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Master Thesis

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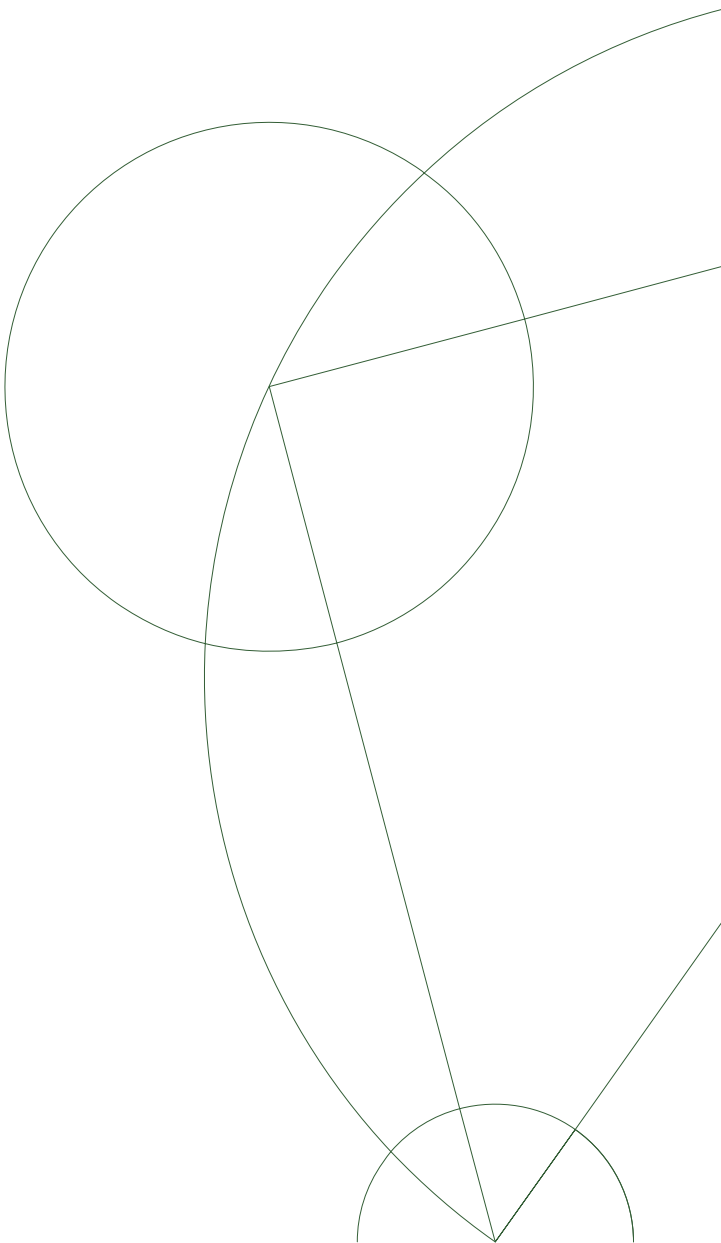
Spontaneous emission in light-atom interactions for atomic ensembles



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Resume

I denne opgave ser vi på Faraday vekselvirkningen mellem et kohært lysfelt og et atomart ensemble. Faraday vekselvirkningen er meget central i kvanteinformation og er ofte anvedt til hukommelesprotokoller. Ved Niels Bohr Institutet har Eugene Polziks gruppe haft succes med at udføre den direkte afbildningsprotol. Indtil videre har der dog ikke være en fyldestgørende beskrivelse af det spontane henfald, som systemet undergår. Det har været denne opgaves formål for første gang at inkludere den fulde niveaustruktur af atomerne og give en fuld beskrivelse af dekohærensen fra den spontane emission. Opgaven er bygget op, på følgende måde: Først betragter vi et enkelt atom og opstillet en effektiv dipol vekselvirkning, der beskriver interaktionen for stærkt ikke-resonant lys. Derefter inkluderer vi koblingen til omgivelserne (som vi tager som vacuum) og ser hvordan den introducerer henfald i vores ideélle system. Dernæst skitserer vi hvordan denne model udvides til et helt ensemble af atomer, der antages ikke at vekselvirke. Til slut ser vi på bevægelsesligningerne for lys- og atom kvadraturerne og ser hvordan den direkte afbildningsprotokol modificeres fra det ideélle tilfælde. Vi finder at for at protokollen skal kunne virke, så skal man have optiske dybder over 10. Vi finder også at $\frac{\pi}{2}$ er den optimale vinkel mellem lys - og atomernes polarisationsretning. For optiske dybder over 20, burde man i princippet kunne opnå en fidelity over 0.75.

Abstract

In this thesis we review the Faraday interaction between a coherent light field and an atomic ensemble. The Faraday interaction is an important tool in the growing field of quantum information, which is widely used to perform quantum memory protocols. At the Niels Bohr Institute, Eugene Polzik's group has had succes with performing the direct mapping protocol based on the Faraday interaction. However so far there has not been a satisfactory description of the spontaneous emission that the system undergoes. In this work we have for the first time included the full level structure of the atoms to get a complete description of the decoherence from spontaneous emission. The thesis is built up as follows: First we consider a single atom and set up an effective dipole interaction that describes the interaction with strongly off-resonant light. After that we include the coupling to the enviroment (which we take as vacuum) and find how it introduces decay to our ideal system. Then we sketch how one extends this interaction to a whole ensemble of atoms, but where the atoms are independent. Finally we look at the equations of motion for light and atom quadratures and see how the direct mapping memory protocol changes from the ideal case. Our results show that for the protocol to be succeful several criteria must be met. First it is important that the detunings be large enough $\sim$ GHz, for the Faraday interaction to be dominant. Secondly one needs optical depths above 10. We also find that $\frac{\pi}{2}$ is the optimal angle between the atomic- and light polarization vector. With optical depths above 20 in principle one should be able to get a fidelity greater than 0.75.

Acknowledgements

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Chapter 1

Introduction

1.1 Quantum information

Since the emergence of quantum mechanics [QM] the technology has experienced a huge boost based on the applications of it. Quantum information [QI] represents another realm which could prove to change our ways of storing and mediating information in a radical way. As is apparent from the name, the theory relies on QM and it seeks to store and manipulate information in states of a quantum system. Loosely said QI consists of two large domains: quantum computation [QC] and quantum cryptography. Things from QM that we usually regard as oddities, such as the uncertainty principle and entanglement, are the very same things that in QI enable us to perform tasks that either would have been impossible or much more time and resource consuming with today's technology. Apart from having a realistic practical value QI also enrichens our understanding of QM and the fundamental laws of nature. It is in topics central to QI that QM has faced the greatest tests, such as Bell inequality, - and passed.
So what is the main difference between classical information and QI? Usually on our computers we store information in bits that can take the values 0 or 1. The QI analog is the qubit, a two-level (spin $\frac{1}{2}$) system:

$$|\psi\rangle \quad = \quad a|0\rangle + b|1\rangle, |a|^2 + |b|^2 = 1. \tag{1.1}$$

QI deals with superpositions of states (or bits) and it produces some essential differences that are worth mentioning. First of all QM tells us that we cannot acces the information in the state without projecting the state to the measured value. More over we cannot clone the state [31] and despite being a superposition, the qubit carries the same amount of information as a classical bit. So what good are these new states? The benefit comes from the processing, which builds on quantum parallelity as Feynman termed it in the 1980's. Simply stated, the principle is that since QM is a linear theory, the action of some operation you want to perform on your state, will work simultaneously on all the substates. That allows a tremendeous speed up that is exploited in QC and we will not go into details, but note that there are many possible utilities, like Shor's factoring algorithm [28] and Grover's search algorithm [8].
Also one can simulate complicated systems, where classical algorithms have proven unable to do so in reasonable (that is polynomial) time. We also mentioned quantum cryptography and it could provide ways to create unconditionally secure keys, meaning that the safety of the protocols would be provided by the laws of quantum mechanics rather than the computational complexity of mathematic problems, as is the case today. There are many possible protocols that could do the job, but the BB84 protocol gives a good description of

the fundamentals [2]. This is not only speculation, the first commercial uses have already appeared on the market, see for instance [1].
Generally one can also say that even though QI as a whole is a very dynamic field, one must say that development for theory and experiment is not parallel. There are still many places where the theoretical foundation has been laid, but practical realizations are absent. One of the biggest unresolved questions is what the best practical way to store the information for longer time periods is. There has been different approaches, one of them coming from the solid state field, where they use electronic states (often in a quantum dot) to keep the information. A recent breakthrough in that field is reported in [12] and it could advance the solid state implementation of QI. But we will look at another alternative, namely the use of alkali atoms as memory holders.

1.2 Atomic ensembles

Initially much attention for implementation of well controlled light-atom interactions was given to systems with few atoms - cavity quantum electrodynamics (Cavity QED). One had one (or few) atoms inside a cavity and the Jaynes-Cummings model interaction [13] could be achieved for single atom - single photon interactions. Despite big advances in Cavity QED, there has been great technical difficulties with making good enough cavities and people started looking for other paths. One of the more succesful of them was the idea that instead of trying to operate with one or few atoms, one could use whole atomic ensembles. In the context of memory, it was realized that one could use the collective spin as the information carrier:

$$J = \sum_{i}^{N} j^{(i)}. \tag{1.2}$$

Here $j^{(i)} = \cdots \otimes 1_{i-1} \otimes j \otimes 1_{i+1} \otimes \cdots$ is the angular momentum operator for the i^{th} atom and through out this thesis we will put $\hbar = c = 1$. The advantage of using many atoms, is that it is much easier to couple light to a whole collection of atoms, rather than a single or few atoms. And besides the collectively enhanced atom-light coupling, it is possible to perform collective operations using simple linear optics. Another aspect is the noise - to be able to perform meaningful operations on the system of interest, it should not be too noise sensitive. And in fact the mean of a big collection of atoms is much more robust with respect to fluctuations than a single or few atoms are - one could compare it to what is known as the law of large numbers in mathematics.
Two possible candidates seem to posses the desired qualities mentioned above. The one we will be looking at, is the room temperature gas in a glass cell, but also much effort is being put into developing systems consisting of cold trapped atoms inside a magneto optical trap [MOT]. In both approaches there are still challenges to be overcome, some are technical, like attenuation in used channels or correcting for the ineffecies of detectors. But also more "fundamental" losses like spontaneuos emission should be adressed. This is what we will opt for in this thesis - to describe how much noise is generated in the system due to spontaneous emission.
But first let us see how exactly we want to use these atomic ensembles to store information. We will assume that the information to be stored is supplied by a coherent light field (laser) and see how the so-called Faraday interaction can give the transfer of information from light to atoms.

Chapter 2

Faraday interaction

In the next chapter we will contruct the off-resonant light-atom interaction for our system, but before starting the full scale analysis we will try to motivate our work by looking at how it can be used to store information. For that we discuss the Faraday interaction which is a simple and yet realistic model of light-atom couplings. We note though that the Faraday interaction is not the only possible interaction for memory purposes. Another very analyzed one is the beamsplitter interaction and it has also proven succeful as in [21]. We will assume that both light and atoms can be described by some quadrature operators $\{X, P\}$ that obey the canonical commutation relations, where i, j labels atoms or light:

$$[X_i, P_j] = i\delta_{ij}, \tag{2.1}$$
$$[X_i, X_j] = [P_i, P_j] = 0. \tag{2.2}$$

As we will see these quadratures are related to the spin of the atoms and polarization states of light. Usually we describe these (vector) quantities using all 3 components, but the idea in this model, is that one of them has a big value and thus may be replaced with it's expectation value. The other two are small and will carry the interesting quantum properties we want to exploit. Precisely how we define these quadratures we will see in chapters 6 and 7, but for now we focus on the Faraday interaction that has the form of the quadrature product:

$$H_F = \kappa P_A P_L. \tag{2.3}$$

Here κ is the interaction strength - a parameter that tells how strongly the light couples to atoms. To get something useful out of the interaction, we want want this quantity to be big enough, which is typically of order 1.
Now what is so special about this interaction? First we note that in our thesis will work in the Heisenberg picture, where the operators A are time dependent and subject to a Hamiltonian H the evolution is governed by:

$$\dot{A} = i[H, A] + \frac{\partial A}{\partial t}, \tag{2.4}$$

where the last term comes from the possible explicit time dependence of A. So according to Heisenberg's equation of motion, together with relations (2.1+2), we get that the light quadratures change as:

$$\dot{X_L} = \kappa P_A, \tag{2.5}$$
$$\dot{P_L} = 0. \tag{2.6}$$

By the same token we have for the atomic variables:

$$\dot{X}_A = \kappa P_L, \tag{2.7}$$
$$\dot{P}_A = 0. \tag{2.8}$$

These first order differential equations for light and atoms couple to each other in a simple way and one quickly finds the solution expressed in terms of input and output:

$$X_L^{out} = X_L^{in} + \kappa P_A^{in}, \tag{2.9}$$
$$P_L^{out} = P_L^{in}, \tag{2.10}$$
$$X_A^{out} = X_A^{in} + \kappa P_L^{in}, \tag{2.11}$$
$$P_A^{out} = P_A^{in}. \tag{2.12}$$

We see that in the interaction the quadrature momenta P_A and P_L do not change and are mapped onto resp. X_L and X_A. Given that the interaction strength κ is big enough we can therefore measure P_A^{in} by measuring X_L^{out}. That way we have a performed a so-called quantum non-demolition measurement - the integrity of the system is preserved after the measurement and it relied on the fact that P_A was not altered in the interaction.
The QND structure of the interaction gives the possibility for creating a memory protocol based on this quadrature formalism. We will consider the direct mapping protocol, as desribed in [14]. After letting the light interact with the atoms through H_F, we measure the value of X_L^{out} and subtract it from the atomic P_A^{out} with a gain g, such that we end up with:

$$X_A^{out} = X_A^{in} + \kappa P_L^{in}, \tag{2.13}$$
$$P_A'^{out} = P_A^{out} - gX_L^{out} = P_A^{in}(1 - \kappa g) - gX_L^{in}. \tag{2.14}$$

If $\kappa = g = 1$ and X_A^{in} has with zero mean, we have thus achieved a faithful storage of both light variables in the atoms:

$$\langle X_A^{out} \rangle = \langle P_L^{in} \rangle, \tag{2.15}$$
$$\langle P_A^{out} \rangle = -\langle X_L^{in} \rangle. \tag{2.16}$$

So how well does this memory protocol work? A number used to describe the efficiency of a memory protocol is the fidelity. It tells how well the state recorded in the atomic quadratures represents the actual state supplied by the light and is simply the overlap of the two. We can calculate the fidelity for this protol using the formula from [10]:

$$\mathcal{F} = (\frac{1}{2} + \Delta X_A^{2,out})^{-\frac{1}{2}} \times (\frac{1}{2} + \Delta P_A^{2,out})^{-\frac{1}{2}}. \tag{2.17}$$

This formula can be used for coherent light input states that have symmetric noise in the quadratures, but does not apply for squeezed states of light that we will mention later. From (2.13+14) we can calculate the variances on the atomic output quadratures:

$$\Delta X_A^{2,out} = \Delta X_A^{2,in} + \Delta P_L^{2,in}, \tag{2.18}$$
$$\Delta P'^{2,out} = \Delta X_L^{2,in}. \tag{2.19}$$

We see that even though X_A^{in} can have a mean zero value, it still contributes to the variance of the output X_A^{out}. Assuming that both light and atoms exhibibit minimum uncertainty, such that the variance of all quadratures is $\frac{1}{2}$, one obtains from (2.17) that the the maximal

fidelity for this protocol is $\sqrt{\frac{2}{3}} \simeq 82\%$. However if we squeeze the input state X_A^{in} such that $\Delta X_A^{2,in} \to 0$, the fidelity approaches 100%. In our work we will usually not include this possibility of squeezing the input atomic state, because it gives some other problems and therefore we usually assume that $\Delta X_A^{2,in} = \Delta P_A^{2,in} = \frac{1}{2}$.

Historically the idea of using a QND Hamiltonian for manipulating quantum states was first made by (among others) [26] in 1989. Since then many experiments have been performed by several groups, confirming this approach. One of them being Eugene Polzik's Quantop at Niels Bohr Institute and in [27] they reported an observed fidelity for the described protocol around 0.70. In this context we want to mention that for our protocol to be succeful, we demand a fidelity that should be higher than the best fidelity that can be achieved classically. If we assume that our light input states could come from the whole (X_L, P_L) phase space, then this classical fidelity is $\frac{1}{2}$. If we only had vacuum or states displaced by small amount from vacuum as input, we could get a high fidelity simply by using vacuum states as memory - we would not need any protocols. So looking at the protocol we think that the states to be stored are not just vacuum, but rather vacuum states displaced by some unspecified amount - coherent states.
In all of this we have assumed our system of atoms to be isolated from everything else. This is of course unrealistic, our atoms will interact with the surroundings, which will give rise to spontaneuos emission. It is already known that even with this complication, the protocols can still be used for memory, but so far there has not been a precise theoretical estimate of how much decay and noise is introduced and this we wish to quantify in this thesis.

Chapter 3

Interaction Hamiltonian

In this chapter we will find the interaction Hamiltonian from which we later will deduce the equations om motion. Our description is composed of three parts, the purely atomic part, the purely photonic part and the interaction part which describes the interaction of atoms with our light field. We will consider these three parts separately first and introduce the relevant notions before going to the interaction part. In most of our analysis we will only consider a single atom, later on we will see how we should approach the situation of a collection of atoms - the atomic ensemble.

3.1 Light

We will quantize the light field in a standard way (for a more detailed description we refer to App.A.1), using the form of the electric field:

$$\mathbf{E}(\mathbf{r},t)=\sum_{k\sigma}\boldsymbol{\epsilon}_{k\sigma}\sqrt{\frac{\omega_k}{2\epsilon_0 V}}(a_{k\sigma}(t)e^{i\mathbf{k}\cdot\mathbf{r}}+a^{\dagger}_{k\sigma}(t)e^{-i\mathbf{k}\cdot\mathbf{r}})=\mathbf{E}^{(+)}(\mathbf{r},t)+\mathbf{E}^{(-)}(\mathbf{r},t). \tag{3.1}$$

We have written the eletric field as a sum of positive- and negative frequency components representing respectively the destruction- and generation of field excitations. The Hamiltonian for the radiation field itself is:

$$H_L \;=\; \sum_{k\sigma}\omega_k a^{\dagger}_{k\sigma}a_{k\sigma}. \tag{3.2}$$

Which simply counts the number of photons in different states and assigns the corresponding energy. For the coherent field we shine onto the atoms, we will assume the electric field to be centralized about the carrier frequency ω_0 and to have a flat profile with area A, with a strongly polarized x component and a weak y component. So we use $1/\sqrt{A}$ as the transverse profile and have in the paraxial approximation with L as the length of the quantization volume, the electric field to be:

$$\begin{aligned}\mathbf{E}(\mathbf{r},t) &= \sqrt{\frac{\omega_0}{2\epsilon_0 A}}\sum_{k\sigma}\boldsymbol{\epsilon}_{\sigma}\frac{1}{\sqrt{L}}(a_{k\sigma}(t)e^{ikz}+a^{\dagger}_{k\sigma}(t)e^{-ikz})\\ &= \sqrt{\frac{\omega_0}{2\epsilon_0 A}}\sum_{\sigma}\boldsymbol{\epsilon}_{\sigma}(a_{\sigma}(z,t)+a^{\dagger}_{\sigma}(z,t)).\end{aligned} \tag{3.3}$$

We have defined the position varying operator $a(z,t)$, such that $n_\sigma(z,t)=a^{\dagger}_{\sigma}(z,t)a_\sigma(z,t)$ gives the flux of photons with polarization σ at position z and time t. Later it will be

useful to describe the fields by using Stokes operators rather than the $a, a^\dagger$-operators. For our laser beam which is travelling in the z direction, we describe the polarization by Stokes operators (also decribed in App.A.2):

$$S_x(z,t) = \frac{1}{2}(n_x(z,t) - n_y(z,t)), \tag{3.4}$$

$$S_y(z,t) = \frac{1}{2}(n_{+45^\circ}(z,t) - n_{-45^\circ}(z,t)), \tag{3.5}$$

$$S_z(z,t) = \frac{1}{2}(n_{\sigma^+}(z,t) - n_{\sigma^-}(z,t)). \tag{3.6}$$

S_x is the difference between the flux of photons being x-polarized and y polarized, S_y is difference between photons having $\pm 45^\circ$ polarization and S_z is the difference between right circularly and left circularly polarized photon flux. Furthermore we will also need the total flux:

$$\phi(z,t) = n_x(z,t) + n_y(z,t). \tag{3.7}$$

Because our light is strongly polarized along x, we can treat S_x as the c-number $\langle S_x \rangle$, while the smaller S_y and S_z retain their quantum properties. Also we mention that the Stokes operators obey the angular momentum like relation:

$$[S_i, S_j] = i \sum_k \epsilon_{ijk} S_k. \tag{3.8}$$

As with spin we cannot know all components of the Stokes vector simultaneously. And because of this commutation relation, they fulfill the uncertainty relation:

$$\Delta S_y^2 \Delta S_z^2 \geq \frac{S_x^2}{4}. \tag{3.9}$$

For a coherent state like the light our laser produces, we have (almost) $\Delta S_y^2 = \Delta S_z^2 = \frac{S_x}{2}$ and we say that the light in this case is shot noise limited. These coherent states can be seen as displaced vacuum states. There are also states that fulfill the equality, but have an uneven distribution of variances and these are called squeezed states and have no classical analog. They will not be used in this work, although they are also very interesting in this context.

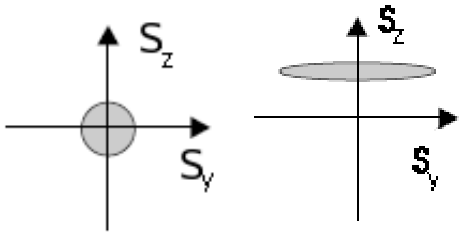

Figure 3.1: Left: Coherent light state with vanishing S_y,S_z and $\Delta S_y^2 = \Delta S_z^2$ Right: Displaced squeezed light state with $\Delta S_y^2 > \Delta S_z^2$.

3.2 Atoms

The atom we will work with in our model is cesium. Cesium is an alkali metal with a single electron in the outer shell. We know that due to fine structure there will be a coupling

of orbital momentum L of the outer electron with its spin S, giving a total of $J = L + S$ where $|J|$ must satisfy the triangle inequality $|L - S| < J < L + S$. Cesium has a ground state with $L = 0$ and $S = \frac{1}{2}$, giving $J = \frac{1}{2}$. The first exited state has $L = 1$ meaning that $J = \frac{1}{2}$ or $J = \frac{3}{2}$. It means that these states will have a shift resulting in two split components and the transitions $6^2S_{\frac{1}{2}} \rightarrow 6^2P_{\frac{1}{2}}$ and $6^2S_{\frac{1}{2}} \rightarrow 6^2P_{\frac{3}{2}}$ from the ground state to this fine structure doublet, we call resp. the D1- and D2 line.
Additionaly we have a hyperfine structure due to the coupling of J to nuclear spin I giving a total angular momentum $F = I + J$ with the similar condition $|J - I| < F < J + I$. Cesium has $I = \frac{7}{2}$ so we have two ground states with $F = 3$ or $F = 4$. For the D1-line the total spin of the exited state can be 3 or 4, while for the D2-line F can be $2, 3, 4, 5$. Because the splitting between the two D-lines is big enough ($\sim$THz), it is possible to lock a laser on the transition of interest. We will be solely investigating the D2 line, but much of the analysis can be applied for the D1 line too.

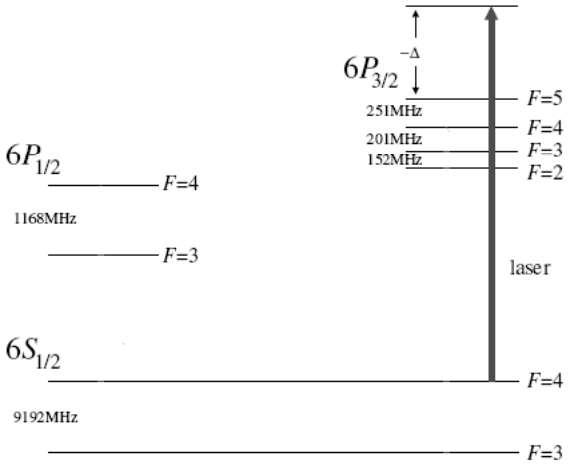

Figure 3.2: D1 and D2 lines of cesium

Now as in [29], we define the projections onto the ground- and exited spin state manifolds(We will use $|F, m\rangle$ and $|\tilde{F}, \tilde{m}\rangle$ to represent ground states and $|F', m'\rangle$ to represent excited states.):

$$P_F = \sum_m |F, m\rangle\langle F, m|, P_{F'} = \sum_{m'} |F', m'\rangle\langle F', m'|, \tag{3.10}$$

$$P_g = \sum_F P_F, P_e = \sum_{F'} P_{F'}, P_g + P_e = 1. \tag{3.11}$$

This projections will be very used for our treatment of the operators and if we specifically apply it to the dipole-operator $\mathbf{d}$ we get (Note that we do not get any contributions from the terms $P_g\mathbf{d}P_g, P_e\mathbf{d}P_e$ since $\mathbf{d}$ is a parity odd operator.):

$$\mathbf{d} = (P_g + P_e)\mathbf{d}(P_g + P_e) = P_e\mathbf{d}P_g + P_g\mathbf{d}P_e = \mathbf{d}^{(+)} + \mathbf{d}^{(-)}. \tag{3.12}$$

The physical intepretation of $\mathbf{d}^{(+)}$ can be seen from it's action on a ground state specified by total- and spin magnetic moment $|F, m\rangle$. It gives an up transition to exited states $|F', m'\rangle$ with amplitudes given by the matrix element of the dipole operator: $\langle F', m'|\mathbf{d}|F, m\rangle$. For exited states $\mathbf{d}^{(+)}$ gives the null ket. In the same way one can see that $\mathbf{d}^{(-)} = (\mathbf{d}^{(+)})^\dagger$ is a lowering from the exited states to the ground states. In the following we will go into

the rotating frame with respect to the laser frequency ω_0, so the energy of the atom is described by the Hamiltonian:

$$H_A = \sum_{F'} \Delta_{F'} P_{F'}. \tag{3.13}$$

Where $\Delta_{F'}$ is the detuning of the exited stated from the laser frequency. For the cesium atom we saw earlier that we have two stable ground states with resp. $F = 3$ and $F = 4$, where we will choose the $F = 4$ as reference:

$$H_0 = -\Delta_{34} P_3. \tag{3.14}$$

Here $\Delta_{34} = 9.192631770$ GHz[1] is the energy difference between the two ground states. As mentioned we will describe the atoms in terms of their spin operators (for a quick review of angular momentum we refer to App.B.1) that have:

$$[j_i, j_j] = i \sum_k \epsilon_{ijk} j_k. \tag{3.15}$$

The same holds for the collective spin as defined in (1.2), that because of Heisenberg's uncertainty relation has:

$$\Delta J_y^2 \Delta J_z^2 \geq \frac{J_x^2}{4}. \tag{3.16}$$

Using the technique of optical pumping [11], it can be arranged such that to a high degree of accuracy, all the atoms have a spin pointing in the same direction, e.g. x, such that $J_x = N_A F$ and $\langle J_y \rangle = \langle J_z \rangle = 0$. In this case we say that the atoms are in a coherent spin state [CSS] and the atoms are independent, so the wavefunction for the system factorizes: $|\psi\rangle = \bigotimes_{n=1}^{N} |\psi\rangle^{(n)}$. The CSS is a minimum uncertainty state: $\Delta J_y^2 = \Delta J_z^2 = \frac{J_x}{2}$ and as with light it is possible to squeeze the uncertainties, such that the equation still holds, but the quarature variances are different. In the experiments they typically have $N_A \sim 10^{12}$ giving an angular uncertainty of order 10^{-6}.

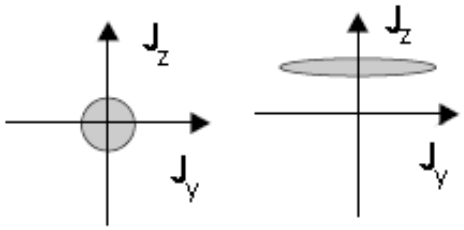

Figure 3.3: Left:Coherent spin state with $\Delta J_y^2 = \Delta J_z^2$, Right: Displaced squeezed state with $\Delta J_y^2 > \Delta J_z^2$.

When we talk about the ensemble polarized along a direction, e.g. x, it means for the collective spin J_x we have $p = \frac{J_x}{NF}$ close to 1, while $p = 0$ describes a completely unpolarized ensemble. In experiments described in [15] using optical pumping they managed to create polarized ensembles with p lying within 10% from the desired value 1.

[1] The transition between the two ground state levels defines the second, which explains the exact value of the splitting.

3.3 Interaction

In this section we find the interaction Hamiltonian. We will work in the dipole approximation, where the interaction between light and atoms is of the form $H_{int} = -\mathbf{d}\cdot\mathbf{E}$. This form works as long as the spatial extent of the atom is much smaller than the wavelength of the light. In the previous sections we wrote the electric field and the dipole operator as a sum of positive and negative components. This we can insert into the expression for H_{int}, and we will affect the rotating wave approximation [RWA] that amounts to neglecting the fast oscillating terms $\mathbf{E}^{(+)}\mathbf{d}^{(+)}, \mathbf{E}^{(-)}\mathbf{d}^{(-)}$ that describe the strongly forbidden processes where e.g. an atom is exited by emitting a photon as for the $\mathbf{E}^{(-)}\mathbf{d}^{(-)}$ term. That way we get:

$$\begin{aligned} H_{int} &= -\mathbf{d}\cdot\mathbf{E} = -(\mathbf{d}^{(+)} + \mathbf{d}^{(-)})(\mathbf{E}^{(+)} + \mathbf{E}^{(-)}) \\ &\simeq -\mathbf{d}^{(+)}\mathbf{E}^{(-)} - \mathbf{d}^{(-)}\mathbf{E}^{(+)} \\ &= -\sum_{F,F'} P_F \mathbf{d} P_{F'} \mathbf{E}^{(-)} + P_{F'}\mathbf{d}P_F\mathbf{E}^{(+)} \\ &= -\sum_{F,F'}\sum_{m,m'} \langle F,m|\mathbf{d}|F',m'\rangle \sigma_{F,m;F',m'}\mathbf{E}^{(-)} + \langle F',m'|\mathbf{d}|F,m\rangle\sigma_{F',m';F,m}\mathbf{E}^{(+)}. \end{aligned} \quad (3.17)$$

We have ended up with a Hamilton that describes the proces where an atom creates (annihilates) a photon as it decays (gets exited) from an exited (ground) state to a ground (excited) state. The matrix element $\langle F,m|\mathbf{d}|F',m'\rangle$ gives the coupling strength of this transition. We want to obtain a Hamiltonian that only involves the ground states and to do that we first form the EOM for the transition matrix, where we use the transition matrix commutation relations from App.D.2 to obtain:

$$\begin{aligned} \frac{d}{dt}\sigma_{F,m;F',m'} &= i[H_{int} + H_A, \sigma_{F,m;F',m'}] \\ &= i(\sum_{\tilde{F},\tilde{m}} \langle F',m'|\mathbf{d}|\tilde{F},\tilde{m}\rangle\mathbf{E}^{(+)}\sigma_{F,m;\tilde{F},\tilde{m}} - \Delta_{F'}\sigma_{F,m;F',m'}). \end{aligned} \quad (3.18)$$

Now we are going to perform the adiabatic elimination. We assume the laser to be far detuned from resonance, such that we can ignore the population in the exited states, more precisely we demand that the saturation parameter satisfies:

$$s = \phi(\frac{\Omega}{\Delta})^2 \ll 1, \quad (3.19)$$

which can be achieved by making the detuning large enough (Ω is the single photon Rabi frequency and ϕ is the laser flux). As a result each atomic dipole will follow the applied field adiabatically and putting the derivative to zero we are left with[2]:

$$\sigma_{F,m;F',m'} = \frac{1}{\Delta_{F'}}\sum_{\tilde{F},\tilde{m}} \langle F',m'|\mathbf{d}|\tilde{F},\tilde{m}\rangle\mathbf{E}^{(+)}\sigma_{F,m;\tilde{F},\tilde{m}}. \quad (3.20)$$

[2]We see that the dispersive part of the interaction goes as $1/\Delta$, while (as we also will see later) the absorptive goes as $1/\Delta^2$. Also we will use the convention that blue detuning is taken as negative.

This expression for σ we can insert into the Hamiltonian (3.17) to obtain:

$$\begin{aligned} H_{int} &= -\sum_{F,\tilde{F},F'}\sum_{m,\tilde{m},m'}\langle F,m|\mathbf{d}|F',m'\rangle\frac{1}{\Delta_{F'}}\langle F',m'|\mathbf{d}|\tilde{F},\tilde{m}\rangle\sigma_{F,m;\tilde{F},\tilde{m}}\mathbf{E}^{(-)}\mathbf{E}^{(+)}+\text{h.c.} \\ &= -\mathbf{E}^{(-)}(\sum_{F,\tilde{F},F'}\sum_{m,\tilde{m},m'}|\tilde{F},\tilde{m}\rangle\frac{\langle\tilde{F},\tilde{m}|\mathbf{d}|F',m'\rangle\langle F',m'|\mathbf{d}|F,m\rangle}{\Delta_{F'}}\langle F,m|)\mathbf{E}^{(+)}+\text{h.c.} \\ &= -\sum_{F'}\mathbf{E}^{(-)}\frac{P_g\mathbf{d}P_e\mathbf{d}P_g}{\Delta_{F'}}\mathbf{E}^{(+)}+\text{h.c.}=\mathbf{E}^{(-)}\alpha\mathbf{E}^{(+)}+\text{h.c.}=2\mathbf{E}^{(-)}\alpha\mathbf{E}^{(+)}. \end{aligned} \tag{3.21}$$

Here we have introduced the atomic polarizability α:

$$\alpha = -\sum_{F'}\frac{P_g\mathbf{d}P_{F'}\mathbf{d}P_g}{\Delta_{F'}}, \tag{3.22}$$

which we will analyze in the following. But first we return to the atomic Hamiltonian H_A that we should add to our H_{int} to get the full system. For H_A we insert the found expression (3.20) for σ (here $|F_0,m_0\rangle$ is some arbitrary ground state that we just insert to use what we have found and it drops out eventually):

$$\begin{aligned} H_A &= \sum_{F',m'}\Delta_{F'}\sigma_{F',m';F',m'}=\sum_{F',m'}\Delta_{F'}\sigma_{F',m';F_0,m_0}\sigma_{F_0,m_0;F',m'} \\ &= \sum_{F',m'}\Delta_{F'}(\frac{1}{\Delta_{F'}})^2\sum_{F,\tilde{F}}\sum_{m,\tilde{m}}\langle\tilde{F},\tilde{m}|\mathbf{d}|F',m'\rangle\langle F',m'|\mathbf{d}|F,m\rangle\mathbf{E}^{(-)} \\ &\times\sigma_{\tilde{F},\tilde{m};F_0,m_0}\sigma_{F_0,m_0;F,m}\mathbf{E}^{(+)}) \\ &= \mathbf{E}^{(-)}(\sum_{F,\tilde{F},F'}\sum_{m,\tilde{m},m'}\frac{\langle\tilde{F},\tilde{m}|\mathbf{d}|F',m'\rangle\langle F',m'|\mathbf{d}|F,m\rangle}{\Delta_{F'}}\sigma_{\tilde{F},\tilde{m};F,m})\mathbf{E}^{(+)} \\ &= -\mathbf{E}^{(-)}\alpha\mathbf{E}^{(+)}. \end{aligned} \tag{3.23, 3.24}$$

So when we add H_A to the interaction Hamiltonian H_{int} we get rid of the factor 2 in (3.21):

$$H_{\text{int}}^{\text{eff}} = H_{\text{int}}+H_A=\mathbf{E}^{(-)}\alpha\mathbf{E}^{(+)}\equiv H_{\text{int}}. \tag{3.25}$$

With our assumption of having light far detuned from resonance we have performed the adiabatic elinination and now have a Hamiltonian that couples ground states to ground states. With this procedure the Hamiltonian works only for timescales longer than $\frac{1}{\Delta_{F'}}\sim ns$, which is fulfilled for the actual experiments, where the pulses typically have a duration of order μs.

