Effects of decoherence on the STIRAP – stimulated raman adiabatic passage

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Abstract

Manifestations of decoherence in the open STIRAP problem is studied in a rather unusual approach. By parametrizing the density operator the Lindblad equation describing the evolution of the open system can be written as a first order linear differential matrix equation. Three matrices, each corresponding to different cases of Lindblad operators are studied. In this real representation, the three-level STIRAP problem is recast into an eight dimensional one, and as a result these matrices are 8×8 and thus eight eigenstates and eigenvalues emerge. It is shown that out of the eight eigenstates only two showed to be real physical states in a closed system, while in an open system several more appeared as the coupling to the environment gets stronger, due to the properties of the matrix it follows that these newly developed states come in pairs. Moreover, a systematic study of how the performance of the STIRAP process is affected by changes of the various system parameters is presented. In particular, it is shown that especially dephasing deteriorates the success rate.

Contents

1 Introduction

In order to get a complete understanding of a quantum system it is necessary to consider its interaction with the environment, i.e. one couples the system to a second quantum system. The second one is often referred to as a bath or environment and is in general much larger than the first system, in such scenarios one speaks of an open quantum system. Open quantum systems serve to be very important to study as most real system are open in one way or the other [1].

In this thesis, open quantum systems will be studied in the realm of the STIRAP (*Stimulated Raman Adiabatic Passage*) method. STIRAP relies on *adiabaticity*, that is making the process slow enough. Within this adiabatic regime a complete population transfer from one atomic Zeeman state,$|1\rangle$, to another one, $|3\rangle$, is accomplished via a third mediating state $|2\rangle$. The third state is a metastable state and is therefore affected by losses and decoherence. Normally such a transfer is intuitively done by coupling $|1\rangle$ to $|2\rangle$ and $|3\rangle$ to $|2\rangle$ by using time-dependent lasers; the lasers amplitudes are taken to have a pulse-shape, in this case Gaussian. However, such a transfer must go through $|2\rangle$ and any population arriving at $|2\rangle$ is subject to loss by spontaneous emission, thus a complete population transfer will not be possible. In the STIRAP method, one instead couples the three states in the 'wrong' order. That is, coupling the initially empty state $|3\rangle$ to $|2\rangle$ first and then the initially populated state $|1\rangle$ to $|2\rangle$. If this is done adiabatically a complete population transfer from $|1\rangle$ to $|3\rangle$ will occur. What is most surprising, the state $|2\rangle$ will never be populated during the process. When coupling the three states in the wrong, counterintuitive order, the population will be trapped in an adiabatic *dark state*. That is a state in a time-dependent superposition of $|1\rangle$ and $|3\rangle$, where state $|1\rangle$ will be the initial state and $|3\rangle$ will be the final state. Thus, a direct route for the population transfer from $|1\rangle$ to $|3\rangle$. Now while the method is very robust if the system is closed, coupling it to an environment will deteriorate its success probability. In the past this has been studied numerically with use of the Lindblad master equation which takes the environment into account [2,3]

$$\dot{\rho} = -i[H,\rho] + \sum_k \gamma(A_k \rho A_k^\dagger - \frac{1}{2}(A_k^\dagger A_k \rho + \rho A_k^\dagger A_k). \tag{1.1}$$

In this thesis a somewhat different approach will be considered. Eq.(1.1) is stated in terms of a density operator ρ which one can always parametrize as a vector. In the three level system one can express the density operator in terms of the eight *Gell-Mann matrices* λ_i as

$$\rho = \frac{1}{3}\left[\hat{I} + \sqrt{3}\sum_{i=1}^{8} c_i(t)\lambda_i\right]. \tag{1.2}$$

Plugging Eq.(1.2) into Eq.(1.1) and one will end up with a first order linear matrix equation for the parameters $c_i(t)$. In the past this method has been mainly utilized for the two-level (qubit) problem, there one instead makes use of the Pauli matrices. In the two level system the result will be a 3×3 matrix and the solutions can be represented by the *Bloch vector* inside or on the so called *Bloch sphere*. However, in the three level STIRAP one will get a 8×8 matrix. In the thesis, this matrix will be studied in more detail, for example how the eigenvalues depend on the system parameters. Because the matrix is 8×8, eight eigenstates will result. Out of these only some will represent 'physical states', and an open question is therefore to try to understand whether the remaining ones have some meaning. Contrary to what one may be used to in the standard course of quantum mechanics, here the matrix to diagonalized will not be Hermitian, that is the eigenvalues will normally be complex, and furthermore one must differ between left and right eigenstates. An open question during the project is also to understand how adiabaticity emerges in an open quantum system. An adiabatic process relies on a slow process, a process which occurs during a long time scale. Now when letting a system interact with an environment dissipation will occur, information will be lost. Letting the system and environment interact over long

time scales only makes the dissipation greater. Thus the question, how does adiabaticity manifest in an open quantum system if at all?

This thesis is aimed for students at a Bachelor's degree level in physics, and at this level some of the math and physics will be very new, such as open quantum systems and the master equation. Due to this some steps in the math will not be written out in its full extent. The thesis will be structured in the ordinary fashion of three parts. A theory section, this section will give the necessary information of the math and physics that is needed to understand the result that follow. The thesis will end with a result and conclusion section.

2 Theory

2.1 Density matrices

In order to understand the chapters that follow the understanding of density matrices, also called density operators, are important. Density operators are in principle the same as the state vector only written in a different form, following the methods used in [4] the density matrix for a *pure state* $|\psi\rangle$ is defined in the following way

$$\rho \coloneqq |\psi\rangle\langle\psi|. \tag{2.1}$$

Now, think of a set of particles. Assume that one has N particles of the same type and knows that N_i of them is in a state $|\psi_i\rangle$, such that $\sum_i N_i = N$. If one would at random pick one of the particles, what will its state be? The probability that the state is in $|\psi_i\rangle$ is $p_i = N_i/N$, where $\sum_i p_i = 1$. It turns out that one cannot represent such a state as a coherent superposition of $|\psi_i\rangle$. Instead one uses density operators and writes the corresponding state as a *mixed state* on the form

$$\rho \coloneqq \sum_i p_i |\psi_i\rangle\langle\psi_i|. \tag{2.2}$$

2.1.1 Time evolution of the density matrices

To find the time evolution of the density matrix one starts from the Schrödinger equation[1]

$$i\hbar \frac{\partial}{\partial t}|\psi\rangle = H|\psi\rangle. \tag{2.4}$$

Taking the Hermitian conjugate of Eq.(2.4)

$$-i\hbar \frac{\partial}{\partial t}\langle\psi| = \langle\psi|H. \tag{2.5}$$

Combining Eq.(2.4) and (2.5) with the time derivative of a mixed state ρ multiplied by $i\hbar$

$$i\hbar \frac{\partial}{\partial t}\rho = i\hbar \sum_i p_i(|\dot{\psi}_i\rangle\langle\psi_i| + |\psi_i\rangle\langle\dot{\psi}_i|) = i\hbar \sum_i p_i(-\frac{i}{\hbar}H|\psi_i\rangle\langle\psi_i| + \frac{i}{\hbar}|\psi_i\rangle\langle\psi_i|H) =$$
$$= \sum_i p_i(H|\psi_i\rangle\langle\psi_i| - |\psi_i\rangle\langle\psi_i|H) = [H, \rho]$$

$$i\hbar \frac{\partial}{\partial t}\rho = [H, \rho]. \tag{2.6}$$

[1] *The "hat" notation on operators will be left out when no confusion can take place.*

Eq.(2.6) is called the *Von Neumann equation* and gives the time evolution of the density matrix. For a time-independent Hamiltonian in Eq.(2.4) one can easily show that

$$|\psi(t)\rangle = e^{-\frac{i}{\hbar}H(t-t_o)}|\psi(t_0)\rangle. \tag{2.7}$$

From Eq.(2.7) one can define the unitary time evolution operator, $U(t, t_0)$

$$U(t, t_0) = e^{-\frac{i}{\hbar}H(t-t_o)}. \tag{2.8}$$

Using Eq.(2.7) and (2.8) one can instead describe the time evolution of a density matrix as

$$\rho(t) = \sum_n p_n U(t, t_0)|\psi(t_0)\rangle\langle\psi(t_0)|U^\dagger(t, t_0),$$

or equivalently

$$\rho(t) = U(t, t_0)\rho(t_0)U^\dagger(t, t_0). \tag{2.9}$$

2.1.2 Properties of the density matrix

A few properties that the density matrix possesses. It always Hermitian

$$\rho = \rho^\dagger,$$

For a pure state it follows that

$$\rho^2 = \rho.$$

Its trace is always one

$$Tr\rho = 1.$$

Where trace is the sum of the diagonal elements of the matrix which can be stated as

$$TrD = \sum_n \langle n|D|n\rangle.$$

Here D is some operator. It should be noted that the trace is independent of bases, i.e. it is unique. The mixed states properties follows to be almost the same, expect the trace of ρ^2

$$\frac{1}{d} \leq Tr\rho^2 < 1.$$

Here, d, which is always greater than zero, is the dimension of the density matrix. Using the definition (2.2) of the density matrix it follows that an expectation value of any operator D becomes

$$\langle D\rangle = \sum_i \langle\psi_i|D|\psi_i\rangle,$$

which can be written as

$$\langle D\rangle = Tr(D\rho).$$

2.1.3 The interaction picture

In deriving the master equation Eq.(1.1), which follows later, one makes use of the so called *interaction picture*. In the interaction picture both the state vectors and the observables carry time dependence. The interaction picture can be seen as a picture in between the more generally used *Schrödinger picture*, where the state vector have time dependence while the observables are static, and *the Heisenberg picture* where only the observables evolve in time.

