{j(l-m)}{N} \right\} + H.c. \right]$$

$$\mp \hbar \sum_{l=1}^{N} \sum_{m=1}^{N} \sum_{j=1}^{N-1} \left[ \hat{\Psi}^\dagger_+ \hat{\Phi}^-_{l-} \hat{\Gamma}^j_A \exp \left\{ 2\pi i \frac{j(l-m)}{N} \right\} + H.c. \right]$$

$$\mp \hbar \sum_{j=1}^{N} \left[ \hat{\Psi}^\dagger_+ \hat{\Psi}_- \hat{\Gamma}^j_A + H.c. \right],$$

(5.34)
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\[ \hat{H}_B \equiv \frac{\hbar}{\sqrt{N}} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \left[ \hat{\Phi}_{l+} \hat{\Gamma}_{Bj} \exp \left\{ 2\pi i \frac{lj}{N} \right\} + H.c. \right] - \sum_{j=1}^{N} \left[ \hat{\Psi}_{l+} \hat{\Gamma}_{Bj} + H.c. \right] \]  

(5.35)

and

\[ \hat{H}_C \equiv \frac{\hbar}{\sqrt{N}} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \left[ \hat{\Phi}_{l-} \hat{\Gamma}_{Cj} \exp \left\{ 2\pi i \frac{lj}{N} \right\} + H.c. \right] - \sum_{j=1}^{N} \left[ \hat{\Psi}_{l-} \hat{\Gamma}_{Cj} + H.c. \right] . \]  

(5.36)

Since the Hamiltonian \( \hat{H}_g \) defined in equation (5.11) only includes system operators it is convenient to add it to the system Hamiltonian \( \hat{H}_S \) by defining

\[ \hat{H}'_S \equiv \hat{H}_S + \hat{H}_g. \]  

(5.37)

The dynamics of the system \( S \) and its environment \( E \) on the Hilbert space \( \mathcal{H}_S \otimes \mathcal{H}_E \) is therefore determined by the Hamiltonian \( \hat{H} \)

\[ \hat{H} = \hat{H}'_S + \hat{H}_B + \hat{H}_{int}, \]  

(5.38)

where \( \hat{H}'_S, \hat{H}_B \) and \( \hat{H}_{int} \) are defined by equations (5.37), (5.12) and (5.13). Assuming that the time evolution of the system \( S \) and the environment \( E \) satisfies the unitary dynamics

\[ \rho_{tot}(t) = U(t)\rho_{tot}(0)U^{-1}(t), \]  

(5.39)

with the unitary operator \( U(t) = \exp\{-i\hat{H}_t\} \) and the total density operator \( \rho_{tot}(t) \) of the combined system \( S + E \), the equation of motion for \( \rho_{tot}(t) \) in the Schrödinger picture is determined by the usual Liouville-von-Neumann equation

\[ \dot{\rho}_{tot}(t) = \mathcal{L}\rho_{tot}(t). \]  

(5.40)

Here the Liouville operator of the combined system \( S + E \) is given by

\[ \mathcal{L}\rho_{tot}(t) = -i\hbar [\hat{H}, \rho_{tot}(t)]. \]  

(5.41)

To get the equation of motion for the system \( S \) under the influence of the environment \( E \) the dynamics of the reduced density operator \( \rho(t) \) is analyzed. \( \rho(t) \) is defined by

\[ \rho(t) = \text{tr}_B\{\rho_{tot}(t)\}, \]  

(5.42)

where \( \text{tr}_B \) is the trace over all bath variables. For further calculations it is convenient to separate the free evolution of the system \( S \) and the environment \( E \) determined by \( \hat{H}'_S \) and \( \hat{H}_E \) from the evolution determined by the interaction Hamiltonian \( \hat{H}_{int} \). Therefore the equation of motion (5.40) is transformed into the interaction picture defined by setting

\[ \rho_{tot}^I(t) \equiv \exp\left\{ i\hbar (\hat{H}'_S + \hat{H}_B)t \right\} \rho_{tot}(t) \exp\left\{ -i\hbar (\hat{H}'_S + \hat{H}_B)t \right\}, \]  

(5.43)
where $\rho'_{\text{tot}}(t)$ is the total density operator in the interaction picture. Together with equation (5.40) $\rho'_{\text{tot}}$ obeys the equation of motion given by

$$\dot{\rho}'_{\text{tot}}(t) = -\frac{i}{\hbar}[\hat{H}'_{\text{int}}(t), \rho'_{\text{tot}}(t)], \quad (5.44)$$

with

$$\hat{H}'_{\text{int}}(t) \equiv \exp \left\{ \frac{i}{\hbar}(\hat{H}'_S + \hat{H}_B)t \right\} \hat{H}_{\text{int}} \exp \left\{ -\frac{i}{\hbar}(\hat{H}'_S + \hat{H}_B)t \right\}. \quad (5.45)$$

Together with the given Hamiltonians $\hat{H}'_S$ and $\hat{H}_B$ and equation (5.33) the explicit time dependent interaction Hamiltonian $\hat{H}'_{\text{int}}(t)$ reads:

$$\hat{H}'_{\text{int}}(t) = \hat{H}'_A(t) + \hat{H}'_B(t) + \hat{H}'_C(t), \quad (5.46)$$

where $\hat{H}'_A(t)$, $\hat{H}'_B(t)$ and $\hat{H}'_C(t)$ are given by

$$\hat{H}'_A(t) = \frac{\hbar}{N} \sum_{l=1}^{N-1} \sum_{m=1}^{N-1} \sum_{j=1}^N \left[ \hat{\Phi}^\dagger_{l+} \hat{\Phi}^\dagger_{m-} \tilde{\Gamma}^j_A(t) \exp \left\{ 2\pi i \frac{j(l-m)}{N} \right\} + \text{H.c.} \right],$$

$$\hat{H}'_B(t) = \frac{\hbar}{N} \sum_{l=1}^{N-1} \sum_{j=1}^N \left[ \hat{\Phi}^\dagger_{l+} \tilde{\Gamma}^j_B(t) \exp \left\{ 2\pi i \frac{j}{N} \right\} + \text{H.c.} \right],$$

$$\hat{H}'_C(t) = \frac{\hbar}{N} \sum_{j=1}^N \left[ \hat{\Phi}^\dagger_{l-} \tilde{\Gamma}^j_C(t) \exp \left\{ 2\pi i \frac{j}{N} \right\} + \text{H.c.} \right],$$

and

$$\hat{H}'_{\text{int}}(t) = \hat{H}'_A(t) + \hat{H}'_B(t) + \hat{H}'_C(t). \quad (5.47)$$
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The operators \( \tilde{M}_\pm(t) \) and \( \tilde{\Gamma}_X^j(t) \), \( X \in \{A, B, C\} \) are hereby defined as

\[
\tilde{M}_\pm(t) \equiv \sum_{n=2}^{\infty} \sqrt{n+1} |D_\pm, n\rangle_N \langle D_\pm, n + 1| + e^{i\omega_g t} |D_\pm, 0\rangle_N \langle D_\pm, 1|
\]

\[
\tilde{\Gamma}_X^j(t) \equiv \exp \left\{ \frac{i}{\hbar} \hat{H}_B t \right\} \tilde{\Gamma}_X^j \exp \left\{ -\frac{i}{\hbar} \hat{H}_B t \right\} .
\]

(5.50)

where \( \omega_g \) is defined by the energy \( E_g \) of the energy gap: \( E_g = \hbar \omega_g \). The details of the calculation leading to equation (5.46) are given in appendix C.

Using equation (5.43) the reduced density operator \( \rho(t) \) given in equation (5.42) can now be expressed by

\[
\rho(t) = \text{tr}_B \left[ \exp \left\{ -\frac{i}{\hbar} (\hat{H}_S + \hat{H}_B) \right\} \rho_{\text{tot}}^I(t) \exp \left\{ \frac{i}{\hbar} (\hat{H}_S + \hat{H}_B) \right\} \right].
\]

(5.52)

Since the Hamilton operator \( \hat{H}_B \) is a function of bath variables only it is allowed to use the cyclic property of the trace to cancel the factors involving \( \hat{H}_B \) which yields

\[
\rho(t) = \exp \left\{ -\frac{i}{\hbar} \hat{H}_S t \right\} \rho^I(t) \exp \left\{ \frac{i}{\hbar} \hat{H}_S t \right\} ,
\]

(5.53)

where \( \rho^I(t) \) is the interaction picture reduced density operator. It is given by

\[
\rho^I(t) = \text{tr}_B \{ \rho_{\text{tot}}^I(t) \}.
\]

(5.54)

As initial conditions it is assumed that the system \( S \) and the environment \( E \) are independent at time \( t = 0 \) so that the total density operator \( \rho_{\text{tot}} \) factorizes into a direct product

\[
\rho_{\text{tot}}(t = 0) = \rho_{\text{tot}}^I(t = 0) = \rho(t = 0) \otimes \rho_B(t = 0).
\]

(5.55)

To take advantage of this condition the equation of motion (5.44) is integrated in time from 0 to \( t \) which gives

\[
\rho_{\text{tot}}^I(t) = \rho_{\text{tot}}^I(0) - \frac{i}{\hbar} \int_0^t dt' [\hat{H}_\text{int}^I(t'), \rho_{\text{tot}}^I(t')] .
\]

(5.56)

Iteration of this equation yields

\[
\rho_{\text{tot}}^I(t) = \rho_{\text{tot}}^I(0) - \frac{i}{\hbar} \int_0^t dt' [\hat{H}_\text{int}^I(t'), \rho_{\text{tot}}^I(0)] - \frac{1}{\hbar^2} \int_0^t dt' \int_0^{t'} dt'' [\hat{H}_\text{int}^I(t''), [\hat{H}_\text{int}^I(t''), \rho_{\text{tot}}^I(t'')]].
\]

(5.57)
Further iteration would lead to a power series in the perturbation $\hat{H}^I_{\text{int}}(t)$. To obtain an integro differential equation for the total density operator $\rho^I_{\text{tot}}(t)$ in the interaction picture one differentiates equation (5.57) with respect to $t$:

$$\dot{\rho}^I_{\text{tot}} = -\frac{i}{\hbar} [\hat{H}^I_{\text{int}}(t), \rho^I_{\text{tot}}(0)] - \frac{1}{\hbar^2} \int_0^t dt' [\dot{\hat{H}}^I_{\text{int}}(t), \dot{\rho}^I_{\text{tot}}(t')] .$$

(5.58)

To obtain an integro differential equation for the reduced density operator $\rho^I(t)$ defined in equation (5.54) both sides of equation (5.58) are traced over all bath variables which results in

$$\text{tr}_B\{\dot{\rho}^I_{\text{tot}}(t)\} = -\frac{1}{\hbar^2} \int_0^t dt' \text{tr}_B\{[\dot{\hat{H}}^I_{\text{int}}(t), \dot{\hat{H}}^I_{\text{int}}(t'), \rho^I_{\text{tot}}(t')]\} ,$$

(5.59)

where the initial condition (5.55) and the relation

$$\text{tr}_B\{\hat{H}^I_{\text{int}}(t)\rho^I_{\text{tot}}(t)\} = 0$$

(5.60)

which is fulfilled in the considered case have been used.

If one furthermore assumes that the reservoir is large enough that its statistical properties are unaffected by the weak coupling to the system then the Born approximation used in the following is justified. Since the interaction in equation (5.13) is a sum of terms like $\hat{X}_S\hat{Y}_B$ where both of these operators are simple operators which act respectively only in the system and the bath spaces, the Born approximation just assumes that the bath correlations are unaffected by the interaction between the system and its environment. In this case it is justified to replace $\rho^I_{\text{tot}}(t')$ on the right hand side of equation (5.59) by the factorized approximation

$$\rho^I_{\text{tot}}(t') \approx \rho^I(t') \otimes \rho_B .$$

(5.61)

Therefore the integro differential equation (5.59) in Born approximation is given by

$$\dot{\rho}^I(t) = -\frac{1}{\hbar^2} \int_0^t dt' \text{tr}_B\{[\hat{H}^I_{\text{int}}(t), \hat{H}^I_{\text{int}}(t'), \rho^I(t') \otimes \rho_B]\} .$$

(5.62)

To turn this equation into a differential equation the Markov approximation is made, which assumes that the rate of change of the system density operator in the interaction picture is quite slow compared to the rate of change of the bath operators. In the case of a thermal bath this assumption is usually fulfilled. Therefore the factor $\rho(t')$ in equation (5.62) does not change significantly over the time over which the correlation functions do not vanish on the right hand side of equation (5.62) and it is justified to make the following replacement

$$\rho^I(t') \rightarrow \rho^I(t) .$$

(5.63)
This leads to the master equation in Born-Markov approximation:

\[
\dot{\rho}(t) = -\frac{1}{\hbar^2} \int_0^t dt' \text{tr}_B\{[\hat{H}_{int}(t), \hat{H}_{int}(t'), \rho(t) \otimes \rho_B]\}. \tag{5.64}
\]

The exact form of \(\text{tr}_B\{[\hat{H}_{int}(t), \hat{H}_{int}(t'), \rho(t) \otimes \rho_B]\}\) is given in appendix C.

### 5.2.4 Results and Discussion

Since only the equations of motion for the matrix elements which describe the dynamics of the subspace \(\mathcal{M} = \{ |D_+, 1\rangle_N, |D_-, 1\rangle_N \}\) are of interest for the existence of quasi decoherence free subspaces, the master equation in Born-Markov approximation (5.64) will only be discussed for the matrix elements \(\langle D_\pm, 1| \rho(t)|D_\pm, 1 \rangle\) and \(\langle D_\pm, 1| \rho(t)|D_{\mp}, 1 \rangle\). These matrix elements describe the dynamics of the population in the collective states \(|D_\pm, 1\rangle\) and of their coherence. Since atom losses are not included in the model, the index \(N\) which counts the atom number has been neglected to simplify the notation. To derive the equations of motion for the matrix elements \(\langle D_\pm, 1| \rho(t)|D_\pm, 1 \rangle\) and \(\langle D_\pm, 1| \rho(t)|D_{\mp}, 1 \rangle\) it is useful to express the dark and bright state polariton operators \(\hat{\Psi}^\pm\) and \(\hat{\Phi}^\pm\) in the orthonormal basis of dark and bright states, which is discussed in detail in appendix C. Furthermore it is assumed that the time \(t\) is much larger than the decay time of the bath correlation functions and therefore the upper bound of the integral in the master equation (5.64) can be replaced by infinity. Together with a change of variable \(\tau = t - t'\) this yields:

\[
\frac{d}{dt}\langle D_+, 1| \rho(t)|D_+, 1 \rangle = -\frac{N-1}{N} \langle D_+, 1| \rho(t)|D_+, 1 \rangle \int_0^\infty d\tau \left(A(\tau)e^{-i\omega_\tau} + \text{c.c.}\right) \\
+ \frac{1}{N} \sum_{l=1}^{N-1} \langle B_{l-1}, 1| \rho(t)|B_{l-1}, 1 \rangle \int_0^\infty d\tau \left(A(\tau)e^{i\omega_\tau} + \text{c.c.}\right) \\
- \frac{1}{N} \langle D_+, 1| \rho(t)|D_+, 1 \rangle \int_0^\infty d\tau \left(A(\tau) + \text{c.c.}\right) \\
+ \frac{1}{N} \langle D_-, 1| \rho(t)|D_-, 1 \rangle \int_0^\infty d\tau \left(A(\tau) + \text{c.c.}\right) \\
+ (\sqrt{2}\langle b^N| \rho(t)|b^N, 2 \rangle + \text{H.c.}) \int_0^\infty d\tau \left(B(\tau)e^{i\omega_\tau} + \text{c.c.}\right) \tag{5.65} \\
- \sqrt{6}e^{-i\omega_\tau}\langle D_+, 3| \rho(t)|D_+, 1 \rangle \int_0^\infty d\tau B(\tau) + \text{H.c.} \\
- 3\langle D_+, 1| \rho(t)|D_+, 1 \rangle \int_0^\infty d\tau \left(B(\tau)e^{-i\omega_\tau} + \text{c.c.}\right) \\
+ 2\langle D_+, 2| \rho(t)|D_+, 2 \rangle \int_0^\infty d\tau \left(B(\tau)e^{i\omega_\tau} + \text{c.c.}\right) \\
+ \langle b^N| \rho(t)|b^N \rangle \int_0^\infty d\tau \left(B(\tau)e^{i\omega_\tau} + \text{c.c.}\right),
\]

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where $A(\tau)$ and $B(\tau)$ are defined as

$$A(\tau) = \int_0^\infty d\omega h(\omega) |g_{c_+c_-}(\omega)|^2 \left[ e^{-i\omega(\tau)}[\bar{n}(\omega) + 1] + e^{i\omega(\tau)}\bar{n}(\omega) \right], \quad (5.66)$$

$$B(\tau) = \int_0^\infty d\omega h(\omega) |g_{c_+b}(\omega)|^2 \left[ e^{-i\omega(\tau)}[\bar{n}(\omega) + 1] + e^{i\omega(\tau)}\bar{n}(\omega) \right]. \quad (5.67)$$

The equation of motion for the matrix element $\langle D_-, 1|\rho^I(t)|D_-, 1 \rangle$ is given by:

$$\frac{d}{dt}\langle D_-, 1|\rho^I(t)|D_-, 1 \rangle = -\frac{N-1}{N} \langle D_-, 1|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau (A(\tau)e^{-i\omega g\tau} + \text{c.c.})$$

$$+ \frac{1}{N} \sum_{l=1}^{N-1} \langle B_{l+}, 1|\rho^I(t)|B_{l+}, 1 \rangle \int_0^\infty d\tau (A(\tau)e^{i\omega g\tau} + \text{c.c.})$$

$$- \frac{1}{N} \langle D_-, 1|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau (A(\tau) + \text{c.c.})$$

$$+ \frac{1}{N} \langle D_+, 1|\rho^I(t)|D_+, 1 \rangle \int_0^\infty d\tau (A(\tau) + \text{c.c.})$$

$$+ (\sqrt{2}\langle b^N|\rho^I(t)|D_-, 2 \rangle + \text{H.c.}) \int_0^\infty d\tau (C(\tau)e^{i\omega g\tau} + \text{c.c.})$$

$$- \sqrt{6}e^{-i\omega g\tau}\langle D_-, 3|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau C(\tau) + \text{H.c.}$$

$$- 3\langle D_-, 1|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau (C(\tau)e^{-i\omega g\tau} + \text{c.c.})$$

$$+ 2\langle D_-, 2|\rho^I(t)|D_-, 2 \rangle \int_0^\infty d\tau (C(\tau)e^{i\omega g\tau} + \text{c.c.})$$

$$+ \langle b^N|\rho^I(t)|b^N \rangle \int_0^\infty d\tau (C(\tau)e^{i\omega g\tau} + \text{c.c.}),$$

where $C(\tau)$ is defined as

$$C(\tau) = \int_0^\infty d\omega h(\omega) |g_{c_-b}(\omega)|^2 \left[ e^{-i\omega(\tau)}[\bar{n}(\omega) + 1] + e^{i\omega(\tau)}\bar{n}(\omega) \right]. \quad (5.69)$$
The equation of motion which describes the time evolution of the coherence between the two states $|D_+, 1\rangle$ and $|D_-, 1\rangle$ is given by:

$$
\frac{d}{dt} \langle D_+, 1|\rho^I(t)|D_-, 1 \rangle = \frac{d}{dt}(\langle D_-, 1|\rho^I(t)|D_+, 1 \rangle)^* - \frac{N-1}{N} \langle D_+, 1|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau (A(\tau)e^{-i\omega_g \tau} + c.c)
$$

$$+ \frac{1}{N} \sum_{l=1}^{N-1} \sum_{l=1}^{N-1} \langle B_{l-1}, 1|\rho^I(t)|B_{l+1}, 1 \rangle \int_0^\infty d\tau (A(\tau)e^{i\omega_g \tau} + c.c)
$$

$$- \frac{1}{N} \langle D_+, 1|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau (A(\tau) + c.c.)
$$

$$+ \frac{1}{N} \langle D_-, 1|\rho^I(t)|D_+, 1 \rangle \int_0^\infty d\tau (A(\tau) + c.c.)
$$

$$- \sqrt{6}e^{-i\omega_g t}\langle D_+, 3|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau B(\tau)
$$

$$- \sqrt{6}e^{i\omega_g t}\langle D_+, 1|\rho^I(t)|D_-, 3 \rangle \int_0^\infty d\tau C^*(\tau)
$$

$$- 3\langle D_+, 1|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau B(\tau)e^{-i\omega_g \tau}
$$

$$- 3\langle D_+, 1|\rho^I(t)|D_-, 1 \rangle \int_0^\infty d\tau C^*(\tau)e^{i\omega_g \tau}.
$$