So far the polarizability α in (3.22) is just some complicated formal expression. We want to examine it closer and to do that we will use the spherical basis[3]:

$$H_{\text{int}} = \mathbf{E}^{(-)}\alpha\mathbf{E}^{(+)}=\sum_{q,q'}E_{q'}^{(-)}\mathbf{e}_{q'}^{*}\alpha\mathbf{e}_qE_q^{(+)}. \tag{3.26}$$

The elements in the polarizability matrix can be simplified by using conservation of momentum to get rid of the irrelevant m-sums. Since we have two ground state spin: F and $\tilde{F}$ and we start in the state with F, we will make the replacement $\Delta_{F'}\to\Delta_{F'F}$ to

[3]This basis is described in App.D.1.

remind ourselves that it is the detuning from the frequency corresponding to the transition $|F,m\rangle \to |F',m'\rangle$ we have in the expressions:

$$\begin{aligned}\alpha_{q,q'} = \mathbf{e}^*_{q'}\cdot\alpha\cdot\mathbf{e}_q &= (-1)^{1+q'}\sum_{F\tilde{F}F'}\frac{P_{\tilde{F}}d_{-q'}P_{F'}d_qP_F}{\Delta_{F'F}} \\ &= (-1)^{1+q'}\sum_{F,\tilde{F},F'}\sum_m \frac{\langle\tilde{F},m+q-q'|d_{-q'}|F',m+q\rangle\langle F',m+q|d_q|F,m\rangle}{\Delta_{F'F}}\sigma_{\tilde{F},m+q-q';F,m}.\end{aligned} \tag{3.27}$$

For the matrix elements $\langle F',m'|d_q|F,m\rangle$ we will use the Wigner-Eckart theorem, which is a very powerful tool for evaluting spherical tensor operators on the basis of angular momentum eigenstates. The details are explained in App.D.3 and one can also look in [5]. As a result the polarizability tensor for given $F,\tilde{F}$, may be written as (In the following we will write the Clebsch-Gordan coefficients $\langle F,m|F',m';1,q\rangle$ as $c^{F,m}_{F',m'}$ etc.):

$$\alpha^{F,\tilde{F}}_{q,q'} = (-1)^{1+F+\tilde{F}}\frac{\alpha_0}{2J+1}\sum_{F'}\frac{f_{F,\tilde{F},F'}}{\Delta_{F'F}}\sum_m c^{F',m+q}_{\tilde{F},m+q-q'}c^{F',m+q}_{F,m}|\tilde{F},m+q-q'\rangle\langle F,m|. \tag{3.28}$$

Where the characteristic polarizability constant for the $|J\rangle \to |J'\rangle$ transition is defined as:

$$\alpha_0 \;=\; |\langle J'||\mathbf{d}||J\rangle|^2\frac{2J+1}{2J'+1}, \tag{3.29}$$

and we have also introduced the generalized relative oscillator strength:

$$f_{F,\tilde{F},F'} = (2J'+1)\sqrt{(2F+1)(2\tilde{F}+1)}\begin{Bmatrix} F & F' & 1\\ J' & J & I\end{Bmatrix}\begin{Bmatrix}\tilde{F} & F' & 1\\ J' & J & I\end{Bmatrix}, \tag{3.30}$$

which for $F=\tilde{F}$ has the sum rule $\sum_F f_{FF'}=1$.
It is easy to understand the physical meaning of the Hamiltonian. We look at processes where an atom initially in state $|F,m\rangle$ absorbs a photon with polarization q and gets exited to the virtual (to stipulate that we are dealing with off resonant processes) state $|F',m'\rangle$. It is accompanied by an emission of a photon with polarization q' after which the atom ends up in the state $|\tilde{F},m+q-q'\rangle$. The strength of the proces is essentially determined by the Clebsch-Gordan coefficients $c^{F',m+q}_{\tilde{F},m+q-q'}c^{F',m+q}_{F,m}$.
Since the atom can end in a different spin state it means that there can be an exchange of energy between the atoms and the light field. For $F=\tilde{F}$ we are looking at an elastic scattering event and will call the interaction coherent.
So far the formalism has been very general, but for the cesium atom we are dealing with, we know from section 3.1 that we just have to consider the two ground states that have $F=3$ or $F=4$. In App.C.1 we have for these choices of F and $\tilde{F}$, calculated the coefficients:

$$C^{F,\tilde{F}}_{q,q'}(\Delta,m) \;=\; (-1)^{F+\tilde{F}}\sum_{F'}\frac{f_{F,\tilde{F},F'}}{(2J'+1)\Delta_{F,F'}}c^{F',m+q}_{\tilde{F},m+q-q'}c^{F',m+q}_{F,m}, \tag{3.31}$$

such that (3.28) can be written as:

$$\alpha^{F,\tilde{F}}_{q,q'} \;=\; -D_0^2\sum_m C^{F,\tilde{F}}_{q,q'}(m)\sigma_{\tilde{F},m+q-q';F,m}. \tag{3.32}$$

Choosing a specific F' as reference, we will take the detuning for this transition to F' outside, and write the polarizability as:

$$\alpha^{F,\tilde{F}}_{q,q'} = -\frac{D_0^2}{\Delta}\sum_m C^{F,\tilde{F}}_{q,q'}(m)\sigma_{\tilde{F},m+q-q';F,m}. \tag{3.33}$$

As is shown in App.C.1 all these coefficients $C^{F,\tilde{F}}_{q,q'}(m)$ can be expressed through some simple expressions involving m and 3 numbers a_0, a_1 and a_2 for $F = \tilde{F}$. For $F \neq \tilde{F}$ we find that besides the m-depedence the coefficients only need two numbers b_1 and b_2. For $F = \tilde{F} = 3$ we get (with Δ being the detuning from the transition to $F' = 2$) the coefficients together with their limit of $|\Delta| \to \infty$:

$$a_0 = \frac{1}{672}(24 + \frac{63}{1+\frac{\Delta_{23}}{\Delta}} + \frac{25}{1+\frac{\Delta_{24}}{\Delta}}) \to \frac{1}{6}, \tag{3.34}$$

$$a_1 = \frac{1}{2688}(-80 - \frac{21}{1+\frac{\Delta_{23}}{\Delta}} + \frac{45}{1+\frac{\Delta_{24}}{\Delta}}) \to -\frac{1}{48}, \tag{3.35}$$

$$a_2 = \frac{1}{2688}(16 - \frac{21}{1+\frac{\Delta_{23}}{\Delta}} + \frac{5}{1+\frac{\Delta_{24}}{\Delta}}) \to 0. \tag{3.36}$$

And for $F = \tilde{F} = 4$ we find:

$$a_0 = \frac{1}{96}(8 + \frac{7}{1-\frac{\Delta_{45}}{\Delta}} + \frac{1}{1-\frac{\Delta_{35}}{\Delta}}) \to \frac{1}{6}, \tag{3.37}$$

$$a_1 = \frac{1}{5760}(176 - \frac{21}{1-\frac{\Delta_{45}}{\Delta}} - \frac{35}{1-\frac{\Delta_{35}}{\Delta}}) \to \frac{1}{48}, \tag{3.38}$$

$$a_2 = \frac{1}{5760}(16 - \frac{21}{1-\frac{\Delta_{45}}{\Delta}} + \frac{5}{1-\frac{\Delta_{35}}{\Delta}}) \to 0. \tag{3.39}$$

We see that that the sign of the a_1 coefficient for $\Delta \to \infty$ is opposite for $F = 3$ and $F = 4$ and they both have $a_2 \to 0$. Now we can construct the coherent Hamiltonian for $F = \tilde{F} = 3, 4$ by evaluting the coefficients $C^{F,\tilde{F}}_{q,q'}(m)$ in terms of a-coefficients as in App.C.1 and applying the sum rules from App.B.1 (a more detailed derivation is found in App.D.4):

$$\begin{aligned}
H_{\rm coh} &= -\frac{D_0^2}{\Delta}\sum_m((a_0 + a_1 m + a_2 m^2)E^{(-)}_+ E^{(+)}_+ + (a_0 - a_1 m + a_2 m^2)E^{(-)}_- E^{(+)}_- \\
&+ (a_0 + F(F+1)a_2 - 2a_2 m^2)E^{(-)}_0 E^{(+)}_0)\sigma_{m,m} \\
&+ \sqrt{\frac{(F+m)(F+1-m)}{2}}(a_2 - a_1 - 2a_2 m)(E^{(-)}_+ E^{(+)}_0 \sigma_{m-1,m} + h.c.) \\
&+ \sqrt{\frac{(F-m)(F+1+m)}{2}}((a_2 - a_1 + 2a_2 m)(E^{(-)}_- E^{(+)}_0 \sigma_{m+1,m} + h.c.)) \\
&+ a_2\sqrt{(F+m)(F+1+m)(F-m)(F+1-m)}(E^{(-)}_+ E^{(+)}_- \sigma_{m-1,m+1} + h.c.) \\
&= -\frac{D_0^2}{\Delta}((a_0 + \frac{1}{3}a_2 \mathbf{j}^2)\mathbf{E}^{(-)}\mathbf{E}^{(+)} + i a_1 \mathbf{E}^{(-)}\cdot\mathbf{j}\times\mathbf{E}^{(+)} \\
&- 2a_2\sum_{i,j} E^{(-)}_i(\frac{j_i j_j + j_j j_i}{2} - \delta_{ij}\frac{\mathbf{j}^2}{3})E^{(+)}_j) \\
&= H^{(0)}_{FF} + H^{(1)}_{FF} + H^{(2)}_{FF}
\end{aligned} \tag{3.40}$$

From the expressions above we see that the Hamiltonian contains parts that changes the atomic angular momentum (m), by 0,1 and 2 and this change is counterbalanced by a polarization change of the light, such that the total angular momentum along z is conserved. We need need three parameters a_0,a_1 and a_2 for this interaction, which reflects that the Hamiltonian is a sum of tensors of rank 0,1 and 2. Essentially what this Hamiltonian describes is the Stark shift experienced by the atoms due to the presence of the light field. For the case with $F \neq \tilde{F}$ we find that to set up the Hamiltonian we need the two b coefficients:

$$b_1 = \frac{1}{384}\left(\frac{3}{1-\frac{\Delta_{35}}{\Delta}} + \frac{5}{1-\frac{\Delta_{45}}{\Delta}}\right) \rightarrow \frac{1}{48} \tag{3.41}$$

$$b_2 = \frac{1}{384}\left(\frac{-1}{1-\frac{\Delta_{35}}{\Delta}} + \frac{1}{1-\frac{\Delta_{45}}{\Delta}}\right) \rightarrow 0 \tag{3.42}$$

But this time we cannot make use of the sum rules to reach expressions involving angular momentum, because σ connects different F. Therefor we simply state the Hamiltonian:

$$\begin{aligned}
H_{34} = &-\frac{D_0^2}{\Delta}\sum_m \sqrt{(4+m)(4-m)}\{b_1(E_+^{(-)}E_+^{(+)} - E_-^{(-)}E_-^{(+)}) \\
&+ b_2 m(\mathbf{E}^{(-)}\cdot\mathbf{E}^{(+)} - 3E_0^{(-)}E_0^{(+)})\}(\sigma_{3,m;4,m} + \sigma_{4,m;3,m}) \\
&+ \sqrt{\frac{(4+m)(5+m)}{2}}\{b_1[(E_+^{(-)}E_0^{(+)} + E_0^{(-)}E_-^{(+)})\sigma_{3,m;4,m+1} + h.c.] \\
&+ (2m-3)b_2[(E_+^{(-)}E_0^{(+)} - E_0^{(-)}E_-^{(+)})\sigma_{3,m;4,m+1} + h.c.]\} \\
&- \sqrt{\frac{(4-m)(5-m)}{2}}\{b_1[(E_-^{(-)}E_0^{(+)} + E_0^{(-)}E_+^{(+)})\sigma_{3,m;4,m-1} + h.c.] \\
&+ (2m+3)b_2[(E_0^{(-)}E_+^{(+)} - E_-^{(-)}E_0^{(+)})\sigma_{3,m;4,m-1} + h.c.]\} \\
&+ b_2\{\sqrt{(3-m)(4-m)(5-m)(4+m)}(E_-^{(-)}E_+^{(+)}\sigma_{3,m+1;4,m-1} + h.c.) \\
&- \sqrt{(3+m)(4+m)(5+m)(4-m)}(E_+^{(-)}E_-^{(+)}\sigma_{3,m-1;4,m+1} + h.c.)\} \\
&= H_{34}^{(1)} + H_{34}^{(2)}
\end{aligned} \tag{3.43}$$

For the case where we go from 3 to 4 or vice versa we only needed two parameters, b_1 and b_2, to describe the interaction. The reason is that we for all processes have a change in the spin state and therefor have no scatic part - a tensor of rank 0 and therefor no b_0 term. Physically the Hamiltonian describes inelastic scattering which gives so-called Raman transitions.
We have plotted the found a and b coefficients and we see that even though they are all different per se, a_1 and b_1 have the same limit value as well as a_2 and b_2.

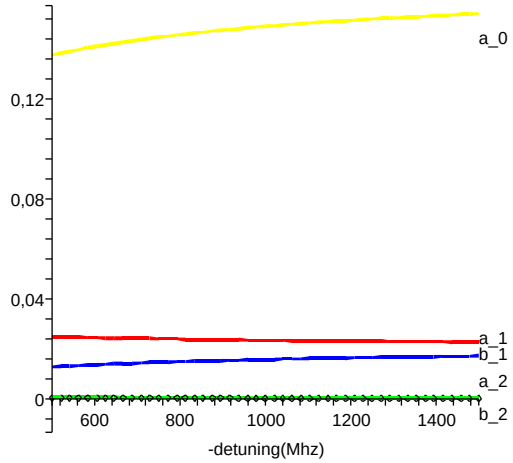

Figure 3.4: a_0, a_1, a_2, b_1 and b_2(black dots) as a function of $-\Delta$.

So far we have gone into the rotating frame of laser field with carrier frequency ω_0, but from section 3.2 we remember that we also have a ground level shift Hamiltonian H_0 (3.14), where we have taken the $F = 4$ as the zero point energy. We will go into the interaction picture with respect to H_0, with the effect that for $\sigma_{3,4}$ and $\sigma_{4,3}$ the operators will oscillate much faster since they gain respectively $\pm i\Delta_{34}t$ in the phase, where we recall that $\Delta \sim$GHz. On the other hand the $\sigma_{3,3}$ and $\sigma_{4,4}$ remain uchanged under this transformation. Because of this, we will neglect the contributions of H_{34} to the coherent part (for both light and atoms), since the involved frequencies are much higher. But we will keep the $\sigma_{3,4}$,$\sigma_{4,3}$ terms in the noise since it will be assumed to be white anyway.

In this section we have found the effective Hamiltonian that descibes the dipole interaction between atoms and strongly off-resonant light. We have included the full level structure of cesium and found that the Hamiltonian consists of two parts. An elastic part that preserves the spin state and describes the Stark shift caused on the atoms by the light and to describe this coherent Hamiltonian we needed the 3 coefficients a_0, a_1, and a_2. The second part was inelastic and desribed Raman scattering and to express this part we needed the two coefficients b_1 and b_2. The involved transitions in the two parts have a big frequency difference allowing us to focus on one of them. For our purpose it is the elastic part and this coherent Hamiltonian will form the basis for the dynamics we investigate. This section has been a bit mathematical, so in the next chapter we comment on the physical meaning of the interaction Hamitonian.

Chapter 4

Coherent interaction

In this chapter we will look at the coherent part of the interaction which is the most interesting. In this case we only consider the light field in the z-direction and our atoms are point like particles from the which the light scatters. Also we will just consider one atom separately and in chapter 5 explain how we adopt the simplest possible model, where we say the dynamics of the sample is the sum of single atom dynamics.
Now let us examine the polarizability Hamiltonian we derived in last section and to make clear that we only look at forward modes of field we put an F on the electric field:

$$H_{\text{coh}} = -\sum_{F,F'} \mathbf{E}_{\text{F}}^{(-)} \cdot \frac{P_F \mathbf{d} P_{F'} \mathbf{d} P_F}{\Delta_{FF'}} \cdot \mathbf{E}_{\text{F}}^{(+)} = \sum_i H_{\text{coh}}^{(i)}. \tag{4.1}$$

As we saw above we can also express the Hamiltonian in terms of a polarizability α, that is a sum of spherical tensors of rank 0,1 and 2:

$$H^{(i)} = \mathbf{E}_{\text{F}}^{(-)} \alpha^{(i)} \mathbf{E}_{\text{F}}^{(+)}, \tag{4.2}$$

$$\alpha = \alpha^{(0)} \oplus \alpha^{(1)} \oplus \alpha^{(2)}. \tag{4.3}$$

We will analyze the 3 three Hamiltonian parts from (3.40), where we just look at a single F state. Instead of electric fields we will express the Hamiltonian in terms of Stokes vectors. We take the single photon strength $|E| = \sqrt{\frac{\omega_0}{2\epsilon_0 A}}$ out of the expression and combine it with the dipole element D_0 to a single photon Rabi frequency $\Omega = D_0|E|$.

4.1 Scalar Hamiltonian

The first term in (3.40) is the scalar Hamiltonian:

$$H^{(0)} = -\frac{\Omega^2}{\Delta}(a_0 + \frac{a_2}{3}\mathbf{j}^2)\phi. \tag{4.4}$$

It is seen that regardless of the internal (magnetic) state the atoms experience a constant shift caused by the light field and proportional to the flux - a constant Stark shift. Similarly because of the atoms, the light sees a change in the index of refraction and the polarization modes of light experience an identical shift. This corresponds to a phase shift and the pulse shape is not changed by this interaction. Since it does not reveal information on the internal atomic state, measuring the pulse will not reveal any information and the state of the atoms is preserved.
This Hamiltonian will not be important for us, since it is "just a constant" and does not

influence the dynamics. However if we were looking at an optical lattice, this term would be very relevant describing a spatially varying (attractive) potential. Also it would be relevant if we had the task of distiguishing between the $F = 3, F = 4$ states, since measuring the outgoing pulses using homodyning we could check the total spin of the atoms.

4.2 Vector Hamiltonian

The second term in (3.40) is the most interaction part of the interaction namely the vector Hamiltonian (the factor 2 in front comes from the definition of the Stokes operators, also see App.A.2):

$$H^{(1)} = -\frac{\Omega^2}{\Delta} 2a_1 S_z j_z. \tag{4.5}$$

It is this Hamiltonian that in terms of quadratures becomes $\kappa P_A P_L$ - the Faraday interaction we discussed in chapter 2. The effect of it on light is that it causes a phase shift on the circular polarization modes. This shift gives a rotation of the Stokes vector about the z axis by an amount that is proportional to the z-component of the atomic spin. So it is a circular birefringence effect and we will analyze it more quantatively in section 6.1. For the atoms we have that the atomic spin also gets rotated about the z axis by an amount proportional to the difference in the circular polarizations. This rotation will be adressed more mathematically in section 7.1.
So the vector Hamiltonian gives us the so-called Faraday rotation.

4.3 Tensor Hamiltonian

Finally we have the complicated tensor Hamiltonian:

$$H^{(2)} = 2a_2 \frac{\Omega^2}{\Delta} (S_x(j_x^2 - j_y^2) + S_y\{j_x, j_y\} + \phi(3j_z^2 - \mathbf{j}^2)/3). \tag{4.6}$$

This Hamiltonian couples atomic spin to light operators in a complicated way. If one was looking at a system with $j = \frac{1}{2}$ we know the spin to be described by Pauli matrices σ_i, with $j_i = \frac{1}{2}\sigma_i$. They obey the relation $\frac{1}{2}\{\sigma_i, \sigma_j\} = \delta_{ij}$ and therefore the elements of the rank 2 polarizability: $j_x^2 - j_y^2, \{j_x, j_y\}, 3j_z^2 - j(j+1)$, all vanish. Conversely for any spin higher than $\frac{1}{2}$, the rank 2 tensor never vanishes exactly. However we have seen that for increasing detuning a_2 goes to zero, so it is a good approximation to neglect this term. The physical intepretation of this Hamiltonian is that it is a dynamic Stark shift, caused by the presence of the laser field. In out work we will only consider the electric fields, but in actual experiments they also use magnetic magnetic fields and the shift induced by this term adds to the Zeeman shifts from the magnetic field. The shift depends on the polarization angle and assuming the light to be polarized linearly with an angle θ relative to the mean atomic spin (being in the xy-plane), it is possible to write (4.6) as [15]:

$$H^{(2)} \propto \frac{\phi}{\Delta}(j_z^2 - (j_x^2 - j_y^2)\cos(2\theta) - \{j_x, j_y\}\sin(2\theta)). \tag{4.7}$$

From this expression it can be shown that the atoms experience an energy shift which goes as $\frac{\phi}{\Delta}(1 + 3\cos(2\theta))$. The shifts have been measured by magneto-optical resonance signals and the results in [15] confirm the form of shift above.

It also interesting to compare the full Hamiltonian[1] to the ideal QND Hamiltonian, which was of the form: $H = \kappa P_A P_L$. When we inlcude the a_2 terms, we also get a $X_A X_L$ term in the Hamiltonian. This gives an unwanted evolution of the quadrature momenta: $\dot{P}_A \propto X_L, \dot{P}_L \propto X_A$. (Actually the a_2 term do even more damage, because they will also produce some constant (but big) drifts on our quadratures.) This is a problem - our interaction is no longer QND, the angular momenta are not preserved and now the protocols from chapter 2 no longer work. So far the way of proceding has been to say that since for big detunings the a_2 goes to zero - this term can neglected. This is a good approximation and we will also make it when we reach the protocols.

In this chapter we have discussed the different components of the interaction Hamiltonian. In the next chapter we will take into account that our atomic system is not completely isolated - it interacts with the environment - a thermal reservoir and it produces decoherence. But we will still think of the coherent interaction to be dominating and the interaction with the environment as a small perturbation that adds some noise, but leaves the form of the dynamics intact.

[1] A mathematical curiosity is that if we had defined the atomic operators along the lines of the Stokes operators for light -(that is by replacing $a_i, a_i^\dagger$ with j_i in the definitions) and denoted them by $\tilde{j}_x, \tilde{j}_y, \tilde{j}_z$, then the full Hamiltonian (apart from a constant term) could be written in a very symmetric form: $H_{int} \propto \tilde{\mathbf{j}} C \mathbf{S}$, where C is a diagonal matrix.

Chapter 5

General EOM

In this section we will find the general evolution of operators (for both atoms and light) and then we will perform some noise analysis that will be important for later discussions. Many of the results in this section are more general than we will need for our treatment of the system and shows that this theory can be extended to other similar systems operating at similar conditions.

5.1 Single atom EOM

First we find the Hamiltonian for a single atom j and then we will say how to extend the results for an ensemble. We will supress the position dependence in the following since all interactions are considered to be point like, so it is understood that the Hamiltonian for atom j should be evaluated at position $\mathbf{r}_j$. Since we will not carry the position dependence with us for light through $e^{\pm i\mathbf{k}\cdot\mathbf{r}_j}$, we will occasionally put a j on the operators b_j,$b_j^\dagger$ to remind that these should be evaluated at the atomic positions. After the derivation of the EOM we will analyze what conditions must hold for the model to be valid. In the next section we will give a more detailed discussion of the position dependence.

Earlier we found the Hamiltonian for a single atom. We will write the electric field as a sum of a forward field, $\mathbf{E}_\mathrm{F}$,(which represents the coherent field created by our laser) and an incoherent field, $\mathbf{E}_\mathrm{S}$,(which represents the environment - the reservoir field). These two kinds of modes of the electric field: forward and "non-forward" we will view as separated and as we already know, it is the forward modes that have the most important role. We will approximate the Hamiltonian by:

$$\begin{aligned} H_j &= \mathbf{E}^{(-)}\alpha_j\mathbf{E}^{(+)} && (5.1)\\ &\simeq \mathbf{E}_\mathrm{F}^{(-)}\alpha_j\mathbf{E}_\mathrm{F}^{(+)} + \mathbf{E}_\mathrm{S}^{(-)}\alpha_j\mathbf{E}_\mathrm{F}^{(+)} + \mathbf{E}_\mathrm{F}^{(-)}\alpha_j\mathbf{E}_\mathrm{S}^{(+)} = H_\mathrm{coh}^j + V_j. && (5.2)\end{aligned}$$

In this approximation we have neglected the much weaker part of the interaction that has reservoir as both input and output. Similarly the process where we have vacuum input and coherent output will be heavily supressed as compared to the conjugate proces. For now we will have both to have a Hermitian Hamiltonian, but the weakness of the former as compared to the latter will enter through the expectation values in the end. Also we remember that we assume that H_coh dominates the dynamics, while V_j is a perturbation. It will be nice to have α dimensionless, so we pull out the factor $\frac{D_0^2}{\Delta}$:

$$H_\mathrm{coh} = \mathbf{E}_\mathrm{F}^{(-)}\alpha\mathbf{E}_\mathrm{F}^{(+)} \equiv \frac{D_0^2}{\Delta}\mathbf{E}_\mathrm{F}^{(-)}\alpha\mathbf{E}_\mathrm{F}^{(+)}. \quad (5.3)$$

Now we form the EOM by absorbing $\mathbf{E}_{\mathrm{F}}^{(+)}$ into α by $\alpha\mathbf{E}_{\mathrm{F}}^{(+)} = |E|\tilde{\alpha}$ (meaning that $\tilde{\alpha}$ now is a polarization vector and not the polarizability matrix). The reason we choose to do so, is that we want the commutators $[\tilde{\alpha}, A], [\tilde{\alpha}^\dagger, A]$ to describe the evolution of both light and atoms. Continuing we can write:

$$V_j = \frac{|E|D_0^2}{\Delta}\sum_{k\sigma}\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}(b_{k\sigma}^\dagger\tilde{\alpha}_j + \tilde{\alpha}_j^\dagger b_{k\sigma}). \tag{5.4}$$

As in last chapter we will introduce the single photon Rabi frequency $\Omega = D_0|E|$. We will model the enviroment by a thermal reservoir[1] in equilibrium, with the standard Hamiltonian:

$$H_R = \sum_{k\sigma}\omega_k b_{k\sigma}^\dagger b_{k\sigma}, \tag{5.5}$$

where $b_{k\sigma}^\dagger b_{k\sigma}$ gives the number of reservoir excitations in the mode $|k\sigma\rangle$ with frequency ω_k. Usually we will assume that the population for $\omega_k = \omega_0$ is negligible, which we also justify later. Before studying the evolution of the operators we really are interested in, it will prove helpful to know the dynamics of these b operators:

$$\frac{\partial}{\partial t}b_{k\sigma}(t) = i[H_R + V_j, b_{k\sigma}] = -i\omega_k b_{k\sigma}(t) - i\frac{\Omega}{\Delta}D_0\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}\tilde{\alpha}_j(t), \tag{5.6}$$

$$\frac{\partial}{\partial t}b_{k\sigma}^\dagger(t) = i[H_R + V_j, b_{k\sigma}^\dagger] = i\omega_k b_{k\sigma}^\dagger(t) + i\frac{\Omega}{\Delta}D_0\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}\tilde{\alpha}_j^\dagger(t). \tag{5.7}$$

These equations have the formal solution:

$$b_{k\sigma}(t) = b_{k\sigma}(0)e^{-i\omega_k t} - i\frac{\Omega}{\Delta}D_0\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}\int_0^t dt'\tilde{\alpha}_j(t')e^{-i\omega_k(t-t')}, \tag{5.8}$$

$$b_{k\sigma}^\dagger(t) = b_{k\sigma}^\dagger(0)e^{i\omega_k t} + i\frac{\Omega}{\Delta}D_0\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}\int_0^t dt'\tilde{\alpha}_j^\dagger(t')e^{i\omega_k(t-t')}. \tag{5.9}$$

The first part is the homogeneous solution and describes the free field evolution. The second part includes the interaction with our quantum system through $\tilde{\alpha}$, which shows that the electromagnetic field (described by the b operators) originates from the atomic polarization $\tilde{\alpha}$. Now we insert the found expression for the reservoir operators into V_j:

$$\begin{aligned}
\frac{d}{dt}A(t) &= i[H^j, A] = i[H_{\mathrm{coh}}^j, A] + i[V_j, A] \\
&= i[H_{\mathrm{coh}}^j, A] + i\frac{\Omega}{\Delta}D_0\sum_{k\sigma}\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}(b_{k\sigma}^\dagger(t)[\tilde{\alpha}_j, A](t) + [\tilde{\alpha}_j^\dagger, A](t)b_{k\sigma}(t)) \\
&= i[H_{\mathrm{coh}}^j, A] + i\frac{\Omega}{\Delta}D_0\sum_{k\sigma}\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}(b_{k\sigma}^\dagger(0)e^{i\omega_k t}[\tilde{\alpha}_j, A](t) + [\tilde{\alpha}_j^\dagger, A](t)b_{k\sigma}(0)e^{-i\omega_k t}) \\
&- (\frac{\Omega}{\Delta})^2 D_0^2\sum_{k\sigma}\frac{\omega_k}{2\epsilon_0 V}\boldsymbol{\epsilon}_{k\sigma}^2\int_0^t dt'\tilde{\alpha}_j^\dagger(t')e^{i\omega_k(t-t')}[\tilde{\alpha}_j, A](t) - [\tilde{\alpha}_j^\dagger, A](t)\tilde{\alpha}_j(t')e^{-i\omega_k(t-t')}.
\end{aligned} \tag{5.10}$$

[1] This is also often referred to as a heat bath in litterature.

Using that the forward electric field has a carrier frequency ω_0, we may introduce the slowly varying operator $\tilde{\alpha_j} \mapsto \tilde{\alpha}_j e^{i\omega_0 t}$ so our equation becomes:

$$\begin{aligned}\frac{d}{dt}A(t) &= i[H_{\text{coh}}, A] \\ &+ i\frac{\Omega}{\Delta}D_0 \sum_{k\sigma} \sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}(b^\dagger_{k\sigma}(0)e^{i(\omega_k-\omega_0)t}[\tilde{\alpha}_j, A](t) + [\tilde{\alpha}^\dagger_j, A](t)b_{k\sigma}(0)e^{-i(\omega_k-\omega_0)t}) \\ &- (\frac{\Omega}{\Delta})^2 D_0^2 \sum_{k\sigma} \frac{\omega_k}{2\epsilon_0 V}\boldsymbol{\epsilon}^2_{k\sigma} \int_0^t dt' \{\tilde{\alpha}^\dagger_j(t')e^{i(\omega_k-\omega_0)(t-t')}[\tilde{\alpha}_j, A](t) \\ &- [\tilde{\alpha}^\dagger_j, A](t)\tilde{\alpha}_j(t')e^{-i(\omega_k-\omega_0)(t-t')}\}. \end{aligned} \tag{5.11}$$

Introducing the unnormalized Langevin noise operator:

$$\mathbf{f}_j(t) = D_0 \sum_{k\sigma} \sqrt{\frac{\omega_k}{2\epsilon_0 V}}\epsilon_{k\sigma} b^j_{k\sigma}(0)e^{-i(\omega_k-\omega_0)t}, \tag{5.12}$$

and making the Markov approximation by assuming that $\tilde{\alpha}_j(t)$ varies little over the inverse reservoir bandwidth, such that we can take it out of the integral in (5.11), we end up with:

$$\frac{d}{dt}A = i[H_{\text{coh}}, A] + i\frac{\Omega}{\Delta}(\mathbf{f}^\dagger_j[\tilde{\alpha}_j, A] + [\tilde{\alpha}^\dagger_j, A]\mathbf{f}_j) - (\frac{\Omega}{\Delta})^2[g^{(+)}\tilde{\alpha}^\dagger_j[\tilde{\alpha}_j, A] - g^{(-)}[\tilde{\alpha}^\dagger_j, A]\tilde{\alpha}_j]. \tag{5.13}$$

We have arrived at a stochastic differential equation, which is called a quantum Langevin equation, because of the similarity with the classical Langevin equation. It has a randomly fluctuating noise term that because of the sum over all reservoir frequencies oscillates quickly over time scales corresponding to the inverse reservoir bandwidth. The last term is the loss, describing the radiation the fluctuations induce $\sim$ spontaneous emission. In deriving it we have assumed that the coupling to the reservoir is weak as compared to the reservoir bandwitdth, such that $\tilde{\alpha}$ was a slowly varying operator in time and we could make the Markov approximation.
It is also worth to mention a few words on ordering. When pursuing this approach one ought be careful with ordering. If A is a light varible we have generally the commutator with the vacuum modes is $[A(t), b(t')] \neq 0$, unless $t = t'$. We have chosen to use normal ordering, however this is not crucial, it just important to be consistent, once the ordering chosen, it should not be changed, also see [20].
Let us now examine the coefficients $g^{(\pm)}$ above, where we will make use of the identity $\sum_{k\sigma} \to \frac{V}{(2\pi)^3}\int d^3k = \frac{V}{(2\pi)^3}\int_0^\infty d\omega_k \omega_k^2 \int d\Omega$. Formally they were defined as:

$$\begin{aligned} g^{(\pm)} &= D_0^2 \sum_{k\sigma} \frac{\omega_k}{2\epsilon_0 V}\boldsymbol{\epsilon}^2_{k\sigma} \int_0^t dt' e^{\pm i(\omega_k-\omega_0)(t-t')} \\ &= D_0^2 \frac{1}{2\epsilon_0 V}(\sum_\sigma \int d\Omega \boldsymbol{\epsilon}^2_{k\sigma})(\frac{V}{(2\pi)^3}\int_0^t dt' \int_0^\infty d\omega_k \omega_k^3 e^{\pm i(\omega_k-\omega_0)(t-t')}). \end{aligned} \tag{5.14}$$

Now we change the variable $t \to t - t'$ and extend the integration limit to infinity (of course the interaction time is finite, but again because the functions are so sharply peaked it is a good approximation), which is consistent with the Markov approximation. If we for now

ignore the imaginary part $\sim$ the Lamb shift, then we find:

$$\begin{aligned} g^{(\pm)} &= D_0^2 \frac{1}{2\epsilon_0 V}(\sum_\sigma \int d\Omega \boldsymbol{\epsilon}_{k\sigma}^2)(\frac{V}{(2\pi)^3}\int_0^\infty dt' \int_0^\infty d\omega_k \omega_k^3 e^{\pm i(\omega_k-\omega_0)t'}) \\ &= D_0^2 \frac{1}{2\epsilon_0 V}\frac{8\pi}{3}\frac{V}{(2\pi)^3}\int_0^\infty d\omega_k \omega_k^3 \pi\delta(\omega_k-\omega_0) = D_0^2\frac{1}{2\epsilon_0}\pi\frac{8\pi}{3}\frac{1}{(2\pi)^3}\omega_0^3 \\ &= D_0^2\frac{\omega_0^3}{6\pi\epsilon_0} = 2\gamma. \end{aligned} \tag{5.15}$$

Here we have defined the population decay rate $\gamma = D_0^2\frac{\omega_0^3}{12\pi\epsilon_0}$, also known as the Wigner-Weisskopf decay rate[2]. Strictly speaking we should not sum over the forward modes, but since this is a few modes out of an infinity, it is an excellent approximation. In fact it can be shown that the corrections are of second order in the opening angle as is noted in [9], (the opening angle being the angular spreading of the coherently emitted forward modes, which we most of the time think as being zero). So since we now have $g^{(+)} = g^{(-)} = 2\gamma$, we may write (5.13) as:

$$\begin{aligned} \frac{d}{dt}A &= i[H^j_{\text{coh}} + \frac{\Omega}{\Delta}(\mathbf{f}_j^\dagger\tilde{\alpha}_j + \tilde{\alpha}_j^\dagger\mathbf{f}_j), A] - (\frac{\Omega}{\Delta})^2 2\gamma(\tilde{\alpha}_j^\dagger[\tilde{\alpha}_j, A] - [\tilde{\alpha}_j^\dagger, A]\tilde{\alpha}_j) \\ &= i[H^j_{\text{coh}} + \frac{\Omega}{\Delta}(\mathbf{f}_j^\dagger\tilde{\alpha}_j + \tilde{\alpha}_j^\dagger\mathbf{f}_j), A] - (\frac{\Omega}{\Delta})^2 2\gamma(\tilde{\alpha}_j^\dagger\tilde{\alpha}_j A + A\tilde{\alpha}_j^\dagger\tilde{\alpha} - 2\tilde{\alpha}_j^\dagger A\tilde{\alpha}_j) \\ &= i[H^j_{\text{coh}} + \frac{\Omega}{\Delta}(\mathbf{f}_j^\dagger\tilde{\alpha}_j + \tilde{\alpha}_j^\dagger\mathbf{f})_j, A] + (\frac{2\Omega}{\Delta})^2\mathscr{L}_j(A). \end{aligned} \tag{5.16}$$

Here we have introduced the Lindblad form:

$$\mathscr{L}_j(A) = \frac{\gamma}{2}(2\tilde{\alpha}_j^\dagger A\tilde{\alpha}_j - \tilde{\alpha}_j^\dagger\tilde{\alpha}_j A - A\tilde{\alpha}_j^\dagger\tilde{\alpha}_j), \tag{5.17}$$

and this form of decay is common to all systems under Markovian conditions. Actually the more general Lindblad form includes the possibility of multichannel decay with distinct rates γ_i, so $\mathscr{L}_j(A) = \sum_i \frac{\gamma_i}{2}(2\tilde{\alpha}_j^\dagger A\tilde{\alpha}_j - \tilde{\alpha}_j^\dagger\tilde{\alpha}_j A - A\tilde{\alpha}_j^\dagger\tilde{\alpha}_j)$. In our case we only have one decay channel due to the coupling to an empty radiation reservoir.
The terms $-\tilde{\alpha}_j^\dagger\tilde{\alpha}_j A, -A\tilde{\alpha}_j^\dagger\tilde{\alpha}_j$ describe the loss experienced due to decay, while $+2\tilde{\alpha}_j^\dagger A\tilde{\alpha}_j$ puts probability back into system - i.e. even though our atoms might decay to another state, they are still there! It can also be seen that without the noise term we would have that for instance the commutator $[a, a^\dagger]$ for light would decay exponentially. So the preservation of the commutator demands the inclusion of noise, as we also motivate in App.D.5.
It is instructive to note that we could write (5.16) in another way:

$$\begin{aligned} \frac{d}{dt}A &= i([H^j_{\text{coh}} + \frac{\Omega}{\Delta}(\tilde{\alpha}_j^\dagger\mathbf{f}_j + \mathbf{f}_j^\dagger\tilde{\alpha}_j), A] + i(\frac{\Omega}{\Delta})^2 2\gamma(\alpha_j^\dagger\tilde{\alpha}_j A + A\tilde{\alpha}_j^\dagger\tilde{\alpha}_j)) + (\frac{2\Omega}{\Delta})^2\gamma\tilde{\alpha}_j^\dagger A\tilde{\alpha}_j \\ &= i(H^j_{\text{eff}}A - AH^j_{\text{eff}}{}^\dagger) + (\frac{2\Omega}{\Delta})^2\mathscr{L}^j_{\text{jump}}(A). \end{aligned} \tag{5.18}$$

Here we have defined the effective Hamiltonian by:

$$H^j_{\text{eff}} = H^j_{\text{coh}} + \frac{\Omega}{\Delta}\tilde{\alpha}_j f_j + i(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\tilde{\alpha}_j^\dagger\tilde{\alpha}_j, \tag{5.19}$$

[2]In fact our derivation has been virtually identical to Weisskopf-Wigner analysis, only we have worked in the Heisenberg picture.

and the jump operator

$$\mathscr{L}^j_{\text{jump}}(A) = \gamma\tilde{\alpha}_j A\tilde{\alpha}_j^\dagger. \tag{5.20}$$

We could have taken the above as a definition of the EOM for observables of this system. The first part of the EOM in (5.19) is continuos and deterministic, albeit not unitary, whilst the second performs "jumps" or projections in a discontinuous way and we see that for increasing detuning we have fewer jumps. This approach is similar to what is used for so-called quantum Monte Carlo simulations and possibly it could have applications, even though we well not dwell more upon it.
In our treatment we only look at population decay, we neglect the added phase decoherence which has it's own decay rate γ_{phase}. We could include it by adding a phase relaxation $\mathscr{L}_{\text{phase}}$ to the EOM. The decoherence could come from thermal collisions or fluctuations in the laser field and it will also increase the total noise in the system. This is a very complicated issue and we assume none of these effects to be present i our system - or at least heavily supressed by the effects we investigate. What makes it more difficult is that there is no simple expression for the phase operator, so it is a formidable task to include the decoherence in the Heisenberg picture, but as an example of how to do it, one can look in [4].
If we were dealing with a non-empty reservoir we would have to modify the decay in (5.16). We simply state the form without further proof, but it can be motivated by the master equation approach as in [17] or the noise correlations we will see later. If we let $\bar{n}$ be the average number of photons given by Planck's law:

$$\bar{n}(\omega_0) = \frac{1}{e^{\omega_0/k_bT} - 1}, \tag{5.21}$$

then the modified decay reads:

$$\frac{d}{dt}A = (\frac{2\Omega}{\Delta})^2([1+\bar{n}]\mathscr{L}_j(A) + \bar{n}\bar{\mathscr{L}_j}(A)), \tag{5.22}$$

with the "conjugate Lindblad form":

$$\bar{\mathscr{L}_j}(A) = \frac{\gamma}{2}(2\tilde{\alpha}_j A\tilde{\alpha}_j^\dagger - \tilde{\alpha}_j\tilde{\alpha}_j^\dagger A - A\tilde{\alpha}_j\tilde{\alpha}_j^\dagger). \tag{5.23}$$

To understand what the difference from the simple decay is, let us consider the decay of the photon flux expectation value:

$$\frac{d}{dt}\langle a^\dagger a\rangle = -\Gamma\langle a^\dagger a\rangle. \tag{5.24}$$

(5.24) is however only true for an empty reservoir where some of the forward modes would decay to the reservoir giving the damping by $\propto e^{-\Gamma t}$. But generally the reverse proces is also possible if there is a significant population of the relevant frequency in the reservoir (in our case ω_0) , so the decay would be modified to:

$$\frac{d}{dt}\langle a^\dagger a\rangle = -\Gamma\langle a^\dagger a\rangle + \Gamma\bar{n}. \tag{5.25}$$

We can write it in the same form as (5.22):

$$\frac{d}{dt}\langle a^\dagger a\rangle = -\Gamma\langle a^\dagger a\rangle(\bar{n}+1) + \Gamma\bar{n}(\langle a^\dagger a\rangle + 1). \tag{5.26}$$

The first term gives the attenuation of the pulse, while the second shows the enhancement of it. The +1 terms describe spontaneous emission and the other terms gives the stimulated emission. Generally we can say that the form (5.22) gives the balance between the system and reservoir, including both loss and growth. But a full and consistent treatment including a non-empty reservoir is beyond the scope of this work and we will usually not include it in the coming analysis.