In a more complicated system one would generally want to split the Hamiltonian into two parts, a more simple Hamiltonian H_0, generally time independent, and a more complicated interacting Hamiltonian V, sometimes time dependent

$$H = H_0 + V. \tag{2.10}$$

Although for this section it is time independent for simplicity. This approach will be studied more in the next section. If the interaction picture is to be equivalent to the Schrödinger picture, then the expectation value of some observable D must not change [5]. The expectation value is given by

$$\langle D(t)\rangle = Tr\big(D\rho(t)\big) = Tr\left(DU(t,t_0)\rho(t_0)U^{\dagger}(t,t_o)\right). \tag{2.11}$$

Splitting the operator $U(t,t_0)$ as

$$U(t,t_0) = U_0(t,t_0)U_I(t,t_0), \tag{2.12}$$

where $U_0(t,t_0)$ and $U_I(t,t_0)$ is

$$U_0(t,t_0) = e^{-iH_0(t-t_0)}, \tag{2.13}$$

$$U_I(t,t_0) = e^{-iV(t-t_0)}. \tag{2.14}$$

Plugging Eq.(2.12) into Eq.(2.11)

$$\langle D(t)\rangle = Tr\left(DU_0(t,t_0)U_I(t,t_0)\rho(t_0)U_I^{\dagger}(t,t_0)U_0^{\dagger}(t,t_0)\right), \tag{2.15}$$

using the cyclic properties of the trace[2] and one finds

$$\langle D(t)\rangle = Tr\left(U_0^{\dagger}(t,t_0)DU_0(t,t_0)U_I(t,t_0)\rho(t_0)U_I^{\dagger}(t,t_0)\right) = Tr\big(O_I(t)\rho_I(t)\big), \tag{2.16}$$

where

$$O_I(t) = U_0^{\dagger}(t,t_0)DU_0(t,t_0) \tag{2.17}$$

$$\rho_I(t) = U_I(t,t_0)\rho(t_0)U_I^{\dagger}(t,t_0). \tag{2.18}$$

Equation.(2.17) shows how interaction picture observables evolve in time. Taking the time derivative of Eq.(2.18) one gets the Von Neumann equation in the interaction frame

$$\dot{\rho}_I(t) = -i[H_I(t),\rho_I(t)]. \tag{2.19}$$

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[2] *The trace of a product is invariant under cyclic permutations;* $Tr(ABC) = Tr(BCA) = Tr(CAB)$*. The proof is straightforward and left as an exercise*

2.2 Time dependent Hamiltonians

This thesis will focus on the methods of STIRAP. That is, as mentioned before, a three level quantum system where the goal is to transfer a population from one Zeeman state to another one through a third intermediate state. When one speaks of such a transition a time dependent Hamiltonian is necessary, time dependence of the Hamiltonian is crucial for STIRAP. Normally one introduces a time dependent potential, in our case the laser pulses coupling the three states in STIRAP, and writes the total Hamiltonian into two parts, following the methods of [6]

$$H(t) = H_0 + V(t). \tag{2.20}$$

Here H_0 is the Hamiltonian for the atom without any laser pulses (the unperturbed Hamiltonian), $V(t)$ comes from the laser pulses and acts as the perturbed Hamiltonian and now carries the time dependence. Now assume that the solution for the atom is known

$$H_0|\psi_n\rangle = E_n|\psi_n\rangle. \tag{2.21}$$

Here E_n are the bare energies of the atom and $|\psi_n\rangle$ are the electronic states. Thus, in the basis of the these states H_0 is diagonal with the elements being the energies E_n. Forming a complete set, any state can be expressed as a linear combination of the states $|\psi_n\rangle$. The time evolution of the combination can then be written with each state having an exponential time dependent factor, such that

$$|\Psi(\mathrm{t})\rangle = \sum_n c_n|\psi_n\rangle e^{-iE_nt/\hbar}. \tag{2.22}$$

Turning on the perturbation $V(t)$, one can still write the states as a linear combination, since the states constitute a complete set, the only difference is the time dependence of the coefficients c_n [7]

$$|\Psi(t)\rangle = \sum_n c_n(t)|\psi_n\rangle. \tag{2.23}$$

Here the exponential factor is combined with the time dependent factor $c_n(t)$. Combining Eq.(2.23) together with the time dependent Schrödinger equation one obtains a set of N coupled differential equations for the coefficients $c_n(t)$. As shown in [7], for the time dependent perturbation theory

$$\dot{c_n}(t) = -\frac{i}{\hbar}\sum_m H_{nm}(t)c_m(t). \tag{2.24}$$

In Eq.(2.24) we have assumed that the diagonal elements of the total Hamiltonian are all zero, which is normally justified. That is because the diagonal elements normally constitutes the so called detuning, as will be shown later. The detuning is normally written as, $\Delta = \omega_{light} - \omega_{transistion}$, where ω_{light} is the frequency of the applied lasers and $\omega_{transition}$ the energy difference between the two involved electronic states. One can of course always choose the detuning to be zero, thus the result of Eq.(2.24). Now let us look at Eq.(2.24) in the absence of any interaction, following the methods of [6]

$$\dot{c_n}(t) = -\frac{i}{\hbar}E_nc_n(t). \tag{2.25}$$

Solving Eq.(2.25) one can easily see that the state vector can be written as

$$|\Psi(t)\rangle = \sum_n e^{-\frac{iE_nt}{\hbar}}c_n(0)|\psi_n\rangle. \tag{2.26}$$

Where $c_n(0)$ remains fixed. Now turning on a time dependent interaction, one can then find it easier to work in the rotating reference frame. The solution can be written on the form

$$|\Psi(t)\rangle = \sum_n e^{-i\phi(t)} C_n(t)|\psi_n\rangle. \tag{2.27}$$

$\phi(t)$ is some phase, to be decided. From here one can express Eq.(2.27) as a superposition of rotating coordinates by introducing

$$|\psi_n'\rangle = e^{-i\phi(t)}|\psi_n\rangle. \tag{2.28}$$

Combining Eq.(2.27) and (2.28) one can obtain a new set of coupled differential equations which can be written in matrix form as

$$\dot{C}(t) = -iW(t)C(t). \tag{2.29}$$

$C(t)$ is a column vector with the amplitudes $C_n(t)$ as components. $W(t)$ is the total Hamiltonian, which is obtained by combining Eq.(2.27) with the time dependent Schrödinger equation

$$i\hbar \frac{\partial|\Psi\rangle}{\partial t} = H(t)|\Psi\rangle. \tag{2.30}$$

Plugging the ansatz (2.27) into the Schrödinger equation one derives

$$i\hbar\left(\dot{C_n}(t)|\psi_n'\rangle + C_n(t)|\dot{\psi}_n'\rangle\right) = H_0 C_n(t)|\psi_n'\rangle + V(t)C_n(t)|\psi_n'\rangle. \tag{2.31}$$

Using the orthogonality of the states

$$\langle\psi_m'|\psi_n'\rangle = \langle\psi_m|\psi_n\rangle = \delta_{mn}, \tag{2.32}$$

the matrix $W(t)$ becomes

$$W(t) = \frac{1}{\hbar}[\langle\psi_m'|H_0|\psi_n'\rangle + \langle\psi_m'|V(t)|\psi_n'\rangle] - \langle\psi_m'|\dot{\psi}_n'\rangle. \tag{2.33}$$

The diagonal elements are, knowing that H_0 is diagonal with E_n as its elements

$$\hbar W_{nn}(t) = E_n + V_{nn} - \hbar\dot{\phi}(t), \tag{2.34}$$

and the off-diagonal

$$\hbar W_{nm}(t) = V_{nm}(t). \tag{2.35}$$

The off diagonal derives entirely from the interaction Hamiltonian $V(t)$. The off diagonal elements are responsible for the transition between states, in this case the laser pulses will couple our states together. In this thesis, these pulses will be called G_1 and G_2. Thus, G_1 is the pulse connecting state $|1\rangle$ with $|2\rangle$ and G_2 connecting state $|3\rangle$ with $|2\rangle$ in the STIRAP model (see section 2.3). They are taken time dependent with a Gaussian form. The diagonal elements consist of the energies E_n together with the diagonals of $V(t)$. The diagonals of $V(t)$ give shifts in the bare energies. However, one can choose the phase in Eq.(2.26) so that the first diagonal element becomes strictly zero. The other diagonal terms will be defined as the detunings after the use of the rotating wave approximation (RWA). That is, neglecting all counter rotating terms which has the form, $e^{\pm i2\omega}$ where ω is some frequency. The RWA states that the counter rotating terms will rotate so fast that they average to zero [1]. Thus, it is justified to neglect them. Summing up, the matrix now has the following diagonal

$$W_{11}(t) = 0, \tag{2.36}$$

$$W_{22}(t) = \Delta, \tag{2.37}$$

$$W_{33}(t) = \Delta_1 - \Delta_2. \tag{2.38}$$

In this thesis there will always be a two photon resonance with state $|1\rangle$ and $|3\rangle$, this means that $\Delta_1 = \Delta_2$. Thus the STIRAP Hamiltonian becomes

$$H_S = \begin{pmatrix} 0 & G_1(t) & 0 \\ G_1(t) & \Delta & G_2(t) \\ 0 & G_2(t) & 0 \end{pmatrix}. \tag{2.39}$$

2.3 The STIRAP method

STIRAP is a widely used method in atomic physics to make complete population transfers between two electronic Zeeman states through a third mediating state, a schematic setup can be seen in figure 1.

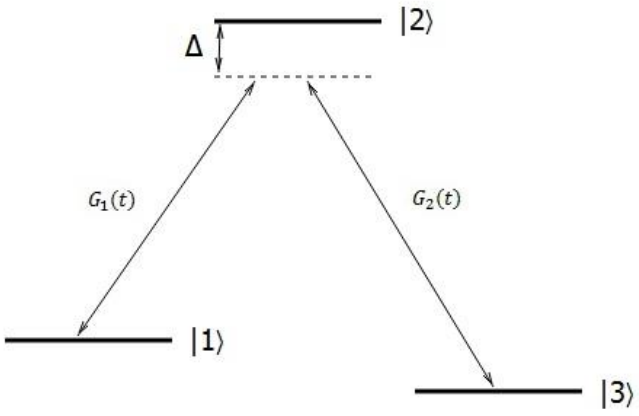

Figure 1: Schematic picture of the STIRAP method with state $|1\rangle$ being initially populated and state $|3\rangle$ being the final target state for the population transfer. Δ is the detuning, $G_1(t)$ and $G_2(t)$ are laser pulses coupling the states in a counterintuitive order. That is, coupling $G_2(t)$ is applied before $G_1(t)$ in order to trap the population in a dark state and make a direct route from $|1\rangle$ to $|3\rangle$.