(5.70)

Using the rotating wave approximation by neglecting the highly oscillatory terms which are proportional to exp{$\pm i\omega_g t$} and calculating the integrals yields the following results:

$$
\frac{d}{dt} \langle D_+, 1|\rho^I(t)|D_+, 1 \rangle =
$$

$$- \frac{N-1}{N} f_A(\omega_g)\bar{n}(\omega_g)\langle D_+, 1|\rho^I(t)|D_+, 1 \rangle + \frac{1}{N} f_A(\omega_g)[\bar{n}(\omega_g) + 1] \sum_{l=1}^{N-1} \langle B_{l-1}, 1|\rho^I(t)|B_{l+1}, 1 \rangle
$$

$$- \frac{1}{N} f_A(0)[2\bar{n}(0) + 1] \langle D_+, 1|\rho^I(t)|D_+, 1 \rangle + \frac{1}{N} f_A(0)[2\bar{n}(0) + 1] \langle D_-, 1|\rho^I(t)|D_-, 1 \rangle
$$

$$- 2f_B(\omega_g)\bar{n}(\omega_g)\langle D_+, 1|\rho^I(t)|D_+, 1 \rangle + 2f_B(\omega_g)[\bar{n}(\omega_g) + 1] \langle D_+, 2|\rho^I(t)|D_+, 2 \rangle
$$

$$- f_B(\omega_g)\bar{n}(\omega_g)\langle D_+, 1|\rho^I(t)|D_+, 1 \rangle + f_B(\omega_g)[\bar{n}(\omega_g) + 1] \langle b^N|\rho^I(t)|b^N \rangle
$$

$$+ \sqrt{2}f_B(\omega_g)[\bar{n}(\omega_g) + 1](b^N|\rho^I(t)|D_+, 2) + H.c.)
$$

(5.71)
The exact calculation of the integrals is given in appendix C. The master equation for the matrix element $\langle D_-, 1| \rho^f(t)|D_-, 1 \rangle$ is given by:

$$
\frac{d}{dt} \langle D_-, 1| \rho^f(t)|D_-, 1 \rangle =
$$

$$
- \frac{N - 1}{N} f_A(\omega_g) \bar{n}(\omega_g) \langle D_+, 1| \rho^f(t)|D_-, 1 \rangle + \frac{1}{N} f_A(\omega_g) [\bar{n}(\omega_g) + 1] \sum_{l=1}^{N-1} \langle B_{l+}, 1| \rho^f(t)|B_{l+}, 1 \rangle
$$

$$
\frac{1}{N} f_A(0) [2\bar{n}(0) + 1] \langle D_-, 1| \rho^f(t)|D_-, 1 \rangle + \frac{1}{N} f_A(0) [2\bar{n}(0) + 1] \langle D_+, 1| \rho^f(t)|D_+, 1 \rangle
$$

$$
- \frac{1}{N} f_C(\omega_g) \bar{n}(\omega_g) \langle D_-, 1| \rho^f(t)|D_-, 1 \rangle + \frac{1}{N} f_C(\omega_g) \bar{n}(\omega_g) \langle D_+, 1| \rho^f(t)|D_+, 1 \rangle
$$

$$
- f_C(\omega_g) \bar{n}(\omega_g) \langle D_+, 1| \rho^f(t)|D_+, 1 \rangle + f_C(\omega_g) \bar{n}(\omega_g) \langle D_-, 1| \rho^f(t)|D_-, 1 \rangle
$$

$$
+ \sqrt{2} f_C(\omega_g) \bar{n}(\omega_g) \langle D_-, 1| \rho^f(t)|D_-, 2 \rangle + \text{H.c.},
$$

where $\bar{n}(\omega)$ is the mean photon number for an oscillator with frequency $\omega$ in thermal equilibrium at temperature $T$ and is given in equation (5.26). The coefficients $f_X(\omega)$, $X \in \{A, B, C\}$, are of the order $\mathcal{O}(f_X(\omega)) \leq 1$ and are defined by:

$$
f_A(\omega) = 2\pi h(\omega)|g_{c\rightarrow e}(\omega)|^2, \quad (5.73)
$$

$$
f_B(\omega) = 2\pi h(\omega)|g_{c\rightarrow b}(\omega)|^2 \quad \text{and} \quad (5.74)
$$

$$
f_C(\omega) = 2\pi h(\omega)|g_{c\rightarrow b}(\omega)|^2, \quad (5.75)
$$

where $h(\omega)$ is the density of oscillators at frequency $\omega$ introduced in section 5.2.2. The master equation for the matrix element $\langle D_+, 1| \rho^f(t)|D_-, 1 \rangle$ is given by:

$$
\frac{d}{dt} \langle D_+, 1| \rho^f(t)|D_-, 1 \rangle =
$$

$$
- \frac{N - 1}{N} f_A(\omega_g) \bar{n}(\omega_g) \langle D_+, 1| \rho^f(t)|D_-, 1 \rangle + \frac{1}{N} f_A(\omega_g) [\bar{n}(\omega_g) + 1] \sum_{l=1}^{N-1} \langle B_{l+}, 1| \rho^f(t)|B_{l+}, 1 \rangle
$$

$$
\frac{1}{N} f_A(0) [2\bar{n}(0) + 1] \langle D_+, 1| \rho^f(t)|D_-, 1 \rangle + \frac{1}{N} f_A(0) [2\bar{n}(0) + 1] \langle D_-, 1| \rho^f(t)|D_+, 1 \rangle
$$

$$
- f_B(\omega_g) \bar{n}(\omega_g) \langle D_+, 1| \rho^f(t)|D_-, 1 \rangle - 3 f_C(\omega_g) \bar{n}(\omega_g) \langle D_+, 1| \rho^f(t)|D_-, 1 \rangle.
$$

The matrix elements $\langle D_\pm, 1| \rho^f(t)|D_\pm, 1 \rangle$ describe the population in the dark states $|D_\pm, 1 \rangle$. Their evolution in time is illustrated in figure 5.4 (a) and (b). Since their interpretations
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Figure 5.4: Illustration of the dynamics of the population in the dark states $|D_+, 1\rangle$ (a) and $|D_-, 1\rangle$ (b). The subspace $\mathcal{M} = \{|D_+, 1\rangle, |D_-, 1\rangle\}$ is energetically separated from all other collective states via the energy gap $E_g$. 

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are similar in the following only the evolution of the matrix element $\langle D_+, |\rho^I(t)| D_+, 1 \rangle$ given by equation (5.71) is described in detail.

The first two summands in equation (5.71), labelled $a_1$ and $a_2$, describe transitions between the dark state $|D_+, 1 \rangle$ and the bright states $|B_{l-1}, 1 \rangle$, $l \in \{1, 2, \ldots, N - 1 \}$. These transitions are caused by a spin flip between the atomic states $|c_+ \rangle$ and $|b \rangle$. The first summand describes the losses out of the population of the dark state $|D_+, 1 \rangle$. Since $\frac{N-1}{N} f_A(\omega_g)$ is of the order one, the first summand scales with $\bar{n}(\omega_g)$, which is the mean photon number of bath oscillators with the frequency $\omega_g$ at temperature $T$. Since the energy gap $E_g = \hbar \omega_g$ is assumed to be sufficiently large, it is justified to conclude that the mean photon number $\bar{n}(\omega_g)$ at the corresponding high frequency $\omega_g$ and at a moderate temperature is zero:

$$\bar{n}(\omega_g) \approx \exp \left\{ - \frac{\hbar \omega_g}{k_b T} \right\} \approx 0.$$  (5.77)

This just states that the bath can not provide the large energy $E_g$ to induce a transition from the dark state $|D_+, 1 \rangle$ to a bright state $|B_{l-1}, 1 \rangle$. Therefore this kind of transition is exponentially suppressed. The second term, labelled $a_2$, scales, due the summation over $l$, as $\bar{n}(\omega_g) + 1 \approx 1$ and describes transitions from the bright states $|B_{l-1}, 1 \rangle$ to the lower dark state $|D_+, 1 \rangle$. According to the storage procedure these bright states are not populated and do therefore not change the population in the dark state $|D_+, 1 \rangle$.

It is important to mention that there is no direct transition between the state $|D_+, 1 \rangle$ and the bright state $|B_{l+1}, 1 \rangle$. This is caused by the fact that such a transition can not be achieved by a single spin flip. For such a transition at least two spin flips are necessary, for example one from an atomic state $|c_+ \rangle$ to the atomic state $|b \rangle$ and one from an atomic state $|b \rangle$ to the atomic state $|c_+ \rangle$.

The third and the fourth summand, labelled $a_3$ and $a_4$, describe transitions within the subspace $\mathcal{M} = \{|D_+, 1 \rangle, |D_-, 1 \rangle\}$ spanned by the two dark states $|D_+, 1 \rangle$ and $|D_-, 1 \rangle$. Since the order of $f_A(0) [2 \bar{n}(0) + 1]$ is smaller or equal than one, these two terms scale like $1/N$ and can therefore be neglected for a large number $N$ of atoms. In the limit of a large number of atoms there are no transitions within the subspace $\mathcal{M}$ which is a necessary condition for a quasi decoherence free subspace.

The fifth and the sixth summand, labelled $a_5$ and $a_6$ in equation (5.71) describe transitions between the dark state $|D_+, 1 \rangle$ with one photonic excitation in the atomic state $|c_+ \rangle$ and the dark state $|D_+, 2 \rangle$ with two atomic excitations in the atomic state $|c_+ \rangle$. Due to the energy necessary to provide a transition from the lower state $|D_+, 1 \rangle$ into the higher state $|D_+, 2 \rangle$ the fifth summand is exponentially suppressed (see equation (5.77)). The sixth summand does not contribute to the population of the dark state $|D_+, 1 \rangle$ since the state $|D_+, 2 \rangle$ is not populated during the loading process.
The summands $a_7$ and $a_8$ describe transitions between the dark state $|D_{+},1\rangle$ and the ground state $|b^N\rangle = |b_1, b_2, \ldots, b_N\rangle$. This former ground state is, due to the additional nonlinear interaction which creates the energy gap, energetically higher than the subspace $\mathcal{M}$. Therefore the transition from the dark state $|D_{+},1\rangle$ into the state $|b^N\rangle$ is also exponentially suppressed (equation (5.77)). After the storage procedure is completed and at the beginning of the storage period the collective state $|b^N\rangle$ is not populated and therefore the eighth summand does not contribute to the population of the dark state $|D_{+},1\rangle$.

The ninth term $a_9$ in equation (5.71) results from an off diagonal element of the density matrix. Those terms can appear by changing from the pure atomic basis into a basis which uses linear combinations of the pure atomic states. Since this matrix element is initially zero at the beginning of the storage period it does not contribute to the population of the dark state $|D_{+},1\rangle$.

The explanation of equation (5.72) which describes the dynamics of the population in the dark state $|D_{-},1\rangle$ is analogue to the above explanation. At the beginning of the storage period only the dark states $|D_{+},1\rangle$ and $|D_{-},1\rangle$ are populated, depending on the stored photon. The equations (5.71) and (5.72) show that all possible transitions out of the subspace $\mathcal{M}$ which is spanned by the two dark states $|D_{+},1\rangle$ and $|D_{-},1\rangle$ caused by an individual one atom spin flip into all other states is exponentially suppressed due to the presence of the energy gap $E_g$. The transitions within the subspace $\mathcal{M}$ scale like $1/N$ and can therefore be neglected for a large number $N$ of atoms which is a necessary condition for the proposed storage technique. But before concluding that the subspace $\mathcal{M}$ is a quasi decoherence free subspace first the dynamics of the matrix elements $\langle D_{\pm},1|\rho(t)|D_{\mp},1\rangle$ has to be considered to justify that the coherence between the two states $|D_{+},1\rangle$ and $|D_{-},1\rangle$ is not destroyed by the interaction of the system with its environment. The dynamics of the matrix elements $\langle D_{\pm},1|\rho(t)|D_{\mp},1\rangle$ are described by equation (5.76). The terms which can lead to decoherence (summands $c_1$ and $c_5$) are suppressed due to the energy gap or scale like $1/N$ (summands $c_3$ and $c_4$) and can therefore be neglected for large $N$. The second and sixth summand do not play a role since these matrix elements are initially zero.

Therefore the conclusion can be drawn that the additional nonlinear interaction which causes the large energy gap creates a decoherence free subspace of dimension two. The transitions out of the subspace $\mathcal{M}$ caused by individual one atom spin flips are energetically suppressed and the transitions within the subspace $\mathcal{M}$ scale as the inverse of the involved number of atoms. Therefore it has been shown that it is possible to construct a quasi decoherence free subspace even if all atoms of the ensemble couple to individual and independent reservoirs which causes one atom spin flips.

It is important to note that the scheme proposed above is not restricted to the storage
5.2. Quasi decoherence free subspaces in collective atomic ensembles

Figure 5.5: Adiabatic transfer of the flying qubits $|\varphi_i\rangle = \alpha_i|1_{i+}\rangle + \beta_i|1_{i-}\rangle$, $i \in \{1, 2, \ldots, m\}$ with two polarization directions $\pm$ to two internal atomic states $|c_{i+}\rangle$ and $|c_{i-}\rangle$. The positions of the levels do not represent relative energies.

of a single qubit. By using different modes for the flying qubits it should be possible to generalize the discussed idea to quasi decoherence free subspaces with a dimension larger than two. In order to do so it is necessary to consider a $2m$-mode quantum memory consisting of $N$ atoms. Each atom has a ground state $|b\rangle$ and $2m$ other meta stable states $|c_{1+}\rangle, |c_{2+}\rangle, \ldots, |c_{m+}\rangle$ and $|c_{1-}\rangle, |c_{2-}\rangle, \ldots, |c_{m-}\rangle$. In such a system $m$ qubits represented by photons

$$|\varphi_i\rangle = \alpha_i|1_{i+}\rangle + \beta_i|1_{i-}\rangle, \ i \in \{1, 2, \ldots, m\}$$

with two possible polarization directions $\pm$ can be stored. The storage can be achieved by a coupling scheme as shown in figure 5.5. By adiabatic following the photons can be transferred into the storage states

$$|D_{i\pm}, 1\rangle = \alpha_i|D_{i+}, 1\rangle + \beta_i|D_{i-}, 1\rangle,$$

where the states $|D_{i\pm}, 1\rangle$ are the states defined in equation (5.4) with the internal atomic states $|c_{i\pm}\rangle$. By using an additional nonlinear interaction to create an energy gap which shifts the subspace spanned by the states $|D_{i\pm}, 1\rangle$, $i \in \{1, 2, \ldots, m\}$, it is possible to generalize the above discussion to create a decoherence free subspace $\mathcal{M}$ of dimension $2m$ spanned by the states $|D_{i\pm}, 1\rangle$.

In summary it has been shown that photonic qubits stored in collective atomic superpositions can be protected from decoherence induced by individual atomic spin flips by creating quasi decoherence free subspaces. Nevertheless it has to be shown in further investigations how the additional interaction described by the Hamiltonian (5.11) to create the necessary energy gap can experimentally be implemented. The experimental realization will depend strongly on the used physical system.
5.3 Suppression of collisional decoherence in atomic vapors

In the previous section the existence of a quasi decoherence free subspace which is immune against individual atomic spin flips caused by the interaction of the system with its environment has been demonstrated. Most experiments which use the proposed light storage technique are performed in atomic vapors [27, 32, 96]. Therefore it is necessary to exclude other sources of decoherence present in those systems. In atomic vapors one possible source of decoherence, besides individual spin flips is magnetic state decoherence caused by elastic collisions. Those collisions redistribute the population among different Zeeman sublevels. Elastic collisions of an atom in a thermal vapor with a bath of foreign gas perturbers for example cause a depolarization of any initial magnetic state coherence present in the considered atom. This leads to a different occupation of the magnetic sublevels as well as dephasing of those. The depolarization caused by elastic collisions can be described in a semiclassical model in which the atoms move along classical trajectories [130]. The interaction itself is modelled as a van der Waals interaction varying as the inverse of the distance between the atoms to the power of six. The collision rate $\Gamma_{\text{col}}$ in this model can be expressed by the impact parameter $b$ and the relative speed $v$ of the colliding atoms and is equal to $v/b$.

One method, well known in NMR setups, to decouple unwanted spin-spin interactions is using open-loop control [106, 131]. This method can also be applied to suppress decoherence in various other systems. First generalizations were done for example by L. Viola and S. Lloyd [107, 132] and M. Ban [133]. They have shown that a series of short $\pi$-pulses can be used to eliminate pure dephasing of a spin caused by the interaction with a quantum reservoir. The suppression of magnetic state decoherence caused by collisions in an atomic vapor has been studied for special cases by C. Search and P.R. Berman [110, 130]. A more general discussion of decoupling for a generic open quantum system from any environmental interaction is proposed by L. Viola et al. [108] and C. Uchiyama and M. Aihara [134, 135]. Unfortunately the noise suppression procedure given in these references is not constructive. Therefore in many realistic cases it is not clear how the dynamical decoupling Hamiltonian has to be chosen.

In the following an atomic vapor is considered to store a flying qubit. The storage procedure is based on the scheme proposed by M. Fleischhauer et al. [1–3], which was discussed in the previous chapters. The goal in this section is to suppress magnetic state decoherence caused by elastic collisions. The idea to suppress decoherence is to perturb the relevant state amplitudes on a time scale which is short compared to the correlation time of the
bath which describes the environment of the system. In the case of an atomic vapor the correlation time of single collision events which causes the magnetic state decoherence is of the order of the collision duration and depends strongly on the specific parameters as the temperature and the atom density of the considered system. The additional periodic perturbation has to be on a time scale which is shorter than the collision duration.

5.3.1 Model

In the following the storage scheme introduced in section 5.2.1 is used to store a photonic qubit $|\varphi\rangle = \alpha_+|1_+\rangle + \alpha_-|1_-\rangle$ with a single photon in two orthogonal modes. To model the collisional dephasing process of the magnetic sublevels the structure of the different sublevels has to be modified. It is assumed that the atomic ground state $|b\rangle$ has the total angular momentum $J_b = 0$ and is not degenerated with respect to its magnetic quantum number $m_b = 0$. In two parallel Λ transitions the orthogonal photonic modes $\hat{a}_+$ and $\hat{a}_-$ couple resonantly the transition between the ground state $|b\rangle$ and the excited states $|a_+\rangle$ and $|a_-\rangle$ respectively. The excited states are assumed to be states with magnetic quantum numbers $m_{a_+} = +1$ and $m_{a_-} = -1$. They belong to a magnetic submanifold with total angular momentum $J_a = 1$. The upper states $|a_+\rangle$ and $|a_-\rangle$ are furthermore coupled to the metastable states $|c_{+2}\rangle$ and $|c_{-2}\rangle$ via two coherent control fields with time dependent Rabi frequencies $\Omega_{+}(t)$ and $\Omega_{-}(t)$. The two states $|c_{+2}\rangle$ and $|c_{-2}\rangle$ which are used for the storage purpose belong to a magnetic submanifold with total angular momentum $J_c = 2$. It is assumed that the states $|c_{\pm 2}\rangle$ have the magnetic quantum numbers $m_{c_{\pm 2}} = \pm 2$. The storage scheme is illustrated in figure 5.6. Initially the ensemble of atoms is assumed to be prepared in the ground state $|b\rangle$. The mixing angles $\theta_{\pm}(t)$ are defined by $\tan \theta_{\pm}(t) = \frac{g_{\pm}\sqrt{N}}{\Omega_{\pm}(t)}$ where $g_{\pm}$ are the Rabi frequencies of the quantum field described by $\hat{a}_{\pm}$. Changing the mixing angles $\theta_{\pm}(t)$ from 0 to $\pi/2$ adiabatically in time, the photonic qubit $|\varphi\rangle$ is transferred to a collective storage state. The relevant collective states of the ensemble can be described in analogy to section 5.2.1 in terms of dark and bright states. The dark and bright state with one excitation $|D_{\pm,1}\rangle_N$ and $|B_{\pm,1}\rangle_N$ read$^3$

$$|D_{\pm,1}\rangle = -\frac{1}{\sqrt{N}} \sum_{j=1}^{N} |b_1, \ldots, c_{\pm 2j}, \ldots, b_N\rangle$$

(5.80)

$$|D_{\pm,1}\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |b_1, \ldots, c_{\pm 2j}, \ldots, b_N\rangle \exp \left\{ -2\pi i \frac{l_j}{N} \right\} .$$

(5.81)

$^3$Here the index $N$ has been suppressed to simplify the notation for the following calculations.
5.3. Suppression of collisional decoherence in atomic vapors

Figure 5.6: Adiabatic transfer of the flying qubits $|\varphi\rangle = \alpha |1_+\rangle + \alpha_- |1_-\rangle$ with two polarization directions $\pm$ to two internal atomic states $|c_{+2}\rangle$ and $|c_{-2}\rangle$.