After having discussed the decay, let us now say a few words on the noise too. In (5.16) we saw that the noisy part of the EOM contained the noise operators for atom j:

$$\mathbf{f}_j(t) = D_0 \sum_{k\sigma} \sqrt{\frac{\omega_k}{2\epsilon_0 V}} \boldsymbol{\epsilon}_{k\sigma} b^j_{k\sigma}(0) e^{-i(\omega_k-\omega_0)t}. \tag{5.27}$$

Assuming that each atom couples to it's own reservoir (we will also discuss it in the next section) and that we possibly have some non-zero populations $\bar{n}(\omega_0)$, it has the correlations for each component $f_\mu = \mathbf{f}\cdot\boldsymbol{\mu}$:

$$\langle f_{i,\mu}(t) f^\dagger_{j,\nu}(t')\rangle = 4\gamma\delta_{ij}\delta_{\mu\nu}\delta(t-t')(1+\bar{n}(\omega_0)), \tag{5.28}$$
$$\langle f^\dagger_{i,\mu}(t) f_{j,\nu}(t')\rangle = 4\gamma\delta_{ij}\delta_{\mu\nu}\delta(t-t')\bar{n}(\omega_0). \tag{5.29}$$

From these relations it is natural to normalize the $\mathbf{f}$ operators, so they now are:

$$\mathbf{f}_j(t) = \frac{D_0}{2\sqrt{\gamma}} \sum_{k\sigma} \sqrt{\frac{\omega_k}{2\epsilon_0 V}} \boldsymbol{\epsilon}_{k\sigma} b^j_{k\sigma}(0) e^{-i(\omega_k-\omega_0)t}. \tag{5.30}$$

And with these we can express the decay rate as:

$$\gamma = \frac{1}{\bar{n}} \int_{-\infty}^{\infty} dt' \langle f^\dagger_{i,\mu}(t) f_{i,\mu}(t')\rangle = \frac{1}{1+\bar{n}} \int_{-\infty}^{\infty} dt' \langle f_{i,\mu}(t) f^\dagger_{i,\mu}(t')\rangle, \tag{5.31}$$

which shows that the decay of our system originates from the reservoir fluctations, as stated in the fluctuation-dissipation theorem.[3] If we go back, we find that the noise operators joined the analysis through the incoherent part of the electric field:

$$\mathbf{E}_{\mathrm{S}} = \sum_{k\sigma} \boldsymbol{\epsilon}_{k\sigma} \sqrt{\frac{\omega_k}{2\epsilon_0 V}} (b_{k\sigma} e^{-i\omega_k t} + b^\dagger_{k\sigma} e^{i\omega_k t}). \tag{5.32}$$

We also remember to have assumed the reservoir to be in thermal equlibrium, which means that the noise is stationary - the reservoir correlations can only depend on the time difference. When dealing with random stationary processes, (as the interaction of our system with the reservoir) the Wiener-Khinchin theorem [19] states that the power spectrum is

[3] This is discussed more together with the regression theorem in App.D.7.

given by the Fourier transform of the autocorrelation function for the radiated field[4]:

$$\begin{aligned} S(\omega) &= \int_0^\infty e^{i\omega t'}\langle E_{\rm S}(t)E_{\rm S}(t-t')\rangle dt' \\ &= \frac{1}{4\pi\epsilon_0}\frac{2}{3}\frac{1}{\pi}\int_0^\infty d\omega_k\omega_k^3\int_0^\infty([1+\bar{n}(\omega_k)]e^{-i(\omega_k-\omega)t'}+\bar{n}(\omega_k)e^{(i\omega_k+\omega)t'})dt' \\ &= \frac{1}{4\pi\epsilon_0}\frac{2}{3}\int_0^\infty d\omega_k\omega_k^3([1+\bar{n}(\omega_k)]\delta(\omega_k-\omega)+\bar{n}(\omega_k)\delta(\omega_k+\omega)) \\ &\quad +\frac{1}{4\pi\epsilon_0}\frac{2}{3}\frac{1}{\pi}P\int_0^\infty d\omega_k\omega_k^3(\frac{[1+\bar{n}(\omega_k)]}{\omega-\omega_k}+\frac{\bar{n}(\omega_k)}{\omega+\omega_k}) \\ &= \frac{1}{4\pi\epsilon_0}\frac{2}{3}\omega^3(1+\bar{n}(\omega))+is=\frac{2\gamma(\omega)}{D_0^2}(1+\bar{n}(\omega))+is. \end{aligned} \tag{5.33}$$

As usual the real part of the power spectrum gives us the decay and here the decay rate is $\gamma(\omega)=\frac{\omega^3}{12\pi\epsilon_0}D_0^2$. The imaginary part modifies the transition frequencies of the system due to a shift of the levels - in our case s is the Lamb shift, which can be incorporated into the transition frequency $\omega_0\to\omega_0+s$. Experimentally this is already this shifted frequency that the lasers are being locked on, so it is assumed that s is already included in our ω_0. Since we neglect the thermal motion of atoms due to conservation of energy both the coherently- and incoherently emitted photons have the same energy ω_0 only the latter have an arbitrary phase and are isotropically distributed in space. So our interest lies in the case where $\omega=\omega_0\sim$ therefor $S(\omega)\simeq S(\omega_0)\sim$ our noise is white.

What we have analyzed here encompasses the most important features of the coupling of a small system with few degrees of freedom [DOF] to a large system with many DOF, which is also known as the quantum theory of dissipation. Now let us try to look at what assumptions must be met for our model to be justified. We have seen that the small system decays exponentially at a constant rate γ and the first natural requirement is that $\gamma\ll\omega_0$. This is fulfilled for the transitions we study, because there $\gamma\sim$ MHz, while $\omega_0\sim$ THz. The second assumtion we make is that the Markov approximation is valid. It enters when we take $\alpha(t')$ out of the integral over $t'\in[0,t]$, thereby assuming implicitly that the coupling to the reservoir is a smoothly and slowly varying function in the range of the resonance frequency ω_0. This is certainly the case for our function $\omega_k^3e^{i(\omega_k-\omega_0)t}$. But it should also be the case for the population $\bar{n}(\omega_k)$. The physical meaning of it is that the reservoir does not have any resonances near ω_0 and therefor in the time domain the reservoir immediately loses memory of which frequency was involved in the transition. This is the essence of the Markov approximation. Had we a situation where the requirements were not met, the non-Markovian evolution would be radically different from the exponential decay we found. Another point worth noting is that in terms of perturbation theory we have only included the first non vanishing element, which in our case is of first order. This is the Born approximation. Going to higher order the corrections would be the processes with emission of 3 or higher odd number of photons[5].
A closer look at the shift s in (5.33) reveals that formally the integral diverges (logarithmically), since we should integrate over all frequencies.This is a very well known divergence called the ultraviolet divergence and it stemns from the incompleteness of the

[4]We make use of the property $\int_0^\infty dte^{i\omega t}=\pi\delta(\omega)+iP\frac{1}{\omega}$, where P denotes the principal part, also known as the Cauchy principal value.

[5]The exclusion of even number of photons come from conservation of parity. Since photons have odd intrinsic parity and initially one photon is absorbed by the atom, consequently odd number of photons have to be emitted.

non-relativistic model. This singularity was later removed by the celebrated renormalization theory developed by Feynman and others [6].
Our description of the noise has been very general. Even though we in our case have that the b_k represented the reservoir mode photons, one could use the same treatment for other bosonic noise sources. For instance one could have situation where the b_k instead were phonons in a solid. We have seen if the reservoir is non-empty, the decay was enhanced by the replacement $\gamma \to (1+n)\gamma$, where n is the mean reservoir occupation. And to be consistent we ought to include the possibility of reservoir induced stimulated emission. However if the reservoir is "cold" enough one could neglect the contributions. For room temperature the thermal energy is about:

$$E_t \simeq k_B T = 2 \cdot 10^{10} \text{ Hz/K} \cdot 300 \text{ K} = 6 \text{ THz}, \tag{5.34}$$

which is roughly a factor 60 smaller than the transition frequency $\omega_0 \approx 350$ THz, meaning that according to the Boltzman distribution the probability having a vacuum photon at the right frequency is less than 10^{-25}. Therefore it is certainly a good approximation to exclude the possibility of excitations from the reservoir as we will do from now on.

The central result of this section is that under some realistic assumptions we have constructed the EOM for our light- and atomic variables that were governed by (5.16) and consisted of a dominating coherent part, a noise part and a spontaneous emission part.

5.2 The ensemble

In the last section we studied the dynamics for a single atom. Our system consists of many atoms (10^{12}) located inside a glass cell of length L ($\sim$cm) and now we want to see how we can generalize the found EOM to be applicable for this situation. Once we have many atoms we obviously have system with spatial extent - that is a 3 dimensional problem. To boil it down to the 1 dimensional model that we want to study we refer to [24]. Along the lines of [23] they present the general reduction from 3- to 1 dimension and identify the parameters that make this approximation work. We will just mention that one key parameter is the Fresnel number, which is desired to be big enough for the model to be valid. That is we have that $F = A/\lambda L \gg 1$ and we will assume that this is the case in the thesis. That it is big enough means that for our purpose the beam is just a plane wave and we can neglect changes in the light profile.
We will introduce continuos operators through a density of atoms $n(\mathbf{r})$:

$$n(\mathbf{r}) = \sum_j \delta(\mathbf{r} - \mathbf{r}_j). \tag{5.35}$$

This density will evaluate the different operators at the right positions - the positions where the atoms are located ($\mathbf{r}_j$) and where the interaction takes place. We may continue and define continous spin operators:

$$j_k(\mathbf{r}) = \sum_j \delta(\mathbf{r} - \mathbf{r}_j) j_{k,j}, k = x, y, z. \tag{5.36}$$

For these continuos j-operators the commutator relation becomes:

$$\begin{aligned}[j_m(\mathbf{r}), j_n(\mathbf{r}')] &= \sum_{jj'} \delta(\mathbf{r}-\mathbf{r}_j)\delta(\mathbf{r}'-\mathbf{r}_{j'})[j_m^j, j_n^{j'}] \\ &= i\sum_{jj'}\sum_k \epsilon_{mnk}\delta(\mathbf{r}-\mathbf{r}_j)\delta(\mathbf{r}'-\mathbf{r}_{j'})j_k^j\delta_{jj'} \\ &= i\sum_k \epsilon_{mnk} j_k(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}'). \end{aligned} \tag{5.37}$$

We generalize this continuos extension for the polarizability too and we write:

$$\alpha(\mathbf{r}) = \sum_j \delta(\mathbf{r}-\mathbf{r}_j)\alpha_j. \tag{5.38}$$

As with the spin in (5.37) we get for an atomic operator A:

$$[\alpha(\mathbf{r}), A(\mathbf{r}')] = [\alpha, A](\mathbf{r})\delta(\mathbf{r}-\mathbf{r}'). \tag{5.39}$$

To continue we will assumme that the averaged density is constant along the sample:

$$n(z) = \frac{1}{A}\int d^2\mathbf{r}_\perp n(\mathbf{r}) = \rho. \tag{5.40}$$

This is a reasonable assumption, but for a more general treatment one can consult [10] where they keep $n(z)$. We define the atomic operators as function of z only, by integrating out the perpendicular part:

$$A(z) = \frac{1}{\rho A}\int d^2\mathbf{r}_\perp A(\mathbf{r}). \tag{5.41}$$

For our purpose we will need the spin $j(z)$ and the polarizability $\alpha(z)$:

$$j(z) = \frac{1}{\rho A}\int d^2\mathbf{r}_\perp j(\mathbf{r}), \tag{5.42}$$

$$\alpha(z) = \frac{1}{\rho A}\int d^2\mathbf{r}_\perp \alpha(\mathbf{r}). \tag{5.43}$$

When we take the commutator we get:

$$[\alpha(\mathbf{r}), A(z)] = \frac{\delta(z-z')}{\rho A}[\alpha, A](\mathbf{r}). \tag{5.44}$$

And therefor:

$$[\alpha(z), A(z')] = \frac{\delta(z-z')}{\rho A}[\alpha, A](z). \tag{5.45}$$

In last chapter we treated light and atoms simultaneously, this becomes troublesome when we are looking at the spatial description of the operators. The reason is that while we for the atomic operators - e.g. spin have that $[j_i(z), j_j(z')] = i\frac{\delta(z-z')}{\rho A}\epsilon_{ijk}j_k(z)$, we at the same time have for light that $[S_i(z), S_j(z')] = i\delta(z-z')\epsilon_{ijk}S(z)$. Now let us first see how the difference shows up in the coherent interaction. To get the Hamiltonian for the ensemble we should sum over all the atoms, but instead with the continuos notation above we turn the Hamiltonian continuos one:

$$H_{\text{int}} = \int d^3\mathbf{r}\mathbf{E}^{(-)}(\mathbf{r})\alpha(\mathbf{r})\mathbf{E}^{(+)}(\mathbf{r}). \tag{5.46}$$

The coherent part of the interaction is simple and becomes according to (5.43):

$$H_{\text{coh}} = \int d^3\mathbf{r}\mathbf{E}_{\text{F}}^{(-)}(z)\alpha(\mathbf{r})\mathbf{E}_{\text{F}}^{(+)}(z) = \int dz\mathbf{E}_{\text{F}}^{(-)}(z)\alpha(z)\mathbf{E}_{\text{F}}^{(+)}(z)\rho A. \quad (5.47)$$

Therefor the only difference from the previous section is that our α is averaged over the transverse dimensions. So for an atomic operator $A(z)$ we get the EOM:

$$\frac{\partial}{\partial t}A(z,t) = i\int dz'\mathbf{E}_{\text{F}}^{(-)}(z')[\alpha(z'),A(z)]\mathbf{E}_{\text{F}}^{(+)}(z')\rho A = i\mathbf{E}_{\text{F}}^{(-)}(z)[\alpha,A](z)\mathbf{E}_{\text{F}}^{(+)}(z). \quad (5.48)$$

Remembering that the electric fields have an $\frac{1}{\sqrt{A}}$ we see that the atomic part of the interaction will go as $\frac{1}{A}$. This is sensible because we know that the light intensity goes as $\frac{1}{A}$, so for a given power, the more focused the light is, (meaning A is smaller) the stronger the interaction should be. If we are dealing with light, say an annilation $a(z)$ operator, then making the EOM we simply get rid of the integral over z. Now the profile areas cancel out and we have that the interaction is proportional to the atomic density ρ. The is also natural, if there were no atoms in the cell, the light would just pass through unaffected, while as we increase the concentration of the atoms it means that light sees a higher refractive index.

Now we will look at the more complicated interaction term V, but omit the details that are found in last section. We will also assume that the operator A is atomic - the derivation for light is very parallel, only one would not have to worry so much about the constants ρA. For the rest terms in the Hamiltonian we write:

$$V(\mathbf{r}) = \frac{\Omega}{\Delta}D_0(\mathbf{E}_{\text{S}}^{(-)}(\mathbf{r})\tilde{\alpha}(\mathbf{r}) + \tilde{\alpha}^\dagger(\mathbf{r})\mathbf{E}_{\text{S}}^{(+)}(\mathbf{r})). \quad (5.49)$$

Forming the EOM along the lines of the previous section we get:

$$\begin{aligned}\frac{\partial}{\partial t}A(z,t) &= i\int d^3\mathbf{r}'[V(\mathbf{r}',t),A(z,t)] \\ &= i\frac{\Omega}{\Delta}D_0\int d^3\mathbf{r}\mathbf{E}_S^{(-)}(\mathbf{r},t)[\tilde{\alpha}(\mathbf{r},t),A(z,t)] + [\tilde{\alpha}^\dagger(\mathbf{r},t),A(z,t)]\mathbf{E}_S^{(+)}(\mathbf{r},t) \\ &= i\frac{\Omega}{\Delta}D_0\frac{1}{\rho A}\int d^3\mathbf{r}\mathbf{E}_S^{(-)}(\mathbf{r},t)[\tilde{\alpha},A](\mathbf{r},t) + [\tilde{\alpha}^\dagger,A](\mathbf{r},t)\mathbf{E}_S^{(+)}(\mathbf{r},t). \quad (5.50)\end{aligned}$$

As in (5.8) we find for the vacuum operators that:

$$b_{k\sigma}(t) = b_{k\sigma}(0)e^{i(\omega_k-\omega_0)t} - i\frac{\Omega}{\Delta}D_0\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\int_0^t dt'\int d^3\mathbf{r}\tilde{\alpha}(\mathbf{r},t)e^{-i(\mathbf{k}\cdot\mathbf{r}-(\omega_k-\omega_0)(t-t'))}. \quad (5.51)$$

This we can insert into the expression for $\mathbf{E}_{\text{S}}$ and one sees that the noisy part of the interaction becomes:

$$\frac{\partial}{\partial t}A(z,t) = \frac{2\Omega}{\Delta}\sqrt{\gamma}(F_A + F_A^\dagger)(z,t). \quad (5.52)$$

With the space averaged noise:

$$F_A(z,t) = \frac{i}{\rho A}\int d^2\mathbf{r}_\perp[\tilde{\alpha},A](\mathbf{r},t)\mathbf{f}(\mathbf{r},t). \quad (5.53)$$

The more complicated decay term will go as:

$$\begin{aligned}\frac{\partial}{\partial t}A(z,t) \propto \sum_{k\sigma}\omega_k\epsilon_{k\sigma}^2\int_0^t dt'\int d^2\mathbf{r}_\perp\int d^3\mathbf{r}\tilde{\alpha}^\dagger(\mathbf{r}',t')[\tilde{\alpha},A](\mathbf{r},t)e^{-i(\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')-(\omega_k-\omega_0)(t-t'))} \\ -[\tilde{\alpha}^\dagger,A](\mathbf{r},t)\tilde{\alpha}(\mathbf{r}',t')e^{i(\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')-(\omega_k-\omega_0)(t-t'))}. \quad (5.54)\end{aligned}$$

Using the definition of the continuos operators we can say loosely that this term is proportional to:

$$\sum_{ij} \delta(\mathbf{r}_i - \mathbf{r}')\delta(\mathbf{r}_j - \mathbf{r})\alpha_i\alpha_j. \tag{5.55}$$

Remembering that α was the atomic polarizability, we see that we here have possible dipole-dipole effects. It descibes interactions between different atoms through the field and can give rise to collective effects, such as superradiance. We ignore these and only consider the decay $\sim$ terms for which $\mathbf{r} = \mathbf{r}'$. That way we can write (5.55) as:

$$\sum_{i} \delta(\mathbf{r}_i - \mathbf{r}')\delta(\mathbf{r} - \mathbf{r}')\alpha_i\alpha_i. \tag{5.56}$$

For that to be realistic the density of atoms ρ should not be too high. After this approximation we can again perform the Markov approximation and use the result from (5.15) and finally write the evolution from V as:

$$\frac{\partial}{\partial t}A(z,t) = \frac{2\Omega}{\Delta}\sqrt{\gamma}(F_A + F_A^\dagger)(z,t) + (\frac{2\Omega}{\Delta})^2\mathscr{L}(A)(z,t). \tag{5.57}$$

With the Lindblad form:

$$\mathscr{L}(A)(z,t) = \frac{\gamma}{2}\frac{1}{\rho A}\int d^2\mathbf{r}_\perp \tilde{\alpha}^\dagger[\tilde{\alpha}, A](\mathbf{r},t) - [\tilde{\alpha}^\dagger, A]\tilde{\alpha}(\mathbf{r},t). \tag{5.58}$$

Looking at the noise operator correlations we get with our assumption of the atoms having independent decay channels:

$$\begin{aligned}\langle F_A(z,t)F_{A'}^\dagger(z',t')\rangle &\approx \delta(z-z')\delta(t-t')\frac{1}{(\rho A)^2}\int d^2\mathbf{r}_\perp([\tilde{\alpha}^\dagger, A][\tilde{\alpha}, A])(\mathbf{r},t) \\ &= \delta(z-z')\delta(t-t')\frac{1}{\rho A}([\tilde{\alpha}^\dagger, A][\tilde{\alpha}, A])(z,t).\end{aligned} \tag{5.59}$$

Here in accordance with the above one should read $([\tilde{\alpha}^\dagger, A][\tilde{\alpha}, A'])(\mathbf{r})$ as the single atom contributions $\sum_i[\tilde{\alpha}_i^\dagger, A][\tilde{\alpha}_i, A']\delta(\mathbf{r}-\mathbf{r}_i)$, etc. Joining (5.57) with the coherent part, the final atomic EOM becomes:

$$\frac{\partial}{\partial t}A(z,t) = i[H_{\text{coh}}, A](z,t) + \frac{2\Omega}{\Delta}\sqrt{\gamma}(F_A(z,t) + F_A^\dagger(z,t)) + (\frac{2\Omega}{\Delta})^2\mathscr{L}(A)(z,t). \tag{5.60}$$

So starting from the single atom interaction model we have constructed the behaviour of the ensemble as the sum of single atom contributions. We have assumed a constant atomic density $n(z) = \rho$ and neglected all cross terms - the atoms are independent entities. For this to hold we must have that the atoms are well separated, the density should not be too high. Otherwise the atoms, that in our model are small dipoles, start to see the other dipoles and one has to deal with dipole-dipole interations. It can lead to collective effects like superradiance, which is very different from the dynamics we expect (want) to see.

We have made the derivation for atomic variables and we will not repeat it for light operators. The reason is that the equations are almost the same - in fact they are more simple and one does not have to worry so much about the factors ρA. The equation (5.60) also describes light variables, but these are defined without the ρA:

$$F(z,t) = i\int d^2\mathbf{r}_\perp[\tilde{\alpha}, A](\mathbf{r},t)\mathbf{f}(\mathbf{r},t), \tag{5.61}$$

$$\mathscr{L}(A)(z,t) = \frac{\gamma}{2}\int d^2\mathbf{r}_\perp \tilde{\alpha}^\dagger[\tilde{\alpha}, A](\mathbf{r},t) - [\tilde{\alpha}^\dagger, A]\tilde{\alpha}(\mathbf{r},t). \tag{5.62}$$

In the next chapters we are going to apply the found results to find the evolution of first light and then atoms. Even though this section has introduced some complications with us having to deal with spatial extension of the system, it is good to keep in mind that with the assumptions made, we have not departed so much from the single atom dynamics. So even though we will see integrals over transverse dimension and factors of ρA one can view the system effectively as a single photon scattering from a single atom. And as we will see later, it is precicely the single particle correlations that describe all wanted quantities.

Chapter 6

Light EOM

As the light passes through the sample, we expect that due to the interaction with atoms, the light experiences a change in polarization and some attenuation due to absorption. We will calculate this absorption and see that it is in fact quite small, just as needed for our use. One should also bear in mind that we assume that we have a flat transverse profile for the ingoing pulses which is roughly $1/\sqrt{A}$. We also assume the profile area to be large, so we have a big Fresnel number and can ignore diffraction of light. When dealing with light we need to include the photonic Hamiltonian H_L itself. As described in App.A.3 we can use H_L to transform the EOM from time (t) to space (z), such that the EOM read:

$$\frac{\partial}{\partial z}A(z,t) \quad = \quad i[H_{\text{coh}}, A](z,t) + \frac{2\Omega}{\Delta}\sqrt{\gamma}(F_A + F_A^\dagger)(z,t) + (\frac{2\Omega}{\Delta})^2\mathscr{L}(A)(z,t). \quad (6.1)$$

As we have seen the EOM consist of 3 parts: the coherent part, noise and associated decay and we will analyze each of them separately and finally combine them in the end. To describe the light we will make use of Stokes-operators S_x, S_y, S_z and they are defined and explained in App.A.1 together with how they are defined as functions of z. The following analysis on the coherent interaction is much inspired by [15] and for a discussion on experimental results we refer to the same.

6.1 Coherent interaction

In chapter 4 we saw that the coherent Hamiltonian can be written as: $H_{\text{coh}} = H^{(0)} + H^{(1)} + H^{(2)}$. We also remember that $H^{(0)}$ represented a static Stark shift, which for the light just will give a phase shift, but otherwise not affect the dynamics. Therefore we can throw away that term and the reduced Hamiltonian thus becomes:

$$\begin{aligned} H_{\text{coh}}^{\text{eff}} &= H^{(1)} + H^{(2)} \\ &= -\frac{2\Omega^2}{\Delta}\int_0^L a_1 j_z(z,t)S_z(z,t) \\ &\quad - a_2[(j_x^2(z,t) - j_y^2(z,t))S_x(z,t) + \{j_x, j_y\}(z,t)S_y(z,t)]\rho A dz. \end{aligned} \quad (6.2)$$

From now on we will use that we can write $2\Omega^2 = \frac{2\sigma_0\gamma}{A}$, where $\sigma_0 = \frac{3\lambda_0^2}{2\pi}$ and it is convenient to write the Hamiltonian as the inner product:

$$H_{\text{coh}}^{\text{eff}} \quad = \quad -\frac{2\sigma_0\gamma}{A\Delta}\int_0^L (\boldsymbol{\gamma}\cdot\mathbf{S})(z,t)\rho A dz. \quad (6.3)$$

Where $\boldsymbol{\gamma} = (-a_2(j_x^2 - j_y^2), -a_2\{j_x, j_y\}, a_1 j_z)$ is a polarization vector. From here it easy to obtain the EOM for the Stokes vector using the canonical commutation relation $[S_i(z), S_j(z')] = i\sum_k \epsilon_{ijk} S_k(z)\delta(z - z')$ combined with the form of the vector product of two operators: $\mathbf{A} \times \mathbf{B} = \sum_{ijk} A_i B_j \mathbf{e}_k \epsilon_{ijk}$:

$$\begin{aligned}
\frac{\partial}{\partial z}\mathbf{S}(z,t) &= -i\frac{2\sigma_0\gamma}{A\Delta}\sum_{ij}\int dz'[\gamma_i(z',t)S_i(z',t), S_j(z,t)\mathbf{e}_j]\rho A, \\
&= \frac{2\sigma_0\gamma}{A\Delta}\sum_{ijk}\gamma_i(z,t)S_k(z,t)\mathbf{e}_j\epsilon_{ijk}\rho A, \\
&= -\frac{2\sigma_0\gamma}{A\Delta}(\boldsymbol{\gamma}\times\mathbf{S})(z,t)\rho A. \qquad (6.4)
\end{aligned}$$

If we replace the operators in $\boldsymbol{\gamma}$ with their expectation values[1], we can interpret that in the interaction the Stokes operator $\mathbf{S}$ gets rotated about the vector $\boldsymbol{\gamma}$, corresponding to the unitary evolution per segment dz:

$$dU = \exp[i\frac{2\sigma_0\gamma}{A\Delta}(\gamma_x S_x + \gamma_y S_y + \gamma_z S_z)\rho A dz]. \qquad (6.5)$$

For clarity we can also write out the expression:

$$\frac{\partial}{\partial z}\begin{pmatrix}S_x\\S_y\\S_z\end{pmatrix}(z,t) = -\frac{2\sigma_0\gamma}{A\Delta}\begin{pmatrix}0 & -a_1 j_z & -a_2\{j_x,j_y\}\\ a_1 j_z & 0 & a_2(j_x^2 - j_y^2)\\ a_2\{j_x,j_y\} & -a_2(j_x^2 - j_y^2) & 0\end{pmatrix}\begin{pmatrix}S_x\\S_y\\S_z\end{pmatrix}(z,t)\rho A. \qquad (6.6)$$

The rotation of $\mathbf{S}$ is seen to be composed of a big rotation (proportional to a_1) about the z-axis and proportional to the atomic spin along z and a small rotation (proportional to a_2) in the (x, y) plane by an angle which depends on the relative angle (this will be shown below) between the mean atomic spin and the Stokes vector.
Also note that by finding the evolution of the $a, a^\dagger$ operators and forming the total flux $\phi = a_x^\dagger a_x + a_y^\dagger a_y$, one can show that the number of photons is conserved as one would expect in the coherent interaction.
We see from the equation for S_z that this component of the Stokes vector is not conserved in general, but changes due to the presence of the a_2-terms $j_x^2 - j_y^2$ and $\{j_x, j_y\}$. Moreover we also know that our system is axially symmetric about z so we should have conservation of angular momentum along z. Since S_z is changing that must mean that j_z is changing too and by the same amount. So for non vanishing S_z we get a rotation of the spin about the z-axis, an effect which complicates the interaction a lot and in principle destroys the protocols we discussed in chapter 2 since the quantities P_A,P_L are no longer conserved.
So what physical interpretation do these annoying terms have? If we look closer at $j_x^2 - j_y^2$ and assume the light to have a neglegible S_x and atoms polarized along x[2], then according to (6.6) we get the evolution:

$$\begin{aligned}
\frac{\partial}{\partial z}S_y(z,t) &= -\frac{2\sigma_0\gamma}{A\Delta}\rho A a_2(j_x^2 - j_y^2)S_z(z,t) = +\frac{\kappa_2}{L}S_z(z,t), \qquad (6.7)\\
\frac{\partial}{\partial z}S_z(z,t) &= +\frac{2\sigma_0\gamma}{A\Delta}\rho A a_2(j_x^2 - j_y^2)S_y(z,t) = -\frac{\kappa_2}{L}S_y(z,t) \qquad (6.8)
\end{aligned}$$

With the dimensionless constant κ_2:

$$\kappa_2 = -\frac{2\sigma_0\gamma}{A\Delta}a_2\frac{F}{2}(2F-1)L\rho A = -\frac{a_2\gamma\rho\sigma_0 L}{\Delta}F(2F-1). \qquad (6.9)$$

[1] How it is done is described in App.B.
[2] Had we chosen the y direction instead we would just have κ_2 with opposite sign.

Here we have replaced the atomic operators $j_x^2 - j_y^2$ with the expectation value $\frac{F}{2}(2F-1)$ and now we can write the solution for the Stokes operators as:

$$S_y(z,t) = S_y(0,t)\cos(\frac{\kappa_2}{L}z) + S_z(0,t)\sin(\frac{\kappa_2}{L}z), \tag{6.10}$$

$$S_z(z,t) = -S_y(0,t)\sin(\frac{\kappa_2}{L}z) + S_z(0,t)\cos(\frac{\kappa_2}{L}z). \tag{6.11}$$

We see that for linearly polarized input photons $S_y(0)$ we build up an amount of circularly polarized photons out and contrary we produce linearly polarized photons for circular polarized input photons. So we conclude that the term $j_x^2 - j_y^2$ gives an alignment in the xy-basis and produces linear birefringence. The effect of $\{j_x, j_y\}$ can be understood in the same way, only the alignment takes place in the $(\pi/4)$ rotated xy-basis.
Now let us assume that the atoms are prepared in a state where the mean spin is parallel to propagation of the light field - the z-direction and replace the operator j_z with it's expectation value. We will have light linearly polarized such that $\langle S_z \rangle = 0$ and we may write:

$$\frac{\partial}{\partial z}\begin{pmatrix} S_x \\ S_y \end{pmatrix}(z,t) = -a_1\frac{\sigma_0\gamma}{2A\Delta}\rho A\langle j_z\rangle \begin{pmatrix} -S_y \\ S_x \end{pmatrix}(z,t). \tag{6.12}$$

I means that as the light pulse travels across the sample we may write the changes with $S^{in} = S(z=0), S^{out} = S(z=L)$:

$$S_x^{out} = S_x^{in}\cos(2\theta_F) - S_y^{in}\sin(2\theta_F), \tag{6.13}$$

$$S_y^{out} = S_x^{in}\sin(2\theta_F) + S_y^{in}\cos(2\theta_F). \tag{6.14}$$

Here we have defined the Faraday rotation angle θ_F:

$$\theta_F = -a_1\frac{\sigma_0\gamma}{A\Delta}\rho AL\langle j_z\rangle. \tag{6.15}$$

This angle is how much the polarization of the light field is rotated due to the presence of the atomic spin pointing along the propagation direction (The factor 2 comes from the fact that if a_x and a_y, are rotated by θ, the Stokes operators are rotated by 2θ, because they are product of these operators.). Expressing it in terms of the collective spin, using that $\langle J_z\rangle = N_A\langle j_z\rangle = \rho AL\langle j_z\rangle$, we can write:

$$\theta_F = -a_1\frac{\sigma_0\gamma}{A\Delta}\langle J_z\rangle. \tag{6.16}$$

The macroscopic spin is a huge quantity so it is a big rotation of the light beam. In section 3.3 we saw that a_1 has different signs and equal magnitude in the two cases $F=3$ and $F=4$, meaning that light will start rotating in different directions depending on whether the atoms are in the $F=3$ state or the $F=4$ state. This also means that for a special distribution of atomic populations one can have that the contributions from $F=3$ and $F=4$ cancelled out, such that there were no overall rotation of the light. These populations p_3 and p_4 (with $p_3+p_4=1$) depend on the detuning of the laser, because the detunings in (6.16) are different for the two states. A few calculations reveal this is achieved by having $p_3 = (1+\frac{3}{4}\frac{1}{1+\frac{\Delta_{34}}{\Delta}})^{-1}$, meaning that $\frac{4}{7} \leq p_3 \leq 1$ and $0 \leq p_4 \leq \frac{3}{7}$. However this rotation of the Stokes operators about z is unwanted, so unless otherwise mentioned we assume the mean atomic spin to be zero along z. More general rotations for this kind of settings are discussed in [7].