The essential trick of STIRAP is having an adiabatic process (see section 2.6 below) and to trap the population in a *dark state*, which is done by making a counterintuitive coupling between the states. That is, coupling the initial unpopulated state $|3\rangle$ to $|2\rangle$ before coupling the initially populated state $|1\rangle$ to $|2\rangle$. Moreover, the excited state $|2\rangle$ never gets populated during the process. The physics behind STIRAP can be seen by studying the eigenvectors and eigenvalues of the corresponding Hamiltonian. Note, however, that we have a time-dependent Hamiltonian so the energy is not conserved. In this respect, to solve the time-dependent Schrödinger equation it is not sufficient to solve the eigenvalue problem. Nevertheless, solving to the *instantaneous* eigenvalues and eigenstates give insight into the physics. To get the eigenvectors and eigenvalues one uses the STIRAP Hamiltonian (Eq.(2.39)) and solves for the following

$$Det\left(H_s - \lambda \hat{I}\right) = 0. \tag{2.40}$$

Where $\hat{I}$ is the unit matrix and the λ's are our eigenvalues. Let us introduce the two angles as done in [3] and [8]

$$\tan(\theta) = \frac{G_1(t)}{G_2(t)}, \tag{2.41}$$

$$\tan(2\phi) = \frac{\Omega(t)}{\Delta}, \tag{2.42}$$

where

$$\Omega(t) = \sqrt{G_1^2(t) + G_2^2(t)}. \tag{2.43}$$

And as before Δ is the detuning. The eigenvectors are found in terms of the angles as (leaving the time dependence for the angles)

$$|\Phi_+(t)\rangle = \sin(\theta)\sin(\phi)\,|1\rangle + \cos(\phi)\,|2\rangle + \cos(\theta)\sin(\phi)\,|3\rangle, \tag{2.44a}$$

$$|\Phi_-(t)\rangle = \sin(\theta)\cos(\phi)\,|1\rangle - \sin(\phi)\,|2\rangle + \cos(\theta)\cos(\phi)\,|3\rangle, \tag{2.44b}$$

$$|\Phi_0(t)\rangle = \cos(\theta)\,|1\rangle - \sin(\theta)\,|3\rangle, \tag{2.44c}$$

with the corresponding eigenvalues

$$\lambda_+(t) = \frac{1}{2}\left(\Delta + \sqrt{\Delta^2 + \Omega(t)^2}\right), \tag{2.45a}$$

$$\lambda_-(t) = \frac{1}{2}\left(\Delta - \sqrt{\Delta^2 + \Omega(t)^2}\right), \tag{2.45b}$$

$$\lambda_0 = 0. \tag{2.45c}$$

The state $|\Phi_0\rangle$ is the dark state[3]. One can now see, from Eq.(2.44c) and Eq.(2.41), by letting G_2 start before G_1 at time $t = -\infty$ that

$$\begin{aligned} \lim_{t\to-\infty} \frac{G_1(t)}{G_2(t)} &= 0, \qquad \theta \to 0, \\ \lim_{t\to+\infty} \frac{G_1(t)}{G_2(t)} &= \infty, \qquad \theta \to \frac{\pi}{2}. \end{aligned} \tag{2.46}$$

Thus, the population goes from $|1\rangle$ to $|3\rangle$ without ever populating $|2\rangle$ by noting that, $|\Phi_0(t=-\infty)\rangle = |1\rangle$ and $|\Phi(t=+\infty)\rangle = -|3\rangle$. The dark state is the more interesting state of the three since it does not contain $|2\rangle$ which is sensitive to losses, and hence a lot of attention in the remaining thesis will go on studying the dark state.

[3] *The name "dark" originates from the fact that the state does not contain the excited* $|2\rangle$ *state which is sensitive to spontaneous emission*

2.4 Derivation of the Born-Markov master equation

In this section we outline the derivation of the Born-Markov master equation. The exact expression for the master equation is complicated to work with and thus, as will be seen in this section, a couple of approximations will be made. In general one uses the fact that the environment couples weakly with the system. The resulting approximations, named after Born and Markov, lead to the Born-Markov master equation. Following from the methods in [1], [5] and [9], one starts by writing the total Hamiltonian as

$$H = H_s + H_E + V. \tag{2.47}$$

Here H_s and H_E are the system and environment Hamiltonian respectively and V denotes the interaction Hamiltonian which is the only part that involves both system and the environment. The system-environment coupling V will be taken small and thereby treated as a perturbation. From here it is convenient to write, $H_o = H_s + H_E$. So that, $H = H_0 + V$, and move into the interaction picture as discussed above

$$V_I(t) = e^{iH_0 t} V e^{-iH_0 t}. \tag{2.48}$$

With the subscript I denoting the interaction picture. Now one makes use of the density matrices, as studied in the previous section 2.1. Knowing that the time evolution of the density matrix can be written on the form of a von Neumann equation. In the interaction picture one then gets (with $\hbar = 1$) according to Eq.(2.19)

$$\dot{\rho}_I(t) = -i[V_I(t), \rho_I(t)]. \tag{2.49}$$

Integrating Eq.(2.49) one finds

$$\rho_I(t) = \rho_I(0) - i \int_0^t [V_I(t'), \rho_I(t')] dt'. \tag{2.50}$$

Substituting Eq.(2.50) back into Eq.(2.49) yields

$$\dot{\rho}_I(t) = -i[V_I(t), \rho_I(0)] - \int_o^t \big[V_I(t), [V_I(t'), \rho_I(t')]\big] dt'. \tag{2.51}$$

Now, Eq.(2.51) is exact and gives us both the system and the environment evolution. Since only the evolution of the system is of interest for us one can make use of the trace operation. Tracing over the environment in Eq.(2.51) yields the systems density operator denoted ρ, that is, $\dot{\rho} = Tr_E(\dot{\rho}_I)$

$$\dot{\rho}(t) = -iTr_E([V_I(t), \rho_I(0)]) - \int_0^t Tr_E\big(\big[V_I(t), [V_I(t'), \rho_I(t')]\big]\big) dt'. \tag{2.52}$$

Equation.(2.52) is still exact and difficult to solve. The final part of this section will focus on making a couple of approximation. First one assumes that there is no interaction between the system and environment at time $t = 0$, no correlation exists initially. Thus, the initial density operator can be written as

$$\rho_I(0) = \rho(0) \otimes \rho_E(0). \tag{2.53}$$

This approximation is only reasonable when working with a weakly interacting environment which is assumed here. Now let us split V_I into two parts

$$V_I(t) = V_S(t) + V_{SE}(t), \tag{2.54}$$

where $Tr\big(V_{SE}(t)\rho_I(0)\big) = 0$. At this time one makes use of the *Born approximation*, using again that the system only interacts weakly with the environment and that the environment is large in comparison to the system, and thus it is permissible to make the substitution $\rho_I(t')$ by $\rho(t') \otimes \rho_E(0)$. Let us also introduces the *Markov approximation* which will make it possible to replace $\rho(t')$ by $\rho(t)$. One can do this by introducing time scales, the Markovian approximation relies on the difference between the memory time scale for the environment and the system evolution time scale. The replacement $\rho(t')$ to $\rho(t)$ can be done when one assumes that the environment memory time scale is much shorter than the systems evolution time scale [5], the equation now becomes

$$\dot{\rho}(t) = -i[V_S(t), \rho(t)] - \int_0^t Tr_E\big(\big[V_I(t), [V_I(t'), \rho(t) \otimes \rho_E(0)]\big]\big)dt'. \tag{2.55}$$

In the final step one makes use of the sharpness of the integrand at $t' \approx t$ and replace the lower limit to $-\infty$, thus getting the final *Born-Markov master equation*

$$\dot{\rho}(t) = -i[V_S(t), \rho(t)] - \int_{-\infty}^t Tr_E\big(\big[V_I(t), [V_I(t'), \rho(t) \otimes \rho_E(0)]\big]\big)dt'. \tag{2.56}$$

2.5 Lindblad master equation

The Born-Markov master equation of the previous subsection is for the general case with no assumptions on the system nor the environment. In this section the derivation will be repeated for a special case and in the end a special master equation on the *Lindblad form* will be presented. *The Lindblad master equation* is such an equation that will be used in the reminder of the thesis. More precisely, the special case of radiative damping of a two-level atom will be considered, following the methods in [1], [9] and [10]. For the start one shall find the two Hamiltonians describing the atom and the environment, i.e. identifying

$$H_0 = H_a + H_E. \tag{2.57}$$

Then one will combine Eq.(2.57) to some interacting Hamiltonian so that the total Hamiltonian

$$H = H_0 + V, \tag{2.58}$$

is determined. One can show that for the radiative damping of a two level atom the Hamiltonian for the environment is

$$H_E = \sum_k \omega_k b_k^\dagger b_k. \tag{2.59}$$

The *wave number k* decodes all of the information specifying the mode coming from the electromagnetic field. $b_k^\dagger$ and b_k are the creation and annihilation operators, sometimes also called the raising and lowering operators. They follow the bosonic commutation relation

$$\left[b_k, b_l^\dagger\right] = \delta_{kl}. \tag{2.60}$$

Let there only be two energy levels (the so called *two-level approximation*) for the atom so that the free Hamiltonian for the atom is

$$H_a = \frac{\omega_a}{2}\sigma_z. \tag{2.61}$$

The two levels represent different electronic Zeeman levels. Here ω_a is the energy difference between the ground state $|0\rangle$ and the excited state $|1\rangle$, and $\sigma_z = |1\rangle\langle 1| - |0\rangle\langle 0|$. The coupling of the atom to an environment, which here represent the electromagnetic field, can be described by the dipole-coupling Hamiltonian [1]

$$V = \sum_k g_k (b_k + b_k^\dagger)(\sigma_+ + \sigma_-). \tag{2.62}$$

Here $\sigma_+ = (\sigma_-)^\dagger = |1\rangle\langle 0|$ and is the raising operator for the atom. g_k are coefficients that will always be assumed real. Now following the methods of section 2.4 and one starts by calculating the interacting Hamiltonian in the interaction picture. At this point the first approximation will be made, in calculating V in the interaction picture one shall once again use the rotating wave approximation (RWA). After imposing the RWA, the interacting Hamiltonian V in the interaction picture is

$$V_I(t) = \sum_k (g_k b_k \sigma_+ e^{-i(\omega_k - \omega_a)t} + g_k b_k^\dagger \sigma_- e^{i(\omega_k - \omega_a)t}). \tag{2.63}$$