In analogy to the existing experiments it is assumed that the atoms which are used for the storage procedure undergo collisions with themselves or with a bath of foreign gas perturbers. These collisions lead to dephasing of the atomic state and to transitions within the submanifolds with the same total angular momentum. These processes are modelled by virtual transitions within an atom. A virtual transition is a transition from the initial state to an higher internal state of the atom and back to the magnetic manifold of the initial state. Collisions between atoms which belong to the storage ensemble can further generate a state exchange during a collision. Since only one photonic excitation is transferred to the atomic ensemble, collisions between two atoms which are both in a state within the manifold with $J_c = 2$ cannot occur. Therefore a state exchange caused by a collision is only possible if one atom in state $|c_s\rangle$, $s \in \mathcal{S} = \{0, \pm 1, \pm 2\}$ collides with an atom in state $|b\rangle$.

After the adiabatic rotation the free dynamics of the atomic ensemble which is used for the storage scheme is described by the following Hamiltonian:

$$\hat{H}_{free} = \hbar \omega_c \sum_{j=1}^{N} \sum_{s \in \mathcal{S}} |c_s\rangle_{jj} \langle c_s|, \text{ with } \mathcal{S} = \{0, \pm 1, \pm 2\}. \tag{5.82}$$

Since the dynamics of the buffer gas atoms are not of interest the possible collisions can be modelled by an effective interaction Hamiltonian $\hat{H}_{int} = \hat{H}_1 + \hat{H}_2 + \hat{H}_3$ which can be separated into three parts. The Hamiltonian $\hat{H}_1$ describes the possible transitions between
the magnetic sublevels $|c_s\rangle$, $s \in S$ and is defined as

$$
\hat{H}_1 = \hbar \sum_{j=1}^{N} \alpha_j^1 [ |c_{j-2}\rangle \langle c_{j-1}| + |c_{j+2}\rangle \langle c_{j+1}| + \text{H.c.} ] 
+ \hbar \sum_{j=1}^{N} \alpha_j^2 [ |c_{j-1}\rangle \langle c_0| + |c_{j+1}\rangle \langle c_0| + \text{H.c.} ] 
+ \hbar \sum_{j=1}^{N} \alpha_j^3 [ |c_{j-1}\rangle \langle c_0| + |c_{j+1}\rangle \langle c_0| + \text{H.c.} ] 
+ \hbar \sum_{j=1}^{N} \alpha_j^4 [ |c_{j-1}\rangle \langle c_{j+1}| + \text{H.c.} ],
$$

(5.83)

where $\alpha_j^1$, $\alpha_j^2$, $\alpha_j^3$ and $\alpha_j^4$ are the transition frequencies between the different magnetic sublevels. Due to the selection rules $\Delta J = J_{\text{final}} - J_{\text{initial}} = \pm 1$ and $\Delta m = m_{\text{final}} - m_{\text{initial}} = 0, \pm 1$ there is no direct transition between the two states $|c_{j+2}\rangle$ and $|c_{j-2}\rangle$ which are used as metastable states for the quantum memory. $J_{\text{initial}}$ and $m_{\text{initial}}$ refer to the initial total angular momentum and magnetic quantum number at the beginning of the virtual transition. $J_{\text{final}}$ and $m_{\text{final}}$ refer to the final total angular momentum and magnetic quantum number at the end of this virtual transition caused by a collision. Furthermore it is assumed that transitions between states with the same total angular momentum and the same absolute value of the magnetic quantum number have the same transition frequencies.

The Hamiltonian $\hat{H}_2$ describes the virtual transitions which end up again in the initial state and is given by

$$
\hat{H}_2 = \hbar \sum_{j=1}^{N} \beta_j^1 |b\rangle \langle b_j| + \hbar \sum_{j=1}^{N} \beta_0^1 |c_0\rangle \langle c_j| 
+ \hbar \sum_{j=1}^{N} \beta_j^2 [ |c_{j-1}\rangle \langle c_{-1}| + |c_{j+1}\rangle \langle c_1| ] 
+ \hbar \sum_{j=1}^{N} \beta_j^3 [ |c_{j-1}\rangle \langle c_{-1}| + |c_{j+1}\rangle \langle c_1| ]
$$

(5.84)

The different transitions described by $\hat{H}_1$ and $\hat{H}_2$ are illustrated in figure 5.7.

The Hamiltonian $\hat{H}_3$ describes the state exchange caused by a collision of an atom in state $|c_s\rangle$, $s \in S$, and an atom in the ground state $|b\rangle$ and reads:

$$
\hat{H}_3 = \frac{\hbar}{2} \sum_{i,j=1}^{N} \sum_{s \in S} g_{ij} [ |b_1, \ldots, c_{s_i}, \ldots, b_N\rangle \langle b_1, \ldots, b_N| + \text{H.c.} ]
$$

(5.85)
5.3.2 Suppression of decoherence caused by atomic collisions

Initially only one excitation is stored in the atomic ensemble. Since it is assumed that the collisions do not create additional excitations, the most general state vector with maximal one atomic excitation can be written in the following form:

$$|\Psi(t)\rangle = b(t)|b_1, \ldots, b_N\rangle + \sum_{j=1}^{N} \sum_{s \in S} c_s^j(t)|b_1, \ldots, c_{s_j}, \ldots, b_N\rangle, \quad S = \{0, \pm 1, \pm 2\}. \quad (5.86)$$

The flying qubit $|\varphi\rangle$ is initially stored in the two magnetic sublevels $|c_{2+}\rangle$ and $|c_{2-}\rangle$ an additional fast periodic interaction is applied which freezes the dynamics of these two states. Therefore an additional fast oscillating electric field is applied which leads to a time dependent Stark shift of the magnetic sublevels. Hereby the magnetic sublevels with the same absolute magnetic quantum number experience the same shift. An illustration of the effect of the additional electric field is given it figure 5.8. For simplicity it is assumed that the electric field can be described by an cosine with frequency $\omega$ and amplitude $V_0$. Therefore the effect of the additional fast oscillating electric field can be described by the
5.3. Suppression of collisional decoherence in atomic vapors

Figure 5.8: Illustration of the Stark shift caused by the additional electric field $V_0$.

effective Hamiltonian $\hat{H}_{\text{eff}}$ which is given by

$$\hat{H}_{\text{eff}} = \hbar f V_0 \cos(\omega t) \sum_{j=1}^{N} |b\rangle_{jj} \langle b| + \hbar f_0 V_0 \cos(\omega t) \sum_{j=1}^{N} |c_0\rangle_{jj} \langle c_0|$$

$$+ \hbar f_1 V_0 \cos(\omega t) \sum_{j=1}^{N} [ |c_{-1}\rangle_{jj} \langle c_{-1}| + |c_{+1}\rangle_{jj} \langle c_{+1}| ]$$

$$+ \hbar f_2 V_0 \cos(\omega t) \sum_{j=1}^{N} [ |c_{-2}\rangle_{jj} \langle c_{-2}| + |c_{+2}\rangle_{jj} \langle c_{+2}| ],$$

where the coefficients $f$, $f_0$, $f_1$ and $f_2$ account for the different Stark shifts of the different magnetic sublevels.

The equations of motion for the state amplitudes are obtained from the Schrödinger equation

$$i\hbar \frac{d}{dt} |\Psi(t)\rangle = (\hat{H}_{\text{int}} + \hat{H}_{\text{eff}}) |\Psi(t)\rangle.$$

They have the following form:

$$c_{\pm 2}(t) = -i \left[ \omega_e + \beta_2^j + \sum_{k=1,k\neq j}^{N} \beta^j + V_0 f_2 \cos(\omega t) + (N - 1)V_0 f \cos(\omega t) \right] c_{\pm 2}(t)$$

$$- \frac{i}{2} \sum_{k=1,k\neq j}^{N} g_{kj} b(t) - i\alpha_1^j c_{\pm 1}(t) - i\alpha_2^j c_0^j(t),$$

(5.89)
\[ c_{\pm 1}(t) = \begin{aligned} & -i \left[ \omega_c + \beta_1^0 + \sum_{k=1, k \neq j}^N \beta_j^k + V_0 f_1 \cos(\omega t) + (N - 1)V_0 f \cos(\omega t) \right] c_{\pm 1}^j(t) \\
& - \frac{i}{2} \sum_{k=1, k \neq j}^N g_k b(t) - i \alpha_1^0 c_{\pm 2}^j(t) - i \alpha_3^0 c_0^j(t) - i \alpha_4^0 c_{\mp 1}^j(t), \\
\end{aligned} \tag{5.90} \]

\[ c_0^j(t) = \begin{aligned} & -i \left[ \omega_c + \beta_0^0 + \sum_{k=1, k \neq j}^N \beta_j^k + V_0 f_0 \cos(\omega t) + (N - 1)V_0 f \cos(\omega t) \right] c_0^j(t) \\
& - \frac{i}{2} \sum_{k=1, k \neq j}^N g_k b(t) - i \alpha_2^0[c_{-2}^j(t) + c_{-1}^j(t)] - i \alpha_3^j[c_{-1}^j(t) + c_{+1}^j(t)], \\
\end{aligned} \tag{5.91} \]

\[ \dot{b}(t) = -i \sum_{j=1}^N \beta_j^j + NV_0 f \cos(\omega t) \left] b(t) - \frac{i}{2} \sum_{j,k=1, j \neq k}^N g_{jk} \sum_{s \in S} c_s^j(t). \right. \tag{5.92} \]

It is convenient to introduce the following rotating amplitudes:

\[ c_{\pm m}^j(t) = c_{\pm 2}^j(t) e^{-i \omega_c t - i \beta_2^j t - i \sum_{k=1, k \neq j}^N \beta_k^1 t - \frac{V_0}{\omega} f_m \sin(\omega t) - i \frac{V_0}{\omega} f(N-1) \sin(\omega t)} \tag{5.93} \]

\[ m \in \{0, 1, 2\}, \tag{5.94} \]

\[ b(t) = \dot{b}(t) e^{-i \sum_{j=1}^N \beta_j^j t - \frac{V_0}{\omega} f(N-1) \sin(\omega t)}. \tag{5.95} \]

With the new rotating variables the equations of motion read

\[ \dot{c}_{\pm 2}^j(t) = \begin{aligned} & - \frac{i}{2} \sum_{k=1, k \neq j}^N g_k \dot{b}(t) e^{i(\omega_c + \beta_2^j - \beta_1^j)} e^{i \frac{V_0}{\omega} (f_2 - f_1) \sin(\omega t)} - i \alpha_1^0 \dot{c}_{\pm 1}^j(t) e^{i \frac{V_0}{\omega} (f_2 - f_1) \sin(\omega t)} \\
& - i \alpha_3^0 \dot{c}_0^j(t) e^{i \frac{V_0}{\omega} (f_2 - f_0) \sin(\omega t)}, \end{aligned} \tag{5.96} \]

\[ \dot{c}_{\pm 1}^j(t) = \begin{aligned} & - \frac{i}{2} \sum_{k=1, k \neq j}^N g_k \dot{b}(t) e^{i(\omega_c + \beta_2^j - \beta_1^j)} e^{i \frac{V_0}{\omega} (f_1 - f_0) \sin(\omega t)} - i \alpha_1^0 \dot{c}_{\pm 2}^j(t) e^{i \frac{V_0}{\omega} (f_1 - f_2) \sin(\omega t)} \\
& - i \alpha_3^0 \dot{c}_0^j(t) e^{i \frac{V_0}{\omega} (f_1 - f_0) \sin(\omega t)} - i \alpha_3^j \dot{c}_{\pm 1}^j(t) + i(\beta_2^j - \beta_1^j) \dot{c}_{\pm 1}^j(t), \end{aligned} \tag{5.97} \]

\[ \dot{c}_0^j(t) = \begin{aligned} & - \frac{i}{2} \sum_{k=1, k \neq j}^N g_k \dot{b}(t) e^{i(\omega_c + \beta_2^j - \beta_1^j)} e^{i \frac{V_0}{\omega} (f_0 - f_1) \sin(\omega t)} - i \alpha_3^j \dot{c}_{-2}^j(t) + i \dot{c}_{+2}^j(t) e^{i \frac{V_0}{\omega} (f_0 - f_2) \sin(\omega t)} \\
& - i \alpha_3^j (c_{-1}^j(t) + c_{+1}^j(t)) e^{i \frac{V_0}{\omega} (f_0 - f_1) \sin(\omega t)} + i(\beta_2^j - \beta_0^j) \dot{c}_0^j(t), \end{aligned} \tag{5.98} \]

\[ \dot{\dot{b}}(t) = \begin{aligned} & - \frac{i}{2} \sum_{j,k=1, j \neq k}^N g_{jk} \sum_{m=1}^2 \dot{c}_{m}^j(t) + \dot{c}_{m}^j(t) e^{-i(\omega_c + \beta_2^j - \beta_1^j)t} e^{i \frac{V_0}{\omega} (f_0 - f_m) \sin(\omega t)} \\
& - \frac{i}{2} \sum_{j,k=1, j \neq k}^N g_{jk} \sum_{m=1}^2 \dot{c}_{m}^j(t) e^{-i(\omega_c + \beta_2^j - \beta_1^j)t} e^{i \frac{V_0}{\omega} (f_0 - f_m) \sin(\omega t)}. \end{aligned} \tag{5.99} \]

Since \( f, f_0, f_1 \) and \( f_2 \) are a measure of the strength of the Stark shift caused by the oscillating electric field, they are different for magnetic sublevels with different absolute
5.3. Suppression of collisional decoherence in atomic vapors

value of the magnetic quantum number. Therefore if the amplitude \( V_0 \) and the frequency \( \omega \) of the oscillating electric field can be chosen large enough that the terms which scale as 
\[ e^{i \frac{V_0}{\omega} (F - F') \sin(\omega t)}, \]
where \( F, F' \in \{ f, f_0, f_1 \} \) and \( F \neq F' \), make many oscillations during the typical time scale over which the state amplitudes changes significantly then in the rotating wave approximation these terms can be neglected and the dynamics of the amplitudes \( \tilde{c}_{\pm 2}(t) \) described by equation (5.96) simplifies to

\[
\dot{\tilde{c}}_{\pm 2}(t) \approx 0, \quad (5.100)
\]
i.e. the dynamics of the state amplitudes \( \tilde{c}_{\pm 2}(t) \) is frozen. However it is important to note that this result can only be obtained for the two atomic levels used for the storage scheme if there is no direct coupling between those. Translating equation (5.100) back into the original state amplitudes \( c_{\pm 2}(t) \) yields:

\[
c_{\pm 2}(t) = c_{\pm 2}(t = 0) e^{i(\beta_j - \beta_j^2)t} e^{-i\phi(t)}, \quad (5.101)
\]
where the phase \( \phi(t) \), which is independent of the atom number \( j \), is defined as:

\[
\phi(t) = -\omega c t + \sum_{j=1}^{N} \beta_j t + \frac{V_0}{\omega} f_2 \sin(\omega t) + \frac{V_0}{\omega} f (N - 1) \sin(\omega t). \quad (5.102)
\]
The result (5.101) show that the dynamics for the collective state \( |b_1, \ldots, c_{\pm 2j}, \ldots, b\rangle \) is just determined by additional phases. In the sense of quantum information however it is important that the dynamics of the storage state \( \alpha_+|D_+, 1\rangle + \alpha_-|D_-, 1\rangle \) is unchanged or just modified by a common phase factor. Therefore it is necessary to translate the results into equations for the dark and bright state amplitudes \( D_\pm(t) \) and \( B_{l\pm}(t) \) which are the amplitudes of the corresponding dark and bright states with one atomic excitation, i.e. \( |D_\pm, 1\rangle \) and \( |B_{l\pm}, 1\rangle \). Since it is assumed that initially the state \( \alpha_+|D_+, 1\rangle + \alpha_-|D_-, 1\rangle \) is stored, the initial values for the time dependent amplitudes are given by

\[
D_\pm(0) = \alpha_\pm, \quad B_{l\pm}(0) = 0, \quad \text{and} \quad c_{\pm 2}(0) = -\frac{\alpha_\pm}{\sqrt{N}}. \quad (5.103)
\]
The relations between the individual amplitudes \( c_{\pm 2}(t) \) and the collective amplitudes \( D_\pm(t) \) and \( B_{l\pm}(t) \) read:

\[
D_\pm(t) = -\frac{1}{\sqrt{N}} \sum_{j=1}^{N} c_{\pm 2}(t), \quad (5.104)
\]
\[
B_{l\pm}(t) = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} c_{\pm 2}(t) \exp \left\{ -2\pi i \frac{l j}{N} \right\}. \quad (5.105)
\]
Together with equation (5.101) this yields for the time evolution of the dark and bright state amplitudes:

\[
D_\pm(t) = -\frac{\alpha_\pm}{N} e^{-i\phi(t)} \sum_{j=1}^{N} e^{i(\beta_j - \beta_j')t},
\]

\[
B_{l\pm}(t) = -\frac{\alpha_\pm}{N} e^{-i\phi(t)} \sum_{j=1}^{N} e^{i(\beta_j - \beta_j')t} e^{-2\pi i lj/N}.
\]

Unfortunately these equations show that even if the two for the storage state relevant dark states, \(|D_+, 1\rangle\) and \(|D_-, 1\rangle\), have a just a common phase factor and the phase relation between them is unchanged, also the bright states \(|B_{l\pm}\rangle\) are populated. Even if it is possible to suppress dephasing processes caused by the collisions of the atoms by the additional fast periodic interaction, it is not possible to suppress a diffusion process which leads to an equal distributed population of all bright states with one atomic excitation. Nevertheless in section 5.2 it has been shown that all transition out of the subspace \(\mathcal{M} = \{|D_+, 1\rangle, |D_-, 1\rangle\}\) which is spanned by the two relevant storage states can be suppressed by using an additional nonlinear interaction which creates a sufficient large energy gap between the dark states \(|D_\pm, 1\rangle\) and all other states of the Hilbertspace. To suppress the diffusion process out of the subspace \(\mathcal{M}\) an additional collective interaction is introduced which can be described by the following Hamiltonian:

\[
\hat{H}_g = -\frac{\hbar \omega_g}{N} \sum_{j,k=1, s \in \mathcal{S}}^{N} (b_1, \ldots, c_{sj}, \ldots, b_N)\langle b_1, \ldots, c_{sk}, \ldots, b_N |.
\]

Here it has been assumed that the additional collective interaction effects all magnetic sublevels of the atomic state \(|c\rangle\) in the same way. The equations of motions for the individual state amplitudes \(c_s(t), s \in \mathcal{S}\), and \(b(t)\) are obtained from the following Schrödinger equation

\[
i \hbar \frac{d}{dt} |\Psi(t)\rangle = (\hat{H}_{\text{int}} + \hat{H}_{\text{eff}} + \hat{H}_g) |\Psi(t)\rangle.
\]