6.2 Noise

In our description we have included the possibility of incoherent scattering of light, this will weaken the pulse and consenquently add some noise to the forward modes. In this section we investigate how much noise is being added to the Stokes operators and ultimately to the light quadratures. We saw in chapter 5 that we could write the noise part of the light EOM as:

$$\frac{\partial}{\partial z}A(z,t) = \frac{2\Omega}{\Delta}\sqrt{\gamma}[F_A + F_A^\dagger](z,t), \tag{6.17}$$

$$F(z,t) = i\int d^2\mathbf{r}_\perp[\tilde{\alpha}, A](\mathbf{r},t)f(\mathbf{r},t). \tag{6.18}$$

With the single atom noise operators:

$$\mathbf{f}(\mathbf{r},t) = \frac{D_0}{2\sqrt{\gamma}}\sum_{k\sigma}\sqrt{\frac{\omega_k}{2\epsilon_0 V}}\boldsymbol{\epsilon}_{k\sigma}b_{k\sigma}(0)e^{i(\mathbf{k}\cdot\mathbf{r}-(\omega_k-\omega_0)t)}. \tag{6.19}$$

And we remember that when forming the noise operator correlations, then only same-atom elements contribute, because each atom atom couples to it's own reservoir. This time we cannot express this part of the interaction directly in terms of Stokes operators. But we can easily find the EOM for the $a, a^\dagger$ operators and then combine them to obtain the EOM for the Stokes operators by using the product rule. That way we obtain that the noise for each component of the Stokes vector is (we will supress the time dependence to simplify the notation):

$$\begin{aligned}\frac{\partial}{\partial z}S_x(z) = \frac{\Omega}{\Delta}i\sqrt{\gamma}\int d^2\vec{r}_\perp f_x^\dagger(\mathbf{r})[\alpha_{xx}(\mathbf{r})a_x(z) - \alpha_{xy}(\mathbf{r})a_y(z)] \\ + f_y^\dagger(\mathbf{r})[\alpha_{yx}(\mathbf{r})a_x(z) - \alpha_{yy}(\mathbf{r},t)a_y(z)] + f_z^\dagger(\mathbf{r})[\alpha_{zx}(\mathbf{r})a_x(z) - \alpha_{zy}(\mathbf{r},t)a_y(z)] + \text{h.c.},\end{aligned} \tag{6.20}$$

$$\begin{aligned}\frac{\partial}{\partial z}S_y(z) = \frac{\Omega}{\Delta}i\sqrt{\gamma}\int d^2\vec{r}_\perp f_x^\dagger(\mathbf{r})[\alpha_{xx}(\mathbf{r})a_y(z) + \alpha_{xy}(\mathbf{r})a_x(z)] \\ + f_y^\dagger(\mathbf{r})[\alpha_{yx}(\mathbf{r})a_y(z) + \alpha_{yy}(\mathbf{r})a_x(z)] + f_z^\dagger(\mathbf{r})[\alpha_{zx}(\mathbf{r})a_y(z) + \alpha_{zy}(\mathbf{r})a_x(z)] + \text{h.c.},\end{aligned} \tag{6.21}$$

$$\begin{aligned}\frac{\partial}{\partial z}S_z(z) = \frac{\Omega}{\Delta}\sqrt{\gamma}\int d^2\vec{r}_\perp f_x^\dagger(\mathbf{r})[\alpha_{xx}(\mathbf{r})a_y(z) - \alpha_{xy}(\mathbf{r})a_x(z)] \\ + f_y^\dagger(\mathbf{r})[\alpha_{yx}(\mathbf{r})a_y(z) - \alpha_{yy}(\mathbf{r})a_x(z)] + f_z^\dagger(\mathbf{r})[\alpha_{zx}(\mathbf{r})a_y(z) - \alpha_{zy}(\mathbf{r})a_x(z)] + \text{h.c.}.\end{aligned} \tag{6.22}$$

This is a new result and even though the expressions are long and complicated they show what the noise of the Stokes operators are expressed in terms of the elements of the polarizability α and vacuum noise operators f. We are interested in the noise of our S_y and S_z from which we will build the light quadatures, while we will treat S_x like a c-number which roughly equals half the total flux ϕ. Therefore we form the new noise operators:

$$\begin{aligned}\tilde{F}_y(z) = \frac{-i}{2}\int d^2\vec{r}_\perp([\alpha_{yx}(\mathbf{r})a_x^\dagger(z) + \alpha_{xx}(\mathbf{r})a_y^\dagger(z)]f_x(\mathbf{r}) \\ + [\alpha_{yy}(\mathbf{r})a_x^\dagger(z) + \alpha_{xy}(\mathbf{r})a_y^\dagger(z)]f_y(\mathbf{r}) + [\alpha_{yz}(\mathbf{r})a_x^\dagger(z) + \alpha_{xz}(\mathbf{r})a_y^\dagger(z)]f_z(\mathbf{r})),\end{aligned} \tag{6.23}$$

$$\begin{aligned}\tilde{F}_z(z) = -\frac{1}{2}\int d^2\vec{r}_\perp([\alpha_{yx}(\mathbf{r})a_x^\dagger(z) - \alpha_{xx}(\mathbf{r})a_y^\dagger(z)]f_x(\mathbf{r}) \\ + [\alpha_{yy}(\mathbf{r})a_x^\dagger(z) - \alpha_{xy}(\mathbf{r})a_y^\dagger(z)]f_y(\mathbf{r}) + [\alpha_{yz}(\mathbf{r})a_x^\dagger(z) - \alpha_{xz}(\mathbf{r})a_y^\dagger(z)]f_z(\mathbf{r})).\end{aligned} \tag{6.24}$$

And write the noise term in short form:

$$\frac{\partial}{\partial z}S_i(z,t) = \frac{2\Omega}{\Delta}\sqrt{\gamma}(\tilde{F}_i + \tilde{F}_i^\dagger)(z,t), i = y, z. \tag{6.25}$$

These $\tilde{F}_i$ have zero mean and below we have listed their correlations[3] where we have made use of the fact that $S_x \simeq \frac{\phi}{2}$: and supressed the (z) dependence that should be on the different α^2:

$$\begin{aligned}
\langle \tilde{F}_y(z,t)\tilde{F}_y^\dagger(z',t')\rangle &= \langle \tilde{F}_z(z,t)\tilde{F}_z^\dagger(z,t)\rangle \\
&= \delta(t-t')\delta(z-z')\frac{\langle S_x\rangle}{2}(\alpha_{yy}^2 - \alpha_{xx}^2 + \frac{1}{2}[\alpha_{xx}^2 + \alpha_{yy}^2]\frac{\langle\phi\rangle}{\langle S_x\rangle})\rho A \\
&\simeq \delta(t-t')\delta(z-z')\langle S_x\rangle\langle\alpha_{yy}^2\rangle\rho A, \qquad (6.26)\\
\langle \tilde{F}_y(z,t)\tilde{F}_z^\dagger(z',t')\rangle &= \langle \tilde{F}_z(z,t)\tilde{F}_y^\dagger(z',t')\rangle^* \\
&= i\delta(t-t')\delta(z-z')\frac{\langle S_x\rangle}{2}(\alpha_{xx}^2 + \alpha_{yy}^2 - \frac{1}{2}[\alpha_{xx}^2 - \alpha_{yy}^2]\frac{\langle\phi\rangle}{\langle S_x\rangle})\rho A \\
&\simeq i\delta(t-t')\delta(z-z')\langle S_x\rangle\langle\alpha_{yy}^2\rangle\rho A. \qquad (6.27)
\end{aligned}$$

From these relatively simple correlations we get:

$$\langle\{\tilde{F}_y(z,t) + \tilde{F}_y^\dagger(z,t), \tilde{F}_z(z',t') + \tilde{F}_z^\dagger(z',t')\}\rangle = 0, \tag{6.28}$$

$$\begin{aligned}
\langle[\tilde{F}_y(z,t) + \tilde{F}_y^\dagger(z,t), \tilde{F}_z(z',t') + \tilde{F}_z^\dagger(z',t')]\rangle &= i\delta(t-t')\delta(z-z')\langle S_x\rangle \\
&\times (\alpha_{xx}^2 + \alpha_{yy}^2 - \frac{1}{2}[\alpha_{xx}^2 - \alpha_{yy}^2]\frac{\langle\phi\rangle}{\langle S_x\rangle})\rho A \\
&\simeq 2i\delta(t-t')\delta(z-z')\langle S_x\rangle\langle\alpha_{yy}^2\rangle\rho A. \qquad (6.29)
\end{aligned}$$

Interestingly all the correlations are effectively desribed by a single number - namely the element $\langle\alpha_{yy}^2\rangle$. The value of it can be written as a constant and a term that depends on the relative angle θ between the light polarization (x) and the mean atomic spin pointing somewhere in the xy-plane:

$$\begin{aligned}
\langle\alpha_{yy}^2\rangle &= (a_0^2 + 4a_1^2 + 340a_2^2 - 24a_0a_2 + 56a_1a_2 + 28b_1^2 + 168b_1b_2 + 252b_2^2 \\
&\quad + 14\cos^2\theta[a_1^2 + 5a_2^2 + 4a_0a_2 - 6a_1a_2 - b_1^2 - 18b_1b_2 + 3b_2^2]) \\
&= \frac{1}{240}(3 + \frac{7}{(1+\frac{251}{\Delta})^2}) + \frac{7}{1440}\cos^2\theta(16 + \frac{5}{(1+\frac{452}{\Delta})^2} - \frac{21}{(1+\frac{251}{\Delta})^2}).
\end{aligned} \tag{6.30}$$

If we focus on the last term then as the detuning becomes large a_2 and b_2 go to zero while a_1^2 and b_1^2 become $(\frac{1}{48})^2$. But because the latter appear in the combination $a_1^2 - b_1^2$ the whole angular part goes to zero. This result is for $F = 4$, but inserting the numbers one finds that the same holds for $F = 3$. Therefore as $\Delta \to \infty$ we are left with the constant term which has the limit $\langle\alpha_{yy}^2\rangle \to \frac{1}{24}$, which is also the limit value for $F = 3$. For many of our quantities it will be nice to have them relative to a_1^2, which is also the case for $\langle\alpha_{yy}^2\rangle$:

[3] In the expressions α_{ij}^2 is the (i,j) element of α^2 and not $\alpha_{ij}\alpha_{ij}$. The explicit values of α^2 and found in App.C.2.

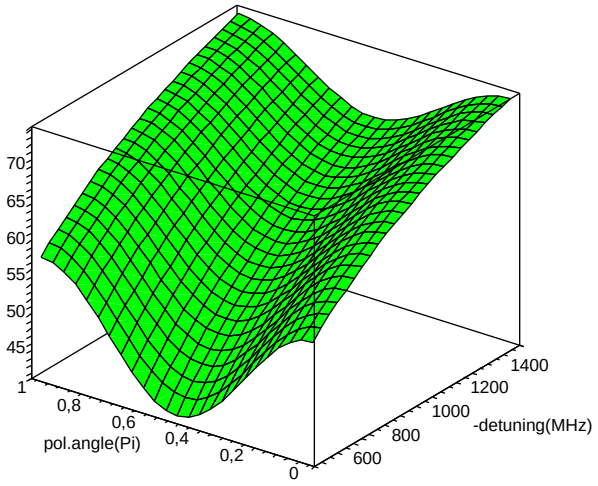

Figure 6.1: Element $\frac{\langle \alpha_{yy}^2 \rangle}{a_1^2}$ as a function of detuning and polarization angle.

We see that $\langle \alpha_{yy}^2 \rangle$ is a smooth function of the angle θ and detuning Δ. One sees that $\langle \alpha_{yy}^2 \rangle$ has period π, as we would expect from symmetry. Also as is both apparent from the expression and the graph, for a given detuning $\langle \alpha_{yy}^2 \rangle$ has maximum at $\theta = 0$ and minimum at $\theta = \frac{\pi}{2}$. This is no coincidence as we will see in the final section on light where we reveal the simple physical interpretation of $\langle \alpha_{yy}^2 \rangle$. Even though it looks like $\langle \alpha_{yy}^2 \rangle$ grows for increasing detuning, we remember that we still have a $\frac{1}{\Delta}$ outside for each of the Stokes operators in (6.25).
If we now choose to normalize the noise operators $\tilde{F}_y$,$\tilde{F}_z$ by $\sqrt{\langle \alpha_{yy}^2 \rangle \rho A \langle S_x \rangle}$ we get that without the tilde the correlations may be written simply:

$$\langle F_i(z,t) F_i^\dagger(z',t') \rangle = \delta(t-t')\delta(z-z'), \tag{6.31}$$

$$\langle [F_i(z,t), F_j^\dagger(z',t')] \rangle = i\epsilon_{ijk}\delta(t-t')\delta(z-z') \ , i,j = y,z. \tag{6.32}$$

And the final form of the noise EOM is:

$$\frac{\partial}{\partial z} S_i(z,t) = \frac{2\Omega}{\Delta} \sqrt{\gamma \langle \alpha_{yy}^2 \rangle \rho A \langle S_x \rangle} (F_i + F_i^\dagger)(z,t). \tag{6.33}$$

6.3 Light attenuation

As the light passes through the atomic sample we expect to see some attenuation of the pulse due to absorption. To find the attenuation it is easiest to use the $a, a^\dagger$ operators by going back to the expression for the decay:

$$\frac{\partial}{\partial z} A(z) = ... - (\frac{2\Omega}{\Delta})^2 \frac{\gamma}{2} \int d^2\mathbf{r}_\perp (\tilde{\alpha}^\dagger [\tilde{\alpha}, A](\mathbf{r}) - [\tilde{\alpha}^\dagger, A]\tilde{\alpha}(\mathbf{r})). \tag{6.34}$$

Only one of the commutators survives when A is our $a, a^\dagger$-operator and we get:

$$\begin{aligned}\frac{\partial}{\partial z}a_x(z) &= -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\int d^2\mathbf{r}_\perp(\alpha^2_{xx}(\mathbf{r})a_x(z)+\alpha^2_{xy}(\mathbf{r})a_y(z)) \\ &= -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}(\alpha^2_{xx}(z)a_x(z)+\alpha^2_{xy}(z)a_y(z))\rho A, \qquad (6.35)\\ \frac{\partial}{\partial z}a_y(z) &= -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\int d^2\mathbf{r}_\perp(\alpha^2_{yx}(\mathbf{r})a_x(z)+\alpha^2_{yy}(\mathbf{r})a_y(z)) \\ &= -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}(\alpha^2_{yx}(z)a_x(z)+\alpha^2_{yy}(z)a_y(z))\rho A. \qquad (6.36)\end{aligned}$$

If we now assume that we shine in circularly polarized light ($a_+ = -\frac{1}{\sqrt{2}}(a_x - ia_y)$) and we have prepared the atoms in the coherent spin state, where all atomic spins are pointing in the z-direction, we have that the light gets attenuated by an amount:

$$\begin{aligned}\frac{\partial}{\partial z}a_+(z) &= -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A\chi a_+, \qquad (6.37)\\ \chi &= \langle\alpha^2_{xx}(z)+i\alpha^2_{xy}(z)\rangle = \langle\alpha^2_{yy}(z)-i\alpha^2_{yx}(z)\rangle. \qquad (6.38)\end{aligned}$$

How big the attenuation is, will depend on the spin state of the atoms. For the case where the atoms after having absorbed the photon decay to the same F state as they started, one finds when putting in the elements of α^2 that the contribution to the attenuation for this proces can be expressed neatly as:

$$\chi = (a_0 + a_1F + a_2F^2)^2. \qquad (6.39)$$

For $F = 3$ and $F = 4$ we therefor get respectively the attenuations:

$$\begin{aligned}\frac{\partial}{\partial z}a_+(z) &= -\frac{25}{2304}(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A a_+(z), \qquad (6.40)\\ \frac{\partial}{\partial z}a_+(z) &= -\frac{1}{16}(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A a_+(z). \qquad (6.41)\end{aligned}$$

For decay to another spin state (H_{34}) we for the $F = 3$ state on the other hand we find:

$$\frac{\partial}{\partial z}a_+(z) = -\frac{35}{2304}(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A a_+(z). \qquad (6.42)$$

While for the $F = 4$ state:

$$\frac{\partial}{\partial z}a_+(z) = 0. \qquad (6.43)$$

This is also what we expected since the transition $|F = 4, m = 4\rangle \rightarrow |F = 5, m = 5\rangle$ is closed, the atom cannot decay to the $F = 3$ state. By adding (6.40) and (6.42) we obtain the total attenuation from the atoms in the $F = 3$ state:

$$\frac{\partial}{\partial z}a_+(z) = -\frac{5}{192}(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A a_+(z). \qquad (6.44)$$

As we expected the attenuation is proportional to ρ - the higher the concentration of atoms is, the higher will the damping of light be. When comparing the attenuations (6.41) and (6.44) one should remember that the detunings are defined differently. This little calculation shows how our model gives us some specific numbers that one can check in the experiments and with earlier work.

Now we return to the general case and find the evolution of the Stokes operators by using the product rule $\frac{\partial}{\partial z}(a_i^\dagger a_j) = \frac{\partial a_i^\dagger}{\partial z} a_j + a_i^\dagger \frac{\partial a_j}{\partial z}$. We obtain that way:

$$\begin{aligned}\frac{\partial}{\partial z}\begin{pmatrix}S_x\\S_y\\S_z\end{pmatrix}(z) &= -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A(\langle\alpha_{xx}^2+\alpha_{yy}^2\rangle\begin{pmatrix}S_x\\S_y\\S_z\end{pmatrix}(z)+\begin{pmatrix}\langle\alpha_{xx}^2-\alpha_{yy}^2\rangle\\\langle\alpha_{xy}^2+\alpha_{yx}^2\rangle\\i\langle\alpha_{xy}^2-\alpha_{yx}^2\rangle\end{pmatrix}\frac{\phi}{2}(z))\\ &= -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A(\langle\alpha_{xx}^2+\alpha_{yy}^2\rangle\begin{pmatrix}S_x\\S_y\\S_z\end{pmatrix}(z)+\begin{pmatrix}\zeta\langle j_x^2-j_y^2\rangle\\\zeta\langle\{j_x,j_y\}\rangle\\\beta\langle j_z\rangle\end{pmatrix}\frac{\phi}{2}(z)). \quad (6.45)\end{aligned}$$

Here β and ζ are some real numbers and when we replace the operators with their expectation values:

$$\begin{aligned}\langle j_x^2-j_y^2\rangle &= \frac{F}{2}(2F-1)\cos(2\theta), &(6.46)\\ \langle\{j_x,j_y\}\rangle &= \frac{F}{2}(2F-1)\sin(2\theta), &(6.47)\\ \langle j_z\rangle &= 0. &(6.48)\end{aligned}$$

We may write the damping as[4]:

$$\frac{\partial}{\partial z}\begin{pmatrix}S_x\\S_y\\S_z\end{pmatrix}(z) = -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A(\Gamma_S\begin{pmatrix}S_x\\S_y\\S_z\end{pmatrix}(z)+\frac{1}{2}\zeta\begin{pmatrix}\cos(2\theta)\\\sin(2\theta)\\0\end{pmatrix}\phi(z)) \quad (6.49)$$

We see that the decay of the Stokes operators consists of a common rate Γ_S, which is independent of the polarization angle and is given by (together with it's limit for $\Delta\to\infty$):

$$\begin{aligned}\Gamma_S &= \langle\alpha_{xx}^2+\alpha_{yy}^2\rangle\\ &= 2a_0^2+22a_1^2+750a_2^2+28a_1a_2+8a_0a_2+42(b_1^2+2b_1b_2+13b_2^2)\\ &= \frac{1}{5760}(256+\frac{35}{(1+\frac{452}{\Delta})^2}+\frac{189}{(1+\frac{251}{\Delta})^2})\to\frac{1}{12} \quad (6.50)\end{aligned}$$

For S_x and S_y we also have a big drift term proportional to the field flux that depends on the polarization angle and starts rotating S_x and S_y by an angle 2θ. This is reflecting that in general the photons are being absorbed in an asymmetric way by the atoms and this absorption depends on the relative polarization angle. The drift term does not have a dependence on θ for S_z since the system is symmetric about the z-axis and therefore cannot have a preferred decay direction. The value of ζ (with the limit $\Delta\to\infty$) is:

$$\begin{aligned}\zeta &= -14(a_1^2+5a_2^2+4a_0a_2-6a_1a_2-b_1^2-18b_1b_2+3b_2^2)\\ &= -\frac{7}{5760}(16+\frac{5}{(1+\frac{452}{\Delta})^2}-\frac{21}{(1+\frac{251}{\Delta})^2})\to 0 \quad (6.51)\end{aligned}$$

We see that apart from the sign this is exactly the angular part of $\langle\alpha_{yy}^2\rangle$ in (6.30) that we saw when discussing noise correlations.

[4]In the following we will absorb the constant $\frac{F}{2}(2F-1)$ into ζ.

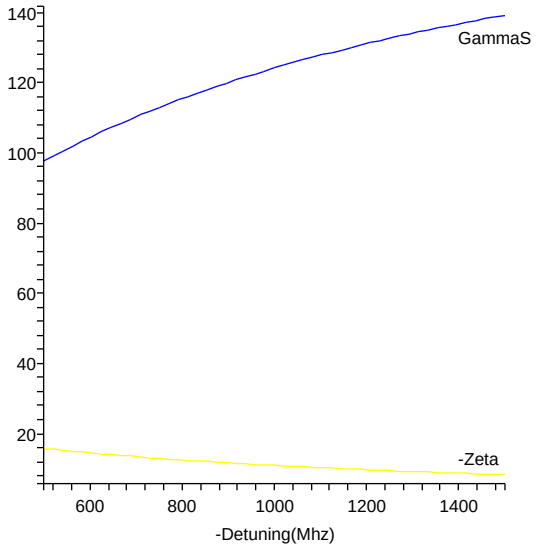

Figure 6.2: $-\frac{\zeta}{a_1^2}$ and $\frac{\Gamma_S}{a_1^2}$ as a function of -Δ.

6.4 Light X,P

Finally we introduce the light quadratures that we spoke about in chapter 2:

$$X_L = \frac{\int_0^T dt S_y(t)}{\sqrt{\langle S_x \rangle T}}, P_L = \frac{\int_0^T dt S_z(t)}{\sqrt{\langle S_x \rangle T}}. \tag{6.52}$$

It is easily checked that they obey $[X_L, P_L] = i$ (Remember that we have assumed that $S_x \approx \langle S_x \rangle$.). Also notice that it reasonable do treat X_L and P_L as continuos variables, because the quantity $S_x \simeq \sqrt{N_p} \simeq 10^7$ is very big as compared to S_y and S_z. We will analyze the EOM of these quadratures, where we include the derivative of $\langle S_x \rangle$:

$$\frac{\partial}{\partial z} X_L = \frac{1}{\sqrt{\langle S_x \rangle T}} \int_0^T dt (\frac{\partial}{\partial z} S_y - \frac{1}{2} \frac{\langle \frac{\partial}{\partial z} S_x \rangle}{\langle S_x \rangle} S_y), \tag{6.53}$$

$$\frac{\partial}{\partial z} P_L = \frac{1}{\sqrt{\langle S_x \rangle T}} \int_0^T dt (\frac{\partial}{\partial z} S_z - \frac{1}{2} \frac{\langle \frac{\partial}{\partial z} S_x \rangle}{\langle S_x \rangle} S_z). \tag{6.54}$$

We will consider each part separately and then combine them in the end for the full EOM. We will omit the details, since most calculations have been done in the sections above.
For the coherent part we have seen in (6.6) that $\langle \frac{\partial}{\partial z} S_x \rangle \simeq 0$ since the contributions from $\langle S_y \rangle$ and $\langle S_z \rangle$ are much smaller. This means that the evolution of S_y and S_z gives the

dynamics:

$$\begin{aligned}\frac{\partial}{\partial z}X_L &= \frac{1}{\sqrt{\langle S_x\rangle T}}\int_0^T dt\frac{\partial}{\partial z}S_y \\ &= -\frac{1}{\sqrt{\langle S_x\rangle T}}\frac{2\sigma_0\gamma}{A\Delta}\rho A\int_0^T dt\ a_1 j_z S_x + a_2(j_x^2 - j_y^2)S_z \qquad (6.55)\\ \frac{\partial}{\partial z}P_L &= \frac{1}{\sqrt{\langle S_x\rangle T}}\int_0^T dt\frac{\partial}{\partial z}S_z \\ &= -\frac{1}{\sqrt{\langle S_x\rangle T}}\frac{2\sigma_0\gamma}{A\Delta}\rho A\int_0^T dt\ a_2(\{j_x, j_y\}S_x - (j_x^2 - j_y^2)S_y) \qquad (6.56)\end{aligned}$$

We can write these equations in compact form:

$$\frac{\partial}{\partial z}\begin{pmatrix}X_L\\P_L\end{pmatrix}(z) = -\frac{2\sigma_0\gamma}{A\Delta}\rho A a_2\langle j_x^2 - j_y^2\rangle\begin{pmatrix}P_L\\-X_L\end{pmatrix}(z) - \frac{1}{\sqrt{\langle S_x\rangle T}}\frac{2\sigma_0\gamma}{A\Delta}\rho A\int_0^T dt\begin{pmatrix}a_1 j_z(z,t)\\a_2\{j_x,j_y\}(z,t)\end{pmatrix} \qquad (6.57)$$

The constant in front of X_L and P_L in (6.57) is precisely the κ_2 we calculated in 6.1. We have seen how light experiences a different index of refraction as it passed through the atomic cell. This change is the same for both S_y and S_z and therefor also X_L and P_L and experimentalists know how to deal with this effect. But for our purpose it is sufficiently to note that by applying the coordinate transformation in App.D.6 we can eliminate this term from the EOM and get:

$$\frac{\partial}{\partial z}\begin{pmatrix}X_L\\P_L\end{pmatrix}(z) = -\frac{2\sigma_0\gamma}{A\Delta}\sqrt{\langle S_x\rangle T}\frac{1}{T}\int_0^T dt\begin{pmatrix}a_1 j_z(z,t)\rho A\\a_2\{j_x,j_y\}(z,t)\rho A\end{pmatrix} \qquad (6.58)$$

Had we only the coherent part we would have the solution with $\kappa = -\frac{2\sigma_0\gamma}{A\Delta}a_1\sqrt{\langle S_x\rangle\langle J_\parallel\rangle T}$ and $X_L^{in} = X_L(z=0)$,$X_L^{out} = X_L(z=L)$ and similarly for P_L:

$$\begin{pmatrix}X_L^{out}\\P_L^{out}\end{pmatrix} = \begin{pmatrix}X_L^{in}\\P_L^{in}\end{pmatrix} + \kappa\begin{pmatrix}\frac{1}{T}\int_0^T dt P_A(t)\\(2F-1)\frac{a_2}{a_1}[\frac{1}{2}\sqrt{\langle J_\parallel\rangle}\sin(2\theta) + \frac{\cos(2\theta)}{T}\int_0^T dt X_A(t)]\end{pmatrix} \qquad (6.59)$$

While as we saw earlier X_L^{out} carries information about the atomic P_A^{in}, we see that a huge quantity ($\sqrt{\langle J_\parallel\rangle}$) is added to P_L^{out}, such that P_L^{out} carries no real information. If we ignore the time dependence of the atomic quadrature variances we obtain:

$$\begin{aligned}\begin{pmatrix}\Delta X_L^{2,out}\\\Delta P_L^{2,out}\end{pmatrix} &= \begin{pmatrix}\Delta X_L^{2,in}\\\Delta P_L^{2,in}\end{pmatrix} + \frac{\kappa^2}{T^2}\begin{pmatrix}Var(\int_0^T dt P_A(t))\\(2F-1)^2(\frac{a_2}{a_1})^2\cos^2(2\theta)Var(\int_0^T dt X_A(t))\end{pmatrix} \\ &\simeq \begin{pmatrix}\Delta X_L^{2,in}\\\Delta P_L^{2,in}\end{pmatrix} + \kappa^2\begin{pmatrix}\Delta P_A^{2,in}\\(2F-1)^2(\frac{a_2}{a_1})^2\cos^2(2\theta)\Delta X_A^{2,in}\end{pmatrix} \qquad (6.60)\end{aligned}$$

We see that if we choose $\theta = \frac{\pi}{4}$ we almost recover the result from the section on the Faraday interaction. Unfortunately this choise has some other problems that we will see in the coming and that render this choice undesired.

The noise operators have zero mean and therefore the evolution of the quadratures is determined by the first part of (6.53) and (6.54), giving that way:

$$\frac{\partial}{\partial z}\begin{pmatrix}X_L\\P_L\end{pmatrix}(z)=\frac{1}{\sqrt{\langle S_x\rangle T}}\int_0^T dt\frac{\partial}{\partial z}\begin{pmatrix}S_y\\S_z\end{pmatrix}(z,t)=\frac{2\Omega}{\Delta}\sqrt{\gamma\langle\alpha_{yy}^2\rangle\rho A}\frac{1}{\sqrt{T}}\int_0^T dt\begin{pmatrix}F_y+F_y^\dagger\\F_z+F_z^\dagger\end{pmatrix}(z,t). \tag{6.61}$$

From here it is natural to define the noise operators by:

$$f_X(z)=\frac{1}{\sqrt{2T}}\int_0^T dt\ F_y(z,t)+F_y^\dagger(z,t), f_P(z)=\frac{1}{\sqrt{2T}}\int_0^T dt\ F_z(z,t)+F_z^\dagger(z,t). \tag{6.62}$$

They inherit the former correlations, just without the time dependence and we can write them in short form:

$$\langle\{f_X(z),f_X(z')\}\rangle = \langle\{f_P(z),f_P(z')\}\rangle=\delta(z-z'), \tag{6.63}$$
$$\langle[f_X(z),f_P(z')]\rangle = i\delta(z-z'). \tag{6.64}$$

This allows to write (6.61) as:

$$\frac{\partial}{\partial z}\begin{pmatrix}X_L\\P_L\end{pmatrix}(z)=\frac{2\Omega}{\Delta}\sqrt{2\gamma\langle\alpha_{yy}^2\rangle\rho A}\begin{pmatrix}f_X\\f_P\end{pmatrix}(z). \tag{6.65}$$

For the decay we have according to (6.49) that the derivative of $\langle S_x\rangle$ has a non-vanishing expectation value and inserting the expressions from (6.49) we find:

$$\frac{\partial}{\partial z}X_L(z)=-(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A(\frac{1}{2}[\Gamma_S-\zeta\cos(2\theta)\frac{\langle\phi\rangle}{2\langle S_x\rangle}]X_L(z)+\frac{\zeta}{2}\sin(2\theta)\int_0^T dt\frac{\phi}{\sqrt{\langle S_x\rangle T}}), \tag{6.66}$$

$$\frac{\partial}{\partial z}P_L(z)=-(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A\frac{1}{2}(\Gamma_S-\zeta\cos(2\theta)\frac{\langle\phi\rangle}{2\langle S_x\rangle})P_L(z). \tag{6.67}$$

We see that X_L and P_L decay at the same rate $\Gamma_L=\frac{1}{2}(\Gamma_S-\zeta\cos(2\theta)\frac{\langle\phi\rangle}{2\langle S_x\rangle})$, but since $\Gamma_S=\langle\alpha_{xx}^2\rangle+\langle\alpha_{yy}^2\rangle$ and $\zeta\cos(2\theta)=\langle\alpha_{xx}^2\rangle-\langle\alpha_{yy}^2\rangle$, we see that $\Gamma_L\simeq\langle\alpha_{yy}^2\rangle$ - the very same element we encountered first time in the noise analysis. X_L has a big term proportional to ϕ expressing that due to the interaction with the atoms the light experiences a drift and starts rotating. This term gives a linear contribution to the decay and it is quite big since it is proportional to $\sqrt{\langle S_x\rangle T}$. Also we see that this term vanishes for $\theta=0$ or $\theta=\frac{\pi}{2}$, motivating the use of extreme polarizations.
So for the decay we have found that:

$$\frac{\partial}{\partial z}\begin{pmatrix}X_L\\P_L\end{pmatrix}(z) = -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\rho A(\Gamma_L\begin{pmatrix}X_L\\P_L\end{pmatrix}(z)+\sqrt{\langle S_x\rangle T}\zeta\sin(2\theta)\begin{pmatrix}1\\0\end{pmatrix}). \tag{6.68}$$

We will in the following neglect the constant term in the decay and keep in mind that we really would like to have $\theta=0$ or $\theta=\frac{\pi}{2}$.

Finally the full light EOM may be written by combining (6.58+61+66+67):

$$\frac{\partial}{\partial z}\begin{pmatrix}X_L\\P_L\end{pmatrix}(z)=\{-\frac{2\sigma_0\gamma}{A\Delta}\sqrt{\langle S_x\rangle T}\frac{1}{T}\int_0^T\begin{pmatrix}a_1j_z(z,t)\\a_2\{j_x,j_y\}(z,t)\end{pmatrix}+\frac{2\Omega}{\Delta}\sqrt{2\gamma\Gamma_L}\begin{pmatrix}f_X\\f_P\end{pmatrix}(z)$$
$$-(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\Gamma_L\begin{pmatrix}X_L\\P_L\end{pmatrix}(z)\}\rho A. \tag{6.69}$$

This is an equation of the form $\dot{x} = -\gamma x + f(t)$ for which the solution is $x(t) = x(0)e^{-\gamma t} + \int_0^t dt' f(t')e^{-\gamma(t-t')}$. Expressing the solution in terms of κ and $\eta = \frac{\kappa^2}{2d}$, where $d = \frac{\sigma_0}{A}N_A$ is the optical depth, we get:

$$\begin{aligned}\begin{pmatrix} X_L^{out} \\ P_L^{out} \end{pmatrix} &= \begin{pmatrix} X_L^{in} \\ P_L^{in} \end{pmatrix} e^{-\eta \frac{N_A}{N_p}\frac{\Gamma_L}{2a_1^2}} \\ &+ \int_0^L dz [\frac{1}{\sqrt{\langle J_\parallel \rangle}} \frac{1}{T}\int_0^T dt\ \kappa \begin{pmatrix} \rho A j_z(z,t) \\ \frac{a_2}{a_1}\rho A\{j_x, j_y\}(z,t) \end{pmatrix} \\ &+ \frac{2\Omega}{\Delta}\sqrt{2\gamma\Gamma_L \rho A}\begin{pmatrix} f_X \\ f_P \end{pmatrix}(z)] e^{-\eta \frac{N_A}{N_p}\frac{\Gamma_L}{2a_1^2}\frac{(z-L)}{L}}. \end{aligned} \tag{6.70}$$

This is the full solution of the problem, including the attenuation and noise. But having in mind that we want to use this for our protocol, we will ignore the a_2 terms in the coherent part (we have seen that j_z changed due to a_2) and can write the simplified form:

$$\begin{pmatrix} X_L^{out} \\ P_L^{out} \end{pmatrix} = (\begin{pmatrix} X_L^{in} \\ P_L^{in} \end{pmatrix} + \kappa \begin{pmatrix} P_A^{in} \\ 0 \end{pmatrix}) e^{-\eta \frac{N_A}{N_p}\frac{\Gamma_L}{2a_1^2}} + \sqrt{\eta \frac{N_A}{N_p}\frac{\Gamma_L}{a_1^2}\frac{1}{L}} \int_0^L dz \begin{pmatrix} f_X(z) \\ f_P(z) \end{pmatrix} e^{-\eta \frac{N_A}{N_p}\frac{\Gamma_L}{2a_1^2}\frac{(z-L)}{L}}. \tag{6.71}$$

The difference from the ideal relations in chapter 2, is that now our coherent part experiences an attenuation and we have added some extra noise terms. For the variances it means that:

$$\begin{pmatrix} \Delta X_L^{2,out} \\ \Delta P_L^{2,out} \end{pmatrix} = \quad (\begin{pmatrix} \Delta X_L^{2,in} \\ \Delta P_L^{2,in} \end{pmatrix} + \kappa^2 \begin{pmatrix} \Delta P_A^{2,in} \\ 0 \end{pmatrix}) e^{-\eta \frac{N_A}{N_p}\frac{\Gamma_L}{a_1^2}} + \frac{1}{2}(1 - e^{-\eta \frac{N_A}{N_p}\frac{\Gamma_L}{a_1^2}}) \begin{pmatrix} 1 \\ 1 \end{pmatrix}. \tag{6.72}$$

As we guessed in the limit of weak attenuation, where the exponential functions in the expressions are approximately 1, we get the desired relations:

$$\begin{pmatrix} X_L^{out} \\ P_L^{out} \end{pmatrix} \quad = \quad \begin{pmatrix} X_L^{in} \\ P_L^{in} \end{pmatrix} + \kappa \begin{pmatrix} P_A^{in} \\ 0 \end{pmatrix}, \tag{6.73}$$

and:

$$\begin{pmatrix} \Delta X_L^{2,out} \\ \Delta P_L^{2,out} \end{pmatrix} \quad = \quad \begin{pmatrix} \Delta X_L^{2,in} \\ \Delta P_L^{2,in} \end{pmatrix} + \kappa^2 \begin{pmatrix} \Delta P_A^{2,in} \\ 0 \end{pmatrix}. \tag{6.74}$$

Even though we have neglected the noise and corresponding decay we note that they have a (almost) symmetric contribution to the variances of X_L and P_L (6.72). We also see that the ratio $\frac{N_A}{N_p}$ characterizes how close we are to the desired situation of weak attenuation, because the rest factor $\frac{\eta\Gamma_L}{a_1^2}$ is approximately of order 1 for the optical depths and detunings that we consider. We can limit the attenuation of light, by making the ratio small enough. Given that one has some fixed amount of atoms in the cell, the easiest way to achieve this, is to increase the number of photons by increasing the power.
Note also that as we expect for very big damping the light quadratures according to (6.71) simply become vacuum operators. They no longer carry information about the atoms and give vacuum noise. We will assume that the light is shot noise limited meaning $\Delta X_L^{2,in} = \Delta P_L^{2,in} = \frac{1}{2}$. Of course by using squeezed states of light one could redistribute the variance in an uneven way, but for the protocols we will consider we will stick to the

coherent states - that is displaced vacuum states. That way we will have a simple expression for the fidelity and we can always use the benchmark $\mathcal{F} = \frac{1}{2}$ to say whether a protocol is succesful. This is not the case for squeezed states where the situation is complicated, but of course the new variance degree of freedom has some advantages.

We conclude this chapter with saying that our analysis have shown that the attenuation of the light quadratures is virtually determined by a single matrix-element $\langle \alpha_{yy}^2 \rangle$. We have seen that by having enough photons the attenuation can be made arbitrarily small. To avoid big drifts we want to have either $\theta = 0$ or $\theta = \frac{\pi}{2}$. We these settings the relations for the light quadratures become the simple ones from chapter 2.

Chapter 7

EOM for atoms

7.1 Simple case EOM

In this section we will embark on the atomic dynamics. Our approach will be similar to the one for light: we will analyze the different parts of the dynamics separately first and then join them in the end. We will still assume the light to be strongly polarized along x and mostly focus on the $F = 4$ state, even though some places we will keep the general F. The results for $F = 3$ can easily be obtained from the procedure of $F = 4$ and all the relevant elements are found in App.C.

Now let us again start from our general dynamical equation:

$$\frac{d}{dt}A(z,t) \quad = \quad i[H_{\rm coh}, A(z,t)] + \frac{2\Omega}{\Delta}(F_A + F_A^\dagger)(z,t) + (\frac{2\Omega}{\Delta})^2 \mathscr{L}(A)(z,t). \tag{7.1}$$

In our case A is atomic spin operator j, from which we later construct the collective spin and then the quadratures X_A and P_A. We will write the coherent Hamiltonian as:

$$H_{\rm coh} \quad = \quad -\frac{2\sigma_0\gamma}{A\Delta}[\boldsymbol{\gamma}\cdot\mathbf{S} + \gamma_0\frac{\phi}{2}], \tag{7.2}$$

where we have $\boldsymbol{\gamma} = (-a_2(j_x^2 - j_y^2), -a_2\{j_x, j_y\}, a_1 j_z)$ and $\gamma_0 = -a_2 j_z^2$. From this we can determine the coherent evolution of the spin vector $\mathbf{j}$:

$$\begin{aligned}\frac{\partial}{\partial t}\mathbf{j}(z,t) = i[H_{\rm coh}, \mathbf{j}](z,t) &= -i\frac{2\sigma_0\gamma}{A\Delta}\sum_{ij}[\gamma_i, j_j](z,t)S_i(z,t)\mathbf{e}_j + [\gamma_0, j_j](z,t)\frac{\phi}{2}\mathbf{e}_j \\ &= -\frac{2\sigma_0\gamma}{A\Delta}\sum_{ij}\tilde{g}_{ij}(z,t)S_i(z,t)\mathbf{e}_j + \tilde{g}_j^0(z,t)\frac{\phi}{2}\mathbf{e}_j. \end{aligned} \tag{7.3}$$

The Hermitian $\tilde{g}$-matrix is defined by:

$$\tilde{g}_{ij} \quad = \quad i[\gamma_i, j_j], \tilde{g}_j^0 = i[\gamma_0, j_j]. \tag{7.4}$$

These elements are listed in App.C.3 and when we insert them we get the equations:

$$\frac{\partial}{\partial t}\begin{pmatrix} j_x \\ j_y \\ j_z \end{pmatrix} = -\frac{2\sigma_0\gamma}{A\Delta}[\begin{pmatrix} a_2\{j_y, j_z\} & -a_2\{j_x, j_z\} & -a_1 j_y \\ a_2\{j_x, j_z\} & a_2\{j_y, j_z\} & a_1 j_x \\ -2a_2\{j_x, j_y\} & 2a_2(j_x^2 - j_y^2) & 0 \end{pmatrix}\begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix} + \frac{a_2}{2}\begin{pmatrix} -\{j_y, j_z\} \\ \{j_x, j_z\} \\ 0 \end{pmatrix}\phi]. \tag{7.5}$$

The equations we have reached are very complicated - they are not closed and couple to each other. Therefor it would be a very difficult task to find an analytical solution for them and will refrain from doing that. But by treating the different terms separately one can gain some insight.
For instance if we look at the a_1 terms in these equations, we see that they represent the rotation of the atomic spin about the S_z component of light (which we take as an expectation value):

$$\frac{\partial}{\partial t}\begin{pmatrix} j_x \\ j_y \end{pmatrix}(z,t) = -\frac{2\sigma_0\gamma}{A\Delta}a_1\begin{pmatrix} -j_y \\ j_x \end{pmatrix}(z,t)S_z. \tag{7.6}$$

We notice the resemblance with (6.12) from the light chapter. The solution is straightforward with $j^{in} = j(t=0), j^{out} = j(t=T)$:

$$\begin{pmatrix} j_x^{out} \\ j_y^{out} \end{pmatrix} = R(\tilde{\theta}_F)\begin{pmatrix} j_x^{in} \\ j_y^{in} \end{pmatrix}. \tag{7.7}$$

Here R is the matrix that rotates the spin by the angle:

$$\tilde{\theta}_F = -a_1\frac{2\sigma_0\gamma}{A\Delta}\langle S_z\rangle T. \tag{7.8}$$

Note that there is no factor 2 this time as compared to (6.16) and we only get a non-zero rotation for circularly polarized light. We see that if the atom starts with the spin in the y direction, then after the interaction it has build up a non-zero value of spin in the x direction and vice versa. But as we would expect, if the spin initially points along z, it stays there. Again if the atom is in $F = 3$ state it will rotate opposite to the atom with $F = 4$.