Next one plugs Eq.(2.63) into Eq.(2.52) and specifies an initial state for the environment. Following the methods of [1], the vacuum state will be used. The vacuum state is the quantum state with the lowest possible energy, or in our case the state with no photons. Using the vacuum state it follows that the first term in Eq.(2.52) will be zero, $-iTr_E([V_I(t), \rho_I(0)]) = 0$. The equation now reads

$$\dot{\rho}(t) = -\int_0^t Tr_E\big([V_I(t), [V_I(t'), \rho_I(t')]]\big) dt'. \tag{2.64}$$

The commutator in Eq.(2.64) becomes

$$\begin{aligned}\big[V_I(t), [V_I(t'), \rho_I(t')]\big] = {} & V_I(t)V_I(t')\rho_I(t') - V_I(t)\rho_I(t')V_I(t') - \\ & - V_I(t')\rho_I(t')V_I(t) + \rho_I(t')V_I(t')V_I(t).\end{aligned} \tag{2.65}$$

Plugging Eq.(2.63) into Eq.(2.65), taking the trace over the environment and the commutator yields the following

$$\begin{aligned} & Tr_E([V_I(t), [V_I(t'), \rho_I(t')]) = \\ = \sum_k \; & g_k^2 \left\langle b_k^\dagger b_k^\dagger \right\rangle e^{-i\Omega(t+t')}(\sigma_-\sigma_-\rho(t') - 2\sigma_-\rho(t')\sigma_- + \rho(t')\sigma_-\sigma_-) + \\ & + g_k^2 \langle b_k^\dagger \, b_k\rangle(\sigma_-\sigma_+\rho(t')e^{i\Omega(t-t')} - \sigma_+\rho(t')\sigma_- e^{i\Omega(t-t')} - \\ & - \sigma_+\rho(t')\sigma_- e^{i\Omega(t'-t)} + \rho(t')\sigma_-\sigma_+ e^{i\Omega(t-t')}) + \\ & + g_k^2 \langle b_k b_k^\dagger\rangle(\sigma_+\sigma_-\rho(t')e^{-i\Omega(t-t')} - \sigma_-\rho(t')\sigma_+ e^{-i\Omega(t-t')} - \\ & - \sigma_-\rho(t')\sigma_+ e^{-i\Omega(t'-t)} + \rho(t')\sigma_+\sigma_- e^{-i\Omega(t'-t)}) + \\ & + g_k^2 \langle b_k b_k\rangle(\sigma_+\sigma_+\rho(t')e^{-i\Omega(t+t')} - 2\sigma_+\rho(t')\sigma_- e^{-i\Omega(t+t')} + \rho(t')\sigma_+\sigma_+ e^{-i\Omega(t-t')}. \end{aligned} \tag{2.66}$$

Here $\Omega = \omega_k - \omega_a$ and the expectation values refer to the initial state of the environment. At this point the vacuum state determines the expectation values as.

$$\begin{aligned}\langle b_k\rangle = \left\langle b_k^\dagger\right\rangle = 0\\ \left\langle b_k^\dagger b_l\right\rangle = 0\\ \left\langle b_k b_l^\dagger\right\rangle = \delta_{kl}\\ \langle b_k b_l\rangle = \left\langle b_k^\dagger b_l^\dagger\right\rangle = 0\end{aligned} \tag{2.67}$$

Most terms in Eq.(2.66) thereby vanish and Eq.(2.64) simplifies to

$$\begin{aligned}\dot{\rho}(t) = -\int_0^t \sum_k\ g_k^2\big(\sigma_+\sigma_-\rho(t')e^{-i\Omega(t-t')} - \sigma_-\rho(t')\sigma_+e^{-i\Omega(t-t')}\\ - \sigma_-\rho(t')\sigma_+e^{-i\Omega(t'-t)} + \rho(t')\sigma_+\sigma_-e^{-i\Omega(t'-t)}\big)dt'.\end{aligned} \tag{2.68}$$

Rewriting it in a more clean way

$$\dot{\rho}(t) = -\int_0^t (\Gamma(t-t')[\sigma_+\sigma_-\rho(t') - \sigma_-\rho(t')\sigma_+] + \text{h.c})dt', \tag{2.69}$$

where h.c stands for the Hermitian conjugate and, $\Gamma(t-t') = \sum_k g_k^2 e^{-i\Omega(t-t')}$. Next step is to make use of the Markovian approximation. First one finds that for an atom in free space Γ can be rewritten as [1]

$$\Gamma(\tau) = \int_0^\infty \rho(\omega)g(\omega)^2 e^{i\Omega\tau}d\omega. \tag{2.70}$$

Where $\rho(\omega)$ is the density of states of the electromagnetic field at frequency ω, not to be confused with the density operator, and Ω is the same as used in Eq.(2.66). This integral will be sharply peaked at $\tau = 0$ provided that $\rho(\omega)g(\omega)^2$ is a smooth function, which will be assumed. Remembering that for the Markovian approximation one has that $t \approx t'$ and that $\rho(t) \approx \rho(t')$, and Eq.(2.69) yields

$$\begin{aligned}\dot{\rho}(t) = -\int_0^t \Gamma(0)dt'(\sigma_+\sigma_-\rho(t) - \sigma_-\rho(t)\sigma_+)\\ -\int_0^t \Gamma^*(0)dt'(\rho(t)\sigma_+\sigma_- - \sigma_-\rho(t)\sigma_+).\end{aligned} \tag{2.71}$$

Defining the integral over Γ as [1]

$$\int_0^\infty \Gamma(\tau)d\tau = i\Delta\omega_a + \frac{\gamma}{2}. \tag{2.72}$$

Here $\Delta\omega_a$ is the *frequency shift* and γ is the *radiative decay rate*. Using Eq.(2.72) in Eq.(2.71), the equation can be written into the following two parts

$$\begin{aligned}\dot{\rho}(t) = -i\Delta\omega_a(\sigma_+\sigma_-\rho(t) - \rho(t)\sigma_+\sigma_-)\\ +\gamma\left(\sigma_-\rho(t)\sigma_+ - \frac{1}{2}(\sigma_+\sigma_-\rho(t) - \rho(t)\sigma_+\sigma_-)\right).\end{aligned} \tag{2.73}$$

Eq.(2.73) can be further rewritten in a more clean way by using the following relation

$$\sigma_+\sigma_- = \frac{1}{2}(\hat{I} + \sigma_z).$$

Eq.(2.73) now becomes

$$\dot{\rho}(t) = \frac{-i\Delta\omega_a}{2}[\sigma_z, \rho] + \gamma\left(\sigma_-\rho(t)\sigma_+ - \frac{1}{2}(\sigma_+\sigma_-\rho(t) - \rho(t)\sigma_+\sigma_-)\right). \tag{2.74}$$

Eq.(2.74) is the master equation for the radiative damping of a two level atom.

Lindblad showed that the most general form of a master equation is [1]

$$\dot{\rho}(t) = -i[H, \rho(t)] + \sum_k \gamma_k D[A_k]\rho(t). \tag{2.75}$$

Where $D[A_k]$ is called the superoperator for any arbitrary operator A_k

$$D[A_k]\rho = A_k\rho A_k^\dagger - \frac{1}{2}\left(A_k^\dagger A_k\rho + \rho A_k^\dagger A_k\right) \tag{2.76}$$

and H is the system Hamiltonian. Eq.(2.75) is the master equation on *Lindblad form* and is the equation that will be further used in the thesis. γ is the radiative decay rate and is a constant, γ can change depending on which state one decays to. Although throughout this thesis γ will be assumed the same for all the states and thus the index k will no longer be written out. Note that when $\gamma = 0$ the master equation returns the original Von Neumann equation (Eq.(2.19)).

2.6 The adiabatic approximation

The STIRAP process relies on the adiabatic approximation. This section briefly discusses the adiabatic theorem and gives the mathematical proof by following the methods of [7]. According to Ref. [7] (page 372); "The adiabatic theorem states that if the particle was initially in the *n*th eigenstate of H^i, it will be carried (under the Schrödinger equation) into the *n*th eigenstate of H^f". What guarantees the applicability of the adiabatic theorem is that the process is slow.

Now consider a Hamiltonian that is time dependent, the (instantaneous) eigenvectors and eigenvalues are thus also time dependent, orthogonal and given by the equation

$$H(t)|\psi_n(t)\rangle = E_n(t)|\psi_n(t)\rangle. \tag{2.77}$$

The general solution to the time dependent Schrödinger equation (Eq.(2.30)) can be written

$$|\Psi(t)\rangle = \sum_n c_n(t)|\psi_n(t)\rangle e^{i\theta_n(t)}, \tag{2.78}$$

where $\theta_n(t)$, the dynamical phase is defined as ($\hbar = 1$)

$$\theta_n(t) = -\int_0^t E_n(t')dt'. \tag{2.79}$$

Combining Eq.(2.78) with the time dependent Schrödinger equation and one finds

$$\begin{aligned} i\sum_n [\dot{c_n}(t)|\psi_n(t)\rangle + c_n(t)\left|\dot{\psi}_n(t)\right\rangle + ic_n(t)|\psi_n(t)\rangle\dot{\theta}_n(t)]e^{i\theta_n(t)} \\ = \sum_n c_n(t)H(t)|\psi_n(t)\rangle e^{i\theta_n(t)}. \end{aligned} \tag{2.80}$$

Having that $|\psi_n(t)\rangle$ is an instantaneous eigenstate and using Eq.(2.77) and (2.79). Eq.(2.80) leads to

$$\sum_n \dot{c}_n(t)|\psi_n(t)\rangle e^{i\theta_n(t)} = -\sum_n c_n(t)\left|\dot{\psi}_n(t)\right\rangle e^{i\theta_n(t)}. \tag{2.81}$$

Multiplying from left with $\langle\psi_m(t)|$ and using the orthogonality of the states

$$\sum_n \dot{c}_n(t)\delta_{nm}e^{i\theta_n(t)} = -\sum_n c_n(t)\left\langle\psi_m(t)\middle|\dot{\psi}_n(t)\right\rangle e^{i\theta_n(t)}. \tag{2.82}$$

The left side is zero unless $n = m$, thus Eq.(2.82) can be rewritten as

$$\dot{c}_m(t) = -\sum_n c_n(t)\left\langle\psi_m(t)\middle|\dot{\psi}_n(t)\right\rangle e^{i(\theta_n(t)-\theta_m(t))}. \tag{2.83}$$

The term $\langle\psi_m|\dot{\psi}_n\rangle$ can be found by starting with the time derivative of Eq.(2.77)

$$\dot{H}(t)|\psi_n(t)\rangle + H(t)\left|\dot{\psi}_n(t)\right\rangle = \dot{E}_n(t)|\psi_n(t)\rangle + E_n(t)\left|\dot{\psi}_n(t)\right\rangle. \tag{2.84}$$