Using the same assumptions of a sufficiently large frequency \(\omega\) and amplitude \(V_0\) of the oscillating electric field yields the following equations of motion for the dark and bright state amplitudes

\[
\dot{D}_\pm(t) = -i[\omega_c - \omega_g + \sum_{j=1}^{N} \beta^j + iV_0 f_2 \cos(\omega t) + iV_0 f(N_1) \cos(\omega t)] + \frac{1}{N} \sum_{j=1}^{n} (\beta_j - \beta_j') |D_\pm(t)\rangle - \frac{i}{N} \sum_{j=1}^{N} \sum_{l=1}^{N-1} (\beta_j - \beta_j') B_{l\pm}(t)e^{2\pi ilj/N},
\]

\[
\dot{B}_{l\pm}(t) = -i[\omega_c - \omega_g + \sum_{j=1}^{N} \beta^j + iV_0 f_2 \cos(\omega t) + iV_0 f(N_1) \cos(\omega t)] + \frac{1}{N} \sum_{j=1}^{N} \sum_{l=1}^{N-1} (\beta_j - \beta_j') B_{l\pm}(t)e^{2\pi ilj/N},
\]

\[
\dot{B}_{l\pm}(t) = -i[\omega_c - \omega_g + \sum_{j=1}^{N} \beta^j + iV_0 f_2 \cos(\omega t) + iV_0 f(N_1) \cos(\omega t)] + \frac{1}{N} \sum_{j=1}^{N} \sum_{l=1}^{N-1} (\beta_j - \beta_j') B_{l\pm}(t)e^{2\pi ilj/N},
\]
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\[
\dot{B}_l(t) = -i \left[ \omega_c + \sum_{j=1}^{N} \beta^j + iV_0 f_2 \cos(\omega t) + iV_0 f(N_1) \cos(\omega t) \right] B_l(t)
\]

\[
\quad + \frac{i}{n} \sum_{j=1}^{N} \sum_{L=1}^{N-1} (\beta^j - \beta^j_2) B_{L\pm}(t) e^{2\pi i j (L-L)/N}
\]

\[
\quad - \frac{i}{N} \sum_{j=1}^{N} (\beta^j - \beta^j_2) D_{\pm}(t).
\]  

(5.111)

From section 5.2 it is known that the energy gap \( E_g = \hbar \omega_g \) which only appears in equation (5.110) suppresses any transition out of the subspace \( \mathcal{M} \). Therefore it is allowed to treat the bright state amplitudes perturbatively which yields for the lowest order the following solution for the dark state amplitudes:

\[
D_{\pm}(t) = \alpha_{\pm} e^{-i\phi(t) + i\omega_g t - i \sum_{j=1}^{N} (\beta^j - \beta^j_2) t/N}
\]

(5.112)

Therefore the time evolution of the storage state \( \alpha_+ |D_+, 1\rangle + \alpha_- |D_-, 1\rangle \) is given by

\[
\alpha_+ |D_+, 1\rangle + \alpha_- |D_-, 1\rangle \rightarrow e^{-i\phi(t) + i\omega_g t - i \sum_{j=1}^{N} (\beta^j - \beta^j_2) t/N} \alpha_+ |D_+, 1\rangle + \alpha_- |D_-, 1\rangle.
\]

(5.113)

This result shows that the time evolution just adds an additional common phase factor to the storage state. The phase relation as well as the population of the initially stored state does not change. By introducing an additional fast oscillating electric field and the additional collective interaction to create the energy gap \( E_g \) as done in the proposed scheme it is possible to suppress decoherence effects caused by individual atomic collisions.
Chapter 6

Summary and outlook

In this thesis a scalable procedure for quantum information processing with collective atomic excitations using electromagnetically induced transparency (EIT) [24] based on the method of M. Fleischhauer and M.D. Lukin [1–3] has been developed. The transfer of the quantum state of a photonic wave packet into symmetric collective states of the $N$ atom system is hereby based on stimulated Raman adiabatic passage (STIRAP) [4]. As previously shown in references [1, 3, 30] in order to achieve a high fidelity of the transfer process an adiabatically varying, explicitly time dependent control field is needed. The requirements for a successful and reversible storage of quantum information using collective atomic excitations have been analyzed in this thesis using quantitative corrections to the previously used adiabatic approximation and the corresponding conditions have been derived. Furthermore the influence of a finite two photon linewidth has been analyzed in view of the narrowing of the transparency window. The results show that for a successful quantum state transfer to the atomic excitations in optically thick media a rather large two photon detuning is tolerable. This is of particular practical importance for current and future experimental implementations where two photon Doppler shifts are not negligible [26–28]. In conjunction with these calculations numerical simulations have been carried out for realistic experimental parameters for the setup in the group of M. Drewsen at the university of Aarhus (Denmark). This setup is currently still under construction but once finished it will enable a direct comparison between the experimental and theoretical results.

One of the major problems of using collective many particle states to store the quantum information is the fact that these highly entangled states are very sensitive to decoherence processes. However it has been shown that by making use of the existence of equivalence classes such a quantum memory does not show an enhanced sensitivity to decoherence as compared to a quantum memory using single atoms. This has been discussed in detail for
different decoherence mechanisms, namely random spin flips, dephasing and atom losses. Even in the case of a non adiabatic read out process of the quantum memory there is no enhanced sensitivity for decoherence processes. Although this is a very promising result for the quantum information storage process quantum logical gates have much stronger requirements. For this purpose decoherence free subspaces or quantum error correction schemes are needed. Within this thesis it has been shown that by choosing appropriate storage states and introducing an additional nonlinear interaction of all atoms it is possible to construct a quasi decoherence free subspace. This quasi decoherence free subspace for the storage states in the atomic ensemble can be realized even if all atoms couple to individual and independent reservoirs. By combining the idea of quasi decoherence free subspaces with the open loop control, known from nuclear magnetic resonances (NMR), the collisional decoherence in atomic vapors can be suppressed. Therefore in addition to the nonlinear interaction of all atoms to create the energy gap a fast oscillating electric field has been introduced to suppress the dephasing between different magnetic sublevels.

The concept of quasi decoherence free subspaces which has been analyzed in the current thesis for the case of quantum computation based on collective atomic excitations is a very powerful tool since there are no restriction on the interaction between the system and its environment. The concept of quasi decoherence free subspaces can therefore be expected to be applied to other quantum computation systems in the future. A concrete physical realization to implement the nonlinear interaction to create the energy gap will be a further step in this direction. Furthermore it would be interesting to generalize the open loop control with respect to its possibilities in quantum memories with collective atomic excitations.

Another interesting approach in quantum information processing is the idea of quantum error corrections. Conventional quantum error correcting schemes have their basis in classical schemes and encode the qubit in larger codevectors consisting of many qubits. But different from the classical scheme the additional qubits cannot be read out directly to detect the error. Instead a measurement on additional ancilla qubits has to be performed which are entangled with the original codevector. Unfortunately those quantum error correcting schemes require a large number of qubits and are therefore extremely difficult to implement experimentally. Many atom systems however have a large number of degrees of freedom. Therefore quantum memories with collective atomic excitations have a large potential for quantum error correcting schemes and further investigations in this direction are very interesting.

A challenging point is to modify the existing storage scheme in a collective atomic ensemble to make it suitable for the storage of shorter light pulses. Due to the transparency
condition the scheme discussed in this thesis only allows pulses with an initial pulse spectrum which lies within the initial transparency window. This restricts the existing scheme to the storage of relatively long pulses, which are for typical experimental parameters in the $\mu$sec regime. Therefore it is of importance to explore alternative ways that allow for the storage of shorter photon wave packets. One possibility to overcome this restriction is to combine the existing storage scheme with photon echo techniques [136] where the pulse length is only limited by the inhomogeneous broadening of the used atomic transition. First calculations in this direction have already been carried out in collaboration with M. Perdian from the university of Torun (Poland). Further investigations have to be done to answer the question whether the pulse length restriction in a radiatively broadened system can be lifted and wave packets with pulse times shorter than the inverse lifetime of the excited state can be stored.

In conclusion the results of this thesis have shown that quantum memories with collective excitations have a large potential in quantum information systems and are competitive to other approaches based for example on Josephson junctions or single atom approaches. The advantages of the many particle excitations have been discussed with respect to decoherence. These ideas can be generalized and extended. Which of the different approaches for quantum computation will eventually succeed is not clear as of today and will be decided by scientific and technological competition.
Appendix A

MATLAB programs

Program main.m:

function [fmain] = main()
%Main program for calculation of the Maxwell-Bloch-equations
z = [-40:1:130]; %spatial variable
t = [0:5:150]; %time variable
m = 0; %symmetry parameter
%Calculation of the solution using pdepe.m:
sol = pdepe(m, 'pdeatomfield', 'pdeatomfieldic', 'pdeatomfieldbc', z, t);
%Storage of data
save solutiondata
main = sol

Program pdeatomfield.m:

function [c, f, s] = pdeatomfield1(z, t, u, dudz)
%Function used as parameter PDEFUN by pdepe.m
%represents the partial differential equations of the system
%for details see description of pdepe.m
c = [1; 1; 1; 1; 1; 1; 1]; %used to create a diagonal matrix
f = [0; 0; 0; 0; 0; 0; 0]; %flux term
%parameters
g1 = 0.1; %decay rate \gamma_a
p = 1000;
g0 = 10.^[p].*g1; %decay rate \gamma_{bc}
c0 = 1; %initial group velocity
A. MATLAB programs

%Initialization of the partial differential equations
s1=-g1.*u(1)+2.*imag(conj(u(7)).*u(4))+2.*field(t).*imag(u(5));
s2=g1./2.*u(1)-2.*imag(conj(u(7)).*u(4));
s3=g1./2.*u(1)-2.*field(t).*imag(u(5));
s4=-1./2.\(g1+g0\).*u(4)+i.*u(7).*(u(2)-u(1))+i.*field(t).*u(6);
s5=-1./2.\(g1+g0\).*u(5)+i.*field(t).*(u(3)-u(1))+i.*u(7).*conj(u(6));
s6=-g0.*u(6)-i.*u(7).*conj(u(5))+i.*field(t).*u(4);
s7=i.*u(4)-c0.*dudz(7);
s=[s1;s2;s3;s4;s5;s6;s7];

Program pdeatomfieldic.m:

function[u0]=pdeatomfieldic1(z)
%Function to implement the initial conditions
u0=[0;1;0;0;0;0;expo(z)];

Program pdeatomfieldbc.m:

function[pl,ql,pr,qr]=pdeatomfieldbc1(zl,ul,zr,ur,t)
%Function to implement the boundary conditions
pl=[ul(1);ul(2)-1;ul(3);ul(4);ul(5);ul(6);ul(7)];
ql=[0;0;0;0;0;0;0];
pr=[ur(1);ur(2)-1;ur(3);ur(4);ur(5);ur(6);ur(7)];
qr=[0;0;0;0;0;0;0];

Program expo.m:

function[fexpo]=expo(z)
%Function to calculate the initial light pulse
z0=0;
dz2=100;
amp=10^-3;
fexpo=amp.*exp(-[z-z0].^2./dz2);

Program field.m:

function[ffield]=field(t)
%Function to calculate cot(theta(t))
%feld01
h1=tanh(0.1.*(t-15));
h2=tanh(0.1.*(t-125));
ffield=100.*(1-0.5.*h1+0.5.*h2);
Appendix B

Calculations to chapter 4

B.1 Calculations to subsection 4.3.2

Calculation of equation (4.36):

\[
|\widehat{D}, n\rangle_N = \sum_{k=0}^{n} \left[ \frac{n!}{(n-k)!k!} \right]^{1/2} p^k (1-p)^{n-k} \left( \cos \theta \hat{a}^\dagger - \sin \theta \frac{1}{\sqrt{N}} \sum_{j=1}^{N} \hat{\sigma}^\dagger b_{c,e} e^{i\varphi_j} \right)^k \frac{1}{\sqrt{(n-k)!}} (\hat{\Psi}^\dagger)^{n-k} |b, 0\rangle_N
\]

\[
= \sum_{k=0}^{n} \left[ \frac{n!}{(n-k)!k!} \right]^{1/2} p^k (1-p)^{n-k} \cdot \sum_{i=0}^{k} \left[ \frac{k!}{(k-l)!!} \right]^{1/2} (-\sin \theta)^l (\cos \theta)^{k-l} \frac{N(N-1)\ldots(N-l+1)}{N^l} \frac{1}{((n-k)-s)!} \left( -\sin \theta \right)^s (\cos \theta)^{(n-k)-s} \frac{1}{(N-l)!(n-k-s)!} \frac{1}{N^s} \right]^1 \sum_{l \geq l_1} \sum_{l_2} \sum_{l_s} \frac{l!}{N(N-1)\ldots(N-l+1) (N-l)\ldots(N-l-s+1)} \cdot \left[ \frac{(n-k-s) + (k-l)}{(k-l)!(n-k-s)!} \right]^{1/2} \cdot |b_1, b_2, \ldots, c_{i_1}, \ldots, c_{i_t}, \ldots, c_{j_1}, \ldots, c_{j_s}, \ldots, b_N, (n-k-s) + (k-l))_N \cdot \exp\{-i(\varphi_{i_1} + \ldots + \varphi_{i_t})\}.
\]
B.1. Calculations to subsection 4.3.2

Explicit form of the density matrix $\tilde{\rho}_{Fock}$ in equation (4.38):

$$
\tilde{\rho}_{Fock} = \sum_{k_1=1}^{n} \sum_{k_2=1}^{n} \left[ \frac{n!}{(n-k_1)!k_1!(n-k_2)!k_2!} \right]^{1/2} p^{k_1+k_2}(1-p)^{2n-k_1-k_2} \\
\cdot \sum_{l_1=0}^{k_1} \sum_{l_2=0}^{k_2} \left[ \frac{k_1!}{(k_1-l_1)!l_1!(k_2-l_2)!l_2!} \right]^{1/2} (-\sin \theta)^{l_1+l_2}(\cos \theta)^{k_1-l_1+k_2-l_2} \\
\cdot \left[ \frac{N(N-1)\ldots(N-l_1+1)}{N_{l_1}} \frac{N(N-1)\ldots(N-l_2+1)}{N_{l_2}} \right]^{1/2} \\
\cdot \sum_{s_1=0}^{n-k_1} \sum_{s_2=0}^{n-k_2} \left[ \frac{(n-k_1)!}{((n-k_1)-s_1)!s_1!((n-k_2)-s_2)!s_2!} \right]^{1/2} (-\sin \theta)^{s_1+s_2}(\cos \theta)^{2n-k_1-s_1-k_2-s_2} \\
\cdot \left[ \frac{(N-l_1)(N-l_1-1)\ldots(N-l_1-s_1+1)}{N^{s_1}} \frac{(N-l_2)(N-l_2-1)\ldots(N-l_2-s_2+1)}{N^{s_2}} \right]^{1/2} \\
\cdot \left[ \frac{((n-k_1-s_1)+(k_1-l_1))((n-k_2-s_2)+(k_2-l_2))}{(n-k_1-s_1)(k_1-l_1)} \frac{(n-k_2-s_2)(k_2-l_2)}{(n-k_2-s_2)(k_2-l_2)} \right]^{1/2} \\
\sum_{l_1 \neq l_1'} \sum_{l_2 \neq l_2'} \sum_{s_1} \sum_{s_2} \left[ \frac{N(N-1)\ldots(N-l_1+1)}{N_{l_1}} \frac{N(N-1)\ldots(N-l_1-1)}{N_{l_1}} \ldots \frac{N(N-1)\ldots(N-l_1-s_1+1)}{N_{l_1}} \right]^{1/2} \\
\cdot \frac{l_1!}{l_2!} \frac{s_1!}{s_2!} \\
\cdot \exp\{-i(\varphi_{l_1} + \ldots + \varphi_{l_2}) + i(\varphi_{l_1} + \ldots + \varphi_{l_2})\} \\
\cdot |b_1, c_1, \ldots, c_{l_1}, \ldots, c_{l_2}, \ldots, c_{l_1'}, \ldots, c_{l_2'}, \ldots, b_{l_1}, c_1, \ldots, c_{l_1}, \ldots, c_{l_2}, \ldots, c_{l_1'}, \ldots, c_{l_2'}, \ldots, b_{l_2}| \\
\otimes |(n-k_1-s_1)+(k_1-l_1)\rangle \langle (n-k_2-s_2)+(k_2-l_2)|.
Calculation of the fidelity (4.40) of the storage device for the stored Fock state after dephasing:

\[ f_{\text{Fock}} = \left[ \sum_{k_2 \leq k_1} \frac{n}{k_2} \sum_{l=0}^{n-k_2-k_1} + \sum_{k=0}^{n} \frac{k}{k_1} \sum_{l=0}^{k_1} + \sum_{k_1} \sum_{k_2} \sum_{l=0}^{k_2} \right] \sum_{s_1} \sum_{s_2} p^{k_1+k_2} (1-p)^{2n-k_1-k_2} \]

\[ \frac{n!}{(n-k_1)!k_1!(n-k_2)!k_2!} \left[ \frac{n!}{(n-k_1)!k_1!(n-k_2)!k_2!} \right]^{1/2} \left[ \frac{k_1!}{(k_1-l)!} \frac{k_2!}{(k_2-l)!!} \right]^{1/2} (-\sin \theta)^{2l}(\cos \theta)^{k_1+k_2-2l} \]

\[ \frac{N!}{(N-l)!N^l} \left[ \frac{n!}{n} \right]^{1/2} \left[ \frac{N!}{(n-k_1-s_1)!s_1!(n-k_2-s_2)!s_2!} \right]^{1/2} (-\sin \theta)^{s_1+s_2}(\cos \theta)^{2n-k_1-s_1-k_2-s_2} \]

\[ \frac{1}{(N-l)!N^{s_2}} \left[ \frac{1}{(N-l-s_1)!N^{s_2}} \right]^{1/2} \left[ \frac{1}{(k_1-l)!}(n-k_1-s_1)(k_2-l)!(n-k_2-s_2)! \right]^{1/2} \]

\[ \frac{1}{(N-l-s_1)!(s_1+l)!(N-l-s_2)!(s_2+l)!} \]

\[ \frac{1}{(N-l-s_1)!(s_1+l)!(N-l-s_2)!(s_2+l)!(N-l-s_2)!(N-l-s_2)!} \]

\[ \frac{s_1!(N-l-s_1)!s_2!(N-l-s_2)!(s_1+l)!(N-l-s_1)!(s_2+l)!(N-l-s_2)!}{N!} \]

\[ \frac{N!}{N!} \frac{N!}{N!} \frac{N!}{N!} \frac{N!}{N!} \]

\[ \left[ \sum_{k_2 \leq k_1} \frac{n}{k_2} \sum_{l=0}^{n-k_2-k_1} + \sum_{k=0}^{n} \frac{k}{k_1} \sum_{l=0}^{k_1} + \sum_{k_1} \sum_{k_2} \sum_{l=0}^{k_2} \right] \sum_{s_1} \sum_{s_2} p^{k_1+k_2} (1-p)^{2n-k_1-k_2} \]

\[ \frac{n!}{(n-k_1)!l!} \frac{n!}{(n-k_2)!l!} \]

\[ (-\sin \theta)^{2l}(\cos \theta)^{k_1+k_2-2l} \]

\[ \sum_{k_1} \sum_{k_2} \sum_{l=0}^{n-k_1-k_2} \frac{1}{(n-k_1-s_1)!s_1!} \frac{1}{(n-k_2-s_2)!s_2!} \]

\[ (-\sin \theta)^{s_1+s_2}(\cos \theta)^{2n-k_1-s_1-k_2-s_2} \]

\[ (-\sin \theta)^{s_1+s_2}(\cos \theta)^{2n-2l-s_1-s_2} \]

\[ \sum_{s_1} \sum_{s_2} \frac{s_1!(N-l-s_1)!s_2!(N-l-s_2)!(s_1+l)!(N-l-s_1)!(s_2+l)!(N-l-s_2)!}{N!} \]

\[ \frac{n!}{(N-l)!!} \]

\[ \frac{n!}{(N-l)!!} \frac{n!}{(N-l)!!} \]