Now let us assume that our light is linearly polarized and we have oriented the mean atomic spin along x. Then we are interested in the behavior of the transverse spin components j_y and j_z. If we again linearize in j_x as we did in (6.7-11) and take $\phi \sim 2S_x$ and only look at the terms involving j_y or j_z, then from (7.5) we are lead to the equations:

$$\begin{aligned}\frac{\partial}{\partial t}\begin{pmatrix} j_y \\ j_z \end{pmatrix}(z,t) &= -\frac{2\sigma_0\gamma}{A\Delta}\begin{pmatrix} (2F-1)a_2 j_z(z,t)S_x + \frac{(2F-1)a_2 j_z(z,t)\phi}{2} \\ -2(2F-1)a_2 j_y(z,t)S_x + F(2F-1)a_2 S_y(z,t) \end{pmatrix} \\ &\simeq \Omega_S\begin{pmatrix} -j_z \\ +j_y \end{pmatrix}(z,t), \Omega_S = \frac{2\sigma_0\gamma}{A\Delta}2(2F-1)a_2\langle S_x\rangle = \frac{\gamma\sigma_0 a_2}{A\Delta}2(2F-1)\langle\phi\rangle.\end{aligned} \tag{7.9}$$

Because of the Stark effect the spin precesses about the mean spin with frequency Ω_S. The related phenomenon, Larmor precession, can be observed with an atom in an applied external magnetic field, where the spin precesses with a frequency proportional to the magnetic field strength. In our case the frequency is proportional to the electric field strength instead. In the actual experiments they use magnetic fields and the calculated effect will be seen as an extra Zeeman shift.

7.2 Atomic X,P

As we have seen from the section above, the general light atom-interaction, is quite complicated. But due to symmetry the interaction can only depend on the relative angle between

the polarization directions of light and atoms. Therefor without loss of generality we have assumed that the light is strongly polarized along x, while the direction of the atomic makes an angle θ with the polarization (the x-axis). But the spin is still assumed to be in the xy-plane - there is no component along z, which as we have seen otherwise gives an unwanted rotation. First we will analyze the two extremal angles $\theta = 0$ and $\theta = \frac{\pi}{2}$ and finally consider general θ.

Parallel configuration ($\theta = 0$)

Now we are ready to look at the EOM for X_A and P_A. These are collective observables for our atomic ensemble and are constructed through the collective atomic spin:

$$J_i(t) \quad = \quad \int_0^L j_i(z,t)\rho A dz. \tag{7.10}$$

From the definition of the continuos spin operators (5.24), we get that the collective spin also has:

$$[J_i, J_j] \quad = \quad i\sum_k \epsilon_{ijk} J_k. \tag{7.11}$$

And this collective spin behaves in the very same way as the usual j operators - basically we have just added the small atomic spins to a large one, but the spin nature has not changed. Assuming that all the atoms have a big component of the spin along $x \sim \langle j_x \rangle = F$, we can treat the collective spin J_x as a classical number. With this setting the light polarization and atomic mean are are parallel $\sim \theta = 0$. From these collective spin we now define the atomic quadratures we talked about in chapter 2:

$$X_A = \frac{J_y}{\sqrt{\langle J_x \rangle}}, P_A = \frac{J_z}{\sqrt{\langle J_x \rangle}}. \tag{7.12}$$

We know that the angular momentum operators have discrete eigenvalues, but since J_x has a very big value while J_y and J_z usually have vanishing or at least small values, the quadratures go approximately as $\frac{1}{\sqrt{N_A}}$ with $N_A \sim 10^{12}$. Therefor it is a very good approximation to treat X_A and P_A as continuos variables. As one can easily check by using (7.11) the X_A and P_A fulfill the canonical commutator relation in the mean:

$$\langle [X_A, P_A] \rangle \quad = \quad i. \tag{7.13}$$

We want to study the dynamics of X_A and P_A from the interaction with light. Even though we treat J_x as the number $\langle J_x \rangle$, we will include that possibly the magnitude of it can change in time. So when we differentiate, we get:

$$\dot{X_A} = \frac{\dot{J}_y}{\sqrt{\langle J_x \rangle}} - \frac{1}{2}\frac{J_y}{\sqrt{\langle J_x \rangle}}\frac{\langle \dot{J}_x \rangle}{\langle J_x \rangle}, \tag{7.14}$$

$$\dot{P_A} = \frac{\dot{J}_z}{\sqrt{\langle J_x \rangle}} - \frac{1}{2}\frac{J_z}{\sqrt{\langle J_x \rangle}}\frac{\langle \dot{J}_x \rangle}{\langle J_x \rangle}. \tag{7.15}$$

We are now in position to see how the dynamics of the atomic system looks in term of X, P - language. First we will study the coherent interation from last section.

Going to back to spin evolution from (7.5) and linearizing by using that j_x is almost[1] a c-number, we obtain for the single atomic spin:

$$\frac{\partial}{\partial t}\begin{pmatrix} j_x \\ j_y \\ j_z \end{pmatrix}(z,t) = -\frac{2\sigma_0\gamma}{A\Delta}[\begin{pmatrix} 0 & -(2F-1)a_2 j_z & -a_1 j_y \\ 2(2F-1)a_2 j_z & 0 & a_1 j_x \\ -2(2F-1)a_2 j_y & (2F-1)a_2 j_x & 0 \end{pmatrix}\begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix}(z,t)$$
$$= -\frac{2\sigma_0\gamma}{A\Delta}\begin{pmatrix} 0 & -a_1 S_z & -(2F-1)a_2 S_y \\ a_1 S_z & 0 & 2(2F-1)a_2 S_x \\ (2F-1)a_2 S_y & -2(2F-1)a_2 S_x & 0 \end{pmatrix}\begin{pmatrix} j_x \\ j_y \\ j_z \end{pmatrix}(z,t). \tag{7.16}$$

Now we can multiply by ρA and integrate over z to get the equations for the collective variables. But first we note from the first row in the matrix (7.16) that we can safely disregard the change in the mean spin j_x. The reason is that the terms in question: $S_z j_y$ and $S_y j_z$ are per contruction much smaller than those involving either S_x or j_x. So when we integrate up the equations, we would have terms that are roughly a factor N_A or N_p smaller than the retained.
Inserting the integrated last two lines of (7.16) into (7.14) and (7.15) we get:

$$\frac{\partial}{\partial t}\begin{pmatrix} X_A \\ P_A \end{pmatrix}(t) = -\frac{2\sigma_0\gamma}{A\Delta}\frac{1}{\sqrt{\langle J_x\rangle}}\int_0^L dz\rho A\begin{pmatrix} 2(2F-1)a_2 S_x j_z(z,t) + a_1 j_x S_z(z,t) \\ -2(2F-1)a_2 S_x j_y(z,t) + (2F-1)a_2 j_x S_y(z,t) \end{pmatrix} \tag{7.17}$$
$$= -\frac{2\sigma_0\gamma}{A\Delta}\begin{pmatrix} 2(2F-1)a_2\langle S_x\rangle P_A(t) + a_1\sqrt{\langle J_x\rangle}\frac{1}{L}\int_0^L dz S_z(z,t) \\ -2(2F-1)a_2\langle S_x\rangle X_A(t) + (2F-1)a_2\sqrt{\langle J_x\rangle}\frac{1}{L}\int_0^L dz S_y(z,t) \end{pmatrix} \tag{7.18}$$

The first term in the equations for X_A and P_A, describes the extra Zeeman shift that the atoms experience. In 7.1 we found how it means that the Larmor frequency experiences a shift by Ω_S, with Ω_S given in (7.9). Generally the Zeeman shift is not symmetric for X_A and P_A and even though it introduces some complications, it can be taken care of experimentally. The way it is done, is to use two cells instead of just one and a smart use of magnetic fields makes it possible to get rid of the effect that way. But then one has to deal with two ensembles, which is beyond the scope of this work and for details we refer to [27].
In this specific case, the shift is identical for X_A and P_A and we can once again perform the coordinate transformation from App.D.6, just for atomic variables this time, and get rid of it that way. With $\kappa = -\frac{2\sigma_0\gamma}{A\Delta}a_1\sqrt{\langle S_x\rangle\langle J_x\rangle}$ the solution for the coherent part will be:

$$\begin{pmatrix} X_A^{out} \\ P_A^{out} \end{pmatrix} = \begin{pmatrix} X_A^{in} \\ P_A^{in} \end{pmatrix} + \kappa\begin{pmatrix} \frac{1}{L}\int_0^L P_L(z)dz \\ \frac{(2F-1)a_2}{a_1}\frac{1}{L}\int_0^L X_L(z)dz \end{pmatrix} \tag{7.19}$$

Now keeping in mind the protocol from chapter 2 we see that we to be able to use it, we need to assume that the a_2 terms do not contribute to the coherent dynamics. We need that for two reasons, first we have seen in last chapter that if $a_2 \neq 0$ then P_L is not conserved, because S_z changes, meaning we cannot save P_L^{in} in X_A. Also in the equation for P_A we have an unwanted position averaged X_L term that messes up our attempt to store X_L^{in}. Ignoring the a_2 terms we obtain the ideal relations (2.11+12) that serve as our point of reference.

[1] But one cannot just replace all the j_x with F - we refer to App.B to see how we make the replacements.

Above we saw how we could translate the coherent spin evolution in terms of X_A and P_A. For the noise we will use start with a the single atom noise form, found in the section on general EOM where the forward field is absorbed into α. That way we get for a single atom that:

$$\frac{\partial}{\partial t}j_i(z,t) = \frac{2\Omega}{\Delta}\sqrt{\gamma}(F_i + F_i^\dagger](z,t), \tag{7.20}$$

where:

$$F_i(z,t) = i\int d^2\mathbf{r}_\perp[\tilde{\alpha}^\dagger, j_i](\mathbf{r},t)\mathbf{f}(\mathbf{r},t). \tag{7.21}$$

If we integrate the single atom evolution (7.20) over the sample we obtain the collective response. Since the noise operators have zero mean, we once again have that only the first term in the EOM (7.14+15) survives:

$$\begin{pmatrix} X_A \\ P_A \end{pmatrix}(z,t) = \frac{2\Omega}{\Delta}\frac{\sqrt{\gamma}}{\sqrt{\langle J_x\rangle}}\int_0^L \begin{pmatrix} [F_y + F_y^\dagger](z,t) \\ [F_z + F_z^\dagger](z,t) \end{pmatrix}\rho A dz = \frac{2\Omega}{\Delta}\sqrt{\gamma}\begin{pmatrix} f_{X_A} \\ f_{P_A} \end{pmatrix}(z,t). \tag{7.22}$$

Here f_{X_A} and f_{P_A} are the noise operators for the atomic quadratures telling what noise they get. Correlations between these kind of noise operators were analyzed in chapter 5 and exploiting the result from (5.59), we see that for f_{X_A} and f_{P_A} we have (only the combinations $\langle F_i F_i^\dagger\rangle$ give something different from zero, because we have assumed the reservoir to be empty):

$$\langle f_{X_A}(t)f_{X_A}(t')\rangle = \frac{\delta(t-t')}{\langle J_x\rangle|E|^2}\mathbf{E}_{\mathrm{F}}^{(-)}[\int d^3\mathbf{r}g_y^2(\mathbf{r})]\mathbf{E}_F^{(+)} = \frac{\delta(t-t')}{\langle J_x\rangle|E|^2}\mathbf{E}_{\mathrm{F}}^{(-)}[\int_0^L dz\rho A g_y^2(z)]\mathbf{E}_{\mathrm{F}}^{(+)}, \tag{7.23}$$

$$\langle f_{P_A}(t)f_{P_A}(t')\rangle = \frac{\delta(t-t')}{\langle J_x\rangle|E|^2}\mathbf{E}_{\mathrm{F}}^{(-)}[\int d^3\mathbf{r}g_z^2(\mathbf{r})]\mathbf{E}_{\mathrm{F}}^{(+)} = \frac{\delta(t-t')}{\langle J_x\rangle|E|^2}\mathbf{E}_{\mathrm{F}}^{(-)}[\int_0^L dz\rho A g_z^2(z)]\mathbf{E}_{\mathrm{F}}^{(+)}. \tag{7.24}$$

We have in (7.23+24) neglected the position dependence of the electric fields. The reason is that we expect this correlation to be propotional to the flux. Moreover we only bother about the dominating x-polarized part of light. Inserting the values of the (x,x) elements of the g^2 matrices from App.C.3, we get explicitly that:

$$\begin{aligned}\langle f_{X_A}(t)f_{X_A}(t')\rangle &= \langle f_{P_A}(t)f_{P_A}(t')\rangle \\ &= \phi\delta(t-t')(4(a_1^2+77a_2^2) + \frac{21}{2}(b_1^2+2b_1b_2+13b_2^2)).\end{aligned} \tag{7.25}$$

This is in effect a number that describes the noisecorrelations of the quadratures and later we will evaluate it for typical values of the detuning. Together with the other correlations we find, it is a new result, describing precisely how much noise we have in the quadratures. That the noise correlations are the same for X_A and P_A is expected. We have x as a symmetry axis and therefor the y and z components of the spin change in a similar way due to interaction with the environment. Therefor the decay of the spin should be the same for and the noise correlations too.
For the commutator we find:

$$[f_{X_A}(t), f_{P_A}(t')] = \frac{\delta(t-t')}{\langle J_x\rangle|E|^2}\mathbf{E}_F^{(-)}[\int d^3\mathbf{r}[g_y,g_z](\mathbf{r})]\mathbf{E}_F^{(+)} \tag{7.26}$$

$$= \frac{\delta(t-t')}{\langle J_x\rangle|E|^2}\mathbf{E}_F^{(-)}\int_0^L dz\rho A[g_y,g_z](z)\mathbf{E}_F^{(+)} \tag{7.27}$$

$$= \phi\delta(t-t')(28(-4a_1a_2+8a_2^2)+21(b_1^2+2b_1b_2+13b_2^2)), \tag{7.28}$$

while the anticommutator $\{f_{X_A}(t), f_{P_A}(t')\} = 0$.
Having looked at the noise it is natural procede with examining the decay for X_A and P_A. To describe the decay of a single atomic spin we use the expression (5.58) from section 5.2 to obtain:

$$\begin{aligned}\frac{\partial}{\partial t}j_i(z,t) &= (\frac{2\Omega}{\Delta})^2\mathscr{L}(j_i)(z,t), &(7.29)\\ \mathscr{L}(j_i)(z) &= -\frac{\gamma}{2}\frac{1}{\rho A}\int d^2\mathbf{r}_\perp[\tilde{\alpha}^\dagger\tilde{\alpha}j_i + j_i\tilde{\alpha}^\dagger\tilde{\alpha} - 2\tilde{\alpha}^\dagger j_i\tilde{\alpha}](\mathbf{r})\\ &= -\frac{\gamma}{2|E|^2}\frac{1}{\rho A}\mathbf{E}_{\mathrm{F}}^{(-)}(z)\int d^2\mathbf{r}_\perp[\alpha^2 j_i + j_i\alpha^2 - 2\alpha j_i\alpha](\mathbf{r})\mathbf{E}_{\mathrm{F}}^{(+)}(z). &(7.30)\end{aligned}$$

We expect this decay to be proportional to the total flux and we can as a good approximation ignore the position dependence of the light field, allowing us to write for the collective spin:

$$\begin{aligned}\mathscr{L}(J_i)(t) &= -\frac{\gamma}{2}\int d^3\mathbf{r}[\tilde{\alpha}^\dagger\tilde{\alpha}j_i + j_i\tilde{\alpha}^\dagger\tilde{\alpha} - 2\tilde{\alpha}^\dagger j_i\tilde{\alpha}](\mathbf{r})\\ &= -\frac{\gamma}{2|E|^2}\mathbf{E}_{\mathrm{F}}^{(-)}\int d^3\mathbf{r}[\alpha^2 j_i + j_i\alpha^2 - 2\alpha j_i\alpha](\mathbf{r})\mathbf{E}_{\mathrm{F}}^{(+)}\\ &= -\frac{\gamma}{2}\phi\Gamma_i J_i(t). &(7.31)\end{aligned}$$

The most notable contribution comes from the x polarized part of light, so since we already expect this effect to be small (remember that it goes as $\frac{1}{\Delta^2}$) we can safely only consider the (x,x) element of the decay matrix $\xi_i = \alpha^2 j_i + j_i\alpha^2 - 2\alpha j_i\alpha$. The decay rate Γ_i is defined as the linearization of the expression:

$$\Gamma_i J_i(t) = \int_0^L dz\rho A\xi_i(z,t). \quad (7.32)$$

This element gives the magnitude of the decay of the respective component of the spin. For the decay to different F (the b terms), we will neglect the term $2\alpha j_i\alpha$, which described the increase in the population of the final state. The reason is that once an atom decays to another F state it will no longer be interesting for us, since we restrict our analysis to collective behaviour of many atoms in the same F state. And as we have seen it reasonable to do since the energy spacing between the two ground spin states is big. For the same reasons we obviously need to keep $2\alpha j_i\alpha$ for the a-terms.
Written in short form the decay reads:

$$\frac{\partial}{\partial t}J_i(t) = -(\frac{2\Omega}{\Delta})^2\frac{\gamma}{2}\phi\Gamma_i J_i(t) = -\frac{\eta}{T}\frac{\Gamma_i}{2a_1^2}J_i(t). \quad (7.33)$$

The different ξ are listed in App.C.4 and are generally quite complicated. But if one just considers the a_1^2 terms, it is seen from the ξ_{xx} that J_x decays twice as fast as J_y and J_z. This holds generally, the component of the spin which points along the polarization of the light, will decay twice as fast as the orthogonal spin components as is also explained in [9]. In the listed ξ-elements one also sees it confirmed from ξ_{yy} for the case where light is polarized along y. But for all other a and b terms we do not have such simple relations. Inserting the elements ξ and using the linearizations from App.B.1, we find the decay for X and P, which again due to symmetry is the same:

$$\frac{\partial}{\partial t}\begin{pmatrix}X_A\\P_A\end{pmatrix} = -\frac{\eta}{T}\frac{1}{2a_1^2}\begin{pmatrix}(\Gamma_y - \frac{1}{2}\Gamma_x)X_A\\(\Gamma_z - \frac{1}{2}\Gamma_x)P_A\end{pmatrix} = -\frac{\eta}{T}\frac{\Gamma}{2a_1^2}\begin{pmatrix}X_A\\P_A\end{pmatrix}, \quad (7.34)$$

with:

$$\Gamma = 112a_2(-a_1+2a_2)+21(b_1^2+2b_1b_2+13b_2^2). \tag{7.35}$$

Comparing the (7.28) with (7.35) we see that (from now on we will take the flux out of the noise correlations, corresponding to us having a $\sqrt{\phi}$ in front in (7.22)):

$$[f_X(t), f_P(t')] = i\Gamma\delta(t-t'). \tag{7.36}$$

This is no coincidence, but holds generally, as we can motivate. If we neglect the coherent part we are dealing with equations for X, P of the form:

$$\frac{\partial}{\partial t}X = -\Gamma_X X + f_X, \tag{7.37}$$

$$\frac{\partial}{\partial t}P = -\Gamma_P P + f_P. \tag{7.38}$$

We can write the quadratures as:

$$X(t+\delta t) = X(t) + \int_t^{t+\delta t} dt' \frac{dX}{dt}(t'), \tag{7.39}$$

$$P(t+\delta t) = P(t) + \int_t^{t+\delta t} dt' \frac{dP}{dt}(t'). \tag{7.40}$$

For the commutator $[X, P]$ we get by using the product rule and the form of X and P in (7.39) and (7.40) that:

$$\dot{[X,P]} = -(\Gamma_X+\Gamma_P)\langle[X,P]\rangle + 2\frac{\langle[f_X(t), f_P(t')]\rangle}{\delta(t-t')}. \tag{7.41}$$

Demanding that the commutation relation $[X, P] = i$ be preserved (as it should for the canonical spin commutation relation to hold), is equivalent to setting the left handside of (7.41) to zero, leading to:

$$\langle[f_X(t), f_P(t')]\rangle = \frac{i}{2}(\Gamma_X+\Gamma_P)\delta(t-t') = i\langle\Gamma\rangle\delta(t-t'). \tag{7.42}$$

In our case we had that $\Gamma_X = \Gamma_P$, which is not always the case as we also will see in the next section. We conclude that the relation (7.36) is equivalent to the fact that the commutator $[X, P]$ is always i.

We have described all the parts of the dynamics of X and P, and now we can join them to find the total EOM. Using that $\eta = \frac{\kappa^2}{2d}$ and collecting the constants, we can write the EOM as:

$$\begin{aligned}\frac{\partial}{\partial t}\begin{pmatrix}X_A\\P_A\end{pmatrix} = &-\frac{2\sigma_0\gamma}{A\Delta}\begin{pmatrix}\frac{a_1}{L}\int_0^L dz\sqrt{\langle J_x\rangle}S_z(z)+2(2F-1)a_2S_xP_A\\(2F-1)a_2\frac{1}{L}\int_0^L\sqrt{\langle J_x\rangle}S_y(z)-2S_xX_A\end{pmatrix}\\ &+\frac{1}{|a_1|}\sqrt{\frac{\eta}{T}}\begin{pmatrix}f_{X_A}\\f_{P_A}\end{pmatrix}-\frac{\eta}{T}\frac{\Gamma}{2a_1^2}\begin{pmatrix}X_A\\P_A\end{pmatrix}.\end{aligned} \tag{7.43}$$

As mentioned earlier we can performing a change of coordinates and eliminate X_A and P_A from the coherent part of the equation system to have:

$$\frac{\partial}{\partial t}\begin{pmatrix}X_A\\P_A\end{pmatrix} = \frac{\kappa}{L}\int_0^L dz\begin{pmatrix}\frac{S_z(z,t)}{\sqrt{\langle S_x\rangle T}}\\\frac{(2F-1)a_2}{a_1}\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}}\end{pmatrix}+\sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\begin{pmatrix}f_{X_A}\\f_{P_A}\end{pmatrix}-\frac{\eta}{T}\frac{\Gamma}{2a_1^2}\begin{pmatrix}X_A\\P_A\end{pmatrix}. \tag{7.44}$$

We see that we have arrived at a differential equation which is very similar to the one we had for light (6.69). Again the solution is of the same form, where the light operators are just some extra noise terms:

$$\begin{pmatrix} X_A^{out} \\ P_A^{out} \end{pmatrix} = e^{-\eta\frac{\Gamma}{2a_1^2}} \begin{pmatrix} X_A^{in} \\ P_A^{in} \end{pmatrix} + \int_0^T dt e^{-\eta\frac{\Gamma}{2a_1^2}\frac{(T-t)}{T}} \{\frac{\kappa}{L}\int_0^L dz \begin{pmatrix} \frac{S_z(z,t)}{\sqrt{\langle S_x\rangle T}} \\ \frac{(2F-1)a_2}{a_1}\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}} \end{pmatrix} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\begin{pmatrix} f_{X_A}(t) \\ f_{P_A}(t) \end{pmatrix}\}. \quad (7.45)$$

We have seen that to use our protocol of mapping light variables onto the atomic variables, we need to get rid of the a_2 term. Therefor we assume that the detuning is sufficiently big such that a_2 contributions are negligible and replace the damping of the light quadrature P_L with a mean damping $e^{-\frac{\eta}{2}\frac{\Gamma}{2a_1^2}}$. This replacement is not trivial and only works in the case where the decay is not too big. We can sketch some details by expanding the exponential to first order and to simplify the expression we use $\beta = \eta\frac{\Gamma}{2a_1^2}$:

$$\begin{aligned} \int_0^T e^{-\beta\frac{(T-t)}{T}} S_z(t)dt &\simeq \int_0^T (1-\beta\frac{(T-t)}{T})S_z(t)dt \\ &= (1-\frac{\beta}{2})\int_0^T S_z(t)dt + \beta\int_0^T dt(\frac{t}{T}-\frac{1}{2})S_z(t). \end{aligned} \quad (7.46)$$

The first term corresponds to the mean damping that we made and is for the simple constant mode that we usually focus on. The last term is a correction that corresponds to another mode of the incoming light and including it would effectively give more noise in the results. We will neglect for our purpose and what is left we recognize as the first order expansion of $e^{-\frac{\beta}{2}}$. One could also have reached the same result simply by saying that for the coherent part in (7.45) we average the time in the exponential to $t = \frac{T}{2}$. So for not too big decay we have that:

$$\begin{pmatrix} X_A^{out} \\ P_A^{out} \end{pmatrix} \simeq \begin{pmatrix} e^{-\eta\frac{\Gamma}{2a_1^2}}X_A^{in} + \kappa e^{-\frac{\eta}{2}\frac{\Gamma}{2a_1^2}}P_L^{in} \\ e^{-\eta\frac{\Gamma}{2a_1^2}}P_A^{in} \end{pmatrix} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\int_0^T dt e^{-\eta\frac{\Gamma}{2a_1^2}\frac{(T-t)}{T}}\begin{pmatrix} f_{X_A}(t) \\ f_{P_A}(t) \end{pmatrix}. \quad (7.47)$$

When we perform the direct mapping protocol described in chapter 2 (subtract X_L^{out} from P_A^{out}), the equations translate to:

$$\begin{pmatrix} X_A^{out} \\ P_A^{out} \end{pmatrix} = \begin{pmatrix} (e^{-\eta\frac{\Gamma}{2a_1^2}}X_A^{in} + \kappa e^{-\frac{\eta}{2}\frac{\Gamma}{2a_1^2}}P_L^{in}) \\ (e^{-\eta\frac{\Gamma}{2a_1^2}} - \kappa g)P_A^{in} - gX_L^{in} \end{pmatrix} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\int_0^T dt e^{-\eta\frac{\Gamma}{2a_1^2}\frac{(T-t)}{T}}\begin{pmatrix} f_{X_A}(t) \\ f_{P_A}(t) \end{pmatrix}. \quad (7.48)$$

Now let us examine what are the optimal values of the parameters g and κ for the protocol to be succeful. We immediately see that we want $g = 1$ so P_A gets the value of X_L. However as we have seen the light also undergoes some decay and in case this decay is non-zero we should put $g = e^{+\eta\frac{N_A}{N_p}\frac{\Gamma_L}{2a_1^2}}$, so the role of g is to correct for the possible decay of the light quadratures. But as we have mentioned we assume that the ratio $\frac{N_A}{N_p} \ll 1$, so we will not take the decay of the light into account. For X_A to get the value of P_L we

need to have $\kappa e^{-\frac{\eta}{2}\frac{\Gamma}{2a_1^2}} = 1$. This equation is of the form $e^{cx^2} = x$ and it cannot be solved in terms of simple functions, but we can write the solution as:

$$\kappa = e^{-\frac{1}{2}\text{LambertW}(-\frac{\Gamma}{4a_1^2 d})}. \tag{7.49}$$

Here LambertW is a special function and it's definition and application in physics can for instance be seen in [3]. The graph of κ:

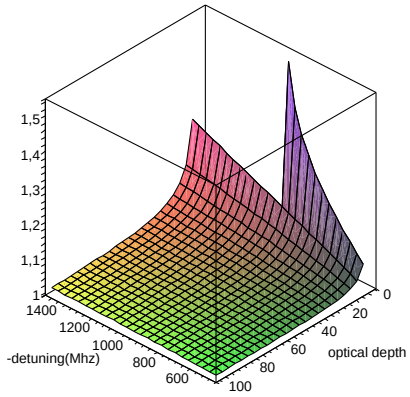

Figure 7.1: $\kappa_0(-\Delta, d)$.

shows that the values of κ are slightly above 1, which is well known from earlier work [27]. Also we see that when the optical depth is too small there is no solution - we cannot map X_L faithfully onto X_A, because the decay is too big. Mathematically it means that the equation $e^{cx^2} = x$ cannot be solved, because c is too big and the exponential function blows up without intersecting x. Remembering that κ is given by (7.49) the equations read:

$$\begin{pmatrix} X_A^{out} \\ P_A^{out} \end{pmatrix} = \begin{pmatrix} e^{-\eta\frac{\Gamma}{2a_1^2}} X_A^{in} + P_L^{in} \\ (e^{-\eta\frac{\Gamma}{2a_1^2}} - e^{\frac{\eta}{2}\frac{\Gamma}{2a_1^2}})P_A^{in} - X_L^{in} \end{pmatrix} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\int_0^T dt e^{-\eta\frac{\Gamma}{2a_1^2}\frac{(T-t)}{T}} \begin{pmatrix} f_{X_A}(t) \\ f_{P_A}(t) \end{pmatrix}. \tag{7.50}$$

These equations represent the memory protocol with included decay. We see that they are not as simple as the ideal ones in (2.13+14) and P_A is no longer conserved. However if X_A and P_A have zero mean input values, we still have achieved the wanted:

$$\langle X_A^{out} \rangle = \langle P_L^{in} \rangle, \tag{7.51}$$

$$\langle P_A^{out} \rangle = -\langle X_L^{in} \rangle. \tag{7.52}$$

So despite the decay and added noise we can still use this protocol for mapping the light quadratures. The variances now become:

$$\Delta X_A^{2,out} = e^{-\eta\frac{\Gamma}{a_1^2}} \Delta X_A^{2,in} + \Delta P_L^{2,in} + \frac{(1 - e^{-\eta\frac{\Gamma}{a_1^2}})}{\Gamma} \langle f_{X_A} f_{X_A} \rangle, \tag{7.53}$$

$$\Delta P_A^{2,out} = (e^{-\eta\frac{\Gamma}{2a_1^2}} - e^{\frac{\eta}{2}\frac{\Gamma}{2a_1^2}})^2 \Delta P_A^{2,in} + \Delta X_L^{2,in} + \frac{(1 - e^{-\eta\frac{\Gamma}{a_1^2}})}{\Gamma} \langle f_{P_A} f_{P_A} \rangle. \tag{7.54}$$

In the limit where the decay vanishes we have that the input-output relations (7.50) and variances (7.53+54), become the original from chapter 2 (2.15-18). This is also what we

expected, but let us now consider the limit where the decay is substantial (but not too big to ruin the protocol) such that we may neglect the terms $e^{-\eta\frac{\Gamma}{a_1^2}}$. Then from (7.25+35) we see that for the b-terms $\frac{\langle f_X f_X\rangle}{\Gamma} = \frac{\langle f_P f_P\rangle}{\Gamma} = \frac{1}{2}$. Therefor this decay does not contribute to the X, P variances. It is reasonable, because if an atom decays to an auxillary level (we remember that the b terms described the processes that changed the spin state), it should not increase the noise in the quadratures - if we started in a minimum uncertainty state and some atoms "decay out" the rest atoms are still in the minimum uncertainty state. So for b-terms it is sensible to model the decay and noise by standard vacuum operators, as is done in [10]. And as one can easily see the same does not hold for the a terms, there the decay will created extra noise - we are no longer in the desired minimum uncertainty state. With some caution one can compare it to what is known in other areas of quantum information: that it is better to lose some part of your system, than to keep it complete, but decohered.
Also note that from (7.35) we see that there are no a_1^2 terms in the decay rate Γ, but we we do have that in the noise correlations (7.25). And actually there is also a non-vanishing contribution to the decay from the b_1 term, which also had a non-zero limit value (equal to the one for a_1). When putting in numbers one finds that the biggest contributions to the decay come from the b terms.
From the variances above we can calculate the fidelity that was given by:

$$\mathcal{F} = (\frac{1}{2} + \Delta X_A^{2,out})^{-\frac{1}{2}} \times (\frac{1}{2} + \Delta P_A^{2,out})^{-\frac{1}{2}}. \tag{7.55}$$

We remember that the fidelity was limited by 82%, but we naturally expect it to be lower due to decay. And as we said $\mathcal{F} = \frac{1}{2}$ is the border between succes or fail of the mapping protocol. Assuming the initial quadratures to be shot noise limited, we get that the fidelity is (to be consistent we have not assumed a constant value of κ, but used the formal expression (7.49)):

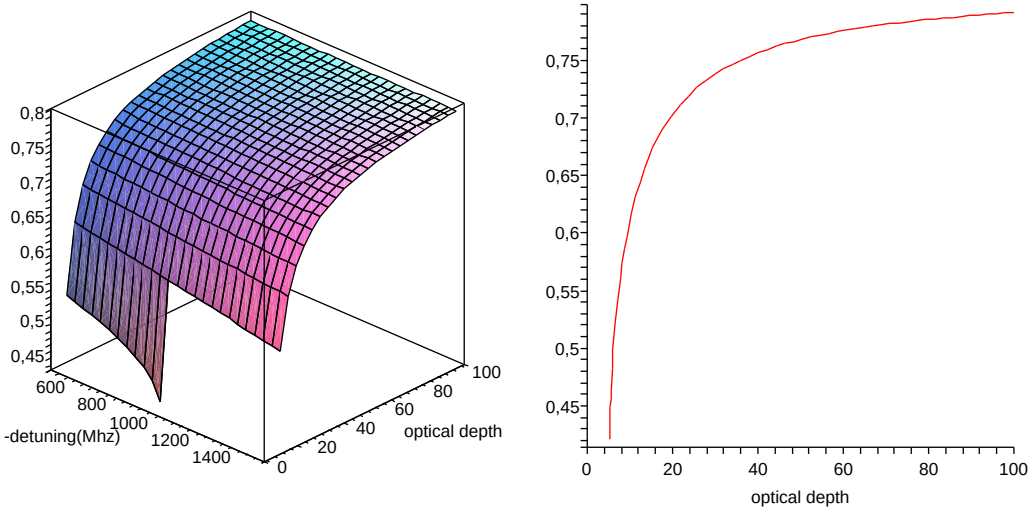

Figure 7.2: Left: $\mathcal{F}_0(\Delta, d)$ and Right: $\mathcal{F}(d)$ at $\Delta = -1$ GHz.

We see that the fidelity is almost independent of the detuning in the considered region, but we keep in mind that we have thrown away the a_2 terms, thereby commiting ourselves

to high detunings. Both the graphs for κ and fidelity show that the protocol can only work for optical depths above 6-7 (value at $\Delta = -1$ GHz). But once it works we (almost) immediately have $\mathcal{F} > \frac{1}{2}$, which was the succes criterion. The fidelity grows as we continue to increase the depth and becomes flat $d = 50$. In the asymptotic regime we approach the maximal value $\mathcal{F}_{max} \simeq 82\%$. Note that because we generally cannot eliminate P_A^{in} from the input-output relations, and there is no reason to squeeze X_A infinitely, because that way we increase the variance of P_A. In our calculations we have simply assumed minimum uncertainty ($\frac{1}{2}$) on both quadratures (as we also do for light), but small squeezings could increase the fidelity.