Taking the inner product with $|\psi_m(t)\rangle$

$$\left\langle\psi_m(t)\middle|\dot{H}(t)\middle|\psi_n(t)\right\rangle + \left\langle\psi_m(t)|H(t)|\dot{\psi}_n(t)\right\rangle = \dot{E}_n(t)\delta_{mn} + E_n(t)\left\langle\psi_m(t)\middle|\dot{\psi}_n(t)\right\rangle. \tag{2.85}$$

Once again one makes use of Eq.(2.77) and Eq.(2.85) can be rewritten as

$$\frac{\left\langle\psi_m(t)\middle|\dot{H}(t)\middle|\psi_n(t)\right\rangle}{E_n(t) - E_m(t)} - \dot{E}_n(t)\delta_{mn} = \left\langle\psi_m(t)\middle|\dot{\psi}_n(t)\right\rangle. \tag{2.86}$$

Now let us assume that the spectrum is non-degenerate such that the denominator is non-zero for any n and m. Inserting Eq.(2.86) into Eq.(2.83) gives

$$\dot{c_m}(t) = -c_m(t)\left\langle\psi_m(t)\middle|\dot{\psi_m}(t)\right\rangle - \sum_{n\neq m} c_n \frac{\left\langle\psi_m(t)\middle|\dot{H}(t)\middle|\psi_n(t)\right\rangle}{E_n(t) - E_m(t)} e^{i(\theta_n(t)-\theta_m(t))}. \tag{2.87}$$

This far we have not imposed any approximations. Having derived this expression, the adiabatic approximation states that as long as the rate of change in the Hamiltonian goes slowly enough the particle will remain in its *n*th eigenstate. Making use of the approximation one can assume that $\dot{H}(t)$ is small and neglect the terms contained in the sum, leaving only

$$\dot{c}_m(t) = -c_m(t)\left\langle\psi_m(t)\middle|\dot{\psi}_m(t)\right\rangle. \tag{2.88}$$

Equation (2.88) has the general solution

$$c_m(t) = c_m(0)e^{i\gamma_m(t)}. \tag{2.89}$$

Introducing the new phase γ (not to be confused with the decay rate)

$$\gamma_m(t) = i\int_0^t \langle\psi_m(t')|\dot{\psi_m}(t')\rangle dt'. \tag{2.90}$$

As an example consider a two state system with the states $|\psi_1\rangle$ and $|\psi_2\rangle$ so that the total state vector becomes, in accordance to Eq.(2.78)

$$|\Psi(t)\rangle = c_1(t)|\psi_1(t)\rangle e^{i\theta_1(t)} + c_2(t)|\psi_2(t)\rangle e^{i\theta_2(t)}. \tag{2.91}$$

Eq.(2.91) can be rewritten using Eq.(2.89), assuming an adiabatic process

$$|\Psi(t)\rangle = c_1(0)e^{i\gamma_1(t)}|\psi_1(t)\rangle e^{i\theta_1(t)} + c_2(0)e^{i\gamma_2(t)}|\psi_2(t)\rangle e^{i\theta_2(t)}. \tag{2.92}$$

Let the particle start in state $|\psi_1\rangle$ at time $t = 0$. That is, $c_1(0) = 1$ and $c_2(0) = 0$. Eq.(2.92) follows

$$|\Psi(t)\rangle = e^{i\gamma_1(t)}|\psi_1(t)\rangle e^{i\theta_1(t)}. \tag{2.93}$$

Thus, the particle remains in $|\psi_1\rangle$ as predicted by the adiabatic theorem.

3 Results

The effects of dissipation in an open system of STIRAP have been studied in the past [11], although most previous studies just solve the Lindblad master equation numerically [2,3]. In this thesis a different view of the STIRAP in an open system will be analyzed. As seen in the previous section 2.5, the Lindblad master equation (Eq.(2.75)) is stated in terms of the density operators (Eq.(2.2)) that was introduced in section 2.1. One can always parametrize the density operator and write it as a vector, this have previously been done in a two-level systems. In the two-level system one uses the three *Pauli operators* [1]

$$\sigma_x = |0\rangle\langle 1| + |1\rangle\langle 0|, \tag{3.1a}$$

$$\sigma_y = i|0\rangle\langle 1| - i|1\rangle\langle 0|, \tag{3.1b}$$

$$\sigma_z = |1\rangle\langle 1| - |0\rangle\langle 0|. \tag{3.1c}$$

The states $|0\rangle$ and $|1\rangle$ are the basis states for the system chosen such that the z-component of the Pauli matrices are diagonal. The state matrix can then be written as

$$\rho(t) = \frac{1}{2}\left[\hat{I} + x(t)\sigma_x + y(t)\sigma_y + z(t)\sigma_z\right]. \tag{3.2}$$

Now, in a three level system, such as STIRAP, one instead uses the eight *Gell-mann matrices (*see appendix B) to parametrize the density operator [12]. The state matrix is then written, as previously seen in Eq.(1.2)

$$\rho(t) = \frac{1}{3}\left[\hat{I} + \sqrt{3}\sum_{i=1}^{8} c_i(t)\lambda_i\right]. \tag{3.3}$$

Inserting Eq.(3.3) into the Lindblad master equation (Eq.(2.75)) one can write the corresponding equation as a first order linear differential matrix equation

$$\dot{\vec{R}} = M\vec{R} + \vec{b}, \tag{3.4}$$

where $\vec{R}$ is a vector with the coefficients $c_i(t)$ as its components. The vector $\vec{b}$ comes as a consequence of the parametrization choice and is widely dependent of the Lindblad operators (see below) in use. One can remove it by instead defining the vector $\vec{R}$ as

$$\vec{R} = \begin{bmatrix} 1 \\ c_1 \\ c_2 \\ c_3 \\ c_4 \\ c_5 \\ c_6 \\ c_7 \\ c_8 \end{bmatrix}, \tag{3.5}$$

and instead get a 9×9 dimension of matrix M. Eq.(3.4) is the equation that will further be used in the thesis where the matrix M will be studied in more detail. We shall also study the effects of the system parameters on STIRAP.

3.1 Calculation of matrix *M*

In order to do the calculation of the matrix M one must first determine the *Lindblad operators* A_k, as seen in Eq.(2.75). In this thesis, three different cases will be studied

$$Case\ 1\text{:}\ A_1 = |1\rangle\langle 2|, \qquad A_2 = |3\rangle\langle 2|, \tag{3.6a}$$

$$Case\ 2\text{:}\ B = |1\rangle\langle 1| - |3\rangle\langle 3|, \tag{3.6b}$$

$$Case\ 3\text{:}\ C = |1\rangle\langle 2| + |3\rangle\langle 2|. \tag{3.6c}$$

Equation (3.5a), case 1, is the case of dissipation from state $|2\rangle$ to either $|1\rangle$ or $|3\rangle$, this comes from spontaneous emission from the excited state $|2\rangle$. Eq.(3.5b), case 2, represent the case of *dephasing*. Dephasing is the loss of coherence between the lower states $|1\rangle$ and $|3\rangle$ as time progresses. Case 3, Eq.(3.5c), represent a process where the decay of state $|2\rangle$ goes into a superposition of $|1\rangle$ and $|3\rangle$. This is not a natural occurrence and cannot be seen in nature itself but could possibly be created in a lab for an experiment. Case 1 and case 3 seem to look very much a like from this point. We have chosen to study case 3 to see what happens if the coherence is preserved in the decay.

In what follows are the following matrices that goes for each case. Since the calculation for the matrix M is very messy the full details of the calculation for every case is presented in appendix *B*. For the three different cases one finds, for case 1

$$M_1 = \begin{pmatrix} -\gamma & \Delta & 0 & 0 & G_2 & 0 & 0 & 0 \\ -\Delta & -\gamma & -2G_1 & -G_2 & 0 & 0 & 0 & 0 \\ 0 & 2G_1 & -3\gamma/2 & 0 & 0 & 0 & -G_2 & \sqrt{3}\gamma/2 \\ 0 & G_2 & 0 & 0 & 0 & 0 & -G_1 & 0 \\ -G_2 & 0 & 0 & 0 & 0 & G_1 & 0 & 0 \\ 0 & 0 & 0 & 0 & -G_1 & -\gamma & -\Delta & 0 \\ 0 & 0 & G_2 & G_1 & 0 & \Delta & -\gamma & -\sqrt{3}G_2 \\ 0 & 0 & \sqrt{3}\gamma/2 & 0 & 0 & 0 & \sqrt{3}G_2 & -\gamma/2 \end{pmatrix}, \tag{3.7}$$

and $\vec{b}_1$ is

$$\overrightarrow{b_1} = \begin{bmatrix} 0 \\ 0 \\ \sqrt{3}\gamma/2 \\ 0 \\ 0 \\ 0 \\ 0 \\ -\gamma/2 \end{bmatrix}. \tag{3.8}$$

For case 2 the vector $\overrightarrow{b_2}$ is zero and M_2 is

$$M_2 = \begin{pmatrix} -\gamma/2 & \Delta & 0 & 0 & G_2 & 0 & 0 & 0 \\ -\Delta & -\gamma/2 & -2G_1 & -G_2 & 0 & 0 & 0 & 0 \\ 0 & 2G_1 & 0 & 0 & 0 & 0 & -G_2 & 0 \\ 0 & G_2 & 0 & -2\gamma & 0 & 0 & -G_1 & 0 \\ -G_2 & 0 & 0 & 0 & -2\gamma & G_1 & 0 & 0 \\ 0 & 0 & 0 & 0 & -G_1 & -\gamma/2 & -\Delta & 0 \\ 0 & 0 & G_2 & G_1 & 0 & \Delta & -\gamma/2 & -\sqrt{3}G_2 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{3}G_2 & 0 \end{pmatrix}. \tag{3.9}$$

And for case 3

$$M_3 = \begin{pmatrix} -\gamma & \Delta & 0 & 0 & G_2 & 0 & 0 & 0 \\ -\Delta & -\gamma & -2G_1 & -G_2 & 0 & 0 & 0 & 0 \\ 0 & 2G_1 & -\frac{3}{2}\gamma & 0 & 0 & 0 & -G_2 & \frac{\sqrt{3}}{2}\gamma \\ 0 & G_2 & -\gamma & 0 & 0 & 0 & -G_1 & \frac{\gamma}{\sqrt{3}} \\ -G_2 & 0 & 0 & 0 & 0 & G_1 & 0 & 0 \\ 0 & 0 & 0 & 0 & -G_1 & -\gamma & -\Delta & 0 \\ 0 & 0 & G_2 & G_1 & 0 & \Delta & -\gamma & -\sqrt{3}G_2 \\ 0 & 0 & \frac{\sqrt{3}}{2}\gamma & 0 & 0 & 0 & \sqrt{3}G_2 & -\frac{\gamma}{2} \end{pmatrix}, \tag{3.10}$$

$$\overrightarrow{b_3} = \begin{bmatrix} 0 \\ 0 \\ \frac{\sqrt{3}}{2}\gamma \\ \frac{\gamma}{\sqrt{3}} \\ 0 \\ 0 \\ 0 \\ -\frac{\gamma}{2} \end{bmatrix}. \tag{3.11}$$

Since the matrices are not Hermitian one has to differ between left and right eigenstates. However, one can notice how matrix M_2 (Eq.(3.9)) is skew-symmetric, while matrix M_1 and M_3 (Eq.(3.7) and (3.10)) are close but not exactly skew-symmetric. For M_2 left and right eigenstate will be the same at all times

while M_1 and M_3 get the same if one lets $\gamma = 0$. In the remaining part of the thesis the right eigenstates will be studied since we only consider the situation when the matrix M operates to the right.