\[ \frac{n!}{(N-l)!!} \frac{n!}{(N-l)!!} \]

\[ = (\cos \theta^{l+2}+\sin \theta^{l+2})^{n-k_1-1} \]

\[ = (\cos \theta^{l+2}+\sin \theta^{l+2})^{n-k_2-1} \]

\[ \sum_{k_1=0}^{n-k_1} \sum_{k_2=0}^{n-k_2} \sum_{l=0}^{n-k_1-k_2} \left[ \frac{(n-k_1)!}{(n-k_1-s_1)!s_1!} \frac{(n-k_2)!}{(n-k_2-s_2)!s_2!} \right] \]

\[ \frac{\sin \theta^{s_1}(\cos \theta)^{n-k_1-s_1}}{\sin \theta^{s_2}(\cos \theta)^{n-k_2-s_2}} \]

\[ \frac{(n-k_1)!}{(n-k_1-s_1)!s_1!} \frac{(n-k_2)!}{(n-k_2-s_2)!s_2!} \frac{(N-l)!!}{N!!} \]

\[ = (\cos \theta^{l+2}+\sin \theta^{l+2})^{n-k_1-1} \]

\[ = (\cos \theta^{l+2}+\sin \theta^{l+2})^{n-k_2-1} \]

\[ \sum_{k_1=0}^{n-k_1} \sum_{k_2=0}^{n-k_2} \sum_{l=0}^{n-k_1-k_2} \left[ \frac{(n-k_1)!}{(n-k_1-s_1)!s_1!} \frac{(n-k_2)!}{(n-k_2-s_2)!s_2!} \right] \]

\[ \frac{(n-k_1)!}{(n-k_1-s_1)!s_1!} \frac{(n-k_2)!}{(n-k_2-s_2)!s_2!} \frac{(N-l)!!}{N!!} \]

\[ = (\cos \theta^{l+2}+\sin \theta^{l+2})^{n-k_1-1} \]

\[ = (\cos \theta^{l+2}+\sin \theta^{l+2})^{n-k_2-1} \]
Calculation of the fidelity (4.46) of the storage device for the stored coherent state $|\alpha\rangle$ after dephasing:

$$f_{\text{coh}} = \lim_{n \to \infty} e^{-2|\alpha|^2} \sum_{m_1}^n \frac{|\alpha|^2(1 - p \sin^2 \theta)}{m_1!} \sum_{m_2}^n \frac{|\alpha|^2(1 - p \sin^2 \theta)}{m_2!}$$

$$= \exp\{-2|\alpha|^2\} \exp\{|\alpha|^2(1 - p \sin^2 \theta)\} \exp\{|\alpha|^2(1 - p \sin^2 \theta)\}$$

$$= \exp\{-2|\alpha|^2 p \sin^2 \theta\}.$$ 

Calculation of the fidelity (4.49) of the storage device for the stored coherent state $\frac{N}{\sqrt{2}}(|i\alpha\rangle + |-i\alpha\rangle)$ after dephasing:

$$f_{\text{Scs}} = \lim_{n \to \infty} N^4 e^{-2|\alpha|^2} \sum_{m_1}^n \frac{(1^{m_1} + (-1)^{m_1}) |\alpha|^2(1 - p \sin^2 \theta)}{m_1!} \sum_{m_2}^n \frac{(1^{m_2} + (-1)^{m_2}) |\alpha|^2(1 - p \sin^2 \theta)}{m_2!}$$

$$= N^4 \exp\{-2|\alpha|^2\} [\exp\{|\alpha|^2\} \exp\{-|\alpha|^2 p \sin^2 \theta\} + \exp\{-|\alpha|^2\} \exp\{|\alpha|^2 p \sin^2 \theta\}]$$

$$= \exp\{-2|\alpha|^2 p \sin^2 \theta\} \left[\frac{1 + \exp\{-2|\alpha|^2(1 - p \sin^2 \theta)\}}{1 + \exp\{-2|\alpha|^2\}}\right]^2.$$
B.2 Calculations to subsection 4.3.3

Calculation of $\tilde{\rho}_{N-1}$ for a stored Fock state in equation (4.52) after the $N$th atom has been lost:

$$\tilde{\rho}_{N-1} = \sum_{k=0}^{n} \sum_{l=0}^{n} \left[ \frac{n!}{k!(n-k)!} \frac{n!}{l!(n-l)!} \right]^{1/2} (-\sin \theta)^{k+l} (\cos \theta)^{2n-k-l}$$

$$\cdot \left[ \frac{N(N-1) \ldots (N-k+1)}{N^k} \frac{N(N-1) \ldots (N-l+1)}{N^l} \right]^{1/2}$$

$$\cdot \sum_{\{i_k \} \{j_l \}} \left[ \frac{k!}{N(N-1) \ldots (N-k+1)} \frac{l!}{N(N-1) \ldots (N-l+1)} \right]^{1/2}$$

$$\cdot \left| b_1, \ldots , c_{i_1}, \ldots , c_{i_k}, \ldots , b_{N-1}, n-k \rangle \langle b_1, \ldots , c_{j_1}, \ldots , c_{j_l}, \ldots , b_{N-1}, n-l \right|$$

$$+ \sum_{k=1}^{n} \sum_{l=1}^{n} \left[ \frac{n!}{k!(n-k)!} \frac{n!}{l!(n-l)!} \right]^{1/2} (-\sin \theta)^{k+l} (\cos \theta)^{2n-k-l}$$

$$\cdot \left[ \frac{N(N-1) \ldots (N-k+1)}{N^k} \frac{N(N-1) \ldots (N-l+1)}{N^l} \right]^{1/2}$$

$$\cdot \sum_{\{i_k \} \{j_l \}} \sum_{\{i_{k-1} \} \{j_{l-1} \}} \left[ \frac{k!}{N(N-1) \ldots (N-k+1)} \frac{l!}{N(N-1) \ldots (N-l+1)} \right]^{1/2}$$

$$\cdot \left| b_1, \ldots , c_{i_1}, \ldots , c_{i_{k-1}}, \ldots , b_{N-1}, n-k \rangle \langle b_1, \ldots , c_{j_1}, \ldots , c_{j_{l-1}}, \ldots , b_{N-1}, n-l \right|$$

$$\equiv S_1 + S_2,$$

where $S_1$ and $S_2$ are given by:

$$S_1 = \sum_{k=0}^{n} \sum_{l=0}^{n} \left[ \frac{N-1}{N} \right]^{(l+k)/2} \left[ \frac{n!}{k!(n-k)!} \frac{n!}{l!(n-l)!} \right]^{1/2} (-\sin \theta)^{k+l} (\cos \theta)^{2n-k-l}$$

$$\cdot \left[ \frac{(N-1)(N-2) \ldots ((N-1)-(k+1))}{(N-1)^k} \frac{(N-1)(N-2) \ldots ((N-1)-(l+1))}{(N-1)^l} \right]^{1/2}$$

$$\cdot \left| c^k, n-k \rangle \langle e^l_{N-1}, c^l_{N-1}, n-l \right|,$$

and

$$S_2 = \sum_{k=0}^{n-1} \sum_{l=0}^{n-1} \frac{n}{N} \left[ \frac{N-1}{N} \right]^{(l+k)/2} \sin^2 \theta \left[ \frac{(n-1)!}{k!(n-1-k)!} \frac{(n-1)!}{l!(n-1-l)!} \right]^{1/2} (-\sin \theta)^{k+l}$$

$$\cdot (\cos \theta)^{2(n-1)-k-l} \left[ \frac{(N-1)(N-2) \ldots ((N-1)-(k+1))}{(N-1)^k} \right]^{1/2}$$

$$\cdot \left[ \frac{(N-1)(N-2) \ldots ((N-1)-(l+1))}{(N-1)^l} \right]^{1/2} \left| c^k, (n-1)-k \rangle \langle e^l_{N-1}, (n-1)-l \right|.$$
The two summands $S_1$ and $S_2$ correspond to the case where the lost atom was in the excited state $|c\rangle$ and in the ground state $|b\rangle$ respectively. With the assumption that the number of stored photons $n$ is always much smaller than the number of atoms, i.e. $k, l \leq n \ll N$, it is possible to expand the coefficients in $S_1$ and $S_2$ in the following way

$$\frac{n}{N} \left[ \frac{N-1}{N} \right]^{(k+l)/2} = \frac{n}{N} \left[ \left( 1 - \frac{1}{N} \right)^{1/2} \right]^{k+l} = \frac{n}{N} + O \left( \frac{1}{N^2} \right),$$

$$\left[ \frac{N-1}{N} \right]^{(k-l)/2} = \left[ \left( 1 - \frac{1}{N} \right)^{1/2} \right]^{k+l} = 1 - \frac{k + l}{2N} + O \left( \frac{1}{N^2} \right).$$

This expansions leads to equation (4.52)

$$\tilde{\rho}_{N-1} = |D, n\rangle_{N-1} \langle D, n|$$

$$- \frac{1}{N} \sum_{k=0}^{n} \sum_{l=0}^{n} \frac{1}{4} [k(k+1) + l(l+1)] \left[ \frac{n!}{k!(n-k)! l!(n-l)!} \right]^{1/2}$$

$$\cdot (-\sin \theta)^{k+l} (\cos \theta)^{2n-k-l} |c^k, n-k\rangle_{N-1} \langle c^l, n-l|$$

$$+ \frac{n}{N} \sin^2 \theta |D, n-1\rangle_{N-1} \langle D, n-1|. $$

Calculation of the fidelity for the case that the $j$th atom is lost for a stored Fock state, see equation (4.53):

$$f_{Fock}^j = 1 - \frac{1}{2N} \sum_{k=0}^{n} \sum_{l=0}^{n} (k + l) \frac{n!}{k!(n-k)! l!(n-l)!} \frac{n!}{(\sin^2 \theta)^{k+l}(\cos^2 \theta)^{2n-l-k}}$$

$$= 1 - \frac{1}{2N} \sum_{k=0}^{n} \sum_{l=0}^{n} \frac{k}{k!(n-k)! l!(n-l)!} \frac{n!}{(\sin^2 \theta)^{k+l}(\cos^2 \theta)^{2n-l-k}}$$

$$- \frac{1}{2N} \sum_{k=0}^{n} \sum_{l=0}^{n} \frac{l}{k!(n-k)! l!(n-l)!} \frac{n!}{(\sin^2 \theta)^{k+l}(\cos^2 \theta)^{2n-l-k}}.$$

Exchanging the summation indices in the last summand $f_{Fock}^j$ is given by

$$f_{Fock}^j = 1 - \frac{1}{N} \sum_{k=0}^{n} \sum_{l=0}^{n} k \frac{n!}{k!(n-k)! l!(n-l)!} \frac{n!}{(\sin^2 \theta)^{k+l}(\cos^2 \theta)^{2n-l-k}}$$

$$= 1 - \frac{1}{N} \sum_{k=0}^{n} \frac{n!}{k!(n-k)!} (\sin^2 \theta)^{k} (\cos^2 \theta)^{n-k} \sum_{l=0}^{n} \frac{n!}{l!(n-l)!} (\sin^2 \theta)^{l} (\cos^2 \theta)^{n-l}$$

$$= 1 - \frac{1}{N} \sum_{k=1}^{n} \frac{n!}{k!(n-k)!} (\sin^2 \theta)^{k} (\cos^2 \theta)^{n-k}. $$

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Changing the summation index via $s = k - 1$ yields:

$$f_{\text{Fock}}^j = 1 - \frac{n}{N} \sin^2 \theta \sum_{j=0}^{n-1} \frac{(n - 1)!}{s!(n - 1 - s)!} (\sin^2 \theta)^s (\cos^2 \theta)^{n-1-s}$$

$$= 1 - \frac{n}{N} \sin^2 \theta.$$
Appendix C

Calculations to chapter 5

Calculation of $\hat{H}_{\text{int}}^l(t)$ in equation (5.46):

To calculate $\hat{H}_{\text{int}}^l(t)$ which is defined in equation (5.45)

$$\hat{H}_{\text{int}}^l(t) \equiv \exp \left\{ -\frac{i}{\hbar} (\hat{H}'_S + \hat{H}_B) t \right\} \hat{H}_{\text{int}} \exp \left\{ -\frac{i}{\hbar} (\hat{H}'_S + \hat{H}_B) t \right\}.$$

it is convenient first to calculate the following expressions:

$$\exp \left\{ \frac{i}{\hbar} \hat{H}'_S t \right\} \hat{\Phi}_l^\pm \exp \left\{ -\frac{i}{\hbar} \hat{H}'_S t \right\} \text{ and } \exp \left\{ \frac{i}{\hbar} \hat{H}'_S t \right\} \hat{\Psi}^\pm \exp \left\{ -\frac{i}{\hbar} \hat{H}'_S t \right\}.$$

Assuming that the number density of photons is much smaller than the density of the atoms the dark and bright state operators fulfill bosonic commutation relations (see section 2.3.1). Therefore the only non vanishing commutation relations are given by:

$$[\hat{\Psi}^\pm, \hat{\Psi}^\dagger_{\mp}] \approx 1, \ [\hat{\Phi}_{l^\pm}, \hat{\Phi}_{m^\pm}^\dagger] \approx \delta_{lm} \text{ and } [\hat{\Phi}_{0^\pm}, \hat{\Phi}_{0^\mp}^\dagger] = 1.$$

To proceed further it is useful to express the dark and bright state polariton operators $\hat{\Psi}^\pm$ and $\hat{\Phi}_{l^\pm}$ in the orthonormal basis of dark and bright states, which is given by

$$|D_{\pm}, n\rangle_N = \frac{1}{\sqrt{n!}} \left( \hat{\Psi}^\dagger_{\mp} \right)^n |b_1, \ldots, b_N, 0\rangle, \ n \in \{1, 2, \ldots\}, \ n \ll N,$$

$$|B_{l\pm}, n\rangle_N = \frac{1}{\sqrt{n!}} \left( \hat{\Phi}_{l^\pm}^\dagger \right)^n |b_1, \ldots, b_N, 0\rangle, \ n \in \{1, 2, \ldots\}, \ n \ll N, \ l \in \{1, 2, \ldots N - 1\},$$

$$|B_{0\pm}, n\rangle_N = \frac{1}{\sqrt{n!}} \left( \hat{a}^\dagger_{\pm} \right)^n |b_1, \ldots, b_N, 0\rangle, \ n \in \{1, 2, \ldots\}, \ n \ll N \text{ and }$$

$$|D_{\pm}, 0\rangle_N = |B_{l\pm}, 0\rangle_N = |B_{0\pm}, 0\rangle_N = |b_1, \ldots, b_N, 0\rangle.$$
Together with the relations
\[ \hat{\Psi}_\pm |D_\pm, n\rangle_N = \sqrt{n + 1} |D_\pm, n + 1\rangle_N, \]
\[ \hat{\Psi}_\pm |D_\pm, n\rangle_N \approx \sqrt{n} |D_\pm, n - 1\rangle_N, \]
\[ \hat{\Phi}_\pm |B_{\pm}, n\rangle_N = \sqrt{n + 1} |B_{\pm}, n + 1\rangle_N \]
\[ \hat{\Phi}_\pm |B_{\pm}, n\rangle_N \approx \sqrt{n} |B_{\pm}, n - 1\rangle_N \]
this yields:
\[ \hat{\Psi}_\pm = \sum_{n=0} \sqrt{n + 1}|D_\pm, n\rangle_N \langle D_\pm, n + 1| \]
\[ \hat{\Phi}_\pm = \sum_{n=0} \sqrt{n + 1}|B_{\pm}, n\rangle_N \langle B_{\pm}, n + 1|. \]

Using the Baker Hausdorff formula and rescaling the energies without loss of generality, i.e. \( E = 0 \), \( \exp \left\{ \frac{i}{\hbar} \hat{H}'_S t \right\} \hat{\Phi}_\pm \exp \left\{ -\frac{i}{\hbar} \hat{H}'_S t \right\} \) and \( \exp \left\{ \frac{i}{\hbar} \hat{H}'_S t \right\} \hat{\Psi} \exp \left\{ -\frac{i}{\hbar} \hat{H}'_S t \right\} \) can be expressed by:
\[ \exp \left\{ \frac{i}{\hbar} \hat{H}'_S t \right\} \hat{\Phi}_\pm \exp \left\{ -\frac{i}{\hbar} \hat{H}'_S t \right\} = \sum_{n=2} \sqrt{n + 1}|D_\pm, n\rangle_N \langle D_\pm, n + 1| + e^{\omega_p t}|D_\pm, 0\rangle_N \langle D_\pm, 1| + e^{-\omega_p t}\sqrt{2}|D_\pm, 1\rangle_N \langle D_\pm, 2| \]
\[ \equiv \hat{M}_\pm(t), \]
\[ \exp \left\{ \frac{i}{\hbar} \hat{H}'_S t \right\} \hat{\Psi} \exp \left\{ -\frac{i}{\hbar} \hat{H}'_S t \right\} = \hat{\Phi}_\pm. \]

Therefore \( \hat{H}'_{int}(t) \) is given by:
\[ \hat{H}'_{int}(t) = \hat{H}'_A(t) + \hat{H}'_B(t) + \hat{H}'_C(t), \]
where \( \hat{H}'_A(t) \), \( \hat{H}'_B(t) \) and \( \hat{H}'_C(t) \) are given by:
\[ \hat{H}'_A(t) = \frac{\hbar}{N} \sum_{l=1}^{N-1} \sum_{m=1}^{N-1} \sum_{j=1}^{N} \left[ \hat{\Phi}_{\pm} \hat{\Phi}_m \hat{\Gamma}_A(t) \exp \left\{ 2\pi i \frac{j(l - m)}{N} \right\} + H.c. \right] \]
\[ - \frac{\hbar}{N} \sum_{l=1}^{N} \sum_{j=1}^{N} \left[ \hat{\Phi}_{\pm} \hat{\tilde{M}}_-(t) \hat{\Gamma}_A(t) \exp \left\{ 2\pi i \frac{j l}{N} \right\} + H.c. \right] \]
\[ - \frac{\hbar}{N} \sum_{l=1}^{N} \sum_{j=1}^{N} \left[ \hat{\Phi}_{\pm} \hat{\tilde{M}}_+(t) \hat{\Gamma}_A(t) \exp \left\{ 2\pi i \frac{j l}{N} \right\} + H.c. \right] \]
\[ + \frac{\hbar}{N} \sum_{j=1}^{N} \left[ \hat{\tilde{M}}_{\pm}(t) \hat{\tilde{M}}_-(t) \hat{\Gamma}_A(t) + H.c. \right] , \]
The operator $\tilde{\Gamma}_X(t)$, $X \in \{A, B, C\}$ is hereby defined as

$$\tilde{\Gamma}_X(t) = \exp \left\{ i \hbar \hat{H}_B t \right\} \Gamma_X(t) \exp \left\{ -i \hbar \hat{H}_B t \right\}.$$ 

The exact form of $\operatorname{tr}_B \left\{ [\hat{H}_{\text{int}}(t), [\hat{H}_{\text{int}}(t'), \rho_{\text{tot}}^{(t')}] ] \right\}$ in equation (5.64): 