Orthogonal configuration ($\theta = \frac{\pi}{2}$)

Now we assume the atoms are prepared in a state with $j_y = F$, so the angle between the light, which is still x-polarized and atomic polarization is $\frac{\pi}{2}$. This is the other extreme - all other cases must lie within this and the previous. This time we define the canonical operators:

$$X_A = -\frac{J_x}{\sqrt{\langle J_y \rangle}}, P_A = \frac{J_z}{\sqrt{\langle J_y \rangle}}. \tag{7.56}$$

And one can check that this operators also obeys $[X, P] = i$ in the mean. We will skip the details, because the derivation is performed similarly to the parallel case and one ends up with a full equation that reads:

$$\begin{aligned}\frac{\partial}{\partial t}\begin{pmatrix} X_A \\ P_A \end{pmatrix}(t) = -\frac{\sigma_0 \gamma}{2A\Delta}\begin{pmatrix} a_1\sqrt{\langle J_y \rangle}\frac{1}{L}\int_0^L dz S_z(z,t) \\ +2(2F-1)a_2\langle S_x \rangle X_A(t) - (2F-1)a_2\sqrt{\langle J_y \rangle}\frac{1}{L}\int_0^L dz S_y(z,t) \end{pmatrix} \\ + \frac{1}{|a_1|}\sqrt{\frac{\eta}{T}}\begin{pmatrix} f_{X_A}(t) \\ f_{P_A}(t) \end{pmatrix} - \frac{\eta}{T}\frac{1}{2a_1^2}\begin{pmatrix} \Gamma_X X_A(t) \\ \Gamma_P P_A(t) \end{pmatrix}.\end{aligned} \tag{7.57}$$

This time our composite system does not posses the same symmetry as last time and it leads to the decays for X_A, P_A:

$$\Gamma_X = \frac{3}{2}a_1^2 + 119a_1a_2 - \frac{245}{2}a_2^2 + 21(b_1^2 + 2b_1b_2 + 13b_2^2), \tag{7.58}$$

$$\Gamma_P = \frac{1}{2}a_1^2 - 7a_1a_2 + \frac{105}{2}a_2^2 + 14(b_1^2 - 6b_1b_2 + 21b_2^2), \tag{7.59}$$

$$\Gamma = \frac{1}{2}(\Gamma_X + \Gamma_P) = a_1^2 + 56a_1a_2 - 35a_2^2 + \frac{7}{2}(5b_1^2 - 6b_1b_2 + 81b_2^2). \tag{7.60}$$

The decays are completely different, not even the b terms are the same. Moreover unlike last time we now have an a_1^2 term in the decays. Similarly the noise correlations are also different now:

$$\langle f_{X_A}(t) f_{X_A}(t') \rangle = \delta(t-t')\frac{1}{2}(9a_1^2 - 14a_1a_2 + 63a_2^2 + \frac{7}{2}(5b_1^2 - 6b_1b_2 + 81b_2^2)), \tag{7.61}$$

$$\langle f_{P_A}(t) f_{P_A}(t') \rangle = \delta(t-t')\frac{1}{2}(a_1^2 + 14a_1a_2 + 651a_2^2 + \frac{7}{2}(5b_1^2 - 6b_1b_2 + 81b_2^2)). \tag{7.62}$$

But they still have:

$$\langle [f_{X_A}(t), f_{P_A}(t')] \rangle = i\delta(t-t')\Gamma, \tag{7.63}$$

$$\langle \{f_{X_A}(t), f_{P_A}(t')\} \rangle = 0. \tag{7.64}$$

Besides that, they have changed a lot from last time, but interestingly the b part is common for the correlations of f_{X_A} and f_{P_A} and is half $\langle\Gamma\rangle$. We can plot these together with the ones for $\theta = 0$:

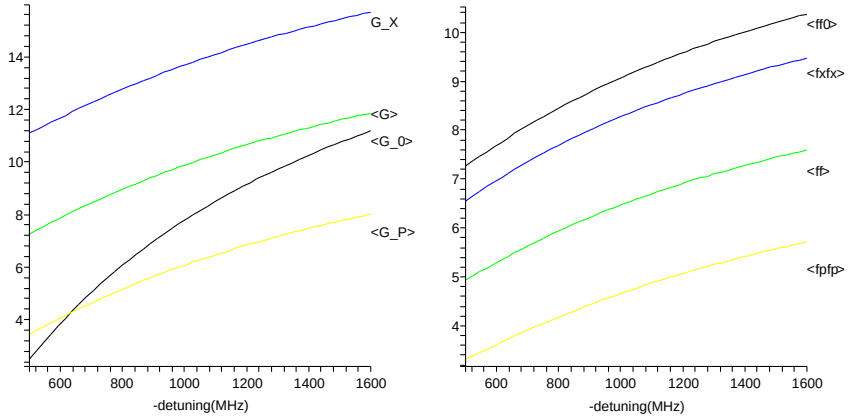

Figure 7.3: Left: Decays $\frac{\Gamma}{a_1^2}$ for $\theta = 0$(black) and $\theta = \frac{\pi}{2}$, Right: Noise correlations $\frac{\langle ff\rangle}{a_1^2}$ for $\theta = 0$(black) and $\theta = \frac{\pi}{2}$.

These graphs show the decay and the noise for the two cases we have analyzed so far. Even though it looks like both the decay and noise grows as we increase the detuning, we must remember that in the actual expressions we also have a $\frac{1}{\Delta^2}$ outside, which damps the decay contributions as we make Δ larger. We see that for $\theta = 0$ we have a bit smaller mean decay than for $\theta = \frac{\pi}{2}$. In the last case we have that Γ_X is more than 2 times as large as Γ_P. A larger Γ_X implies that we expect the critical (for which the protocol begins to work) optical depth to be higher than earlier. For the noise we see that interestingly all the noise correlations for $\theta = \frac{\pi}{2}$ are smaller than for $\theta = 0$. From first sight it can be surprising that even though our decay rates as whole are larger, then we still have less noise on the quadratures. And because the noise correlations are smaller we indeed will find a higher fidelity.
Now let us we return to the EOM. We can no longer make a unitary transformation in (7.57) to eliminate the X_A term from the coherent part of P_A, since this is no longer a Hermitian matrix we ought to make diagonal. This Zeeman shift can still be accounted for and we throw away this term, so we again can write:

$$\frac{\partial}{\partial t}\begin{pmatrix} X_A \\ P_A \end{pmatrix}(t) = \frac{\kappa}{L}\int_0^L dz \begin{pmatrix} \frac{S_z(z,t)}{\sqrt{\langle S_x\rangle T}} \\ -\frac{(2F-1)a_2}{a_1}\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}} \end{pmatrix} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\begin{pmatrix} f_{X_A}(t) \\ f_{P_A}(t) \end{pmatrix} - \frac{\eta}{T}\frac{1}{2a_1^2}\begin{pmatrix} \Gamma_X X_A(t) \\ \Gamma_P P_A(t) \end{pmatrix}. \tag{7.65}$$

So we see that for the coherent part we have a sign change for the coherent evolution of P_L relative to the former case, but the overall structure is the same. However we keep in mind that this time the noise operators and decay have changed asymmetrically and give different correlations. Solving the equation we get:

$$\begin{pmatrix} X_A^{out} \\ P_A^{out} \end{pmatrix} = \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{2a_1^2}} X_A^{in} \\ e^{-\eta\frac{\Gamma_P}{2a_1^2}} P_A^{in} \end{pmatrix} + \int_0^T dt \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{2a_1^2}\frac{(T-t)}{T}}[\frac{\kappa}{L}\int_0^L dz \frac{S_z(z,t)}{\sqrt{\langle S_x\rangle T}} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}f_{X_A}(t)] \\ e^{-\eta\frac{\Gamma_P}{2a_1^2}\frac{(T-t)}{T}}[-\frac{(2F-1)a_2}{a_1}\frac{\kappa}{L}\int_0^L dz \frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}f_{P_A}(t)] \end{pmatrix}. \tag{7.66}$$

Now to use the protocol we again ignore the a_2 terms in the coherent part and affect a mean damping of P_L:

$$\begin{pmatrix} X_A^{out} \\ P_A^{out} \end{pmatrix} = \begin{pmatrix} (e^{-\eta\frac{\Gamma_X}{2a_1^2}} X_A^{in} + \kappa e^{-\frac{\eta}{2}\frac{\Gamma_X}{2a_1^2}} P_L^{in}) \\ e^{-\eta\frac{\Gamma_P}{2a_1^2}} P_A^{in} \end{pmatrix} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\int_0^T dt \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{2a_1^2}\frac{T-t}{T}} f_{X_A}(t) \\ e^{-\eta\frac{\Gamma_P}{2a_1^2}\frac{T-t}{T}} f_{P_A}(t) \end{pmatrix}. \tag{7.67}$$

Performing the protocol and optimizing the parameter κ (we ignore the attenuation of light, meaning that g is always 1) we get for this case that to map P_L onto X_A we need:

$$\kappa \quad = \quad e^{-\frac{1}{2}\mathrm{LambertW}(-\frac{\Gamma_X}{4a_1^2 d})}. \tag{7.68}$$

This gives a graph that is very similar to the last one:

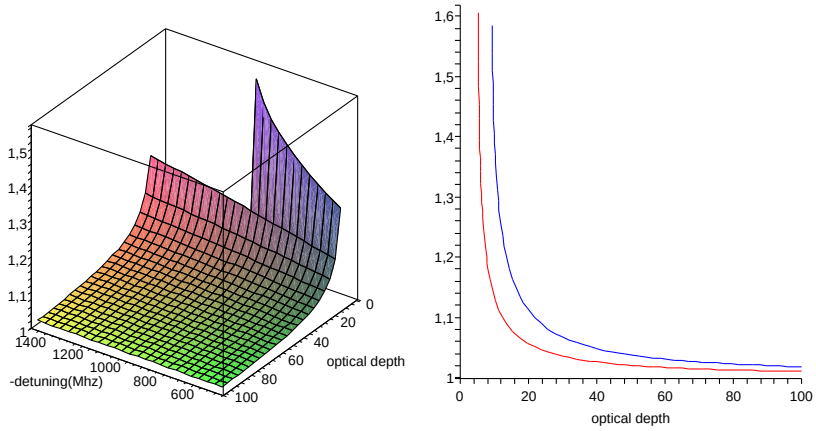

Figure 7.4: Left: $\kappa_{\frac{\pi}{2}}(-\Delta, d)$ for $\theta = \frac{\pi}{2}$, Right: κ_0(red) and $\kappa_{\frac{\pi}{2}}$(blue) at $\Delta = -1$ GHz.

Once again we see how κ diverges as $d \to 0$, while as we increase d we have κ close to 1. Again κ depends very weakly on the detuning and much more on d.
Again performing our memory protocol, we arrive at:

$$\begin{pmatrix} X_A^{out} \\ P_A'^{out} \end{pmatrix} = \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{2a_1^2}} X_A^{in} + P_L^{in} \\ (e^{-\eta\frac{\Gamma_P}{2a_1^2}} - e^{\frac{\eta}{2}\frac{\Gamma_X}{2a_1^2}}) P_A^{in} - X_L^{in} \end{pmatrix} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\int_0^T dt \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{2a_1^2}\frac{T-t}{T}} f_{X_A}(t) \\ e^{-\eta\frac{\Gamma_P}{2a_1^2}\frac{T-t}{T}} f_{P_A}(t) \end{pmatrix}. \tag{7.69}$$

The only difference from (7.50) is that the noise and decay for X_A,P_A are no longer the same. In the limit of vanishing decay we again obtain (2.13+14) and in the mean we again have accomplished the mapping (7.51+52). This time the variances are:

$$\begin{pmatrix} \Delta X_A^{2,out} \\ \Delta P_A^{2,out} \end{pmatrix} = \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{a_1^2}} \Delta X_A^{2,in} + \Delta P_L^{2,in} + \frac{(1-e^{-\eta\frac{\Gamma_X}{a_1^2}})}{\Gamma_X}\langle f_{X_A} f_{X_A}\rangle \\ (e^{-\eta\frac{\Gamma_P}{2a_1^2}} - e^{\frac{\eta}{2}\frac{\Gamma_X}{2a_1^2}})^2 \Delta P_A^{2,in} + \Delta X_L^{2,in} + \frac{(1-e^{-\eta\frac{\Gamma_P}{a_1^2}})}{\Gamma_P}\langle f_{P_A} f_{P_A}\rangle \end{pmatrix}. \tag{7.70}$$

This is almost what we found before, but now we have different decay rates and noise contributions for X and P because of the asymmetry. From the variances we can find the fidelity, which is:

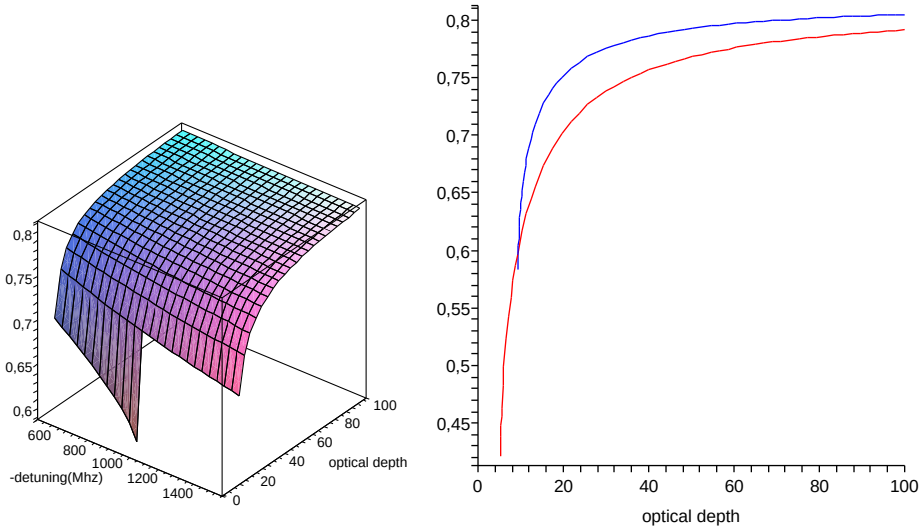

Figure 7.5: Left: $\mathcal{F}_{\frac{\pi}{2}}(-\Delta, d)$, Right: $\mathcal{F}_0(d)$ (red) and $\mathcal{F}_{\frac{\pi}{2}}(d)$ (blue) at $\Delta = -1$ GHz.

This time we see that the protocol only starts working for optical depths above 10. This is because X_A decays at a highter rate, as we saw in the fig 7.3. On the other hand, once it works it immediately gives $\mathcal{F} > \frac{1}{2}$ and the fidelities produced are higher than for $\theta = 0$. This can also be explained by the graphs from fig.7.3. Because even though we have to deal with higher decay rates, we on the other hand have smaller noise contributions, which is more important for the fidelity. At an optical depth around 20, the fidelity is around 0.75 and it continues to grow as we increase d, but is virtually unaffected by the detuning. Actually as was also the case last time, the fidelity is even slightly larger for smaller detunings. But we remember that for the protocol to be meaningful we needed small a_2 and therefor it is the higher detunings that interest us. In (7.58-62) we saw the decays and correlations explicitly and from them we see that both $\frac{\langle f_{X_A} f_{X_A} \rangle}{\Gamma_X}$ and $\frac{\langle f_{X_A} f_{X_A} \rangle}{\Gamma_X}$ are no longer $\frac{1}{2}$. Actually when putting in numbers one finds that the uncertainty for X in below shot noise, while for P it is above. This means that if we once again start in a minimum uncertainty state (and again neglect the $e^{-\Gamma}$ terms), then if the atoms decay to another spin state, it produces a squeezing of X_A and stretching of P_A. We conclude that the orthogonal configuration requires a higher critical depth (10), but overall gives higher fidelities (0-5%) than the parallel configuration.

General configuration

Now we will consider the general case with two orthogonal components of the spin $J_\perp$ and J_z and a parallel component $J_\parallel$, so:

$$X_A = \frac{J_\perp}{\sqrt{\langle J_\parallel \rangle}}, P_A = \frac{J_z}{\sqrt{\langle J_\parallel \rangle}}. \tag{7.71}$$

We can write the spin in terms of cartesian components:

$$J_x = \cos\theta J_\parallel - \sin\theta J_\perp, \tag{7.72}$$
$$J_y = \sin\theta J_\parallel + \cos\theta J_\perp. \tag{7.73}$$

We see that J_x,J_y are nothing but a rotation of the $(J_\parallel, J_\perp)$ by an angle θ around the z-axis. When $\theta = 0$ the atoms are polarized along the x-direction and the y-direction when $\theta = \frac{\pi}{2}$. So we can express the general X_A,P_A through:

$$X_A = \frac{\langle J_x\rangle J_y - \langle J_y\rangle J_x}{(\langle J_x\rangle^2 + \langle J_y\rangle^2)^{\frac{3}{4}}}, P_A = \frac{J_z}{(\langle J_x\rangle^2 + \langle J_y\rangle^2)^{\frac{1}{4}}}. \tag{7.74}$$

It is seen that they fulfill:

$$\begin{aligned}[X_A, P_A] &= \frac{1}{\langle J_x\rangle^2 + \langle J_y\rangle^2}(\langle J_x\rangle[J_y, J_z] - \langle J_y\rangle[J_x, J_z]) \\ &= \frac{i}{\langle J_x\rangle^2 + \langle J_y\rangle^2}(\langle J_x\rangle J_x + \langle J_y\rangle J_y).\end{aligned} \tag{7.75}$$

So on average we have again that $\langle[X_A, P_A]\rangle = i$. Now the analysis in the general case is very parallel to the two special cases $\theta = 0$ and $\theta = \frac{\pi}{2}$, only now the evolution reads:

$$\dot{X_A} = \frac{\dot{J_\perp}}{\sqrt{\langle J_\parallel\rangle}} - \frac{1}{2}\frac{J_\perp}{\sqrt{J_\parallel}}\frac{\langle\dot{J_\parallel}\rangle}{\langle J_\parallel\rangle}, \tag{7.76}$$

$$\dot{P_A} = \frac{\dot{J_z}}{\sqrt{\langle J_\parallel\rangle}} - \frac{1}{2}\frac{J_z}{\sqrt{J_\parallel}}\frac{\langle\dot{J_\parallel}\rangle}{\langle J_\parallel\rangle}. \tag{7.77}$$

The most notable difference is that when we linearize the atomic operators we obtain an angular dependence:

$$\frac{\partial}{\partial t}j_\parallel = -\frac{2\sigma_0\gamma}{A\Delta}(2F-1)a_2\sin(2\theta)(-\frac{1}{2}\cos\theta j_\parallel S_y + S_x j_z), \tag{7.78}$$

$$\frac{\partial}{\partial t}j_\perp = -\frac{2\sigma_0\gamma}{A\Delta}(a_1 j_\parallel S_z + (2F-1)a_2(\frac{1}{2}\sin\theta\sin(2\theta)j_\parallel S_y + 2\cos^2\theta S_x j_z), \tag{7.79}$$

$$\frac{\partial}{\partial t}j_z = -\frac{2\sigma_0\gamma}{A\Delta}(2F-1)a_2(\cos(2\theta)j_\parallel S_y - \frac{1}{2}\sin(2\theta)j_\parallel S_x - 2\cos(2\theta)S_x j_\perp). \tag{7.80}$$

Here we have kept those terms where at least one of the variables has a big mean value ($j_\parallel$ or S_x), but the full equations can be found in App.B.2. The equations for the momenta are quite intricate and have many terms that go as $\sin(2\theta)$, which we therefore did not see in the parallel and orthogonal case.

We really want the mean spin to be fixed during the interaction, but according to (7.78) it is not the case in general. Even though the magnitude $\langle j_\parallel\rangle$ might be preserved or change by a only small amount, the fact that the right-hand side of (7.78) is non-zero, means that we at best only have a rotation of $j_\parallel$. This however is already bad enough and is the first indication of that we really are only interested in the choices $\theta = 0, \frac{\pi}{2}$.

As we expected, the Faraday term (a_1) in (7.78) does not depend on θ, because it described hos the z component of atomic spin and S_z coupled to each other, while θ is an angle in the xy-plane. Furthermore we see from (7.79) and (7.80) how the much the a_2 terms generally complicate the interaction. The last term in (7.79) and (7.80) is the Zeeman shift and as we saw for the orthogonal case, it as a rule is not identical for the two spin components. Nonetheless the biggest problem is the second term in (7.80). Both S_x and $j_\parallel$ (when we change to collective spin) are very big quantities, meaning that the product $S_x j_\parallel$ will produce a gigantic drift of j_z, which we want to be conserved. Again this can be solved (besides setting $a_2 = 0$), by having $\theta = 0, \frac{\pi}{2}$.

Keeping in mind that the coherent interaction is the central, our discussion leads to the conclusion that for the interaction to be as close to the Faraday form we (besides having big detunings) really want $\theta = 0$ or $\theta = \frac{\pi}{2}$. As a consequence of that we could stop with the general considerations here and solely rely on the results from the parallel- and ortogonal configuration. But there are some interesting points worth noting for the noise and decay, so we continue.
First we look at the evolution of X_A and P_A, where we neglect the changes in $j_\parallel$:

$$\begin{aligned}\frac{\partial}{\partial t}\begin{pmatrix}X_A\\P_A\end{pmatrix}(t) &= \kappa\{\frac{1}{L}\int_0^L dz\begin{pmatrix}\frac{S_z(z,t)}{\sqrt{\langle S_x\rangle T}}+\frac{(2F-1)a_2}{a_1}\sin\theta\sin(2\theta)\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}}\\ \frac{2(2F-1)a_2}{a_1}(\cos(2\theta)\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}}-\sin(2\theta)\frac{S_x}{\sqrt{\langle S_x\rangle T}})\end{pmatrix}\\ &+\frac{2(2F-1)a_2}{a_1}\sqrt{\frac{\langle S_x\rangle}{\langle J_\parallel\rangle}}\begin{pmatrix}\cos^2\theta P_A(t)\\-\cos(2\theta)X_A(t)\end{pmatrix}\}.\end{aligned} \tag{7.81}$$

Ignoring the Zeeman shift, gives the solution:

$$\begin{pmatrix}X_A^{out}\\P_A^{out}\end{pmatrix}=\begin{pmatrix}X_A^{in}\\P_A^{in}\end{pmatrix}+\kappa\frac{1}{L}\int_0^L dz\begin{pmatrix}P_L(z)+\frac{(2F-1)a_2}{a_1}\sin\theta\sin(2\theta)X_L(z)\\ \frac{2(2F-1)a_2}{a_1}(\cos(2\theta)X_L(z)-\sqrt{\langle S_x\rangle T}\sin(2\theta))\end{pmatrix}. \tag{7.82}$$

The last term in the expression of P_A^{out} is very big since $\langle S_x\rangle T \simeq N_p$ and it means that unless $\theta = 0$ or $\frac{\pi}{2}$, it practically erases any information we want to have in P_A by adding this big quantity. Choosing $a_2 = 0$ we return to the ideal relations.

We can find the noise operators in the general case, by using that $j_\perp = -\sin\theta j_x + \cos\theta j_y$ and that we in this chapter already have discussed how they were formed for the different Cartesian spin components. As with the former cases using the g elements from App.C.3. one can construct the noise operators:

$$\begin{pmatrix}f_{X_A}\\f_{P_A}\end{pmatrix} = \frac{1}{\sqrt{\langle J_\parallel\rangle}}\int_0^L\begin{pmatrix}(F_\perp+F_\perp^\dagger)(z)\\(F_z+F_z^\dagger)(z)\end{pmatrix}\rho A dz, \tag{7.83}$$

where the noise operator F_i was defined in (7.21) and $F_\perp = -\sin\theta F_x + \cos\theta F_y$. One could fear that the correlations between these operators would be very complicated due to the θ dependence, both explicitly from the definition of $F_\perp$ and implicitly when we should evaluate the elements of g_i^2. But as in the former cases due to the assumption of light being strongly polarized along x we will only keep the (x,x) terms of the decay and the noise correlations. After some calculations where we use the values of g^2 from App.C.3. and the linearizations from B.1, we find that the result is quite simple:

$$\langle f_{X_A}(t)f_{X_A}(t')\rangle(\theta) = \delta(t-t')(\langle f_{X_A}f_{X_A}\rangle(0)\cos^2\theta + \langle f_{X_A}f_{X_A}\rangle(\frac{\pi}{2})\sin^2\theta), \tag{7.84}$$

$$\langle f_{P_A}(t)f_{P_A}(t')\rangle(\theta) = \delta(t-t')(\langle f_{P_A}f_{P_A}\rangle(0)\cos^2\theta + \langle f_{P_A}f_{P_A}\rangle(\frac{\pi}{2})\sin^2\theta). \tag{7.85}$$

Apparently the noise correlations can be written as $f(\theta) = f(0)\cos^2\theta + f(\frac{\pi}{2})\sin^2\theta$. Thinking about it, it is quite reasonable. Our quantities should be invariant under rotations of π and moreover symmetry implies the quantities to be the same for $\pm\theta$, which makes the form above quite plausible. That allows us to plot the different noise correlations:

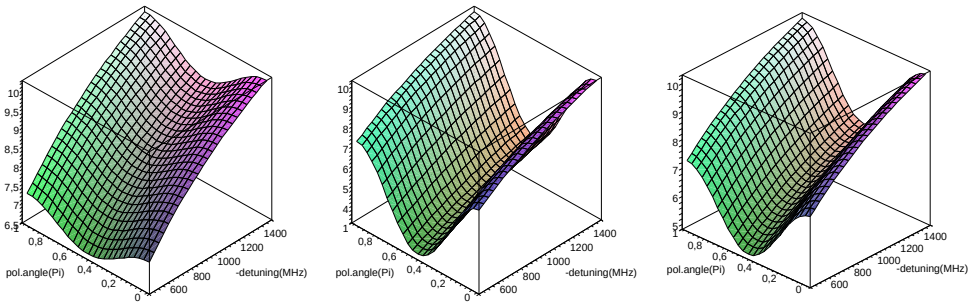

Figure 7.6: Left:$\frac{\langle f_X f_X \rangle}{a_1^2}(-\Delta,\theta)$, Center:$\frac{\langle f_P f_P \rangle}{a_1^2}(-\Delta,\theta)$, Right:$\frac{\langle f f \rangle}{a_1^2}(-\Delta,\theta)$.

We see that for a give detuning, both the X-and P noise correlations are smooth functions of θ. They have minimum for orthogonal configuration and maximum for parallel-, which is also confirmed in fig.7.3. So loosely stated we get most noise for $\theta = 0$ and least noise for $\theta = \frac{\pi}{2}$.

To construct the decay of X, P in the general case we follow the same path as for the noise. We express $j_\perp$ and $j_\parallel$ in terms of $\cos\theta, \sin\theta$ combinations of j_x and j_y and use the (x,x) elements of the decay matrices ξ_i in App.C.4. That way we can find the decay of $J_\parallel, J_\perp$ and J_z, and again we find the general form to be a harmonic combination of $\theta = 0$ and $\theta = \frac{\pi}{2}$:

$$\Gamma_\parallel(\theta) = \Gamma_x(0)\cos^2\theta + \Gamma_y(\frac{\pi}{2})\sin^2\theta, \tag{7.86}$$

$$\Gamma_\perp(\theta) = \Gamma_y(0)\cos^2\theta + \Gamma_x(\frac{\pi}{2})\sin^2\theta, \tag{7.87}$$

$$\Gamma_z(\theta) = \Gamma_z(0)\cos^2\theta + \Gamma_z(\frac{\pi}{2})\sin^2\theta. \tag{7.88}$$

Here each of components $\Gamma_i, i = x, y, z$ is obtained from the corresponding ξ_i according to (7.32). From (7.76+77) we form the decay of the quadratures:

$$\Gamma_X = \Gamma_\perp - \frac{1}{2}\Gamma_\parallel = \Gamma_X(0)\cos^2\theta + \Gamma_X(\frac{\pi}{2})\sin^2\theta, \tag{7.89}$$

$$\Gamma_P = \Gamma_z - \frac{1}{2}\Gamma_\parallel = \Gamma_P(0)\cos^2\theta + \Gamma_P(\frac{\pi}{2})\sin^2\theta. \tag{7.90}$$

The following graphs show the decays for different angles:

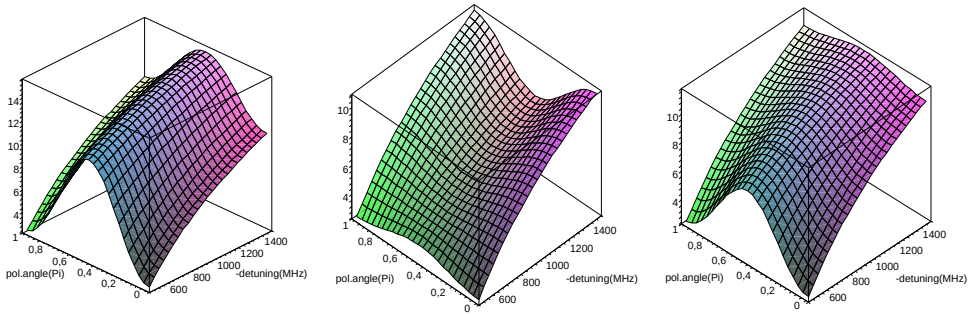

Figure 7.7: Left:$\frac{\Gamma_X}{a_1^2}(-\Delta,\theta)$, Center:$\frac{\Gamma_P}{a_1^2}(-\Delta,\theta)$, Right:$\frac{\langle\Gamma\rangle}{a_1^2}(-\Delta,\theta)$.

The first graph shows that X_A decays slowest at $\theta = 0$ and fastest $\theta = \frac{\pi}{2}$. From the second graph we see that the situation is reversed for P_A, which decays fastest for $\theta = 0$. Because Γ_X overall is larger than Γ_P, we get that the mean decay $\langle\Gamma\rangle$ has the same extremal points as Γ_X, but for increasing detuning the angular dependence disappears. Again this we also saw earlier in Fig.7.3.
We remember that the b part of Γ_X was the same for both configurations and together with (7.68) it means that it is independent of the polarization angle. Possibly we could also have guessed that it was so - after all we decay "out", so how precisely our atoms are oriented relative to the polarization of the light field should not matter. But on the other hand, we remember that the same did not hold for Γ_P, the b part of the decay was different in the two situations. The fact that we have some kind of angular dependence for Γ_P must mean that we are dealing with a kind of interference effect. The decay interference comes from the interference between the possible decay paths and is not something new - it can for instance serve to induce transparency as described in [22].
When it comes to the b parts of the noise correlations, we had in both cases that $\langle f_X f_X\rangle = \langle f_P f_P\rangle = \frac{1}{2}\langle\Gamma^b\rangle$, which therefor must hold for all θ. But note that they can only be directly related to the mean decay rate, not the individual Γ_X and Γ_P. We have not talked so much about the a-terms, because they have no simple relations between noise correlations and decay.
Together with the noise correlations above we have seen that there somehow is a tradeoff between noise correlations and decay, if you want one of them to be smaller, the other one grows correspondingly. Noise is smallest for orthogonal configuration, while we have least decay for parallel.

Now we are ready to join the results in the full general EOM. Including the noise and decay in (7.80) we get without the Zeeman shift:

$$\begin{aligned}\frac{\partial}{\partial t}\begin{pmatrix}X_A\\P_A\end{pmatrix}(t) &= \frac{\kappa}{L}\int_0^L dz\begin{pmatrix}\frac{S_z(z,t)}{\sqrt{\langle S_x\rangle T}} + (2F-1)a_2\sin\theta\cos(2\theta)\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}}\\ 2(2F-1)a_2(\cos(2\theta)\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}} - \sin(2\theta)\frac{S_x}{\sqrt{\langle S_x\rangle T}})\end{pmatrix}\\ &\quad+\sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\begin{pmatrix}f_{X_A}\\f_{P_A}\end{pmatrix}(t) - \frac{\eta}{2a_1^2T}\begin{pmatrix}\Gamma_X X_A\\\Gamma_P P_A\end{pmatrix}(t).\end{aligned} \tag{7.91}$$

Once again the solution is of the form:

$$\begin{aligned}\begin{pmatrix}X_A^{out}\\P_A^{out}\end{pmatrix} &= \begin{pmatrix}e^{-\eta\frac{\Gamma_X}{2a_1^2}}X_A^{in}\\e^{-\eta\frac{\Gamma_P}{2a_1^2}}P_A^{in}\end{pmatrix}\\ &\quad+\int_0^T dt\begin{pmatrix}e^{-\eta\frac{\Gamma_X}{2a_1^2}\frac{T-t}{T}}[\frac{\kappa}{L}\int_0^L dz a_1\frac{S_z(z,t)}{\sqrt{\langle S_x\rangle T}} + (2F-1)a_2\sin\theta\cos(2\theta)\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}f_{X_A}]\\ e^{-\eta\frac{\Gamma_P}{2a_1^2}\frac{T-t}{T}}[\frac{\kappa}{L}\frac{2(2F-1)a_2}{a_1}\int_0^L dz\cos(2\theta)\frac{S_y(z,t)}{\sqrt{\langle S_x\rangle T}} - \sin(2\theta)\frac{S_x}{\sqrt{\langle S_x\rangle T}} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}f_{P_A}]\end{pmatrix}.\end{aligned} \tag{7.92}$$

To use the protocol we again ignore the a_2 terms in the coherent part and assume a mean damping of P_L:

$$\begin{pmatrix}X_A^{out}\\P_A^{out}\end{pmatrix} = \begin{pmatrix}(e^{-\eta\frac{\Gamma_X}{2a_1^2}}X_A^{in} + \kappa e^{-\frac{\eta}{2}\frac{\Gamma_X}{2a_1^2}}P_L^{in})\\ e^{-\eta\frac{\Gamma_P}{2a_1^2}}P_A^{in}\end{pmatrix} + \sqrt{\frac{\eta}{T}}\frac{1}{|a_1|}\int_0^T dt\begin{pmatrix}e^{-\eta\frac{\Gamma_X}{2a_1^2}\frac{T-t}{T}}f_{X_A}(t)\\ e^{-\eta\frac{\Gamma_P}{2a_1^2}\frac{T-t}{T}}f_{P_A}(t)\end{pmatrix}. \tag{7.93}$$

The resulting equation is identical to the one for $\theta = \frac{\pi}{2}$, only now our decay and noise have an angular dependency. Again performing the protocol and optimizing κ ($g = 1$) we get that κ should be as in (7.59). This time we choose to plot κ as a function of θ and d and set the detuning to -1 GHz:

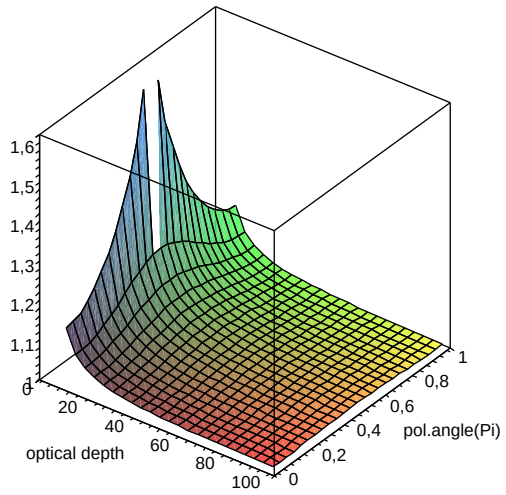

Figure 7.8: $\kappa(d, \theta)$ at $\Delta = -1$ GHz.

For high optical depths κ is close to 1, no matter what θ is. But for small depths κ diverges and the closer the angle is to $\frac{\pi}{2}$ the more pronounced is the divergence. With this setting of κ the protol yields:

$$\begin{pmatrix} X_A^{out} \\ P_A'^{out} \end{pmatrix} = \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{2a_1^2}} X_A^{in} + P_L^{in} \\ (e^{-\eta\frac{\Gamma_P}{2a_1^2}} - e^{\frac{\eta}{2}\frac{\Gamma_X}{2a_1^2}}) P_A^{in} - X_L^{in} \end{pmatrix} + \sqrt{\frac{\eta}{T}} \frac{1}{|a_1|} \int_0^T dt \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{2a_1^2}\frac{T-t}{T}} f_{X_A}(t) \\ e^{-\eta\frac{\Gamma_P}{2a_1^2}\frac{T-t}{T}} f_{P_A}(t) \end{pmatrix}. \tag{7.94}$$

One sees that we again for vanishing atomic input, have succesfully performed the mapping:

$$\langle X_A^{out} \rangle = \langle P_L^{in} \rangle, \tag{7.95}$$

$$\langle P_A^{out} \rangle = -\langle X_L^{in} \rangle. \tag{7.96}$$

The variances are also formally the same as for $\theta = \frac{\pi}{2}$:

$$\begin{pmatrix} \Delta X_A^{2,out} \\ \Delta P_A^{2,out} \end{pmatrix} = \begin{pmatrix} e^{-\eta\frac{\Gamma_X}{a_1^2}} \Delta X_A^{2,in} + \Delta P_L^{2,in} + \frac{(1-e^{-\eta\frac{\Gamma_X}{a_1^2}})}{\Gamma_X} \langle f_{X_A} f_{X_A} \rangle \\ (e^{-\eta\frac{\Gamma_P}{2a_1^2}} - e^{\frac{\eta}{2}\frac{\Gamma_X}{2a_1^2}})^2 \Delta P_A^{2,in} + \Delta X_L^{2,in} + \frac{(1-e^{-\eta\frac{\Gamma_P}{a_1^2}})}{\Gamma_P} \langle f_{P_A} f_{P_A} \rangle \end{pmatrix}, \tag{7.97}$$

and inserting these into the expression for the fidelity we get the fidelity as a function of optical depth and polarization angle:

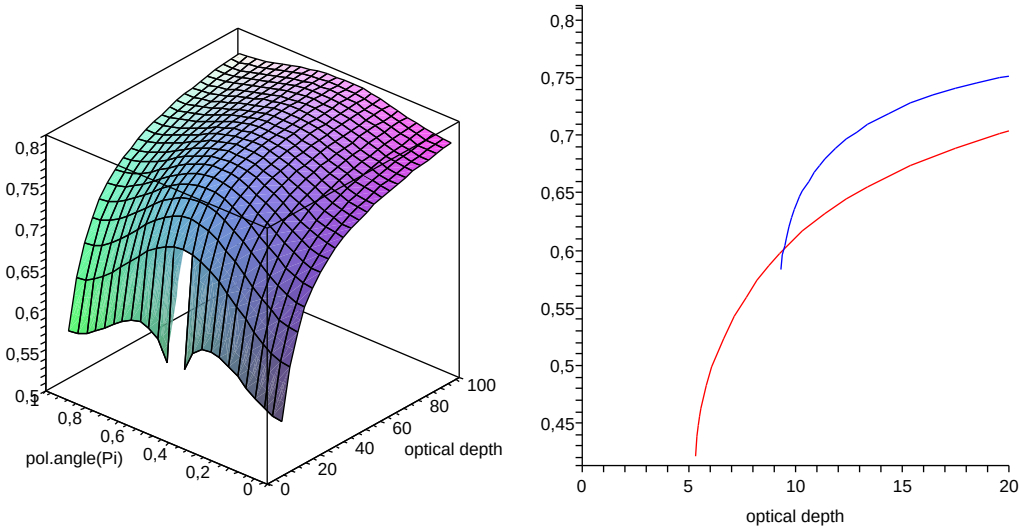

Figure 7.9: Left: $\mathcal{F}(d,\theta)$ at $\Delta = -1$ GHz, Right: $\mathcal{F}_0(d)$(red) and $\mathcal{F}_{\frac{\pi}{2}}(d)$(blue) at $\Delta = -1$ GHz.

The graph shows that the fidelity does not depend so much on θ, but much more on d. For optical depths above 10 we can make the protocol work for all angles. The fidelity has a small bulge for $\theta = \frac{\pi}{2}$, but as we increase d it smears out and becomes flat around $d = 50$. Increasing d further we approach the maximal fidelity of 82%.

In this chapter we have found the fidelity $\mathcal{F}$ as a continuos function of polarization angle θ and optical depth d. During that we have stressed that we actually only have the choices $\theta = 0$ or $\theta = \frac{\pi}{2}$. The reason is that for general θ we encountered big drift terms going as $\sin(2\theta)$ that vanish only for these two settings. And unless one has optical depths below 10 (in principle the protocol should work for optical depths as low as 6-7 for the parallel configuration), the highest fidelity is achieved for $\theta = \frac{\pi}{2}$, where it is about 0.60 at $d = 10$ and grows as d is increased and is 0.75 already at $d = 20$.