3.2 The eigenvalues and eigenstates of matrix *M*

In this section a more detailed study on the eigenvalues and eigenstates for each matrix will be presented. In the case of Eq.(3.7) and (3.10) the extra $\vec{b}$ will be left out and our focus will be left on the corresponding matrix M. As usual one solves for the eigenvalues and eigenstates by the equation

$$\mathrm{Det}\left(M - \epsilon \hat{I}\right) = 0, \tag{3.14}$$

where the ϵ's are the eigenvalues and $\hat{I}$ is, as before, the unit matrix, only now in the dimension of 8×8. The matrix M, as one knows, has the dimension 8×8, therefore eight eigenvalues and eight eigenstates comes as a result. However, only a few of these states can be real physical states. The criteria determining whether a solution is physical or not is determined from the fact that the resulting state (Eq.(3.3)) has to be a proper density operator. That is (as seen in section 2.1.2), it has to be Hermitian, have unit trace, and be positive definite. The unit trace is guaranteed from its construction. Hermiticity follows if $\vec{R}$ is real. A real $\vec{R}$ does not automatically result in positivity. However $\left|\vec{R}\right| < 1$ is necessary in order to warrant positivity. Nevertheless, provided $\vec{R}$ is real, one can always make $|\vec{R}|$ sufficiently small and ρ will be positive definite. For $\gamma = \Delta = 0$, out of the 8 eigenvalues only two are real, the rest are complex. Thus, only two eigenstates out of the eight are real physical states. In figure 2, 3 and 4 below one can see how the real eigenvalues depend on γ with the pulses held constant, $G_1 = G_2 = 1$ and the detuning[4] $\Delta = 0$.

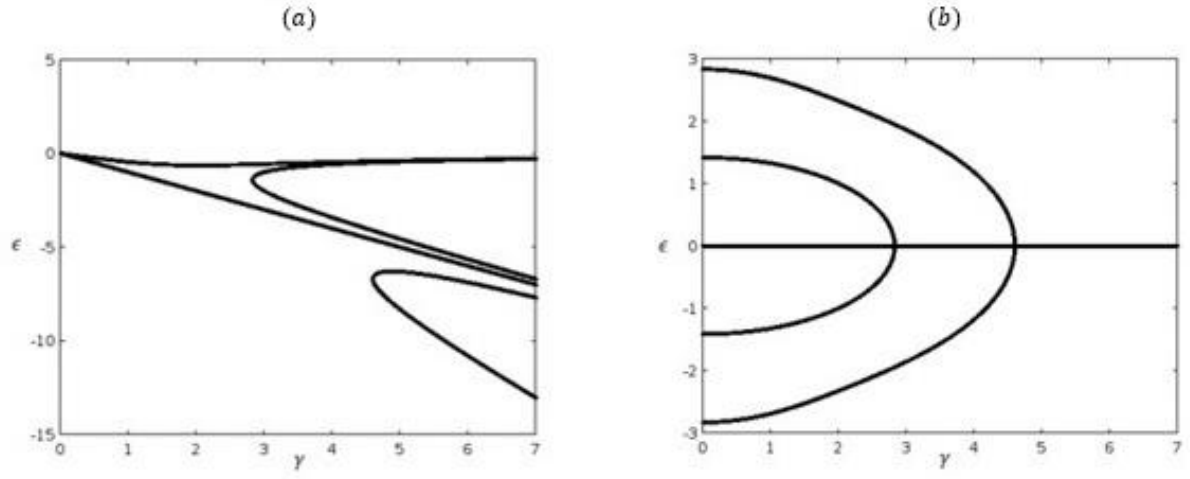

Figure 2: The eigenvalues for matrix M_1 (Eq.(3.7)) and their dependence on γ. (a) The real eigenvalues for matrix M_1. One can see how more real eigenvalues appear, thus more physical states, as γ gets bigger and that they appear in pairs due to the properties of the matrix M_1. (b) The imaginary parts of the eigenvalues for matrix M_2. One can note how the imaginary parts of the eigenvalues decay in pairs, the amplitude drops and goes to zero. At the same time purely real eigenvalues emerge.

[4] *The detuning will be zero throughout the numerical analysis as the qualitative result will not depend on Δ.*

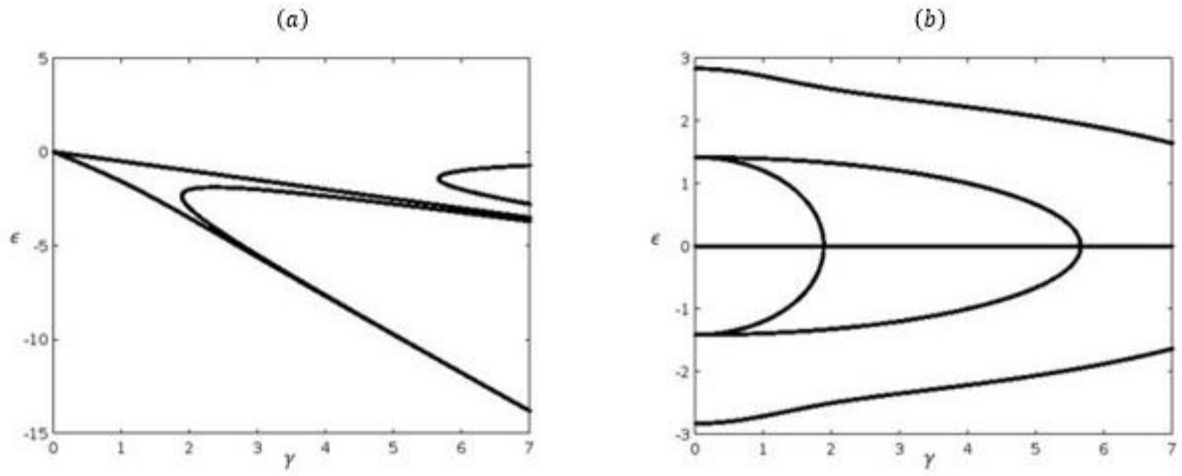

Figure 3: The eigenvalues of matrix M_2 (Eq.(3.9)) and their dependence on γ. (a) The real eigenvalues for matrix M_2. One can see how more physical states appear as γ gets bigger, eventually all eight becomes real. New purely real eigenvalues emerge in pairs and not as single ones. This is a property of the matrix M_2 as the trace is real. (b) The imaginary parts of the eigenvalues for matrix M_2 where can see how the imaginary parts of the eigenvalues decay in pairs, at the same time real eigenvalues emerge.

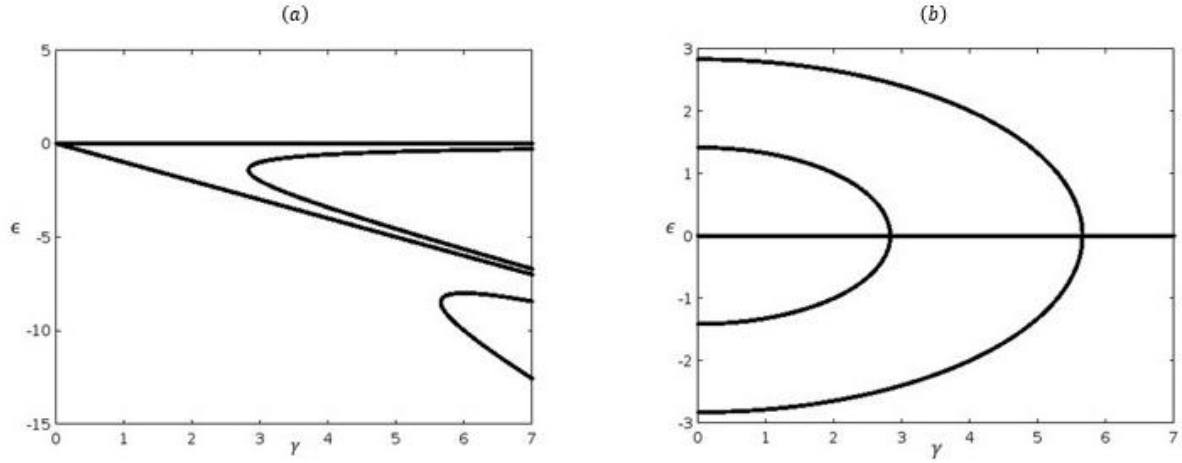

Figure 4: The eigenvalues for matrix M_3 (Eq.(3.10)) and their dependence on γ (a) The real eigenvalues for matrix M_3. More physical states appear as γ gets bigger, they appear in pairs as a consequence of the properties in matrix M_3. (b) The imaginary parts of the eigenvalues for matrix M_3 where one can see how the imaginary parts of the eigenvalues decay in pairs, at the same time real eigenvalues emerge.