Using that the different reservoirs are uncorrelated the exact form of $\operatorname{tr}_B \left\{ [\hat{H}_{\text{int}}(t), [\hat{H}_{\text{int}}(t'), \rho_{\text{tot}}^{(t')}] ] \right\}$ in the integro differential equation (5.64) in Born approximation is given by:

$$\operatorname{tr}_B \left\{ [\hat{H}_{\text{int}}(t), [\hat{H}_{\text{int}}(t'), \rho_{\text{tot}}^{(t')}] ] \right\} = \sum_{a=1}^{4} \sum_{b=1}^{8} S_{ab} + \sum_{a=5}^{8} \sum_{b=1}^{4} S_{ab},$$

where the summands $S_{ab}$ are defined by:

$$S_{11} = \frac{\hbar^2}{N^2} \sum_{L,l,M,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(L - M) + j(l - m)}{N} \right\} \cdot \left[ \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{M} - \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{m} - \rho(t) \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{m} - \rho(t) \hat{\Phi}_{M} \right] A + \text{H.c.}$$

$$+ \frac{\hbar^2}{N^2} \sum_{L,l,M,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(L - M) + j(l - m)}{N} \right\} \cdot \left[ \rho'(t) \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{m} - \rho'(t) \hat{\Phi}_{l}^{\dagger} \hat{\Phi}_{M} - \rho(t) \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{M} \right] A' + \text{H.c.},$$

$$S_{12} = \frac{\hbar^2}{N^2} \sum_{L,l,M,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(L - M) + j(l - m)}{N} \right\} \cdot \left[ \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{M} - \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{m} - \rho(t) \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{m} - \rho(t) \hat{\Phi}_{M} \right] A + \text{H.c.}$$

$$+ \frac{\hbar^2}{N^2} \sum_{L,l,M,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(L - M) + j(l - m)}{N} \right\} \cdot \left[ \rho'(t) \hat{\Phi}_{m} - \rho'(t) \hat{\Phi}_{l}^{\dagger} \hat{\Phi}_{M} - \rho(t) \hat{\Phi}_{L}^{\dagger} \hat{\Phi}_{m} \right] A' + \text{H.c.},$$
\[ S_{13} = - \frac{\hbar^2}{N^2} \sum_{L,l,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j (L - M) + j l}{N} \right\} \]
\[
\cdot \left[ \hat{\Phi}_{L+}^j \hat{\Phi}_{M-} - \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} + \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} (t') \rho^j (t') - \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} (t') \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} \right] A + \text{H.c.} \]
\[
- \frac{\hbar^2}{N^2} \sum_{L,l,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j (L - M) + j l}{N} \right\} \]
\[
\cdot \left[ \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} (t') \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} - \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} \right] A' + \text{H.c.,} \]
\[ S_{14} = - \frac{\hbar^2}{N^2} \sum_{L,l,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j (L - M) - j l}{N} \right\} \]
\[
\cdot \left[ \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} (t') \hat{\Phi}_{L+}^j \rho^j (t) - \hat{\tilde{M}}_{-} (t') \hat{\Phi}_{L+}^j \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} \right] A + \text{H.c.} \]
\[
- \frac{\hbar^2}{N^2} \sum_{L,l,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j (L - M) - j l}{N} \right\} \]
\[
\cdot \left[ \rho^j (t) \hat{\tilde{M}}_{-} (t') \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} - \hat{\tilde{M}}_{-} \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} \right] A' + \text{H.c.,} \]
\[ S_{15} = - \frac{\hbar^2}{N^2} \sum_{L,l,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j (L - M) - j l}{N} \right\} \]
\[
\cdot \left[ \hat{\Phi}_{L+}^j \hat{\Phi}_{M-} \hat{\tilde{M}}_{-} (t') \hat{\Phi}_{L+}^j \rho^j (t) - \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} (t') \hat{\Phi}_{L+}^j \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} \right] A + \text{H.c.} \]
\[
- \frac{\hbar^2}{N^2} \sum_{L,l,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j (L - M) - j l}{N} \right\} \]
\[
\cdot \left[ \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} (t') \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} - \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{-} \right] A' + \text{H.c.,} \]
\[ S_{16} = - \frac{\hbar^2}{N^2} \sum_{L,l,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j (L - M) + j l}{N} \right\} \]
\[
\cdot \left[ \hat{\Phi}_{L+}^j \hat{\Phi}_{M-} \hat{\tilde{M}}_{+} (t') \rho^j (t) - \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{+} (t') \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{+} \right] A + \text{H.c.} \]
\[
- \frac{\hbar^2}{N^2} \sum_{L,l,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j (L - M) + j l}{N} \right\} \]
\[
\cdot \left[ \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{+} (t') \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{+} - \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{+} \rho^j (t) \hat{\Phi}_{L+}^j \hat{\tilde{M}}_{+} \right] A' + \text{H.c.,} \]
\[ S_{17} = \frac{\hbar^2}{N^2} \sum_{L,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(L - M)}{N} \right\} \]  
\[ \cdot \left[ \hat{\Phi}_L^+ \hat{\Phi}_M \hat{M}_L^+(t') \hat{M}_M^-(t') \rho^I(t) - \hat{M}_L^+(t') \hat{M}_M^-(t') \rho^I(t) \hat{\Phi}_L^+ \hat{\Phi}_M \right] A + \text{H.c.} \]  
\[ + \frac{\hbar^2}{N^2} \sum_{L,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(L - M)}{N} \right\} \]  
\[ \cdot \left[ \rho^I(t) \hat{M}_L^+(t') \hat{M}_M^-(t') \hat{\Phi}_L^+ \hat{\Phi}_M - \hat{\Phi}_L^+ \hat{\Phi}_M \rho^I(t) \hat{M}_L^+(t') \hat{M}_M^-(t') \right] A' + \text{H.c.}, \]  
\[ S_{18} = \frac{\hbar^2}{N^2} \sum_{L,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(L - M)}{N} \right\} \]  
\[ \cdot \left[ \hat{\Phi}_L^+ \hat{\Phi}_M \hat{M}_L^+(t') \hat{M}_M^-(t') \rho^I(t) - \hat{M}_L^+(t') \hat{M}_M^-(t') \rho^I(t) \hat{\Phi}_L^+ \hat{\Phi}_M \right] A + \text{H.c.} \]  
\[ + \frac{\hbar^2}{N^2} \sum_{L,M=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(L - M)}{N} \right\} \]  
\[ \cdot \left[ \rho^I(t) \hat{M}_L^+(t') \hat{M}_M^-(t') \hat{\Phi}_L^+ \hat{\Phi}_M - \hat{\Phi}_L^+ \hat{\Phi}_M \rho^I(t) \hat{M}_L^+(t') \hat{M}_M^-(t') \right] A' + \text{H.c.}, \]  
\[ S_{21} = -\frac{\hbar^2}{N^2} \sum_{L,l,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL + j(l - m)}{N} \right\} \]  
\[ \cdot \left[ \hat{\Phi}_L^+ \hat{M}_m^-(t') \hat{\Phi}_l^+ \hat{\Phi}_m - \hat{\Phi}_l \hat{\Phi}_m^+ \rho^I(t) \hat{\Phi}_L^+ \hat{M}_m^-(t') \right] A + \text{H.c.} \]  
\[ - \frac{\hbar^2}{N^2} \sum_{L,l,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL + j(l - m)}{N} \right\} \]  
\[ \cdot \left[ \rho^I(t) \hat{\Phi}_l^+ \hat{\Phi}_m \hat{M}_m^+(t') - \hat{\Phi}_l \hat{\Phi}_m^+ \rho^I(t) \hat{\Phi}_l^+ \hat{\Phi}_m \right] A' + \text{H.c.}, \]  
\[ S_{22} = -\frac{\hbar^2}{N^2} \sum_{L,l,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL - j(l - m)}{N} \right\} \]  
\[ \cdot \left[ \hat{\Phi}_L^+ \hat{M}_m^-(t') \hat{\Phi}_l^+ \hat{\Phi}_m \rho^I(t) - \hat{\Phi}_l \hat{\Phi}_m^+ \rho^I(t) \hat{\Phi}_L^+ \hat{M}_m^-(t') \right] A + \text{H.c.} \]  
\[ - \frac{\hbar^2}{N^2} \sum_{L,l,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL - j(l - m)}{N} \right\} \]  
\[ \cdot \left[ \rho^I(t) \hat{\Phi}_l^+ \hat{\Phi}_m \hat{M}_m^+(t') - \hat{\Phi}_l \hat{\Phi}_m \rho^I(t) \hat{\Phi}_l^+ \hat{\Phi}_m \right] A' + \text{H.c.}, \]  

C. Calculations to chapter 5
\begin{align*}
S_{23} &= \frac{\hbar^2}{N^2} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL + jl}{N} \right\} \\
&\quad \cdot \left[ \Phi_L^+ M_-(t) \Phi_L^+ M_-(t') \rho' \left( t \right) - \Phi_L^+ \tilde{M}_-(t') \rho' \left( t \right) \right] A + \text{H.c} \\
&\quad + \frac{\hbar^2}{N^2} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL + jl}{N} \right\} \\
&\quad \cdot \left[ \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t') \Phi_L^+ \tilde{M}_-(t) - \Phi_L^+ \tilde{M}_-(t) \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t') \right] A' + \text{H.c}, \\
S_{24} &= \frac{\hbar^2}{N^2} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL - jl}{N} \right\} \\
&\quad \cdot \left[ \Phi_L^+ \tilde{M}_-(t) \Phi_L^+ \tilde{M}_-(t') \Phi_L^+ \rho' \left( t \right) - \Phi_L^+ \tilde{M}_-(t') \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t) \right] A + \text{H.c} \\
&\quad + \frac{\hbar^2}{N^2} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL - jl}{N} \right\} \\
&\quad \cdot \left[ \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t') \Phi_L^+ \tilde{M}_-(t) - \Phi_L^+ \tilde{M}_-(t) \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t') \right] A' + \text{H.c}, \\
S_{25} &= \frac{\hbar^2}{N^2} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL - jl}{N} \right\} \\
&\quad \cdot \left[ \Phi_L^+ \tilde{M}_-(t) \Phi_L^+ \tilde{M}_-(t') \Phi_L^+ \rho' \left( t \right) - \Phi_L^+ \tilde{M}_-(t') \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t) \right] A + \text{H.c} \\
&\quad + \frac{\hbar^2}{N^2} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL - jl}{N} \right\} \\
&\quad \cdot \left[ \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t') \Phi_L^+ \tilde{M}_-(t) - \Phi_L^+ \tilde{M}_-(t) \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t') \right] A' + \text{H.c}, \\
S_{26} &= \frac{\hbar^2}{N^2} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL + jl}{N} \right\} \\
&\quad \cdot \left[ \Phi_L^+ \tilde{M}_-(t) \Phi_L^+ \tilde{M}_+(t') \rho' \left( t \right) - \Phi_L^+ \tilde{M}_+(t') \rho' \left( t \right) \Phi_L^+ \tilde{M}_-(t) \right] A + \text{H.c} \\
&\quad + \frac{\hbar^2}{N^2} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL + jl}{N} \right\} \\
&\quad \cdot \left[ \rho' \left( t \right) \Phi_L^+ \tilde{M}_+(t') \Phi_L^+ \tilde{M}_-(t) - \Phi_L^+ \tilde{M}_-(t) \rho' \left( t \right) \Phi_L^+ \tilde{M}_+(t') \right] A' + \text{H.c},
\end{align*}
\[ S_{2s} = - \frac{\hbar^2}{N^2} \sum_{L=1}^{N-1} s \sum_{j=1}^{N} \exp \left\{ \frac{2\pi i j L}{N} \right\} \]
\[ \cdot \left[ \hat{\Phi}_{L+}^\dagger \hat{M}_{-} \left( t \right) \hat{M}_{+}^\dagger \left( t' \right) \rho \left( t \right) - \hat{M}_{+}^\dagger \left( t' \right) \hat{M}_{-} \left( t \right) \rho \left( t' \right) \right] A + \text{H.c.} \]
\[
S_{3a} = \frac{\hbar^2}{N^2} \sum_{L, l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL + jl}{N} \right\} \]

\[
\cdot \left[ \tilde{M}_L^+(t)\hat{\Phi}_{L-} \hat{\Phi}_L(t) + \tilde{M}_L^+(t')\hat{\Phi}_{L-} \hat{\Phi}_L(t') \right] A + \text{H.c.}
\]

\[
+ \frac{\hbar^2}{N^2} \sum_{L, l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL + jl}{N} \right\} \]

\[
\cdot \left[ \tilde{M}_L^+(t)\hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) + \tilde{M}_L^+(t')\hat{\Phi}_{L-} \hat{\Phi}_L(t') \rho_l(t') \hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) \hat{\Phi}_{L-} \hat{\Phi}_L(t') \rho_l(t') \right] A' + \text{H.c.},
\]

\[
S_{3b} = + \frac{\hbar^2}{N^2} \sum_{L, l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL + jl}{N} \right\} \]

\[
\cdot \left[ \tilde{M}_L^+(t)\hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) + \tilde{M}_L^+(t')\hat{\Phi}_{L-} \hat{\Phi}_L(t') \rho_l(t') \hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) \right] A + \text{H.c.}
\]

\[
+ \frac{\hbar^2}{N^2} \sum_{L, l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL + jl}{N} \right\} \]

\[
\cdot \left[ \tilde{M}_L^+(t)\hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) + \tilde{M}_L^+(t')\hat{\Phi}_{L-} \hat{\Phi}_L(t') \rho_l(t') \hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) \hat{\Phi}_{L-} \hat{\Phi}_L(t') \rho_l(t') \right] A' + \text{H.c.},
\]

\[
S_{3c} = \frac{\hbar^2}{N^2} \sum_{L, l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL + jl}{N} \right\} \]

\[
\cdot \left[ \tilde{M}_L^+(t)\hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) + \tilde{M}_L^+(t')\hat{\Phi}_{L-} \hat{\Phi}_L(t') \rho_l(t') \hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) \right] A + \text{H.c.}
\]

\[
+ \frac{\hbar^2}{N^2} \sum_{L, l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL + jl}{N} \right\} \]

\[
\cdot \left[ \tilde{M}_L^+(t)\hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) + \tilde{M}_L^+(t')\hat{\Phi}_{L-} \hat{\Phi}_L(t') \rho_l(t') \hat{\Phi}_{L-} \hat{\Phi}_L(t) \rho_l(t) \right] A' + \text{H.c.},
\]
\[ S_{3r} = -\frac{\hbar^2}{N^2} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL}{N} \right\} \]
\[ \cdot \left[ \hat{M}_+(t) \hat{\Phi}_{L+} \hat{M}_-(t') \rho^I(t) - \hat{M}_-(t') \hat{\Phi}_{L-} \rho^I(t) \hat{M}_+(t') \right] A + \text{H.c.} \]
\[ -\frac{\hbar^2}{N^2} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL}{N} \right\} \]
\[ \cdot \left[ \rho^I(t) \hat{M}_+(t') \hat{\Phi}_{L-} - \hat{M}_+(t') \rho^I(t) \hat{\Phi}_{L-} \right] A' + \text{H.c.}, \]

\[ S_{3s} = -\frac{\hbar^2}{N^2} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL}{N} \right\} \]
\[ \cdot \left[ \hat{M}_+(t) \hat{\Phi}_{L-} \hat{M}_-(t') \rho^I(t) - \hat{M}_+(t') \hat{\Phi}_{L-} \rho^I(t) \hat{M}_-(t') \right] A + \text{H.c.} \]
\[ -\frac{\hbar^2}{N^2} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jL}{N} \right\} \]
\[ \cdot \left[ \rho^I(t) \hat{M}_-(t') \hat{\Phi}_{L+} - \hat{M}_-(t') \rho^I(t) \hat{\Phi}_{L+} \right] A' + \text{H.c.}, \]

\[ S_{41} = +\frac{\hbar^2}{N^2} \sum_{l,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(l-m)}{N} \right\} \]
\[ \cdot \left[ \hat{M}_+^\dagger(t) \hat{\Phi}_{l+} \hat{M}_-(t') \rho^I(t) - \hat{M}_-(t') \hat{\Phi}_{l-} \rho^I(t) \hat{M}_+^\dagger(t) \right] A + \text{H.c.} \]
\[ +\frac{\hbar^2}{N^2} \sum_{l,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{j(l-m)}{N} \right\} \]
\[ \cdot \left[ \rho^I(t) \hat{\Phi}_{l+} \hat{\Phi}_{m-} - \hat{\Phi}_{l+} \rho^I(t) \hat{\Phi}_{m-} \hat{M}_+(t') \right] A' + \text{H.c.}, \]

\[ S_{42} = +\frac{\hbar^2}{N^2} \sum_{l,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-j(l-m)}{N} \right\} \]
\[ \cdot \left[ \hat{M}_+^\dagger(t) \hat{\Phi}_{l-} \hat{M}_-(t') \rho^I(t) - \hat{M}_-(t') \hat{\Phi}_{l+} \rho^I(t) \hat{M}_+^\dagger(t) \right] A + \text{H.c.} \]
\[ +\frac{\hbar^2}{N^2} \sum_{l,m=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-j(l-m)}{N} \right\} \]
\[ \cdot \left[ \rho^I(t) \hat{\Phi}_{m-} \hat{\Phi}_{l+} \hat{M}_-(t') \rho^I(t) \hat{\Phi}_{m-} \hat{\Phi}_{l+} \hat{M}_+(t') \right] A' + \text{H.c.}, \]
\[ S_{4s} = - \frac{\hbar^2}{N^2} \sum_{l=1}^{N-1} \sum_{j=1}^N \exp \left\{ \frac{2\pi i j l}{N} \right\} \]
\[ \cdot \left[ \hat{M}_+^l(t) \hat{M}_{-}(t) \hat{\Phi}^l_{t+} \hat{M}_{-}(t') \rho^l(t) - \hat{\Phi}^l_{t+} \hat{M}_{-}(t') \rho^l(t) \hat{M}_+^l(t) \hat{M}_{-}(t) \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N^2} \sum_{l=1}^{N-1} \sum_{j=1}^N \exp \left\{ \frac{2\pi i j l}{N} \right\} \]
\[ \cdot \left[ \rho^l(t) \hat{\Phi}^l_{t+} \hat{M}_{-}(t') \hat{M}_{-}(t) \hat{M}_+^l(t) \hat{M}_{-}(t) - \hat{M}_+^l(t) \hat{M}_{-}(t) \rho^l(t) \hat{\Phi}^l_{t+} \hat{M}_{-}(t') \right] A' + \text{H.c.,} \]
\[ S_{4a} = - \frac{\hbar^2}{N^2} \sum_{l=1}^{N-1} \sum_{j=1}^N \exp \left\{ \frac{2\pi i j l}{N} \right\} \]
\[ \cdot \left[ \hat{M}_+^l(t) \hat{M}_{-}(t) \hat{\Phi}^l_{t-} \rho^l(t) - \hat{\Phi}^l_{t-} \hat{M}_{-}(t') \rho^l(t) \hat{M}_+^l(t) \hat{M}_{-}(t) \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N^2} \sum_{l=1}^{N-1} \sum_{j=1}^N \exp \left\{ \frac{2\pi i j l}{N} \right\} \]
\[ \cdot \left[ \rho^l(t) \hat{M}_+^l(t') \hat{\Phi}^l_{t-} \hat{M}_-^l(t) \hat{M}_{-}(t) - \hat{M}_+^l(t) \hat{M}_{-}(t) \rho^l(t) \hat{M}_+^l(t') \hat{\Phi}^l_{t-} \right] A' + \text{H.c.,} \]
\[ S_{4b} = - \frac{\hbar^2}{N^2} \sum_{l=1}^{N-1} \sum_{j=1}^N \exp \left\{ \frac{2\pi i j l}{N} \right\} \]
\[ \cdot \left[ \hat{M}_+^l(t) \hat{\Phi}^l_{t-} \hat{M}_-^l(t') \rho^l(t) - \hat{\Phi}^l_{t-} \hat{M}_-^l(t') \rho^l(t) \hat{M}_+^l(t) \hat{M}_{-}(t) \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N^2} \sum_{l=1}^{N-1} \sum_{j=1}^N \exp \left\{ \frac{2\pi i j l}{N} \right\} \]
\[ \cdot \left[ \rho^l(t) \hat{\Phi}^l_{t-} \hat{M}_-^l(t') \hat{M}_-^l(t) \hat{M}_{-}(t) - \hat{M}_-^l(t) \hat{M}_{-}(t) \rho^l(t) \hat{\Phi}^l_{t-} \hat{M}_-^l(t') \right] A' + \text{H.c.,} \]
\[ S_{4r} = + \frac{\hbar^2}{N^2} \sum_{j=1}^N \left[ \hat{M}_+^j(t) \hat{M}_{-}(t) \hat{M}_-^j(t') \hat{M}_{-}(t') \rho^j(t) - \hat{M}_+^j(t') \hat{M}_{-}(t') \rho^j(t) \hat{M}_-^j(t) \hat{M}_{-}(t) \right] A + \text{H.c.} \]
\[ + \frac{\hbar^2}{N^2} \sum_{j=1}^N \left[ \rho^j(t) \hat{M}_+^j(t') \hat{M}_{-}(t') \hat{M}_-^j(t) \hat{M}_{-}(t) - \hat{M}_+^j(t) \hat{M}_{-}(t) \rho^j(t) \hat{M}_-^j(t') \hat{M}_{-}(t') \right] A' + \text{H.c.,} \]
\[ S_{4s} = + \frac{\hbar^2}{N^2} \sum_{j=1}^{N} \left[ \tilde{M}_j^+ (t) \tilde{M}_- (t) \tilde{M}_{j-1}^+ (t') \tilde{M}_{j+1}^- (t') \rho^I (t) - \tilde{M}_j^- (t') \tilde{M}_+ (t) \rho^I (t) \tilde{M}_j^+ (t) \tilde{M}_- (t) \right] A + \text{H.c.} \]

\[ + \frac{\hbar^2}{N^2} \sum_{j=1}^{N} \left[ \rho^I (t) \tilde{M}_j^+ (t') \tilde{M}_+ (t') \tilde{M}_{j-1}^+ (t') \tilde{M}_{j+1}^- (t) - \tilde{M}_j^- (t) \tilde{M}_+ (t) \rho^I (t) \tilde{M}_j^+ (t) \tilde{M}_- (t) \right] A' + \text{H.c.}, \]

\[ S_{51} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL + jl}{N} \right\} \left[ \hat{\Phi}_{L+}^I \hat{\Phi}_{L+}^I \rho^I (t) - \hat{\Phi}_{L+}^I \rho^I (t) \hat{\Phi}_{L+}^I \right] B + \text{H.c.} \]