Chapter 8

Conclusion

Let us now sum up what we have done in this thesis. Our work has been motivated by the direct mapping protocol, which builds on the Faraday interaction. To understand the system we have set up an effective dipole Hamiltonian that describes the interaction of atoms with highly off-resonant light. We have included the full level structure of the atoms and treated the light field in a simple way, assuming the transverse mode to be constant and having high Fresnel numbers to avoid diffraction effects. Moreover we have assumed it to have a strong classical field along x and a weak quantum field along y. From this Hamiltonian we have looked at the dynamics of light- and atomic variables and seen how one can include the spontaneous emission arising from couplings to environment. For the atoms we have sketched how one extends the dynamics from a single atom to a whole ensemble of atoms.

After that we were ready to investigate the equations of motion for the quadratures. We studied the cases where the atomic ensemble and light have parallel- and orthogonal polarizations. We have explicitly calculated the amount of noise and decay in these cases and seen that for the mapping protocol to work, one generally needs optical depths above 10 (for the parallel configuration we have found that in principle it should still work for optical depths around 6-7). The highest fidelity can be achieved by having atoms polarized orthogonal to light although the difference from the parallel is just a few percent. The fidelities we find are well above the limit $\frac{1}{2}$, rendering the protocol in theory to be useful and for optical depths around 20 one should be able to reach fidelities around 0.75. Of course our assumptions of having everything all interesting operators shot noise limited is difficult to realize in real lifem, so one can expect the actual fidelities to be lower.

Besides the two extremal situations, we have also looked at a general angle between the polarization of light and atoms. Our calculations show that there is no reason to have other configurations of light and atoms, because otherwise the protocols get distorted by big drift terms.

Generally we find that a simple way to increase the fidelity, is to increase the optical depth. That could be done by increasing the concentration of atoms, but as we have seen in chapter 5 then one can get in trouble because then the dipole-dipole interaction between neighbouring atoms can no longer be ignored and the simple model breaks down. How much the concentration can be increased is likely to be guided by the experiments at hand. The noise correlations and decays we have calculated include the full level structure of cesium and these numbers should for the first time give a realistic theoretical comparison with the measured values. From the elements in App.C one can find even more correlations than we have needed.

So what future analysis could be worth making? For instance one could try other atomic transitions. Most of the time we have only been looking at $F = 4$, one could go through the same calculations and check whether there is less noise for $F = 3$. Of course we could also look at the D1 line, but there the problem is that the a_2 terms are bigger and since the whole protocol builds on the assumption that $a_2 \to 0$, it is not likely to be a better candidate. Most of the theoretical framework we have seen can also be carried over to other alkali atoms. Here we mainly think of rubidium, which a recent paper describes [16]. A degree of freemdom we have not exploited is the squeezing of the atomic input quadratures X_A and P_A. They could also be important tools for increasing the fidelity. For instance we have seen how in the pure Faraday interaction one could increase the fidelity by squeezing X_A. Once we introduced decay, it was no longer desirable to have infinite squeezing, but it could be worth determining how much squeezing is optimal for the fidelity with included decoherence.
Finally it could also be worth to set up a theory that works as the detunings approach zero. In our framework the noise and decay diverges at that point. Possibly it could be done by waiting with the adiabatic elimination, but the precise path is yet to be found, even though many things already fit well in [10].

Appendix A

Light

A.1 Radiation theory

We make a little detour into the general theory of radiation to remind ourselves of the basic concepts. As always when dealing with electromagnetic fields the natural starting point are Maxwell's equations:

$$\begin{aligned} \boldsymbol{\nabla} \times \mathbf{E} + \frac{\partial \mathbf{B}}{\partial t} &= 0, & \text{(A.1)} \\ \boldsymbol{\nabla} \times \mathbf{H} - \frac{\partial \mathbf{D}}{\partial t} &= \mathbf{J}, & \text{(A.2)} \\ \boldsymbol{\nabla} \cdot \mathbf{D} &= \sigma, & \text{(A.3)} \\ \boldsymbol{\nabla} \cdot \mathbf{B} &= 0. & \text{(A.4)} \end{aligned}$$

Here $\mathbf{J}$ is the current density, while σ is the free charge density, which generates our fields. In vacuum they are zero, while $\mathbf{D} = \epsilon_0 \mathbf{E}$, $\mathbf{H} = \mu_0 \mathbf{B}$. A standard way of expressing the fields, is to exploit the forms of the equations and write the electric and magnetic field as:

$$\mathbf{E} = -\boldsymbol{\nabla}\phi - \frac{\partial \mathbf{A}}{\partial t}, \mathbf{B} = \boldsymbol{\nabla} \times \mathbf{A}. \tag{A.5}$$

As one can check Maxwell's equations are invariant under gauge transformations, so if we make the transformations:

$$\phi' = \phi - \frac{\partial \chi}{\partial t}, \mathbf{A}' = \mathbf{A} + \boldsymbol{\nabla}\chi. \tag{A.6}$$

(here χ is any given scalar function of position and space), Maxwell's equation remain unchanged, that is $\mathbf{A}'$ and ϕ' also fulfill them if $\mathbf{A}$ and ϕ do. We will adopt the Coulomb gauge for which we set $\phi = 0$ and $\boldsymbol{\nabla} \cdot \mathbf{A} = 0$, so the field equations simplify:

$$\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t}, \mathbf{B} = \boldsymbol{\nabla} \times \mathbf{A}. \tag{A.7}$$

So the vector potential uniquely determines the electric- and magnetic field. Using the vector identity $\boldsymbol{\nabla} \times (\boldsymbol{\nabla} \times \mathbf{A}) = \boldsymbol{\nabla}(\boldsymbol{\nabla} \cdot \mathbf{A}) - \nabla^2 \mathbf{A}$ we get the wave equation:

$$(\nabla^2 - \frac{\partial}{\partial t^2})\mathbf{A} = 0. \tag{A.8}$$

In open space we write the potential as:

$$\mathbf{A}(\mathbf{r}, t) = \sum_{k\sigma} \boldsymbol{\epsilon}_{k\sigma} \sqrt{\frac{1}{2\epsilon_0 V \omega_k}} (a_{k\sigma} e^{i(\mathbf{k}\cdot\mathbf{r} - \omega_k t)} + a_{k\sigma}^\dagger e^{-i(\mathbf{k}\cdot\mathbf{r} - \omega_k t)}). \tag{A.9}$$

From which the fields follow according to (11.7):

$$\mathbf{E}(\mathbf{r},t) = i\sum_{k\sigma}\boldsymbol{\epsilon}_{k\sigma}\sqrt{\frac{\omega_k}{2\epsilon_0 V}}(a_{k\sigma}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} - a^\dagger_{k\sigma}e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}), \tag{A.10}$$

$$\mathbf{B}(\mathbf{r},t) = i\sum_{k\sigma}\hat{\mathbf{k}}\times\boldsymbol{\epsilon}_{k\sigma}\sqrt{\frac{\omega_k}{2\epsilon_0 V}}(a_{k\sigma}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} - a^\dagger_{k\sigma}e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}). \tag{A.11}$$

Here $\hat{\mathbf{k}} = \mathbf{k}/|\mathbf{k}|$ and we will employ the useful separation:

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}^{(+)}(\mathbf{r},t) + \mathbf{E}^{(-)}(\mathbf{r},t). \tag{A.12}$$

Here $\mathbf{E}^{(+)}$ contains the annihilation operators that oscillate as $e^{-i\omega_k t}$ and $\mathbf{E}^{(-)}(\mathbf{r},t) = (\mathbf{E}^{(+)}(\mathbf{r},t))^\dagger$ contains the creation operators oscillating with the conjugate frequency. Another common way of writing the electric field omits the i and corresponds to an overall phase change, so:

$$\mathbf{E}(\mathbf{r},t) = \sum_{k\sigma}\boldsymbol{\epsilon}_{k\sigma}\sqrt{\frac{\omega_k}{2\epsilon_0 V}}(a_{k\sigma}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)} + a^\dagger_{k\sigma}e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}). \tag{A.13}$$

This is the form of the electric field that we will use in this work. We will not need the magnetic field for our purpose.

A.2 Stokes operators

The Stokes operators are very practical for descriptions of light and are more easy to measure from an experimental point of view, than the creation/annihilation operators. It is also useful to introduce the circular annihilation (and creation) operators:

$$a_+ = -\frac{a_x - ia_y}{\sqrt{2}}, \tag{A.14}$$

$$a_- = \frac{a_x + ia_y}{\sqrt{2}}. \tag{A.15}$$

They describe light having right (a_+) - or left (a_-) circular polarization. The Stokes operators can be seen as a new operator basis, where S_x gives the number of photons polarized along $\mathbf{e}_x$ minus those along $\mathbf{e}_y$, S_y the number of photons polarized along $\mathbf{e}_{+45}$ minus those along $\mathbf{e}_{-45}$ and finally S_z gives the difference between σ_+- and σ_- polarizations. They can be expressed in terms of the linear and circular operators:

$$S_x = \frac{1}{2}(a^\dagger_+ a_- + a^\dagger_- a_+) = \frac{1}{2}(a^\dagger_x a_x - a^\dagger_y a_y), \tag{A.16}$$

$$S_y = \frac{i}{2}(a^\dagger_- a_+ - a^\dagger_+ a_-) = \frac{1}{2}(a^\dagger_x a_y + a^\dagger_y a_x), \tag{A.17}$$

$$S_z = \frac{1}{2}(a^\dagger_+ a_+ - a^\dagger_- a_-) = \frac{1}{2i}(a^\dagger_x a_y - a^\dagger_y a_x). \tag{A.18}$$

The Stokes operators obey the canonical commutation relation:

$$[S_i, S_j] = i\sum_k \varepsilon_{ijk} S_k. \tag{A.19}$$

If light is strongly polarized along the x-axis we may treat the operator a_x as a classical number A_x and now we may write the Stokes operators:

$$S_x \approx \frac{A_x^2}{2}, \tag{A.20}$$

$$S_y \approx \frac{A_x}{2}(a_y + a_y^\dagger), \tag{A.21}$$

$$S_z \approx \frac{A_x}{2i}(a_y - a_y^\dagger). \tag{A.22}$$

So all the interesting properties of the light are encoded in the weak y-polarized mode. Finally we note that we also will use the total number of photons:

$$\phi = a_+^\dagger a_+ + a_-^\dagger a_- = a_x^\dagger a_x + a_y^\dagger a_y. \tag{A.23}$$

In the thesis our Stokes operators are actually fluxes, so they instead count the number of photons per time.

A.3 Transformation of the light EOM

We will work in the paraxial approximation assuming a flat transverse profile and write the forward electric field as:

$$\begin{aligned}\mathbf{E}_{\mathrm{F}}(z,t) &= \sum_{k\sigma} \boldsymbol{\epsilon}_\sigma \sqrt{\frac{\omega_0}{2\epsilon_0 V}}(a_{k\sigma}(t)e^{ikz} + a_{k\sigma}^\dagger(t)e^{-ikz}), \\ &= |E| \sum_\sigma \boldsymbol{\epsilon}_\sigma (a_\sigma(z,t) + a_\sigma^\dagger(z,t)).\end{aligned} \tag{A.24}$$

Here $|E| = \sqrt{\frac{\omega_0}{2\epsilon_0 A}}$ and we have defined the space dependent operators $a_\sigma(z,t) = \frac{1}{\sqrt{L}} \sum_k a_{k\sigma}(t)e^{ikz}$, where it is assumed that the different k are close to k_0, so the operators oscillate at the common frequency $\omega_0 = |k_0|$. For the radiation field we have that:

$$H_{\mathrm{L}} = \sum_{k\sigma} \omega_k a_{k\sigma}^\dagger a_{k\sigma}. \tag{A.25}$$

Now we are to ready to form the EOM:

$$\frac{\partial}{\partial t} a_\sigma(z,t) = i[H_{\mathrm{int}} + H_{\mathrm{L}}, a_\sigma(z,t)], \tag{A.26}$$

$$[H_{\mathrm{L}}, a_\sigma(z,t)] = \frac{1}{\sqrt{L}} \sum_k [H_{\mathrm{L}}, a_{k\sigma}(t)]e^{ikz} = -\frac{1}{\sqrt{L}} \sum_k \omega_k a_{k\sigma}(t)e^{ikz}. \tag{A.27}$$

But also have from the explicit z-dependence that:

$$\frac{\partial}{\partial z} a_\sigma(z,t) = \frac{1}{\sqrt{L}} \sum_k ika_{k\sigma}(t)e^{ikz}. \tag{A.28}$$

Which we recognize as $-i[H_{\mathrm{L}}, a_\sigma(z,t)]$. So we replace the time evolution from the radiation field by minus derivative with respect to z and end up with:

$$(\frac{\partial}{\partial t} + \frac{\partial}{\partial z}) a_\sigma(z,t) = i[H_{int}, a_\sigma(z,t)]. \tag{A.29}$$

This procedure is directly applicable for Stokes operators too. Now we throw away the time derivative, which is the same as ignoring retardation effects. In fact it can be shown that by introcucing a rescaled time the equality of the last equation without the time derivative can be made exact. So totally we have an equation in position and not time for light observables.

Appendix B

Atoms

B.1 Angular momentum

Throughout the thesis we are going to make an extensive use of the angular momentum operators. Here I only present the relevant details - for a full explanation one can consult any elementary book on quantum mechanics, such as [25]. The z-axis is conventionally taken as the quantization axis and the defining properties of angular momentum operators are:

$$j_z|m\rangle = m|m\rangle, \tag{B.1}$$
$$j_\pm|m\rangle = \sqrt{F(F+1)-m(m\pm 1)}|m\pm 1\rangle. \tag{B.2}$$

Where we have $j_\pm = j_x \pm i j_y$. These operators are closely related to the rotation group $SO(3)$ and the mathematical structure of it is well explained in [30] and also [18]. We will need the operators expressed in terms of density operators $\sigma_{ab} = |a\rangle\langle b|$, that is:

$$j_x = \frac{1}{2}\sum_m \sqrt{F(F+1)-m(m+1)}(\sigma_{m+1,m}+\sigma_{m,m+1}), \tag{B.3}$$
$$j_y = \frac{1}{2i}\sum_m \sqrt{F(F+1)-m(m+1)}(\sigma_{m+1,m}-\sigma_{m,m+1}), \tag{B.4}$$
$$j_z = \sum_m m\sigma_{m,m}, \tag{B.5}$$
$$j_+ = \sum_m \sqrt{F(F+1)-m(m+1)}\sigma_{m+1,m}, \tag{B.6}$$
$$j_- = \sum_m \sqrt{F(F+1)-m(m+1)}\sigma_{m,m+1}. \tag{B.7}$$

By combining these we may form new expressions:

$$j_z^2 = \sum_m m^2\sigma_{m,m} \tag{B.8}$$
$$j_+^2 = \sum_m \sqrt{(F-m)(F+m)(F+1+m)(F+1-m)}\sigma_{m+1,m-1}, \tag{B.9}$$
$$j_-^2 = \sum_m \sqrt{(F-m)(F+m)(F+1+m)(F+1-m)}\sigma_{m-1,m+1}, \tag{B.10}$$
$$j_+ j_z = \sum_m \sqrt{F(F+1)-m(m+1)}m\sigma_{m+1,m}, \tag{B.11}$$
$$j_- j_z = \sum_m \sqrt{F(F+1)-m(m-1)}m\sigma_{m-1,m}. \tag{B.12}$$

And it is useful to know that:

$$j_+|F\rangle = 0, j_+|F-1\rangle = \sqrt{2F}|F\rangle, j_+|F-2\rangle = \sqrt{2}\sqrt{2F-1}|F-1\rangle, \quad \text{(B.13)}$$

$$j_-|F\rangle = \sqrt{2F}|F-1\rangle, j_-|F-1\rangle = \sqrt{2}\sqrt{2F-1}|F-2\rangle. \quad \text{(B.14)}$$

Often we will linearize the atomic spin operators and here we describe how we have found the given linearizations. Let $j_\parallel$ denote the parallel component of the spin, along which we assume the collective spin is pointing and $j_\perp$ be a component perpindicular to $j_\parallel$. We will assume that the atom is almost in a coherent spin state and write the state as $|\psi\rangle = \frac{1}{\sqrt{1+\epsilon^2}}(|F\rangle + \epsilon|F-1\rangle)$. That way we find that to first order in ϵ:

$$\langle\psi|j_\parallel|\psi\rangle = \frac{1}{\sqrt{1+\epsilon^2}}\langle\psi|F|F\rangle + (F-1)\epsilon|F-1\rangle) = F, \quad \text{(B.15)}$$

$$\begin{aligned}\langle\psi|j_\perp|\psi\rangle &= \frac{1}{2}\langle\psi|j_+ + j_-\rangle|\psi\rangle = \frac{1}{2}\frac{1}{\sqrt{1+\epsilon^2}}\langle\psi|\sqrt{2F}\epsilon|F\rangle + \sqrt{2F}|F-1\rangle, \\ &= \frac{\sqrt{2F}}{2}\frac{1}{\sqrt{1+\epsilon^2}}(\langle F| + \epsilon\langle F-1|)(\epsilon|F\rangle + |F-1\rangle) = \frac{\epsilon}{\sqrt{1+\epsilon^2}}\sqrt{2F}. \quad \text{(B.16)}\end{aligned}$$

For instance the linearization for $\langle j_\parallel j_\perp j_\parallel\rangle$ becomes this way:

$$\begin{aligned}j_\parallel j_\perp j_\parallel|\psi\rangle &= \frac{1}{\sqrt{1+\epsilon^2}}\frac{1}{2}j_\parallel(j_+ + j_-)(F|F\rangle + (F-1)\epsilon|F-1\rangle), \\ &= \frac{1}{\sqrt{1+\epsilon^2}}\frac{1}{2}j_\parallel(\sqrt{2F}F|F-1\rangle + (F-1)\sqrt{2F}\epsilon|F\rangle + (F-1)\sqrt{4F-2}\epsilon|F-2\rangle), \\ &= \frac{1}{\sqrt{1+\epsilon^2}}\frac{1}{2}(\sqrt{2F}F(F-1)(|F-1\rangle + \epsilon|F\rangle) + (F-1)(F-2)\sqrt{4F-2}\epsilon|F-2\rangle).\end{aligned} \quad \text{(B.17)}$$

Taking the overlap with $\langle\psi|$ we get this way:

$$\langle\psi|j_\parallel j_\perp j_\parallel|\psi\rangle = \frac{\epsilon}{\sqrt{1+\epsilon^2}}F(F-1)\sqrt{2F} = F(F-1)j_\perp. \quad \text{(B.18)}$$

Using this approach one finds the matrix elements:

$$\langle\{j_\perp, j'_\perp\}\rangle \approx 0, \langle\{j_\parallel, j_\perp\}\rangle \approx (2F-1)j_\perp, \langle j_\parallel^2 - j_\perp^2\rangle \approx \frac{F}{2}(2F-1), \quad \text{(B.19)}$$

$$\langle j_\parallel j_\perp j_\parallel\rangle \approx F(F-1)j_\perp, \langle j_\perp^2\rangle \approx \frac{F}{2}, \langle j_\perp^3\rangle \approx \frac{3F-1}{2}j_\perp, \langle j_\perp^4\rangle \approx \frac{F(3F-1)}{4}, \quad \text{(B.20)}$$

$$\langle j_\perp j_\parallel^2 j_\perp\rangle \approx \frac{F(F-1)^2}{2}, \langle j_\perp j_\parallel j_\perp\rangle \approx \frac{1}{2}(F-1)j_\parallel, \quad \text{(B.21)}$$

$$\langle j_\perp j'_\perp j_\perp\rangle \approx \frac{F-1}{2}j'_\perp, \langle j_\perp {j'_\perp}^2 j_\perp\rangle \approx \frac{F}{4}(3F-1). \quad \text{(B.22)}$$

Together with these relations and that:

$$j_x = \cos\theta j_\parallel - \sin\theta j_\perp, \quad \text{(B.23)}$$

$$j_y = \sin\theta j_\perp + \cos\theta j_\parallel. \quad \text{(B.24)}$$

one obtains for instance the expectation values:

$$\begin{aligned}
\langle j_z j_x j_z \rangle &= \frac{F-1}{2}(\cos\theta j_{\parallel} - \sin\theta j_{\perp}), && \text{(B.25)}\\
\langle j_z j_y j_z \rangle &= \frac{F-1}{2}(\sin\theta j_{\parallel} + \cos\theta j_{\perp}), && \text{(B.26)}\\
\langle j_x j_z j_x \rangle &= \frac{F-1}{2} j_z(1 + (2F-1)\cos^2\theta), && \text{(B.27)}\\
\langle j_y j_z j_y \rangle &= \frac{F-1}{2} j_z(1 + (2F-1)\sin^2\theta), && \text{(B.28)}\\
\langle \{j_x, j_y\} \rangle &= (2F-1)(\frac{j_{\parallel}}{2}\sin(2\theta) + \cos(2\theta) j_{\perp}), && \text{(B.29)}\\
\langle \{j_y, j_z\} \rangle &= = (2F-1)\sin\theta j_z, && \text{(B.30)}\\
\langle \{j_x, j_z\} \rangle &= (2F-1)\cos\theta j_z && \text{(B.31)}\\
\langle j_x^2 \rangle &= \frac{j_{\parallel}}{2}(1 + (2F-1)\cos^2\theta) - (2F-1)\cos\theta\sin\theta j_{\perp}, && \text{(B.32)}\\
\langle j_y^2 \rangle &= \frac{j_{\parallel}}{2}(1 + (2F-1)\sin^2\theta) + (2F-1)\cos\theta\sin\theta j_{\perp}. && \text{(B.33)}
\end{aligned}$$

Using the same principles one can find expectation values of other combinations and these are useful when we in chapter 6 and 7 want to have expectation values of matrix elements, such as α^2, etc.

B.2 General coherent interaction

The equations describe the general coherent evolution of spin:

$$\begin{aligned}
\frac{\partial}{\partial t} j_{\parallel} = &-\frac{2\sigma_0\gamma}{A\Delta}(-\frac{(2F-1)}{2} a_2 \cos\theta\sin(2\theta) S_y j_{\parallel} + (-(2F-1) a_2 \cos\theta\cos(2\theta) S_y - a_1 S_z) j_{\perp}\\
&+ (2F-1) a_2(\sin(2\theta) S_x + \sin^2\theta S_y) j_z) && \text{(B.34)}\\
\frac{\partial}{\partial t} j_{\perp} = &-\frac{2\sigma_0\gamma}{A\Delta}(\frac{(2F-1)}{2} a_2 \sin\theta\sin(2\theta) S_y + a_1 S_z) j_{\parallel} + (2F-1) a_2 \sin\theta\cos(2\theta) S_y j_{\perp}\\
&+ (2F-1) a_2(2\cos^2(\theta) S_x + \frac{1}{2}\sin(2\theta) S_y) j_z && \text{(B.35)}\\
\frac{\partial}{\partial t} j_z = &-\frac{2\sigma_0\gamma}{A\Delta}(2F-1) a_2[(\cos(2\theta) S_y - \frac{\sin(2\theta) S_x}{2}) j_{\parallel} - 2(\cos(2\theta) S_x + \sin(2\theta) S_y) j_{\perp}] && \text{(B.36)}
\end{aligned}$$

Appendix C

Interesting matrix elements

C.1 $C^{F,\tilde{F}}_{q,q'}(m)$, coefficients

When setting up the coherent Hamiltonian we need the coefficients $C^{F,\tilde{F}}_{q,q'}(m)$ explicitly. They are calculated using the formula from (3.31):

$$C^{F,\tilde{F}}_{q,q'}(m) \quad = \quad (-1)^{F+\tilde{F}} \sum_{F'} \frac{f_{F,\tilde{F},F'}}{(2J'+1)\Delta_{F'F}} c^{F'm+q}_{\tilde{F},m+q-q'} c^{F',m+q}_{F,m}. \tag{C.1}$$

Explicitly they are:

$$F = \tilde{F} = 3 :$$

$$C_{\pm,\pm}(m) = \frac{1}{40320\Delta}(240m^2 \mp 1200m + 1440 + \frac{-315m^2 \mp 315m + 3780}{1+\frac{\Delta_{23}}{\Delta}} + \frac{75m^2 \pm 675m + 1500}{1+\frac{\Delta_{24}}{\Delta}}), \tag{C.2}$$

$$C_{0,0}(m) = \frac{1}{40320\Delta}(-480m^2 + 4320 + \frac{630m^2}{1+\frac{\Delta_{23}}{\Delta}} + \frac{-150m^2+2400}{1+\frac{\Delta_{24}}{\Delta}}), \tag{C.3}$$

$$C_{\pm,0}(m) = \frac{1}{40320\Delta}\sqrt{\frac{(3\pm m)(4\mp m)}{2}}(\pm 480m - 1440 \mp \frac{630m}{1+\frac{\Delta_{23}}{\Delta}} + \frac{\pm 150m + 600}{1+\frac{\Delta_{24}}{\Delta}}), \tag{C.4}$$

$$C_{0,\pm}(m) = C_{\pm,0}(m \pm 1), \tag{C.5}$$

$$C_{\pm,\mp}(m \mp 1) = \frac{1}{40320\Delta}\sqrt{(3+m)(4+m)(3-m)(4-m)} \times (240 - \frac{315}{1+\frac{\Delta_{23}}{\Delta}} + \frac{75}{1+\frac{\Delta_{24}}{\Delta}}). \tag{C.6}$$

They lead to the definition of the a-coefficients together with their limit value of $\Delta \to \infty$:

$$a_0 = \frac{1}{40320}(1440 + \frac{3780}{1+\frac{\Delta_{23}}{\Delta}} + \frac{1500}{1+\frac{\Delta_{24}}{\Delta}}) \to \frac{1}{6}, \tag{C.7}$$

$$a_1 = \frac{1}{40320}(-1200 - \frac{315}{1+\frac{\Delta_{23}}{\Delta}} + \frac{675}{1+\frac{\Delta_{24}}{\Delta}}) \to -\frac{1}{48}, \tag{C.8}$$

$$a_2 = \frac{1}{40320}(240 - \frac{315}{1+\frac{\Delta_{23}}{\Delta}} + \frac{75}{1+\frac{\Delta_{24}}{\Delta}}) \to 0. \tag{C.9}$$

The a terms give the C-coefficients through the relations:

$$C_{\pm,\pm}(m) = \frac{1}{\Delta}(a_0 \pm a_1 m + a_2 m^2), C_{0,0}(m) = \frac{1}{\Delta}(a_0 + 12a_2 - 2a_2 m^2), \tag{C.10}$$

$$C_{\pm,0}(m) = \frac{1}{\Delta}\sqrt{\frac{(3 \pm m)(4 \pm m)}{2}}(a_1 - a_2 \pm 2a_2 m), \tag{C.11}$$

$$C_{+,-}(m-1) = \frac{1}{\Delta}\sqrt{(3+m)(4+m)(3-m)(4-m)}a_2. \tag{C.12}$$

For $F = 4$ one obtains:

$$F = \tilde{F} = 4 :$$

$$C_{\pm,\pm}(m) = \frac{1}{40320\Delta}(112m^2 \pm 1232m + 3360 + \frac{-147m^2 \mp 147m + 2940}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{35m^2 \mp 245m + 420}{1 - \frac{\Delta_{35}}{\Delta}}), \tag{C.13}$$

$$C_{0,0}(m) = \frac{1}{40320\Delta}(-224m^2 + 5600 + \frac{294m^2}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{-70m^2 + 1120}{1 - \frac{\Delta_{35}}{\Delta}}), \tag{C.14}$$

$$C_{\pm,0}(m) = \frac{1}{40320\Delta}\sqrt{\frac{(4 \pm m)(5 \mp m)}{2}}(\pm 244m + 1120 \mp \frac{294m}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{\pm 70m - 280}{1 - \frac{\Delta_{35}}{\Delta}}), \tag{C.15}$$

$$C_{\pm,\mp}(m \mp 1) = \frac{1}{40320\Delta}\sqrt{(4+m)(5+m)(4-m)(5-m)} \times (112 - \frac{147}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{35}{1 - \frac{\Delta_{35}}{\Delta}}). \tag{C.16}$$

This time the a-coefficients become:

$$a_0 = \frac{1}{40320}(3360 + \frac{2940}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{420}{1 - \frac{\Delta_{35}}{\Delta}}) \to \frac{1}{6}, \tag{C.17}$$

$$a_1 = \frac{1}{40320}(1232 - \frac{147}{1 - \frac{\Delta_{45}}{\Delta}} - \frac{245}{1 - \frac{\Delta_{35}}{\Delta}}) \to -\frac{1}{48}, \tag{C.18}$$

$$a_2 = \frac{1}{40320}(112 - \frac{147}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{35}{1 - \frac{\Delta_{35}}{\Delta}}) \to 0. \tag{C.19}$$

$$\tag{C.20}$$

The C-coefficients can be expressed as:

$$C_{\pm,\pm}(m) = \frac{1}{\Delta}(a_0 + a_1 m + a_2 m^2), C_{0,0}(m) = \frac{1}{\Delta}(a_0 + 20a_2 - 2a_2 m^2), \tag{C.21}$$

$$C_{\pm,0}(m) = \frac{1}{\Delta}\sqrt{\frac{(4 \pm m)(5 \pm m)}{2}}(a_1 - a_2 \pm 2a_2 m), \tag{C.22}$$

$$C_{+,-}(m-1) = \frac{1}{\Delta}\sqrt{(4+m)(5+m)(4-m)(5-m)}a_2.$$

For different F, namely $F = 3$ and $F = 4$ we get the coefficients:

$$F = 3, \tilde{F} = 4 :$$

$$C_{\pm,\pm}(m) = \frac{1}{384}\sqrt{(4+m)(4-m)}(\frac{-m\pm 3}{\Delta_3} + \frac{m\pm 5}{\Delta_4}) = (\pm b_1 + b_2 m)\sqrt{(4+m)(4-m)}, \tag{C.23}$$

$$C_{0,0}(m) = \frac{2}{384} m\sqrt{(4+m)(4-m)}(\frac{1}{\Delta_3} - \frac{1}{\Delta_4}) = -2b_2 m\sqrt{(4+m)(4-m)}, \tag{C.24}$$

$$\begin{aligned} C_{\pm,0}(m) &= \frac{2}{384}\sqrt{\frac{(4\pm m)(5\pm m)}{2}}(\frac{-m\pm 3}{\Delta_3} + \frac{m\pm 1}{\Delta_4}) \\ &= (\pm(b_1 - b_2) + 2b_2 m)\sqrt{(4\pm m)(5\pm m)/2}, \end{aligned} \tag{C.25}$$

$$\begin{aligned} C_{0,\pm}(m) &= \frac{2}{384}\sqrt{\frac{(4\mp m)(5\mp m)}{2}}(\frac{m}{\Delta_3} + \frac{-m\mp 4}{\Delta_4}) \\ &= (\mp(b_1 + 3b_2) - 2b_2 m)\sqrt{(4\mp m)(5\mp m)/2}, \end{aligned} \tag{C.26}$$

$$\begin{aligned} C_{\pm,\mp}(m) &= \pm\frac{2}{384}\sqrt{(3\pm m)(4\pm m)(5\pm m)(4\mp m)}(\frac{1}{\Delta_3} - \frac{1}{\Delta_4}), \\ &= \mp b_2\sqrt{(3\pm m)(4\pm m)(5\pm m)(4\mp m)}. \end{aligned} \tag{C.27}$$

This time we define just two b-coefficients:

$$b_1 = \frac{1}{384}(\frac{3}{\Delta_3} + \frac{5}{\Delta_4}), \tag{C.28}$$

$$b_2 = \frac{1}{384}(\frac{-1}{\Delta_3} + \frac{1}{\Delta_4}). \tag{C.29}$$

C.2 Polarizability α and α^2 elements

In section 3 and 4 we contruct the coherent Hamiltonian which we write as:

$$H_{\text{coh}} = \frac{D_0^2}{\Delta}\mathbf{E}_{\text{F}}^{(-)}\alpha\mathbf{E}_{\text{F}}^{(+)}. \tag{C.30}$$

The elements of the polarizability α_{FF} are:

$$\alpha = \begin{pmatrix} (a_0 + a_2\mathbf{j}^2) - 2a_2 j_x^2 & -ia_1 j_z - a_2\{j_x, j_y\} & ia_1 j_y - a_2\{j_x, j_z\} \\ ia_1 j_z - a_2\{j_x, j_y\} & (a_0 + a_2\mathbf{j}^2) - 2a_2 j_y^2 & -ia_1 j_x - a_2\{j_y, j_z\} \\ -ia_1 j_y - a_2\{j_x, j_z\} & ia_1 j_x - a_2\{j_y, j_z\} & (a_0 + a_2\mathbf{j}^2) - 2a_2 j_z^2 \end{pmatrix}. \tag{C.31}$$

In chapter 7 we rewrite the Hamiltonian in terms of Stokes operators:

$$H_{\text{coh}} = \frac{D_0^2}{\Delta}\mathbf{E}_{\text{F}}^{(-)}\alpha\mathbf{E}_{\text{F}}^{(+)} = -2\frac{\Omega^2}{\Delta}(\boldsymbol{\gamma}\cdot\mathbf{S} + \gamma_0\frac{\phi}{2}). \tag{C.32}$$

This somehow simplifies the epxression in terms of a vector

$$\boldsymbol{\gamma} = \begin{pmatrix} -a_2(j_x^2 - j_y^2) \\ -a_2\{j_x, j_y\} \\ a_1 j_z \end{pmatrix}, \tag{C.33}$$

and the term $\gamma_0 = -a_2 j_z^2$.
The elements of polarizability for different F, α_{34} are:

$$\alpha_{xx} = b_2 \sum_m m\sqrt{(4+m)(4-m)}\sigma_{3,m;4.m} + \frac{1}{2}\sqrt{(3+m)(4+m)(5+m)(4-m)}\sigma_{3,m-1;4,m+1} - \frac{1}{2}\sqrt{(3-m)(4-m)(5-m)(4+m)}\sigma_{3,m+1;4,m-1} + h.c., \quad \text{(C.34)}$$

$$\alpha_{xy} = i\sum_m -b_1\sqrt{(4+m)(4-m)}\sigma_{3,m;4.m} + \frac{b_2}{2}(\sqrt{(3+m)(4+m)(5+m)(4-m)}\sigma_{3,m-1;4,m+1} + \sqrt{(3-m)(4-m)(5-m)(4+m)}\sigma_{3,m+1;4,m-1}) + h.c., \quad \text{(C.35)}$$

$$\alpha_{yx} = -i\sum_m -b_1\sqrt{(4+m)(4-m)}\sigma_{3,m;4.m} + \frac{b_2}{2}(\sqrt{(3+m)(4+m)(5+m)(4-m)}\sigma_{3,m-1;4,m+1} + \sqrt{(3-m)(4-m)(5-m)(4+m)}\sigma_{3,m+1;4,m-1}) + h.c., \quad \text{(C.36)}$$

$$\alpha_{yy} = b_2 \sum_m m\sqrt{(4+m)(4-m)}\sigma_{3,m;4.m} - \frac{1}{2}\sqrt{(3+m)(4+m)(5+m)(4-m)}\sigma_{3,m-1;4,m+1} + \frac{1}{2}\sqrt{(3-m)(4-m)(5-m)(4+m)}\sigma_{3,m+1;4,m-1} + h.c., \quad \text{(C.37)}$$

$$\alpha_{xz} = \frac{1}{2}\sum_m \sqrt{(4+m)(5+m)}((-b_1+b_2(3-2m))\sigma_{3,m;4,m+1} + (b_1+b_2(3-2m))\sigma_{4,m+1;3,m}) + \sqrt{(4-m)(5-m)}((-b_1+b_2(3+2m))\sigma_{3,m;4,m-1} + (b_1+b_2(3+2m))\sigma_{4,m-1;3,m}), \quad \text{(C.38)}$$

$$\alpha_{zx} = \frac{1}{2}\sum_m \sqrt{(4+m)(5+m)}((b_1+b_2(3-2m))\sigma_{3,m;4,m+1} + (-b_1+b_2(3-2m))\sigma_{4,m+1;3,m}) + \sqrt{(4-m)(5-m)}((b_1+b_2(3+2m))\sigma_{3,m;4,m-1} + (-b_1+b_2(3+2m))\sigma_{4,m-1;3,m}), \quad \text{(C.39)}$$

$$\alpha_{yz} = \frac{i}{2}\sum_m \sqrt{(4+m)(5+m)}((-b_1+b_2(3-2m))\sigma_{3,m;4,m+1} + (-b_1-b_2(3-2m))\sigma_{4,m+1;3,m}) + \sqrt{(4-m)(5-m)}((b_1-b_2(3+2m))\sigma_{3,m;4,m-1} + (b_1+b_2(3+2m))\sigma_{4,m-1;3,m}), \quad \text{(C.40)}$$

$$\alpha_{zy} = \frac{-i}{2}\sum_m \sqrt{(4+m)(5+m)}((-b_1-b_2(3-2m))\sigma_{3,m;4,m+1} + (-b_1+b_2(3-2m))\sigma_{4,m+1;3,m}) + \sqrt{(4-m)(5-m)}((b_1+b_2(3+2m))\sigma_{3,m;4,m-1} + (b_1-b_2(3+2m))\sigma_{4,m-1;3,m}), \quad \text{(C.41)}$$

$$\alpha_{zz} = -2b_2 \sum_m m\sqrt{(4+m)(4-m)}\sigma_{3,m;4,m}. \quad \text{(C.42)}$$

Values of α^2 (for both $F = 3$ and $F = 4$):

$$\alpha_{xx}^2 = (a_0 + a_2\mathbf{j}^2)^2 - 4a_0a_2j_x^2 + a_1a_2(6j_x^2 - 2\mathbf{j}^2) + a_1^2(\mathbf{j}^2 - j_x^2) + a_2^2(\mathbf{j}^2 - 5j_x^2), \tag{C.43}$$

$$\alpha_{yy}^2 = (a_0 + a_2\mathbf{j}^2)^2 - 4a_0a_2j_y^2 + a_1a_2(6j_y^2 - 2\mathbf{j}^2) + a_1^2(\mathbf{j}^2 - j_y^2) + a_2^2(\mathbf{j}^2 - 5j_y^2), \tag{C.44}$$

$$\begin{aligned}\alpha_{xy}^2 = {} & -2a_2(a_0 + a_2\mathbf{j}^2)\{j_x, j_y\} - 2ia_1(a_0 + a_2\mathbf{j}^2)j_z + a_1a_2(3\{j_x, j_y\} + ij_z) \\ & - a_1^2j_yj_x + a_2^2(4(\mathbf{j}^2 - 1)j_xj_y - j_yj_x),\end{aligned} \tag{C.45}$$

$$\begin{aligned}\alpha_{yx}^2 = {} & -2a_2(a_0 + a_2\mathbf{j}^2)\{j_x, j_y\} + 2ia_1(a_0 + a_2\mathbf{j}^2)j_z + a_1a_2(3\{j_x, j_y\} - ij_z) \\ & - a_1^2j_xj_y + a_2^2(4(\mathbf{j}^2 - 1)j_yj_x - j_xj_y).\end{aligned} \tag{C.46}$$