Whenever an eigenvalue is purely real, it follows that the corresponding eigenvectors will be real. Thus, figure 2,3 and 4 demonstrate that it does not necessary need to be just two real states, as γ gets bigger more and more real physical states appear. At the same time the imaginary parts of the eigenvalues dissapear in pairs, the amplitude drops and goes to zero. This is expected, as a property of M is that the eigenvalues/eigenstates come in pairs if they are complex. This has to be so since the trace of M is real, and the trace is the sum of eigenvalues. This is also reflected in figure 2, 3 and 4, new purely real eigenvalues emerge in pairs, not as single ones. At the end in figure 4, with $\gamma = 7$, one has a total of six physical states. For even larger γ's, eventually all eight eigenvalues become real. Even though we call the real eigenvalues physical when they are real it should be remembered that it is not enough that they

are real for generating a physical state. However, it is found that for sufficiently small norm of the eigenstate they will represent proper physical states.

3.3 The system parameters and their effects on population transfer

As mentioned in section 2.3, the STIRAP relies on the fact that the laser pulses couples to the states in a counterintuitive order, as seen in figure 5.

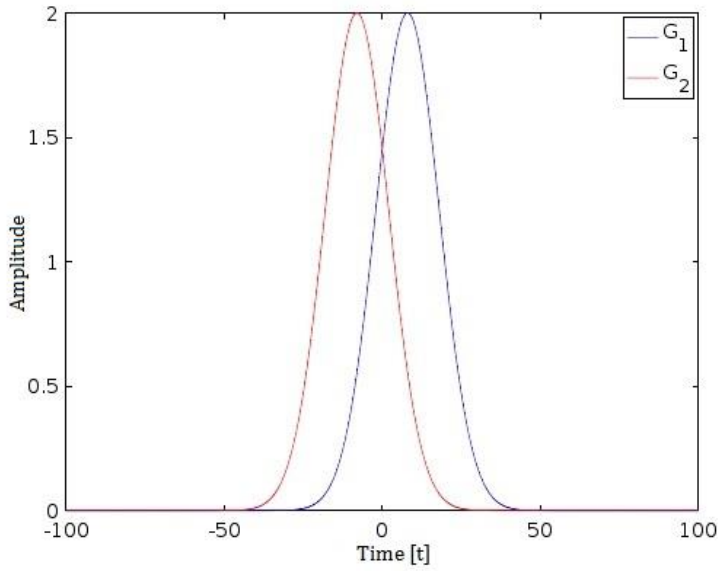

Figure 5: The counterintuitive pulse sequence for the laser pulses $G_1(t)$ and $G_2(t)$. That is, $G_2(t)$ starts before $G_1(t)$ in order to trap the population in a dark state induce a complete population transfer.

The pulses $G_1(t)$ and $G_2(t)$ have a Gaussian form

$$G_1(t) = a\exp\left(-\frac{(t-k\tau)^2}{2k\sigma^2}\right),$$
$$G_2(t) = a\exp\left(-\frac{(t+k\tau)^2}{2k\sigma^2}\right), \tag{3.15}$$

where a is the pulse amplitude, $k\sigma$ the pulse width and $2k\tau$ the distance between the pulses. The parameter k is an *'adiabaticity parameter'* that has been introduced to study the effects of having different widths of the pulses. a, τ and σ are all held constant ($a = 2, \tau = 8, \sigma = 10$) throughout the thesis. For an initial $|1\rangle$ state and provided the evolution is adiabatic, when one makes such a coupling as seen in figure 5, the population becomes trapped in the adiabatic dark state (see section 2.3). As a result, a complete population transfer is possible without ever going through the mediating state. Indeed, in a closed system ($\gamma = 0$) such a behavior is seen (figure 6 below). Throughout the thesis the time scale will be, initial time $t = -100$ and final time $t = 100$. The results will not be affected by making the scale larger and hence there is no need to make a very large time scale.

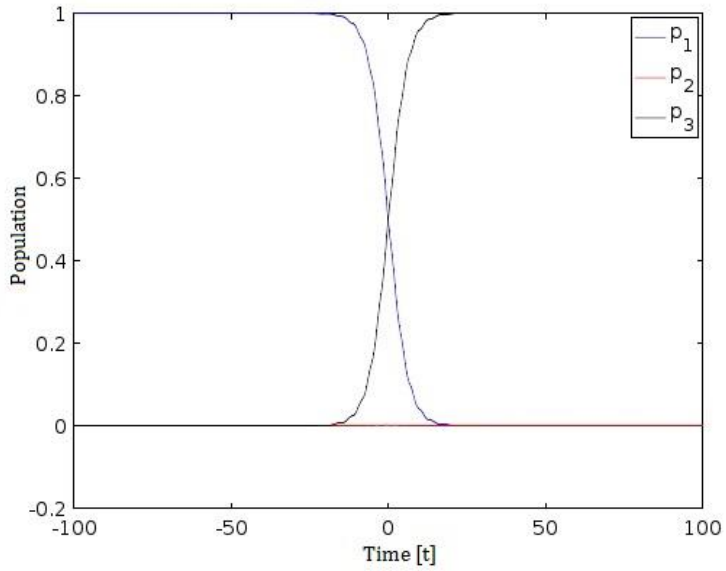

Figure 6: Population transfer in a closed system ($\gamma = 0$). A direct route between state $|1\rangle$ and $|3\rangle$ is made due to the coupling in a counterintuitive order and thus trapping the population in the dark state, one can see this by noting that throughout the process state $|2\rangle$ never gets populated and a complete population transfer is made.

Coupling to an environment quickly deteriorates the success. These effects can be seen in figure 7, for case 2 (Eq.(3.6b)) with a small coupling to the environment ($\gamma = 0.1$) .

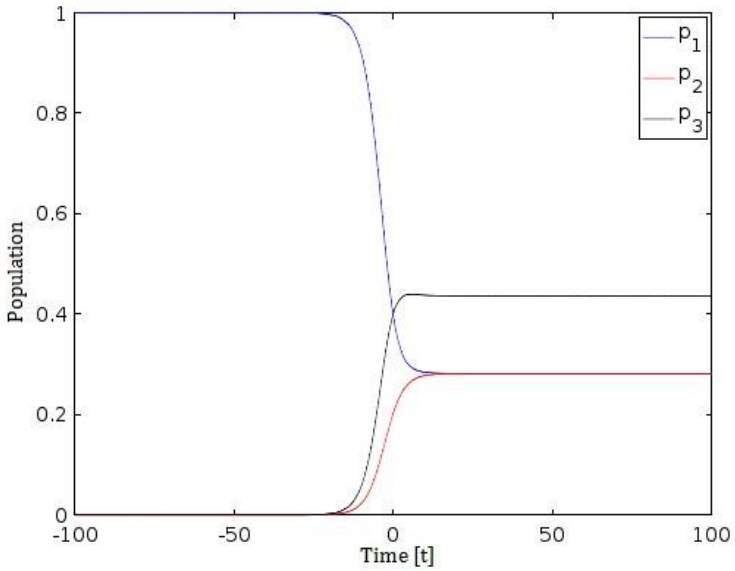

Figure 7: Population transfer in an open quantum system ($\gamma = 0.1$) in the case of dephasing, case 2. The environment greatly affects the performance of STIRAP. Note, state $|2\rangle$ now gets populated during the process.

One can clearly see in figure 7 how the environment affects the performance of STIRAP. Note that state $|2\rangle$ now gets populated during the process. Figure 8 shows how the final population in each state varies as a function of γ for the two cases of Eq.(3.6a) and (3.6b), i.e. for spontaneous emission and for the dephasing of the lower states.

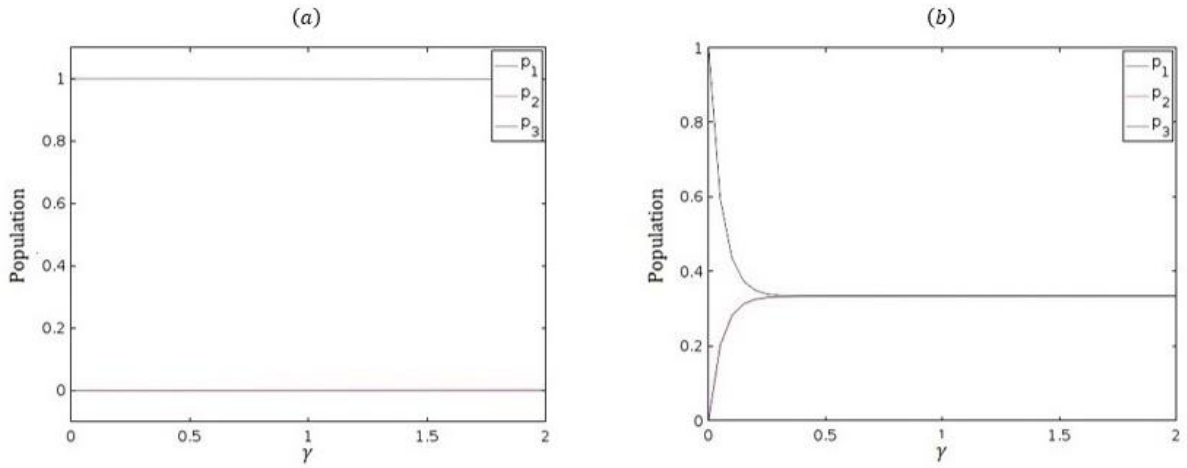

Figure 8: The final population for the different states. (a) is showing the case of spontaneous emission from state $|2\rangle$, case 1, as a function of γ. That is, one has dissipation from state $|2\rangle$ to either state $|3\rangle$ or $|1\rangle$. STIRAP does not seem to care about the decoherence that comes with case 1, even with a strong coupling to the environment a complete population transfer is made. (b) is showing the case of dephasing between the lower two states, $|1\rangle$ and $|3\rangle$. That is, loss of coherence between the two lower states. Due to the decay between the lowering states, state $|2\rangle$ will get populated and at large enough γ every state will have a final average population of $1/3$.