\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL + jl}{N} \right\} \left[ \rho^I (t) \hat{\Phi}_{L+}^I \hat{\Phi}_{L+}^I - \hat{\Phi}_{L+}^I \rho^I (t) \hat{\Phi}_{L+}^I \right] B' + \text{H.c.}, \]

\[ S_{52} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL - jl}{N} \right\} \left[ \hat{\Phi}_{L+}^I \hat{\Phi}_{L+}^I \rho^I (t) - \hat{\Phi}_{L+}^I \rho^I (t) \hat{\Phi}_{L+}^I \right] B + \text{H.c.} \]

\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL - jl}{N} \right\} \left[ \rho^I (t) \hat{\Phi}_{L+}^I \hat{\Phi}_{L+}^I - \hat{\Phi}_{L+}^I \rho^I (t) \hat{\Phi}_{L+}^I \right] B' + \text{H.c.}, \]

\[ S_{53} = - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL}{N} \right\} \left[ \hat{\Phi}_{L+}^I \tilde{M}_{j+1}^-(t') \rho^I (t) - \tilde{M}_{j+1}^+(t') \rho^I (t) \hat{\Phi}_{L+}^I \right] B + \text{H.c.} \]

\[ - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL}{N} \right\} \left[ \rho^I (t) \tilde{M}_{j+1}^+(t') \hat{\Phi}_{L+}^I - \tilde{M}_{j+1}^-(t') \rho^I (t) \hat{\Phi}_{L+}^I \right] B' + \text{H.c.}, \]

\[ S_{54} = - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL}{N} \right\} \left[ \hat{\Phi}_{L+}^I \tilde{M}_{j+1}^- (t') \rho^I (t) - \tilde{M}_{j+1}^+ (t') \rho^I (t) \hat{\Phi}_{L+}^I \right] B + \text{H.c.} \]

\[ - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL}{N} \right\} \left[ \rho^I (t) \tilde{M}_{j+1}^+ (t') \hat{\Phi}_{L+}^I - \tilde{M}_{j+1}^- (t') \rho^I (t) \hat{\Phi}_{L+}^I \right] B' + \text{H.c.}, \]

\[ S_{61} = - \frac{\hbar^2}{N} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL}{N} \right\} \left[ \tilde{M}_j^+ (t) \hat{\Phi}_{L+}^I \rho^I (t) - \hat{\Phi}_{L+}^I \rho^I (t) \tilde{M}_j^+ (t) \right] B + \text{H.c.} \]

\[ - \frac{\hbar^2}{N} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{jL}{N} \right\} \left[ \rho^I (t) \hat{\Phi}_{L+}^I \tilde{M}_j^+ (t) - \tilde{M}_j^+ (t) \rho^I (t) \hat{\Phi}_{L+}^I \right] B' + \text{H.c.}, \]

\[ S_{62} = - \frac{\hbar^2}{N} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{-jl}{N} \right\} \left[ \tilde{M}_j^- (t) \hat{\Phi}_{L+}^I \rho^I (t) - \hat{\Phi}_{L+}^I \rho^I (t) \tilde{M}_j^- (t) \right] B + \text{H.c.} \]

\[ - \frac{\hbar^2}{N} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2 \pi i \frac{-jl}{N} \right\} \left[ \rho^I (t) \hat{\Phi}_{L+}^I \tilde{M}_j^- (t) - \tilde{M}_j^- (t) \rho^I (t) \hat{\Phi}_{L+}^I \right] B' + \text{H.c.}, \]
\[ S_{63} = \frac{\hbar^2}{N} \sum_{j=1}^{N} \left[ \tilde{M}_+^I(t) \tilde{M}_+^I(t') \rho^I(t) - \tilde{M}_-^I(t') \rho^I(t) \tilde{M}_-^I(t) \right] B + \text{H.c} \]
\[ + \frac{\hbar^2}{N} \sum_{j=1}^{N} \left[ \rho^I(t) \tilde{M}_+^I(t') \tilde{M}_+^I(t) - \tilde{M}_+^I(t') \rho^I(t) \tilde{M}_+^I(t) \right] B' + \text{H.c.} , \]
\[ S_{64} = \frac{\hbar^2}{N} \sum_{j=1}^{N} \left[ \tilde{M}_+^I(t) \tilde{M}_+^I(t') \rho^I(t) - \tilde{M}_-^I(t') \rho^I(t) \tilde{M}_-^I(t) \right] B + \text{H.c} \]
\[ + \frac{\hbar^2}{N} \sum_{j=1}^{N} \left[ \rho^I(t) \tilde{M}_-^I(t') \tilde{M}_-^I(t) - \tilde{M}_-^I(t') \rho^I(t) \tilde{M}_-^I(t) \right] B' + \text{H.c.} , \]
\[ S_{71} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL + jl}{N} \right\} \left[ \tilde{\Phi}_L^I \tilde{\Phi}_L^I \rho^I(t) - \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I \right] C + \text{H.c} \]
\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL + jl}{N} \right\} \left[ \rho^I(t) \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I - \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I \right] C' + \text{H.c.} , \]
\[ S_{72} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL - jl}{N} \right\} \left[ \tilde{\Phi}_L^I \rho^I(t) - \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I \right] C + \text{H.c} \]
\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL - jl}{N} \right\} \left[ \rho^I(t) \tilde{\Phi}_L^I - \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I \right] C' + \text{H.c.} , \]
\[ S_{73} = - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL}{N} \right\} \left[ \tilde{\Phi}_L^I \tilde{\Phi}_L^I \rho^I(t) - \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I \right] C + \text{H.c} \]
\[ - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL}{N} \right\} \left[ \rho^I(t) \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I - \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I \right] C' + \text{H.c.} , \]
\[ S_{74} = - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL}{N} \right\} \left[ \tilde{\Phi}_L^I \tilde{\Phi}_L^I \rho^I(t) - \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I \right] C + \text{H.c} \]
\[ - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jL}{N} \right\} \left[ \rho^I(t) \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I - \tilde{\Phi}_L^I \rho^I(t) \tilde{\Phi}_L^I \right] C' + \text{H.c.} , \]
\[ S_{81} = - \frac{\hbar^2}{N} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jl}{N} \right\} \left[ \tilde{\Phi}_l^I \tilde{\Phi}_l^I \rho^I(t) - \tilde{\Phi}_l^I \rho^I(t) \tilde{\Phi}_l^I \right] C + \text{H.c} \]
\[ - \frac{\hbar^2}{N} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{jl}{N} \right\} \left[ \rho^I(t) \tilde{\Phi}_l^I \rho^I(t) \tilde{\Phi}_l^I - \tilde{\Phi}_l^I \rho^I(t) \tilde{\Phi}_l^I \right] C' + \text{H.c.} , \]
C. Calculations to chapter 5

\[ S_{82} = - \frac{\hbar^2}{N} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jl}{N} \right\} \left[ \bar{M}^\dagger_l(t) \Phi_{l-} \rho^l(t) - \hat{\Phi}_{l-} \rho^l(t) \bar{M}^\dagger_l(t) \right] C + \text{H.c} \]

\[- \frac{\hbar^2}{N} \sum_{l=1}^{N-1} \sum_{j=1}^{N} \exp \left\{ 2\pi i \frac{-jl}{N} \right\} \left[ \rho^l(t) \Phi_{l-} \bar{M}^\dagger_l(t) - \bar{M}^\dagger_l(t) \rho^l(t) \hat{\Phi}_{l-} \right] C' + \text{H.c.}, \]

\[ S_{83} = \frac{\hbar^2}{N} \sum_{j=1}^{N} \left[ \bar{M}^\dagger_l(t) \bar{M}^\dagger_j(t) \rho^j(t) - \bar{M}^\dagger_j(t) \rho^j(t) \bar{M}^\dagger_l(t) \right] C + \text{H.c} \]

\[ + \frac{\hbar^2}{N} \sum_{j=1}^{N} \left[ \rho^j(t) \bar{M}^\dagger_l(t) \bar{M}^\dagger_j(t) - \bar{M}^\dagger_j(t) \rho^j(t) \bar{M}^\dagger_l(t) \right] C' + \text{H.c.} \]

and

\[ S_{84} = \frac{\hbar^2}{N} \sum_{j=1}^{N} \left[ \bar{M}^\dagger_l(t) \bar{M}^\dagger_j(t) \rho^j(t) - \bar{M}^\dagger_j(t) \rho^j(t) \bar{M}^\dagger_l(t) \right] C + \text{H.c} \]

\[ + \frac{\hbar^2}{N} \sum_{j=1}^{N} \left[ \rho^j(t) \bar{M}^\dagger_l(t) \bar{M}^\dagger_j(t) - \bar{M}^\dagger_j(t) \rho^j(t) \bar{M}^\dagger_l(t) \right] C' + \text{H.c.} \]

Here \( A, A', B, B', C \) and \( C' \) are defined by

\[ X = \text{tr}_B \{ \bar{\Gamma}^j_X(t) \tilde{\Gamma}^j_X(t') \rho_B \} = \sum_k |g_{jk}^X|^2 \langle \hat{X}_{kj} \hat{X}^\dagger_{kj} \rangle \times e^{-i\omega_k(t-t')} + \langle \hat{X}^\dagger_{kj} \hat{X}_{kj} \rangle \times e^{i\omega_k(t-t')} \]

\[ X' = \text{tr}_B \{ \tilde{\Gamma}^j_X(t') \bar{\Gamma}^j_X(t) \rho_B \} = \sum_k |g_{jk}^X|^2 \langle \hat{X}^\dagger_{kj} \hat{X}_{kj} \rangle \times e^{i\omega_k(t-t')} + \langle \hat{X}_{kj} \hat{X}^\dagger_{kj} \rangle \times e^{-i\omega_k(t-t')} \]

with \( X \in \{ A, B, C \} \).

This expressions for \( X \) and \( X' \) can be further simplified by using equations (5.19)-(5.24) in section 5.2.2 to calculate the correlation functions of the bath variables. Furthermore the correspondence (5.27) in section 5.2.2 is used to convert the sum over \( k \) into an integral over \( \omega \). This results in

\[ A = \int_0^\infty d\omega h(\omega) |g_{c+i-c}(\omega)|^2 \left[ e^{-i\omega(t-t')} [\bar{n}(\omega) + 1] + e^{i\omega(t-t')} \bar{n}(\omega) \right], \]

\[ A' = \int_0^\infty d\omega h(\omega) |g_{c+i-c}(\omega)|^2 \left[ e^{i\omega(t-t')} [\bar{n}(\omega) + 1] + e^{-i\omega(t-t')} \bar{n}(\omega) \right] = A^*, \]

\[ B = \int_0^\infty d\omega h(\omega) |g_{c+i-b}(\omega)|^2 \left[ e^{-i\omega(t-t')} [\bar{n}(\omega) + 1] + e^{i\omega(t-t')} \bar{n}(\omega) \right], \]

\[ B' = \int_0^\infty d\omega h(\omega) |g_{c+i-b}(\omega)|^2 \left[ e^{i\omega(t-t')} [\bar{n}(\omega) + 1] + e^{-i\omega(t-t')} \bar{n}(\omega) \right] = B^*, \]

\[ C = \int_0^\infty d\omega h(\omega) |g_{c+i-b}(\omega)|^2 \left[ e^{-i\omega(t-t')} [\bar{n}(\omega) + 1] + e^{i\omega(t-t')} \bar{n}(\omega) \right], \]

\[ C' = \int_0^\infty d\omega h(\omega) |g_{c+i-b}(\omega)|^2 \left[ e^{i\omega(t-t')} [\bar{n}(\omega) + 1] + e^{-i\omega(t-t')} \bar{n}(\omega) \right] = C^*. \]
Since $A$, $A'$, $B$, $B'$, $C$ and $C'$ are independent of the atom number $j$ the summation over $j$ in the summands $S_{ab}$ can be carried out, which yields:

\[
S_{11} = \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+l,M+m} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A + \text{H.c.}
\]

\[
+ \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+l,M+m} \left[ \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \right] A' + \text{H.c.}
\]

\[
S_{12} = \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+m,L+M} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \rho(t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \right] A + \text{H.c.}
\]

\[
- \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+l,M+m} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A' + \text{H.c.}
\]

\[
S_{13} = - \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+m,L+M} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A + \text{H.c.}
\]

\[
- \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+l,M+m} \left[ \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A' + \text{H.c.}
\]

\[
S_{14} = - \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+m,L+M} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A + \text{H.c.}
\]

\[
- \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+l,M+m} \left[ \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A' + \text{H.c.}
\]

\[
S_{15} = - \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+m,L+M} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A + \text{H.c.}
\]

\[
- \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+l,M+m} \left[ \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A' + \text{H.c.}
\]

\[
S_{16} = - \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+m,L+M} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A + \text{H.c.}
\]

\[
- \frac{\hbar^2}{N} \sum_{L,l,M,m=1}^{N-1} \delta_{L+l,M+m} \left[ \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A' + \text{H.c.}
\]

\[
S_{17} = \frac{\hbar^2}{N} \sum_{L,M=1}^{N-1} \delta_{L,M} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A + \text{H.c.}
\]

\[
+ \frac{\hbar^2}{N} \sum_{L,M=1}^{N-1} \delta_{L,M} \left[ \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \rho(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-}^\dagger (t) \right] A' + \text{H.c.}
\]

C. Calculations to chapter 5
\[ S_{1s} = \frac{\hbar^2}{N} \sum_{L,M=1}^{N-1} \delta_{L,M} \left[ \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{m-} \hat{M}_+^\dagger(t') \hat{M}_+(t') \rho^L(t) - \hat{M}_+^\dagger(t') \hat{M}_+^\dagger(t') \rho^L(t) \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{M-} \right] A + \text{H.c.} \]

\[ + \frac{\hbar^2}{N} \sum_{L,M=1}^{N-1} \delta_{L,M} \left[ \rho^L(t) \hat{M}_+^\dagger(t') \hat{M}_+(t') \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{m-} - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{m-} \rho^L(t) \hat{M}_+^\dagger(t') \hat{M}_+(t') \right] A' + \text{H.c.}, \]

\[ S_{21} = -\frac{\hbar^2}{N} \sum_{L,l,m=1}^{N-1} \delta_{L+l,m} \left[ \hat{\Phi}_{L+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{l+}^\dagger \hat{M}_+(t) \rho^L(t) - \hat{\Phi}_{l+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) \right] A + \text{H.c} \]

\[ - \frac{\hbar^2}{N} \sum_{L,l,m=1}^{N-1} \delta_{L+l,m} \left[ \rho^L(t) \hat{\Phi}_{l+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{l+}^\dagger \hat{M}_-^\dagger(t) \hat{M}_+(t) \right] A' + \text{H.c}, \]

\[ S_{22} = -\frac{\hbar^2}{N} \sum_{L,l,m=1}^{N-1} \delta_{L+m,l} \left[ \hat{\Phi}_{L+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{m-}^\dagger \hat{M}_+(t) \rho^L(t) - \hat{\Phi}_{m-}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) \right] A + \text{H.c} \]

\[ - \frac{\hbar^2}{N} \sum_{L,l,m=1}^{N-1} \delta_{L+m,l} \left[ \rho^L(t) \hat{\Phi}_{m-}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{m-}^\dagger \hat{M}_-^\dagger(t) \hat{M}_+(t) \right] A' + \text{H.c}, \]

\[ S_{23} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{L+l,0} \left[ \hat{\Phi}_{L+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{l+}^\dagger \hat{M}_+(t) \rho^L(t) - \hat{\Phi}_{l+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) \right] A + \text{H.c} \]

\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{L+l,0} \left[ \rho^L(t) \hat{\Phi}_{l+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) - \hat{\Phi}_{L+}^\dagger \hat{\Phi}_{l+}^\dagger \hat{M}_-^\dagger(t) \hat{M}_+(t) \right] A' + \text{H.c} , \]

\[ = 0 \text{ since } l, L > 0, \]

\[ S_{24} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{l,l} \left[ \hat{\Phi}_{l+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{l+}^\dagger \hat{M}_+(t) \rho^L(t) - \hat{M}_+^\dagger(t') \hat{\Phi}_{l+}^\dagger \hat{M}_+(t) \right] A + \text{H.c} \]

\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{l,l} \left[ \rho^L(t) \hat{M}_+^\dagger(t') \hat{\Phi}_{l+}^\dagger \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) - \hat{\Phi}_{l+}^\dagger \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) \right] A' + \text{H.c}, \]

\[ S_{25} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{l,l} \left[ \hat{\Phi}_{l+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{l+}^\dagger \hat{M}_+(t) \rho^L(t) - \hat{M}_+^\dagger(t') \hat{\Phi}_{l+}^\dagger \hat{M}_+(t) \right] A + \text{H.c} \]

\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{l,l} \left[ \rho^L(t) \hat{M}_+^\dagger(t') \hat{\Phi}_{l+}^\dagger \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) - \hat{\Phi}_{l+}^\dagger \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) \right] A' + \text{H.c}, \]

\[ S_{26} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{L+l,0} \left[ \hat{\Phi}_{L+}^\dagger \hat{M}_-^\dagger(t) \hat{\Phi}_{l+}^\dagger \hat{M}_+(t) \rho^L(t) - \hat{\Phi}_{l+}^\dagger \hat{M}_+^\dagger(t') \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) \right] A + \text{H.c} \]