It is possible to find α_{34}^2 and choosing z as the quantization axis we get:

$$\begin{aligned}\alpha_{xx}^2 = {} & P_3(20(b_1 + 3b_2)^2 + (b_1^2 - 30b_1b_2 + 45b_2^2)j_x^2 \\ & + P_4(12(b_1 - 5b_2)^2 + (b_1^2 + 18b_1b_2 - 3b_2^2)j_x^2),\end{aligned} \tag{C.47}$$

$$\begin{aligned}\alpha_{yy}^2 = {} & P_3(20(b_1 + 3b_2)^2 + (b_1^2 - 30b_1b_2 + 45b_2^2)j_y^2 \\ & + P_4(12(b_1 - 5b_2)^2 + (b_1^2 + 18b_1b_2 - 3b_2^2)j_y^2),\end{aligned} \tag{C.48}$$

$$\begin{aligned}\alpha_{xy}^2 = {} & P_3(-\frac{9i}{2}(b_1^2 + 10b_1b_2 + 5b_2^2)j_z + (\frac{1}{2}b_1^2 - 15b_1b_2 + \frac{45}{2}b_2^2)\{j_x, j_y\}) \\ & + P_4(\frac{7i}{2}(b_1^2 - 6b_1b_2 + 21b_2^2)j_z + (\frac{1}{2}b_1^2 + 9b_1b_2 - \frac{3}{2}b_2^2)\{j_x, j_y\}),\end{aligned} \tag{C.49}$$

$$\begin{aligned}\alpha_{yx}^2 = {} & P_3(\frac{9i}{2}(b_1^2 + 10b_1b_2 + 5b_2^2)j_z + (\frac{1}{2}b_1^2 - 15b_1b_2 + \frac{45}{2}b_2^2)\{j_x, j_y\}) \\ & + P_4(P_4(-\frac{7i}{2}(b_1^2 - 6b_1b_2 + 21b_2^2)j_z + (\frac{1}{2}b_1^2 + 9b_1b_2 - \frac{3}{2}b_2^2)\{j_x, j_y\}).\end{aligned} \tag{C.50}$$

C.3 Elements of g and g^2

The Hermitian matrix $g_i = i[\alpha, j_i]$ desribes the coherent evolution of the atomic spin component i:

$$g_x = \begin{pmatrix} 0 & ia_1j_y - a_2\{j_x, j_z\} & ia_1j_z + a_2\{j_x, j_y\} \\ -ia_1j_y - a_2\{j_x, j_z\} & -2a_2\{j_y, j_z\} & 2a_2(j_y^2 - j_z^2) \\ -ia_1j_z + a_2\{j_x, j_y\} & 2a_2(j_y^2 - j_z^2) & 2a_2\{j_y, j_z\} \end{pmatrix}, \tag{C.51}$$

$$g_y = \begin{pmatrix} 2a_2\{j_x, j_z\} & -ia_1j_x + a_2\{j_y, j_z\} & 2a_2(j_z^2 - j_x^2) \\ ia_1j_x + a_2\{j_y, j_z\} & 0 & ia_1j_z - a_2\{j_x, j_y\} \\ 2a_2(j_z^2 - j_x^2) & -ia_1j_z - a_2\{j_x, j_y\} & -2a_2\{j_x, j_z\} \end{pmatrix}, \tag{C.52}$$

$$g_z = \begin{pmatrix} -2a_2\{j_x, j_y\} & 2a_2(j_x^2 - j_y^2) & -ia_1j_x - a_2\{j_x, j_y\} \\ 2a_2(j_x^2 - j_y^2) & 2a_2\{j_x, j_y\} & -ia_1j_y + a_2\{j_x, j_z\} \\ ia_1j_x - a_2\{j_y, j_z\} & ia_1j_y + a_2\{j_x, j_z\} & 0 \end{pmatrix}. \tag{C.53}$$

Again if one instead expresses the Hamiltonian in terms of $\boldsymbol{\gamma}$ one gets the $\tilde{g}$-matrices from section 7.1:

$$\tilde{g} = \begin{pmatrix} a_2\{j_y, j_z\} & -a_2\{j_x, j_z\} & -a_1j_y \\ a_2\{j_x, j_z\} & a_2\{j_y, j_z\} & a_1j_x \\ -2a_2\{j_x, j_y\} & 2a_2(j_x^2 - j_y^2) & 0 \end{pmatrix}, \tag{C.54}$$

$$\tilde{g}^0 = a_2 \begin{pmatrix} -\{j_y, j_z\} \\ \{j_x, j_z\} \\ 0 \end{pmatrix}. \tag{C.55}$$

We need elements of g^2 when we consider the noise correlations for f_X and f_P. These hold for both $F = 3$ and $F = 4$:

$$g_x^2(x,x) = a_1^2(\mathbf{j}^2 - j_x^2) + 4a_1a_2j_x^2 + a_2^2(4j_x(\mathbf{j}^2 - j_x^2 - 1) + \mathbf{j}^2 - j_x^2), \tag{C.56}$$
$$g_x^2(y,y) = a_1^2j_y^2 + 2a_1a_2(\mathbf{j}^2 - 2j_x^2) + a_2^2(2\mathbf{j}^2(2\bar{j}^2 - 3) + 4(3 - \mathbf{j}^2)j_x^2 + j_y(1 - 4j_x^2)j_y), \tag{C.57}$$
$$g_x^2(x,y) = 2a_1a_2(j_xj_y - i(\mathbf{j}^2 - j_x^2)j_z) + a_2^2(4j_x(\mathbf{j}^2 - j_x^2 - 2)j_y + 2i(\mathbf{j}^2j_z - 3j_xj_zj_x)), \tag{C.58}$$
$$g_x^2(y,x) = 2a_1a_2(j_yj_x + ij_z(\mathbf{j}^2 - j_x^2)) + a_2^2(4j_y(\mathbf{j}^2 - j_x^2 - 2)j_x - 2i(\mathbf{j}^2j_z - 3j_xj_zj_x)), \tag{C.59}$$

$$g_y^2(x,x) = a_1^2j_x^2 + 2a_1a_2(\mathbf{j}^2 - 2j_y^2) + a_2^2(2\mathbf{j}^2(2\mathbf{j}^2 - 3) + 4(3 - \mathbf{j}^2)j_y^2 + j_x(1 - 4j_y^2)j_x), \tag{C.60}$$
$$g_y^2(y,y) = a_1^2(\mathbf{j}^2 - j_y^2) + 4a_1a_2j_y^2 + a_2^2(4j_y(\mathbf{j}^2 - j_y^2 - 1) + \mathbf{j}^2 - j_y^2), \tag{C.61}$$
$$g_y^2(x,y) = 2a_1a_2(j_xj_y - i(\mathbf{j}^2 - j_x^2)j_z) + a_2^2(4j_y(\mathbf{j}^2 - j_y^2 - 1)j_y + \mathbf{j}^2 - j_y^2), \tag{C.62}$$
$$g_y^2(y,x) = 2a_1a_2(j_yj_x + i(\mathbf{j}^2 - j_y^2)j_z) + a_2^2(4j_y(\mathbf{j}^2 - j_y^2 - 1)j_y + \mathbf{j}^2 - j_y^2), \tag{C.63}$$

$$g_z^2(x,x) = a_1^2j_x^2 + 2a_1a_2(\mathbf{j}^2 - 2j_z^2) + a_2^2(2\mathbf{j}^2(2\mathbf{j}^2 - 3) + 4(3 - \mathbf{j}^2)j_z^2 + j_x(1 - 4j_z^2)j_x), \tag{C.64}$$
$$g_z^2(y,y) = a_1^2j_x^2 + 2a_1a_2(\mathbf{j}^2 - 2j_z^2) + a_2^2(2\mathbf{j}^2(2\mathbf{j}^2 - 3) + 4(3 - \mathbf{j}^2)j_z^2 + j_y(1 - 4j_z^2)j_y), \tag{C.65}$$
$$\begin{aligned} g_z^2(x,y) &= a_1^2j_xj_y - 2ia_1a_2(2\mathbf{j}^2 - j_z^2 - 1)j_z \\ &\quad + a_2^2(6i(j_xj_zj_x + j_yj_zj_y) + 4i\mathbf{j}^2j_z - 4j_xj_z^2j_y + j_xj_y), \end{aligned} \tag{C.66}$$
$$\begin{aligned} g_z^2(x,y) &= a_1^2j_yj_x - 2ia_1a_2(2\mathbf{j}^2 - j_z^2 - 1)j_z \\ &\quad + a_2^2(-6i(j_xj_zj_x + j_yj_zj_y) + 4i\mathbf{j}^2j_z - 4j_yj_z^2j_x + j_yj_x). \end{aligned} \tag{C.67}$$

And for the α_{34} part we only state the part which has $F = 4$:

$$g_x^2(x,x) = 12(b_1 - 5b_2)^2j_x^2 + (b_1^2 + 18b_1b_2 - 3b_2^2)j_x^4, \tag{C.68}$$
$$g_x^2(y,y) = 12(b_1 - 5b_2)^2j_x^2 + (b_1^2 + 18b_1b_2 - 3b_2^2)j_xj_y^2j_x, \tag{C.69}$$
$$g_x^2(x,y) = (\frac{1}{2}b_1^2 + 9b_1b_2 - \frac{3}{2}b_2^2)j_x\{j_x,j_y\}j_x + \frac{7i}{2}(b_1^2 - 6b_1b_2 + 21b_2^2)j_xj_zj_x, \tag{C.70}$$
$$g_x^2(y,x) = (\frac{1}{2}b_1^2 + 9b_1b_2 - \frac{3}{2}b_2^2)j_x\{j_x,j_y\}j_x - i\frac{7}{2}(b_1^2 - 6b_1b_2 + 21b_2^2)j_xj_zj_x, \tag{C.71}$$

$$g_y^2(x,x) = 12(b_1 - 5b_2)^2j_y^2 + (b_1^2 + 18b_1b_2 - 3b_2^2)j_yj_x^2j_y, \tag{C.72}$$
$$g_y^2(y,y) = 12(b_1 - 5b_2)^2j_y^2 + (b_1^2 + 18b_1b_2 - 3b_2^2)j_y^4 \tag{C.73}$$
$$g_y^2(x,y) = (\frac{1}{2}b_1^2 + 9b_1b_2 - \frac{3}{2}b_2^2)j_y\{j_x,j_y\}j_y + \frac{7i}{2}(b_1^2 - 6b_1b_2 + 21b_2^2)j_yj_zj_y, \tag{C.74}$$
$$g_y^2(y,x) = (\frac{1}{2}b_1^2 + 9b_1b_2 - \frac{3}{2}b_2^2)j_y\{j_x,j_y\}j_y - \frac{7i}{2}(b_1^2 - 6b_1b_2 + 21b_2^2)j_yj_zj_y, \tag{C.75}$$

$$g_z^2(x,x) = 12(b_1-5b_2)^2 j_z^2 + (b_1^2+18b_1b_2-3b_2^2) j_z j_x^2 j_z, \tag{C.76}$$
$$g_z^2(y,y) = 12(b_1-5b_2)^2 j_z^2 + (b_1^2+18b_1b_2-3b_2^2) j_z j_y^2 j_z, \tag{C.77}$$
$$g_z^2(x,y) = (\frac{1}{2}b_1^2+9b_1b_2-\frac{3}{2}b_2^2) j_z\{j_x,j_y\}j_z + \frac{7i}{2}(b_1^2-6b_1b_2+21b_2^2) j_z^3, \tag{C.78}$$
$$g_z^2(y,x) = (\frac{1}{2}b_1^2+9b_1b_2-\frac{3}{2}b_2^2) j_z\{j_x,j_y\}j_z - \frac{7i}{2}(b_1^2-6b_1b_2+21b_2^2) j_z^3. \tag{C.79}$$

C.4 Decay matrix ξ elements

When we consider the decay we the elements of $\xi_i = \alpha^2 j_i + j_i\alpha^2 - 2\alpha j\alpha = i[\alpha, g_i]$. In our work we have only used the (x,x)-element, but the other could also be interesting:

$$\xi_x(x,x) = 2a_1^2 j_x + 4a_1a_2(2(\mathbf{j}^2-j_x^2)-1)j_x + 2a_2^2(8j_x^2-4\mathbf{j}^2+1)j_x, \tag{C.80}$$
$$\xi_x(y,y) = a_1^2 j_x - 2a_1a_2(4j_yj_xj_y+j_x) + a_2^2(8j_x^3-4\mathbf{j}^2j_x+9j_x+24j_yj_xj_y+8j_zj_xj_z), \tag{C.81}$$
$$\begin{aligned}\xi_x(x,y) = {} & -ia_1^2 j_zj_x + 2a_1a_2((2\mathbf{j}^2-1)j_y - 4j_xj_yj_x - 3ij_xj_z) + \\ & a_2^2(-4j_y^3+5j_y+12j_xj_yj_x-4j_zj_yj_z+i(9-4\mathbf{j}^2)j_xj_z),\end{aligned} \tag{C.82}$$
$$\begin{aligned}\xi_x(y,x) = {} & ia_1^2 j_xj_z + 2a_1a_2((2\mathbf{j}^2-1)j_y - 4j_xj_yj_x + 3ij_zj_x) + \\ & a_2^2(-4j_y^3+5j_y+12j_xj_yj_x-4j_zj_yj_z-i(9-4\mathbf{j}^2)j_zj_x),\end{aligned} \tag{C.83}$$

$$\xi_y(x,x) = a_1^2 j_y - 2a_1a_2(4j_xj_yj_x+j_y) + a_2^2(8j_y^3-4\mathbf{j}^2j_y+9j_y+24j_xj_yj_x+8j_zj_yj_z), \tag{C.84}$$
$$\xi_y(y,y) = 2a_1^2 j_y + 4a_1a_2(2(\mathbf{j}^2-j_y^2)-1)j_y + 2a_2^2(8j_y^2-4\mathbf{j}^2+1)j_y, \tag{C.85}$$
$$\begin{aligned}\xi_y(x,y) = {} & -ia_1^2 j_yj_z + 2a_1a_2((2\mathbf{j}^2-1)j_x - 4j_yj_xj_y - 3ij_zj_y) \\ & + a_2^2(-4j_x^3+12j_yj_xj_y+12ij_zj_y+3ij_yj_z-4j_zj_xj_z-4i\mathbf{j}^2j_zj_y),\end{aligned} \tag{C.86}$$
$$\begin{aligned}\xi_y(y,x) = {} & ia_1^2 j_zj_y + 2a_1a_2((2\mathbf{j}^2-1)j_x - 4j_yj_xj_y + 3ij_yj_z) \\ & + a_2^2(-4j_x^3+12j_yj_xj_y-12ij_yj_z-3ij_zj_y-4j_zj_xj_z+4i\mathbf{j}^2j_yj_z),\end{aligned} \tag{C.87}$$

$$\xi_z(x,x) = a_1^2 j_z - 2a_1a_2(4j_xj_zj_x+j_z) + a_2^2(8j_z^3-4\mathbf{j}^2j_z+9j_z+24j_xj_zj_x+8j_yj_zj_y), \tag{C.88}$$
$$\xi_z(y,y) = a_1^2 j_z - 2a_1a_2(4j_yj_zj_y+j_z) + a_2^2(8j_z^3-4\mathbf{j}^2j_z+9j_z+24j_yj_zj_y+8j_xj_zj_z), \tag{C.89}$$
$$\begin{aligned}\xi_z(x,y) = {} & ia_1^2(\mathbf{j}^2-j_z^2) - 2a_1a_2(2(j_xj_yj_z+j_zj_yj_x)-i(\mathbf{j}^2-3j_z^2)) \\ & + ia_2^2(4\{j_x,j_y\}^2+\{j_x,j_z\}^2+\{j_y,j_z\}^2+4j_x^4+4j_y^4-8j_x^2j_y^2),\end{aligned} \tag{C.90}$$
$$\begin{aligned}\xi_z(y,x) = {} & -ia_1^2(\mathbf{j}^2-j_z^2) - 2a_1a_2(2(j_xj_yj_z+j_zj_yj_x)+i(\mathbf{j}^2-3j_z^2)) \\ & - ia_2^2(4\{j_x,j_y\}^2+\{j_x,j_z\}^2+\{j_y,j_z\}^2+4j_x^4+4j_y^4-8j_y^2j_x^2).\end{aligned} \tag{C.91}$$

And for the α_{34} we again only give the $F=4$ part:

$$\xi_x(x,x) = 24(b_1-5b_2)^2 j_x + 2(b_1^2+18b_1b_2-3b_2^2)j_x^3, \tag{C.92}$$
$$\xi_x(y,y) = (25b_1^2-222b_1b_2+597b_2^2)j_x + 2(b_1^2+18b_1b_2-3b_2^2)j_yj_xj_y, \tag{C.93}$$
$$\xi_x(x,y) = \frac{1}{2}(b_1^2+18b_1b_2-3b_2^2)(4j_xj_yj_x+j_y) + i\frac{7}{2}(b_1^2-6b_1b_2+21b_2^2)\{j_x,j_z\}, \tag{C.94}$$
$$\xi_x(y,x) = \frac{1}{2}(b_1^2+18b_1b_2-3b_2^2)(4j_xj_yj_x+j_y) - i\frac{7}{2}(b_1^2-6b_1b_2+21b_2^2)\{j_x,j_z\}, \tag{C.95}$$

$$\xi_y(x,x) = (25b_1^2 - 222b_1b_2 + 597b_2^2)j_y + 2(b_1^2 + 18b_1b_2 - 3b_2^2)j_xj_yj_x, \tag{C.96}$$

$$\xi_y(y,y) = 24(b_1 - 5b_2)^2 j_y + 2(b_1^2 + 18b_1b_2 - 3b_2^2)j_y^3, \tag{C.97}$$

$$\xi_y(x,y) = \frac{1}{2}(b_1^2 + 18b_1b_2 - 3b_2^2)(4j_yj_xj_y + j_x) + \frac{7i}{2}(b_1^2 - 6b_1b_2 + 21b_2^2)\{j_y, j_z\}, \tag{C.98}$$

$$\xi_y(y,x) = \frac{1}{2}(b_1^2 + 18b_1b_2 - 3b_2^2)(4j_yj_xj_y + j_x) - \frac{7i}{2}(b_1^2 - 6b_1b_2 + 21b_2^2)\{j_y, j_z\}, \tag{C.99}$$

$$\xi_z(x,x) = (25b_1^2 - 222b_1b_2 + 597b_2^2)j_z + 2(b_1^2 + 18b_1b_2 - 3b_2^2)j_xj_zj_x, \tag{C.100}$$

$$\xi_z(y,y) = (25b_1^2 - 222b_1b_2 + 597b_2^2)j_z + 2(b_1^2 + 18b_1b_2 - 3b_2^2)j_yj_zj_y, \tag{C.101}$$

$$\xi_z(x,y) = \frac{1}{2}(b_1^2 + 18b_1b_2 - 3b_2^2)(j_z\{j_x, j_y\} + \{j_x, j_y\}j_z) + 7i(b_1^2 - 6b_1b_2 + 21b_2^2)j_z^2, \tag{C.102}$$

$$\xi_z(y,x) = \frac{1}{2}(b_1^2 + 18b_1b_2 - 3b_2^2)(j_z\{j_x, j_y\} + \{j_x, j_y\}j_z) - 7i(b_1^2 - 6b_1b_2 + 21b_2^2)j_z^2. \tag{C.103}$$

Appendix D

Miscellaneous

D.1 Subsection - Spherical basis

The spherical basis is given by:

$$\mathbf{e}_+ = -(\mathbf{e}_x + i\mathbf{e}_y)/\sqrt{2}, \tag{D.1}$$

$$\mathbf{e}_- = (\mathbf{e}_x - i\mathbf{e}_y)/\sqrt{2}, \tag{D.2}$$

$$\mathbf{e}_0 = \mathbf{e}_z. \tag{D.3}$$

The different spherical basis vectors obey:

$$\mathbf{e}_q^* = \mathbf{e}_{-q}(-1)^q, \mathbf{e}_q \cdot \mathbf{e}_{q'}^* = \delta_{q,q}. \tag{D.4}$$

An arbitrary vector $\mathbf{A}$ may be written as:

$$\mathbf{A} = \sum_q A_q \mathbf{e}_q^* = \sum_q (-1)^q A_q \mathbf{e}_{-q}, \tag{D.5}$$

$$\mathbf{A} \cdot \mathbf{e}_q = A_q, \mathbf{A} \cdot \mathbf{e}_q^* = (-1)^q A_{-q}. \tag{D.6}$$

This is used for the contruction of the interaction Hamiltonian in section 3.1.

D.2 Useful commutation relations

When we contruct the interaction Hamiltonian we find the evolution of such transition elements as $\sigma_{F,m;F',m'}$. We will use primes for exited states and tildes and bare symbols for ground states. First we observe that:

$$[\sigma_{\mu,\nu}; \sigma_{\mu'\nu'}] = \sigma_{\mu,\nu'}\delta_{\mu,\nu'} - \sigma_{\mu',\nu}\delta_{\nu',\mu}. \tag{D.7}$$

Using that the exited states are weakly populated it is straightforward to contruct the commutators needed for $[H_{int}, \sigma_{F,m,F',m'}]$:

$$[\sigma_{F,m;F',m'}; \sigma_{\tilde{F},\tilde{m};F'',m''}] = \sigma_{F,m;F'',m''}\delta_{F',\tilde{F}}\delta_{m',\tilde{m}} - \sigma_{\tilde{F},\tilde{m};F',m'}\delta_{F'',F}\delta_{m'',m} = 0, \tag{D.8}$$

$$\begin{aligned}[\sigma_{F,m;F',m'}; \sigma_{F'',m'';\tilde{F},\tilde{m}}] &= \sigma_{F,m;\tilde{F},\tilde{m}}\delta_{F',F''}\delta_{m',m''} - \sigma_{F'',m'';F',m'}\delta_{\tilde{F},F}\delta_{\tilde{m},m} \\ &\approx \delta_{F',F''}\delta_{m',m''}\sigma_{F,m;\tilde{F},\tilde{m}}.\end{aligned} \tag{D.9}$$

For the commutator needed for $[H_A, \sigma_{F,m;F',m'}]$ we find:

$$\begin{aligned}[\sigma_{F,m;F',m'}, \sigma_{F'',m'';F'',m''}] &= \sigma_{F,m;F'',m''}\delta_{F',F''}\delta_{m',m''} - \sigma_{F'',m'',F',m'}\delta_{F,F''}\delta_{m,m''} \\ &= \delta_{F',F''}\delta_{m',m''}\sigma_{F,m;F'',m''}.\end{aligned} \tag{D.10}$$

D.3 Wigner-Eckart

The Wigner-Eckart theorem [WE] is a deep result of QM and states that the matrix elements for interactions can be factored in a geometric part and a pure interaction part. For a discussion we refer to chapter 3 in [25]. We make use of the WE in the form:

$$\langle F', m'|d_q|F, m\rangle = \frac{\langle F'||\mathbf{d}||F\rangle\langle F', m'|F, m; 1, q\rangle}{\sqrt{2J+1}}. \tag{D.11}$$

And because the dipole operator only talks to the electron, we can factor out the nuclear part and express the reduced element as [7]:

$$\langle F'||\mathbf{d}||F\rangle = \langle J'||\mathbf{d}||J\rangle(-1)^{F+J+I+1}\sqrt{(2F+1)(2J+1)}\left\{\begin{array}{ccc} F & F' & 1 \\ J' & J & I \end{array}\right\}. \tag{D.12}$$

Here the last term is a Wigner 6j-coefficient[30]. Combining the two expressions we get:

$$\langle F', m'|d_q|F, m\rangle = (-1)^{F+J+I+1}\sqrt{2F+1}\langle J'||\mathbf{d}||J\rangle\langle F, m; 1, q|F', m'\rangle\left\{\begin{array}{ccc} F & F' & 1 \\ J' & J & I \end{array}\right\}. \tag{D.13}$$

For our purpose it is:

$$\langle F', m+q|d_q|F, m\rangle = (-1)^{F+J+I+1}\sqrt{2F+1}\langle J'||\mathbf{d}||J\rangle c^{F',m+q}_{F,m}\left\{\begin{array}{ccc} F & F' & 1 \\ J' & J & I \end{array}\right\}. \tag{D.14}$$

In a similar fashion one obtains:

$$\langle \tilde{F}, m+q-q'|d_{-q'}|F', m+q\rangle = (-1)^{\tilde{F}+J+I+1+q'}\sqrt{2\tilde{F}+1}\langle J||\mathbf{d}||J'\rangle c^{F',m+q}_{\tilde{F},m+q-q'}\left\{\begin{array}{ccc} \tilde{F} & F' & 1 \\ J' & J & I \end{array}\right\}. \tag{D.15}$$

Using that we may write the product of the two matrix elements from (3.28):

$$\langle F', m+q|d_q|F, m\rangle\langle \tilde{F}, m+q-q'|d_{-q'}|F', m+q\rangle, \tag{D.16}$$

as:

$$\begin{aligned} C &= (-1)^{F+\tilde{F}+q'}c^{F',m+q}_{F,m}c^{F',m+q}_{\tilde{F},m+q-q'}|\langle J|||\mathbf{d}||J'\rangle|^2 \times \\ &\quad \sqrt{(2F+1)(2\tilde{F}+1)}\left\{\begin{array}{ccc} F & F' & 1 \\ J' & J & I \end{array}\right\}\left\{\begin{array}{ccc} \tilde{F} & F' & 1 \\ J' & J & I \end{array}\right\} \\ &= \frac{\alpha_0}{2J+1}(-1)^{F+\tilde{F}+q'}f_{F,\tilde{F},F'}c^{F',m+q}_{F,m}c^{F',m+q}_{\tilde{F},m+q-q'}. \end{aligned} \tag{D.17}$$

Note that the $(-1)^{q'}$ cancels the same factor in (3.28).

D.4 Detailed derivation of the interaction Hamiltonian

In chapter 3 we find that the interaction Hamiltonian may be written as:

$$H_{\text{int}} = \sum_{q,q'} E^{(-)}_{q'}C^{F,F}_{q,q'}(m)E^{(+)}_{q}\sigma_{\tilde{F},m+q-q';F,m}. \tag{D.18}$$

We are just looking at the case $F = \tilde{F}$ are insert the elements from App.B.2. That way we get:

$$\begin{aligned}
H_{\text{int}} = \frac{1}{\Delta}\sum_m [&(a_0 + a_1 m + a_2 m^2)E_+^{(-)}E_+^{(+)} + (a_0 - a_1 m + a_2 m^2)E_-^{(-)}E_-^{(+)} \\
&+ (a_0 + F(F+1)a_2 - 2a_2 m^2)E_0^{(-)}E_0^{(+)}]\sigma_{F,m;F,m} + \\
&\sqrt{\frac{(F+m)(F+1-m)}{2}}((a_1 - a_2 + 2a_2 m)(E_+^{(-)}E_0^{(+)}\sigma_{F,m-1;F,m} + E_0^{(-)}E_+^{(+)}\sigma_{F,m;F,m-1})) + \\
&\sqrt{\frac{(F-m)(F+1+m)}{2}}((a_1 - a_2 - 2a_2 m)(E_-^{(-)}E_0^{(+)}\sigma_{F,m+1;F,m} + E_0^{(-)}E_-^{(+)}\sigma_{F,m;F,m+1})) + \\
&a_2\sqrt{(F+1)(F+1+m)(F-m)(F+1-m)}(E_-^{(-)}E_+^{(+)}\sigma_{F,m+1;F,m-1} + E_+^{(-)}E_-^{(+)}\sigma_{F,m-1;F,m+1}).
\end{aligned} \tag{D.19}$$

Applying the sum rules from App.B.1. one can make the m-sums and express them through angular momentum operators:

$$\begin{aligned}
H_{\text{int}} = \frac{1}{\Delta}&(a_0\mathbf{E}^{(-)}\mathbf{E}^{(+)} + a_1 j_z(E_+^{(-)}E_+^{(+)} - E_-^{(-)}E_-^{(+)}) + a_2 j_z^2\mathbf{E}^{(-)}\mathbf{E}^{(+)} + a_2(F(F+1) - 3j_z^2)\mathbf{E}^{(-)}\mathbf{E}^{(+)} + \\
&\frac{1}{\sqrt{2}}([a_1 - a_2](j_- E_+^{(-)}E_0^{(+)} + j_+ E_0^{(-)}E_+^{(+)} + j_+ E_-^{(-)}E_0^{(+)} + j_- E_0^{(-)}E_-^{(+)}) \\
&+ 2a_2(j_- j_z E_+^{(-)}E_0^{(+)} + j_z j_+ E_0^{(-)}E_+^{(+)} - j_+ j_z E_-^{(-)}E_0^{(+)} - j_z j_- E_0^{(-)}E_-^{(+)})) \\
&+ a_2(j_+^2 E_-^{(-)}E_+^{(+)} + j_-^2 E_+^{(-)}E_-^{(+)})).
\end{aligned} \tag{D.20}$$

Collecting the terms and expressing the spherical components of the electric fields and angular momentum with cartesian ones, we get finally after some rearranging:

$$\begin{aligned}
H_{\text{int}} &= \frac{1}{\Delta}((a_0 + \frac{a_2}{3}\mathbf{j}^2)\mathbf{E}^{(-)}\mathbf{E}^{(+)} + ia_1\mathbf{E}^{(-)}\cdot\mathbf{j}\times\mathbf{E}^{(+)} - 2a_2\sum_{ij}E_i^{(-)}(\frac{\{j_i,j_j\}}{2} - \delta_{ij}\frac{\mathbf{j}^2}{3})E_j^{(+)}) \\
&= H^{(0)} + H^{(1)} + H^{(2)}.
\end{aligned} \tag{D.21}$$

D.5 Noise inclusion

Let us forget about the coherent part for a moment and focus on the decay and associated noise. In chapter 5 and 6 we EOM of the form:

$$\dot{b} = \sqrt{\gamma}f(t) - \frac{\gamma}{2}b. \tag{D.22}$$

What would happen if we would throw away the noise and keep the decay?

$$\dot{b} \;=\; -\frac{\gamma}{2}b. \tag{D.23}$$

It would mean that $b(t) = b(0)e^{-\frac{\gamma}{2}t}$ giving that the commutator $[b(t), b^\dagger(t)]$ would decay exponentially at rate γ. This is a big violation of the principles of QM, the commutator should be preserved at all times. But if we now include the noise $f(t)$ in the EOM we can write the solution as:

$$b(t) \;=\; b(0)e^{-\frac{\gamma}{2}t} + \sqrt{\gamma}\int_0^t f(t')e^{-\frac{\gamma}{2}(t-t')}dt'. \tag{D.24}$$

And this time the commutator is preserved:

$$[b(t), b^\dagger(t)] = e^{-\gamma t} + \gamma \int_0^t dt' \int_0^t dt'' e^{-\frac{\gamma}{2}(2t-t'-t'')}[f(t'), f^\dagger(t'')], \quad \text{(D.25)}$$

$$= e^{-\gamma t} + \gamma \int_0^t e^{-\gamma(t-t')} dt' = e^{-\gamma t} + (1 - e^{-\gamma t}) = 1. \quad \text{(D.26)}$$

Here we have used that the noise operators are delta-correlated in time:

$$[f(t), f^\dagger(t')] = \delta(t - t'). \quad \text{(D.27)}$$

So we see that it really nessecary to include the noise to have meaningfull description of the system - neglecting the noise gives decaying commutators.

D.6 X,P coordinateshift

The following transformations will prove to be useful in our calculations. We will have equations of the form:

$$\frac{d}{dt}\begin{pmatrix} X \\ P \end{pmatrix} = \begin{pmatrix} -\Gamma & -C \\ C & -\Gamma \end{pmatrix}\begin{pmatrix} X \\ P \end{pmatrix} = A\begin{pmatrix} X \\ P \end{pmatrix}. \quad \text{(D.28)}$$

Here C and Γ are real numbers and for our purposes the determinant of A will always be non-zero. Not we perform the unitary transformation:

$$\begin{pmatrix} \tilde{X} \\ \tilde{P} \end{pmatrix} = \begin{pmatrix} \cos(Ct) & \sin(Ct) \\ -\sin(Ct) & \cos(Ct) \end{pmatrix}\begin{pmatrix} X \\ P \end{pmatrix} = B\begin{pmatrix} X \\ P \end{pmatrix}. \quad \text{(D.29)}$$

These shifted X and P obey:

$$\frac{d}{dt}\begin{pmatrix} \tilde{X} \\ \tilde{P} \end{pmatrix} = \dot{B}\begin{pmatrix} X \\ P \end{pmatrix} + B\frac{d}{dt}\begin{pmatrix} X \\ P \end{pmatrix} = C\begin{pmatrix} \tilde{P} \\ -\tilde{X} \end{pmatrix} + A\begin{pmatrix} \tilde{X} \\ \tilde{P} \end{pmatrix} = -\Gamma\begin{pmatrix} \tilde{X} \\ \tilde{P} \end{pmatrix}. \quad \text{(D.30)}$$

So by using the unitary transform we have eliminated the coherent part and are left with the decay. Note that since the transformation is unitary, the noise operators satisfy the same relations as the untransformed ones.

D.7 Fluctuation-dissipation theorem

In this section we give a small discussion of the fluctuation-dissipation theorem, but for a deeper analysis we refer to [19]. To get a more broad description we consider at set of operators $\{A_\mu\}$, where μ is a suitable label for the operator in question - labelling whether we deal with a light- or atomic operator and for instance polarization. When we neglect the coherent part (that ideally does not introduce noise above shot noise level), as we have seen in the thesis we end up with the Langevin equation, where F and D is the drift (decay):

$$\dot{A}_\mu = D_\mu(t) + F_\mu(t). \quad \text{(D.31)}$$

The noise operators have zero mean and are delta-correlated:

$$\langle \dot{A}_\mu \rangle = \langle D_\mu \rangle, \quad \text{(D.32)}$$

$$\langle F_\mu(t) F_\nu(t') \rangle = 2\langle D_{\mu\nu} \rangle \delta(t - t'). \quad \text{(D.33)}$$

Here $D_{\mu\nu}$ is the diffusion element and it gives the strength of the correlated fluctuations. First we start with the general identity:

$$A_\mu(t) = A_\mu(t-\delta t) + \int_{t-\delta}^{t} dt' \dot{A}_\mu(t'). \tag{D.34}$$

From here we get the (same time) correlation between noise and the observables of interest:

$$\langle A_\mu(t)F_\nu(t)\rangle = \langle A_\mu(t-\delta t)F_\mu(t)\rangle + \int_{t-\delta}^{t} dt' \langle (D_\mu(t') + F_\mu(t'))F_\nu(t)\rangle. \tag{D.35}$$

In the Markov approximation the operator A cannot depend on future noise, so the first term vanishes. Same argument applies for $\langle D_\mu(t')F_\nu(t)\rangle$, which is only possibly nonzero when $t = t'$, but then the integral gives zero anyway. As a result we are left with:

$$\langle A_\mu(t)F_\nu(t)\rangle = \int_{t-\delta}^{t} dt' \langle (F_\mu(t'))F_\nu(t)\rangle. \tag{D.36}$$

Assuming that our noise is stationary we extend the limits of integration:

$$\langle A_\mu(t)F_\nu(t)\rangle = \frac{1}{2}\int_{-\infty}^{\infty} dt' \langle F_\mu(t')F_\nu(t)\rangle. \tag{D.37}$$

Which according to (C.30) gives:

$$\langle A_\mu(t)F_\nu(t)\rangle = \langle D_{\mu\nu}\rangle, \tag{D.38}$$

$$\langle F_\mu(t)A_\nu(t)\rangle = \langle D_{\mu\nu}\rangle. \tag{D.39}$$

Now examining the mean of a product of operators:

$$\begin{aligned} \frac{d}{dt}\langle A_\mu A_\nu\rangle &= \langle \dot{A}_\mu A_\nu\rangle + \langle A_\mu \dot{A}_\nu\rangle, \\ &= \langle D_\mu A_\nu\rangle + \langle F_\mu A_\nu\rangle + \langle A_\mu D_\nu\rangle + \langle A_\mu F_\nu\rangle. \end{aligned} \tag{D.40}$$

We can insert what we found above and that way get the generalized Einstein relation:

$$2\langle D_{\mu\nu}\rangle = -\langle A_\mu D_\nu\rangle - \langle D_\mu A_\nu\rangle + \frac{d}{dt}\langle A_\mu A_\nu\rangle. \tag{D.41}$$

It is a quantum fluctuation dissipation theorem and gives the connection between the diffusion coefficients $\langle D_{\mu\nu}\rangle$ and drift D_μ and D_ν. If one had a method to calculate the time evolution of $\langle A_\mu A_\nu\rangle$ one could find the diffusion elements from the drift coefficients. A related theorem is the quantum regression theorem, which also can be easily obtained from the equations above. If we are interested in the two time correlation between two operators A_μ and A_ν, we obtain immediately ($t' < t$):

$$\frac{d}{dt}\langle A_\mu(t)A_\nu(t')\rangle = \langle D_\mu(t)A_\nu(t')\rangle + \langle F_\mu(t)A_\nu(t')\rangle. \tag{D.42}$$

Again we affect the Markov approximation and eliminate the term $\langle F_\mu(t)A_\nu(t')\rangle$ since the operator cannot depend on future noise. We are left with the desired result, namely:

$$\frac{d}{dt}\langle A_\mu(t)A_\nu(t')\rangle = \langle D_\mu(t)A_\nu(t')\rangle. \tag{D.43}$$

It shows that the two time correlation function $\langle A_\mu(t)A_\nu(t')\rangle$ obeys the same EOM as the single time $\langle A_\mu\rangle$ does. We have not made use of these theorems in our work, but they are actually very powerful tools and possibly they could provide more insight and derive correlations in a more elegant way, than we have done it.

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