\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{L+l,0} \left[ \rho^L(t) \hat{\Phi}_{l+}^\dagger \hat{M}_+^\dagger(t') \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) - \hat{\Phi}_{l+}^\dagger \hat{\Phi}_{L+}^\dagger \hat{M}_+(t) \right] A' + \text{H.c}, \]

\[ = 0 \text{ since } l, L > 0, \]
\[ S_{2r} = - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \delta_{L,0} \left[ \hat{\Phi}_{L+}^\dagger \bar{M}_{-} (t) \hat{M}_{+}^\dagger (t') \bar{M}_{-} (t') \rho^I (t) - \hat{M}_{+}^\dagger (t') \bar{M}_{-} (t') \rho^I (t) \hat{\Phi}_{L+}^\dagger \bar{M}_{-} (t) \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \delta_{L,0} \left[ \rho^I (t) \hat{M}_{+}^\dagger (t') \bar{M}_{-} (t') \hat{\Phi}_{L+}^\dagger \bar{M}_{-} (t) - \hat{\Phi}_{L+}^\dagger \bar{M}_{-} (t) \rho^I (t) \hat{M}_{+}^\dagger (t') \bar{M}_{-} (t') \right] A' + \text{H.c.} \]

\[ = 0 \text{ since } L > 0, \]

\[ S_{2s} = - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \delta_{L,0} \left[ \hat{\Phi}_{L+}^\dagger \bar{M}_{-} (t) \hat{M}_{+}^\dagger (t') \bar{M}_{-} (t') \rho^I (t) - \hat{M}_{+}^\dagger (t') \bar{M}_{-} (t') \rho^I (t) \hat{\Phi}_{L+}^\dagger \bar{M}_{-} (t) \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \delta_{L,0} \left[ \rho^I (t) \hat{M}_{+}^\dagger (t') \bar{M}_{-} (t') \hat{\Phi}_{L+}^\dagger \bar{M}_{-} (t) - \hat{\Phi}_{L+}^\dagger \bar{M}_{-} (t) \rho^I (t) \hat{M}_{+}^\dagger (t') \bar{M}_{-} (t') \right] A' + \text{H.c.} \]

\[ = 0 \text{ since } L > 0, \]

\[ S_{3i} = - \frac{\hbar^2}{N} \sum_{L,L,m=1}^{N-1} \delta_{i,L+m} \left[ \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \hat{\Phi}_{i+}^\dagger \hat{\Phi}_{m-} \rho^I (t) - \hat{\Phi}_{i+}^\dagger \hat{\Phi}_{m-} \rho^I (t) \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N} \sum_{L,L,m=1}^{N-1} \delta_{i,L+m} \left[ \rho^I (t) \hat{\Phi}_{i+}^\dagger \hat{\Phi}_{m-} \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} - \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \rho^I (t) \hat{\Phi}_{i+}^\dagger \hat{\Phi}_{m-} \right] A' + \text{H.c.} \]

\[ S_{3s} = - \frac{\hbar^2}{N} \sum_{L,l,m=1}^{N-1} \delta_{m,L+l} \left[ \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \hat{\Phi}_{m+}^\dagger \hat{\Phi}_{l+} \rho^I (t) - \hat{\Phi}_{m+}^\dagger \hat{\Phi}_{l+} \rho^I (t) \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N} \sum_{L,l,m=1}^{N-1} \delta_{m,L+l} \left[ \rho^I (t) \hat{\Phi}_{m+}^\dagger \hat{\Phi}_{l+} \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} - \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \rho^I (t) \hat{\Phi}_{m+}^\dagger \hat{\Phi}_{l+} \right] A' + \text{H.c.} \]

\[ S_{3o} = - \frac{\hbar^2}{N} \sum_{L,l,m=1}^{N-1} \delta_{l,0} \left[ \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \hat{\Phi}_{l+} \hat{\Phi}_{-} \rho^I (t) - \hat{\Phi}_{l+} \hat{\Phi}_{-} \rho^I (t) \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \right] A + \text{H.c.} \]
\[ + \frac{\hbar^2}{N} \sum_{L,l,m=1}^{N-1} \delta_{l,0} \left[ \rho^I (t) \hat{\Phi}_{l+} \hat{\Phi}_{-} \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} - \hat{M}_{+}^\dagger (t) \hat{\Phi}_{L-} \rho^I (t) \hat{\Phi}_{l+} \hat{\Phi}_{-} \right] A' + \text{H.c.} \]

\[ = 0 \text{ since } l, L > 0, \]
\[ S_{35} = + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{0,L+l} \left[ \tilde{M}^1_L(t) \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A + \text{H.c.} \]
\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{0,L+l} \left[ \rho^I(t) \tilde{M}^1_L(t') \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A' + \text{H.c.} \]
\[ = 0 \text{ since } l, L > 0, \]
\[ S_{36} = \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{L,l} \left[ \tilde{M}^1_L(t) \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A + \text{H.c.} \]
\[ + \frac{\hbar^2}{N} \sum_{L,l=1}^{N-1} \delta_{L,l} \left[ \rho^I(t) \tilde{M}^1_L(t') \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A' + \text{H.c.}, \]
\[ S_{37} = - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \delta_{L,0} \left[ \tilde{M}^1_L(t) \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \delta_{L,0} \left[ \rho^I(t) \tilde{M}^1_L(t') \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A' + \text{H.c.} \]
\[ = 0 \text{ since } L > 0, \]
\[ S_{38} = - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \delta_{L,0} \left[ \tilde{M}^1_L(t) \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A + \text{H.c.} \]
\[ - \frac{\hbar^2}{N} \sum_{L=1}^{N-1} \delta_{L,0} \left[ \rho^I(t) \tilde{M}^1_L(t') \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A' + \text{H.c.} \]
\[ = 0 \text{ since } L > 0, \]
\[ S_{41} = + \frac{\hbar^2}{N} \sum_{L,m=1}^{N-1} \delta_{l,m} \left[ \tilde{M}^1_L(t) \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A + \text{H.c.} \]
\[ + \frac{\hbar^2}{N} \sum_{L,m=1}^{N-1} \delta_{l,m} \left[ \rho^I(t) \tilde{M}^1_L(t') \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A' + \text{H.c.}, \]
\[ S_{42} = + \frac{\hbar^2}{N} \sum_{L,m=1}^{N-1} \delta_{l,m} \left[ \tilde{M}^1_L(t) \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A + \text{H.c.} \]
\[ + \frac{\hbar^2}{N} \sum_{L,m=1}^{N-1} \delta_{l,m} \left[ \rho^I(t) \tilde{M}^1_L(t') \hat{\Phi}_L - \tilde{M}^1_L(t') \hat{\Phi}_L \right] A' + \text{H.c.}, \]
\[ S_{4_3} = -\frac{\hbar^2}{N} \sum_{l=1}^{N-1} \delta_{l,0} \left[ \tilde{M}_+^l(t) \tilde{M}_-(t) \tilde{\Phi}_L^l \tilde{\Phi}_R^l \rho^l(t) - \tilde{\Phi}_L^l \tilde{\Phi}_R^l (t) \rho^l(t) \tilde{M}_+^l(t) \tilde{M}_-(t) \right] A + \text{H.c.} \]

\[ S_{4_4} = -\frac{\hbar^2}{N} \sum_{l=1}^{N-1} \delta_{l,0} \left[ \rho^l(t) \tilde{M}_+^l(t) \tilde{M}_-(t') \tilde{\Phi}_L^l \rho^l(t') - \tilde{M}_+^l(t') \tilde{\Phi}_L^l \rho^l(t) \tilde{M}_+^l(t) \tilde{M}_-(t) \right] A' + \text{H.c.} \]

\[ S_{4_5} = -\frac{\hbar^2}{N} \sum_{l=1}^{N-1} \delta_{l,0} \left[ \tilde{\Phi}_R^l(t) \tilde{\Phi}_R^l(t') \tilde{\Phi}_L^l \rho^l(t) - \tilde{\Phi}_R^l(t') \tilde{\Phi}_L^l \rho^l(t) \tilde{\Phi}_R^l(t) \tilde{\Phi}_L^l \tilde{\Phi}_R^l(t) \right] A' + \text{H.c.} \]

\[ S_{4_6} = -\frac{\hbar^2}{N} \sum_{l=1}^{N-1} \delta_{l,0} \left[ \tilde{\Phi}_R^l(t) \tilde{\Phi}_R^l(t') \tilde{\Phi}_L^l \tilde{\Phi}_R^l(t) \tilde{\Phi}_L^l \tilde{\Phi}_R^l(t) \right] A + \text{H.c.} \]

\[ S_{4_7} = \frac{\hbar^2}{N} \left[ \tilde{\Phi}_R^l(t) \tilde{\Phi}_R^l(t') \tilde{\Phi}_L^l \tilde{\Phi}_R^l(t) \tilde{\Phi}_L^l \tilde{\Phi}_R^l(t) \right] A + \text{H.c.} \]

\[ S_{4_8} = \frac{\hbar^2}{N} \left[ \tilde{\Phi}_R^l(t) \tilde{\Phi}_R^l(t') \tilde{\Phi}_L^l \tilde{\Phi}_R^l(t) \tilde{\Phi}_L^l \tilde{\Phi}_R^l(t) \right] A + \text{H.c.} \]

\[ S_{5_1} = \delta_{L+1,0} \left[ \left( \tilde{\Phi}_L^l \tilde{\Phi}_R^l \rho^l(t) - \tilde{\Phi}_L^l \rho^l(t) \tilde{\Phi}_R^l \right) B + \left[ \rho^l(t) \tilde{\Phi}_L^l \tilde{\Phi}_R^l \tilde{\Phi}_L^l \tilde{\Phi}_R^l \rho^l(t) \tilde{\Phi}_R^l \right] \right] B' + \text{H.c.} \]

\[ S_{5_2} = \delta_{L+1,0} \left[ \left( \tilde{\Phi}_L^l \tilde{\Phi}_R^l \rho^l(t) - \tilde{\Phi}_L^l \rho^l(t) \tilde{\Phi}_R^l \right) B + \left[ \rho^l(t) \tilde{\Phi}_L^l \tilde{\Phi}_R^l \tilde{\Phi}_L^l \tilde{\Phi}_R^l \rho^l(t) \tilde{\Phi}_R^l \right] \right] B' + \text{H.c.} \]
\[ S_{53} = -\hbar^2 \sum_{L=1}^{N-1} \delta_{L,0} \left[ \hat{\Phi}^\dagger_{L+} \hat{M}^\dagger_+(t') \rho^I(t) - \hat{M}^\dagger_+(t') \rho^I(t) \hat{\Phi}^\dagger_{L+} \right] B + H.c \]

\[ - \hbar^2 \sum_{L=1}^{N-1} \delta_{L,0} \left[ \rho^I(t) \hat{M}^\dagger_+(t') \hat{\Phi}^\dagger_{L+} - \hat{\Phi}^\dagger_{L+} \rho^I(t) \hat{M}^\dagger_+(t') \right] B' + H.c. \]

= 0 since \( L > 0 \),

\[ S_{54} = -\hbar^2 \sum_{L=1}^{N-1} \delta_{L,0} \left[ \hat{\Phi}^\dagger_{L+} \hat{M}^\dagger_+(t') \rho^I(t) - \hat{M}^\dagger_+(t') \rho^I(t) \hat{\Phi}^\dagger_{L+} \right] B + H.c \]

\[ - \hbar^2 \sum_{L=1}^{N-1} \delta_{L,0} \left[ \rho^I(t) \hat{M}^\dagger_+(t') \hat{\Phi}^\dagger_{L+} - \hat{\Phi}^\dagger_{L+} \rho^I(t) \hat{M}^\dagger_+(t') \right] B' + H.c. \]

= 0 since \( L > 0 \),

\[ S_{61} = -\hbar^2 \sum_{l=1}^{N-1} \delta_{l,0} \left[ \hat{M}^\dagger_+(t) \hat{\Phi}^\dagger_{l+} \rho^I(t) - \hat{\Phi}^\dagger_{l+} \rho^I(t) \hat{M}^\dagger_+(t) \right] B + H.c \]

\[ - \hbar^2 \sum_{l=1}^{N-1} \delta_{l,0} \left[ \rho^I(t) \hat{\Phi}^\dagger_{l+} \hat{M}^\dagger_+(t) - \hat{M}^\dagger_+(t) \rho^I(t) \hat{\Phi}^\dagger_{l+} \right] B' + H.c. \]

= 0 since \( l > 0 \),

\[ S_{62} = -\hbar^2 \sum_{l=1}^{N-1} \delta_{l,0} \left[ \hat{M}^\dagger_+(t) \hat{\Phi}^\dagger_{l+} \rho^I(t) - \hat{\Phi}^\dagger_{l+} \rho^I(t) \hat{M}^\dagger_+(t) \right] B + H.c \]

\[ - \hbar^2 \sum_{l=1}^{N-1} \delta_{l,0} \left[ \rho^I(t) \hat{\Phi}^\dagger_{l+} \hat{M}^\dagger_+(t) - \hat{M}^\dagger_+(t) \rho^I(t) \hat{\Phi}^\dagger_{l+} \right] B' + H.c. \]

= 0 since \( l > 0 \),

\[ S_{63} = \hbar^2 \left[ \hat{M}^\dagger_+(t) \hat{\Phi}^\dagger_{l+} \rho^I(t) - \hat{\Phi}^\dagger_{l+} \rho^I(t) \hat{M}^\dagger_+(t) \right] B + H.c \]

\[ + \hbar^2 \left[ \rho^I(t) \hat{\Phi}^\dagger_{l+} \hat{M}^\dagger_+(t') - \hat{M}^\dagger_+(t') \rho^I(t) \hat{\Phi}^\dagger_{l+} \right] B' + H.c., \]

\[ S_{64} = \hbar^2 \left[ \hat{M}^\dagger_+(t) \hat{\Phi}^\dagger_{l+} \rho^I(t) - \hat{\Phi}^\dagger_{l+} \rho^I(t) \hat{M}^\dagger_+(t) \right] B + H.c \]

\[ + \hbar^2 \left[ \rho^I(t) \hat{\Phi}^\dagger_{l+} \hat{M}^\dagger_+(t') - \hat{M}^\dagger_+(t') \rho^I(t) \hat{\Phi}^\dagger_{l+} \right] B' + H.c., \]

\[ S_{71} = \hbar^2 \sum_{L,l=1}^{N-1} \delta_{L+l,0} \left[ \left[ \hat{\Phi}^\dagger_{L-} \hat{\Phi}^\dagger_{l-} \rho^I(t) - \hat{\Phi}^\dagger_{l-} \rho^I(t) \hat{\Phi}^\dagger_{L-} \right] C + \left[ \rho^I(t) \hat{\Phi}^\dagger_{L-} \hat{\Phi}^\dagger_{l-} - \hat{\Phi}^\dagger_{l-} \rho^I(t) \hat{\Phi}^\dagger_{L-} \right] C' \right] + H.c. \]

= 0 since \( L, L > 0 \),

\[ S_{72} = \hbar^2 \sum_{L,l=1}^{N-1} \delta_{L,l} \left[ \left[ \hat{\Phi}^\dagger_{L-} \hat{\Phi}^\dagger_{l-} \rho^I(t) - \hat{\Phi}^\dagger_{l-} \rho^I(t) \hat{\Phi}^\dagger_{L-} \right] C + \left[ \rho^I(t) \hat{\Phi}^\dagger_{L-} \hat{\Phi}^\dagger_{l-} - \hat{\Phi}^\dagger_{l-} \rho^I(t) \hat{\Phi}^\dagger_{L-} \right] C' \right] + H.c., \]

\[ \text{C. Calculations to chapter 5} \]
C. Calculations to chapter 5

\[ S_{73} = -\hbar^2 \sum_{L=1}^{N-1} \delta_{L,0} \left[ \tilde{\Phi}_{L,-}^I \tilde{M}_{-}^I(t') \rho^I(t) - \tilde{M}_{-}^I(t') \rho^I(t) \tilde{\Phi}_{L,-}^I \right] C + \text{H.c} \]

\[-\hbar^2 \sum_{L=1}^{N-1} \delta_{L,0} \left[ \rho^I(t) \tilde{M}_{-}^I(t') \tilde{\Phi}_{L,-}^I - \tilde{\Phi}_{L,-}^I \rho^I(t) \tilde{M}_{-}^I(t') \right] C' + \text{H.c.} \]

=0 since \( L > 0 \),

\[ S_{74} = -\hbar^2 \sum_{L=1}^{N-1} \delta_{L,0} \left[ \tilde{\Phi}_{L,-}^I \tilde{M}_{-}^I(t') \rho^I(t) - \tilde{M}_{-}^I(t') \rho^I(t) \tilde{\Phi}_{L,-}^I \right] C + \text{H.c} \]

\[-\hbar^2 \sum_{L=1}^{N-1} \delta_{L,0} \left[ \rho^I(t) \tilde{\Phi}_{L,-}^I \tilde{M}_{-}^I(t) - \tilde{\Phi}_{L,-}^I \rho^I(t) \tilde{M}_{-}^I(t) \right] C' + \text{H.c.} \]

=0 since \( L > 0 \),

\[ S_{81} = -\hbar^2 \sum_{l=1}^{N-1} \delta_{l,0} \left[ \tilde{M}_{1}^I(t) \tilde{\Phi}_{l}^I \rho^I(t) - \tilde{\Phi}_{l}^I \rho^I(t) \tilde{M}_{1}^I(t) \right] C + \text{H.c} \]

\[-\hbar^2 \sum_{l=1}^{N-1} \delta_{l,0} \left[ \rho^I(t) \tilde{\Phi}_{l}^I \tilde{M}_{1}^I(t) - \tilde{\Phi}_{l}^I \rho^I(t) \tilde{M}_{1}^I(t) \right] C' + \text{H.c.} \]

=0 since \( l > 0 \),

\[ S_{82} = -\hbar^2 \sum_{l=1}^{N-1} \delta_{l,0} \left[ \tilde{M}_{1}^I(t) \tilde{\Phi}_{l}^I \rho^I(t) - \tilde{\Phi}_{l}^I \rho^I(t) \tilde{M}_{1}^I(t) \right] C + \text{H.c} \]

\[-\hbar^2 \sum_{l=1}^{N-1} \delta_{l,0} \left[ \rho^I(t) \tilde{\Phi}_{l}^I \tilde{M}_{1}^I(t) - \tilde{\Phi}_{l}^I \rho^I(t) \tilde{M}_{1}^I(t) \right] C' + \text{H.c.} \]

=0 since \( l > 0 \),

\[ S_{83} = \hbar^2 \left[ \tilde{M}_{1}^I(t) \tilde{M}_{1}^I(t') \rho^I(t) - \tilde{M}_{1}^I(t') \rho^I(t) \tilde{M}_{1}^I(t) \right] C + \text{H.c} \]

\[ + \hbar^2 \left[ \rho^I(t) \tilde{M}_{1}^I(t') \tilde{M}_{1}^I(t) - \tilde{M}_{1}^I(t') \rho^I(t) \tilde{M}_{1}^I(t) \right] C' + \text{H.c.}, \]

\[ S_{84} = \hbar^2 \left[ \tilde{M}_{1}^I(t) \tilde{M}_{-}^I(t') \rho^I(t) - \tilde{M}_{-}^I(t') \rho^I(t) \tilde{M}_{1}^I(t) \right] C + \text{H.c} \]

\[ + \hbar^2 \left[ \rho^I(t) \tilde{M}_{-}^I(t') \tilde{M}_{1}^I(t) - \tilde{M}_{-}^I(t') \rho^I(t) \tilde{M}_{1}^I(t) \right] C' + \text{H.c.}. \]

**Calculation of the integrals used in equations (5.65)-(5.68):**

To calculate the integrals in equations (5.65)-(5.68) it is used that

\[ \int_{0}^{\infty} \exp \{ \pm ixy \} dy = \pi \delta(x) \pm iP \left[ \frac{1}{x} \right], \]

where \( P \) indicates the Cauchy principal value. Since the imaginary part of the integral just leads to small energy shifts in the atomic level scheme, it is neglected in the following.
Therefore the integrals are given by:

\[ \int_{0}^{\infty} X(\tau) \, d\tau = \int_{0}^{\infty} X^*(\tau) = \pi h(0) |g_{e_+ e_-}(0)|^2 [2\bar{n}(0) + 1] \]

\[ \int_{0}^{\infty} X(\tau) e^{i\omega g \tau} d\tau = \int_{0}^{\infty} X^*(\tau) e^{-i\omega g \tau} d\tau = \pi h(\omega_g) \bar{n}(\omega_g) + 1 \],

\[ \int_{0}^{\infty} X(\tau) e^{-i\omega g \tau} d\tau = \int_{0}^{\infty} X^*(\tau) e^{i\omega g \tau} d\tau = \pi h(\omega_g) \bar{n}(\omega_g), \]

with \( X \in \{A, B, C\} \).
Publications

Parts of this thesis and other work in context of the thesis have been already published or are in preparation for publication:

1. **Mewes, C.; Unanyan, R.; Fleischhauer, M.:** *Suppression of decoherence caused by atomic collisions in quantum memories with collective atomic excitations*, in preparation.

2. **Mewes, C.; Fleischhauer, M.:** *Quasi decoherence free subspaces in collective quantum memories*, in preparation.

3. **Mewes, C.; Fleischhauer, M.:** *Decoherence in collective quantum memories*, in preparation.


Bibliography


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