While some of the behavior in figure 8 are expected it is instructive to see the quantitative parameter dependence. Figure 8a shows case 1, the case of spontaneous emission from the excited state (Eq.(3.6a)). That is, one have a dissipation to either state $|1\rangle$ or $|3\rangle$ from $|2\rangle$. As seen in figure 8a, an interesting result emerge, as case 1 still makes a complete population transfer even with a strong coupling to the environment. In figure 8b one sees the case of dephasing (Eq.(3.6b)). In this case, state $|1\rangle$ and state $|3\rangle$ get out of phase throughout the evolution. As γ gets non-zero a decay process between state $|1\rangle$ and $|3\rangle$ occurs. Due to this, state $|2\rangle$ will get populated and as γ gets bigger all the states decay to the average population value of $1/3$. The last case (Eq.(3.6c)), which can never be found in nature, shows a similar result (figure 9 below) to case 1 (figure 8a). The method of STIRAP do not seem to care about the type of decoherence that comes with Eq.(3.6c) or Eq.(3.6a). Therefore, even with a strong coupling to the environment, STIRAP makes a complete population transfer from $|1\rangle$ to $|3\rangle$ without ever populating $|2\rangle$, just as before. We should point out, however, that if there is some population of the intermediate state throughout the process, due to non-adiabatic excitations, the dissipation of this level will lower the success rate of the STIRAP. The reason for exploring this more 'artificial' situation is to explore whether decoherence plays an important role for the STIRAP in case 1: Case 1 gives incoherent dissipation while case 3 represent coherent loss of $|2\rangle$.

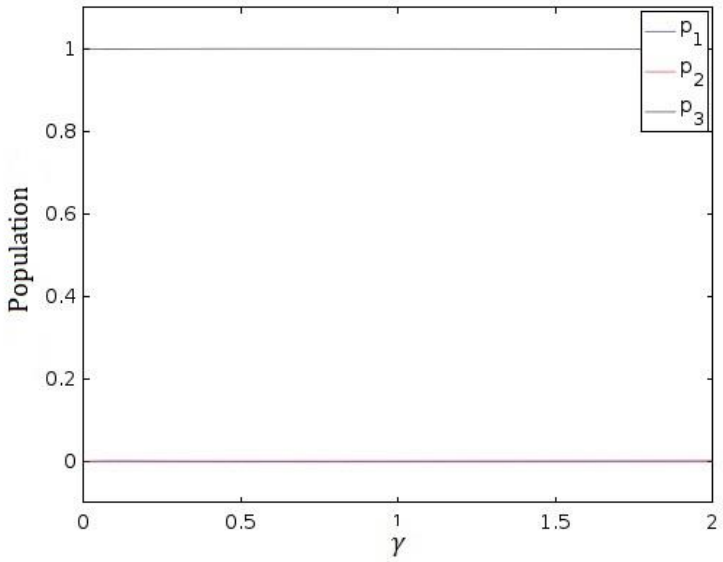

Figure 9: The same as figure 8, i.e. final population of state $|3\rangle$ in case 3. Case 3 have no visible effect on STIRAP even with a strong coupling to the environment. Instead the population transfer process is exactly the same as in the closed system ($\gamma = 0$), where the entire population goes from state $|1\rangle$ to state $|3\rangle$ without ever populating state $|2\rangle$.

The width and the ordering of the pulses effects STIRAP, therefore the introduction of the adiabaticity parameter k in Eq.(3.15). Figure 10 shows the population for state $|3\rangle$ at the final time of Eq.(3.6a) and (3.6b) for different values of k and γ. $k = 1$, is the 'default' width used in figure 5. Lower values of k means a narrower pulse.

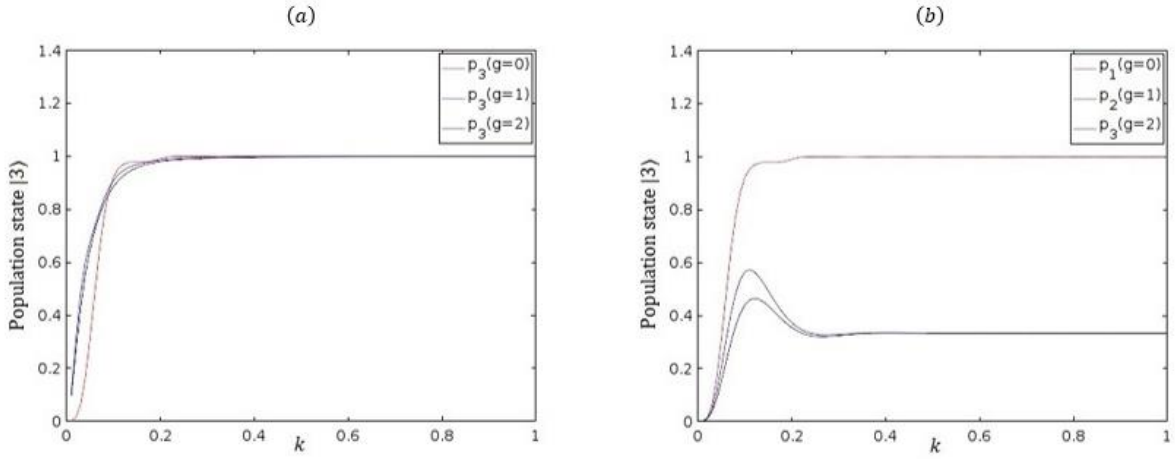

Figure 10: Population of state $|3\rangle$ at the final time t for different values of γ (denoted g in the insets) as a function of the adiabaticity parameter k. (a) shows the case of spontaneous emission from state $|2\rangle$. One can see how the population transfer, in an open system ($\gamma > 0$), is more successful than a closed system ($\gamma = 0$) if a very narrow pulse is used, i.e. small k. (b) shows the case of dephasing between the lower two states. Yet again the population transfer is more successful, in the open system, if the pulse is narrow.

One can in figure 10b envision how the population transfer is adiabatic from $k \approx 0.3$ to $k = 1$ and $k \approx 0.1$ to $k = 1$ for figure 10a in the closed case. Interesting result are show in figure 10a, which represent case 1. One can see how the population transfer is more successful in an open system than in the closed

system for a very narrow pulse (small k). Hence, a small pulse width will improve the success of STIRAP in an open system. Similar effects can also be seen in figure 10b, that if the system is subject to losses making the process implies higher success rate. However, the closed system is still more successful at all different values of k .The unnatural case, case 3, displays a very similar behavior to case 1 in figure 10a.

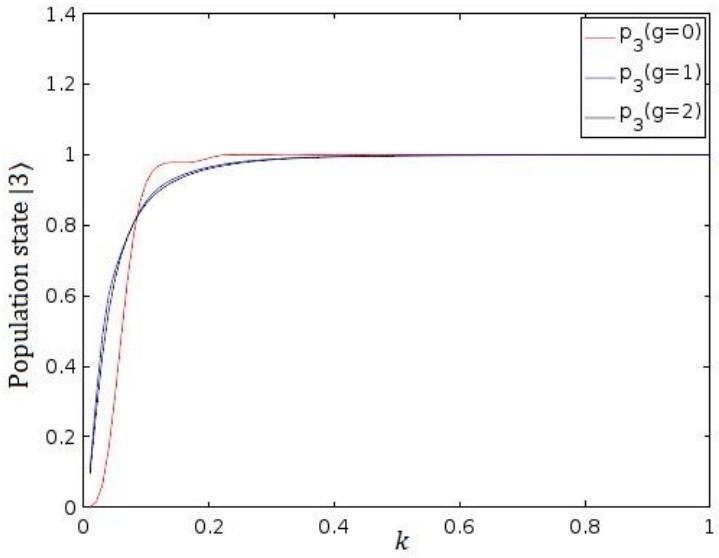

Figure 11: The final population of state $|3\rangle$ at large t for different values of γ (denoted g in the insets) as a function of the adiabaticity parameter k. Here, one can see an unexpected event, that for small k, an open system can improve the success of the STIRAP scheme.

In figure 11 one can see a similar behavior as case 1 do in figure 10a. The final population transfer, at small values of k, for state $|3\rangle$ with $\gamma > 0$ turns out to be more successful than $\gamma = 0$. For both case 1 and case 3 the STIRAP process is favored by the coupling to an environment at small k.

3.4 Purity

As seen in section 2.1, the density matrix can be either pure or mixed. One can calculate the *purity* of a density matrix as

$$P = Tr(\rho^2). \tag{3.16}$$

For a mixed state, as seen in section 2.1.2, the purity follow

$$\frac{1}{d} \leq P \leq 1, \tag{3.17}$$

where d is the dimension of the density matrix. Thus, in this case $d = 3$. A completely mixed state is $P = 1/3$ and a completely pure state is $P = 1$.

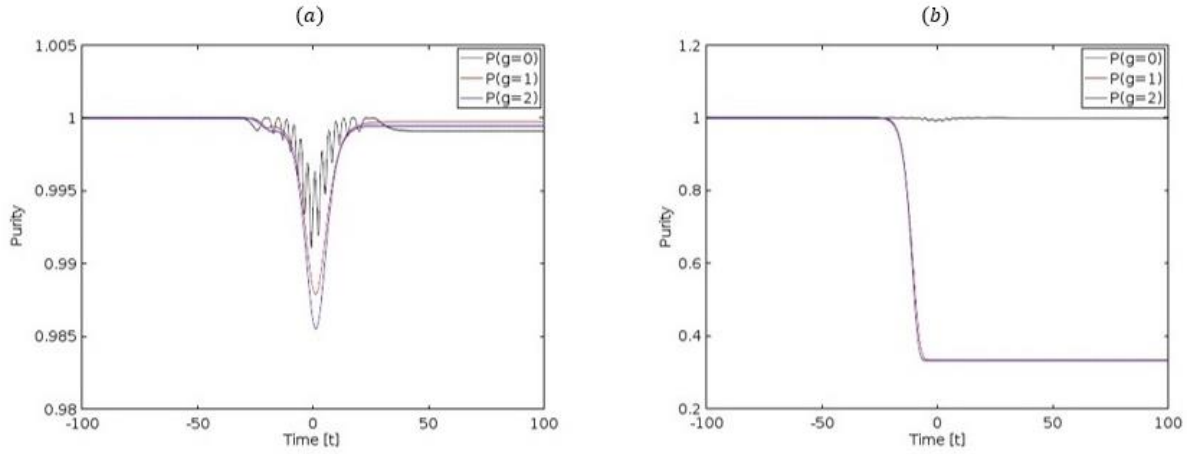

Figure 12: Purity, for different values of γ (denoted g in the insets), as a function of time, (a) shows the purity for case 1. One can see that the state ends very slightly mixed, yet again showing how the environment have little effect on case 1. (b) purity for case 2 where it is clear that the state at the end of the process becomes completely mixed at $P = 1/3$.

Figure 12a shows that the state for case 1 is partly mixed at the final time $t = 100$, that is, $P > 1/3$. Figure 12b shows how for case 2 the system evolves into a completely mixed state at the final time. That is, the purity $P = 1/3$.

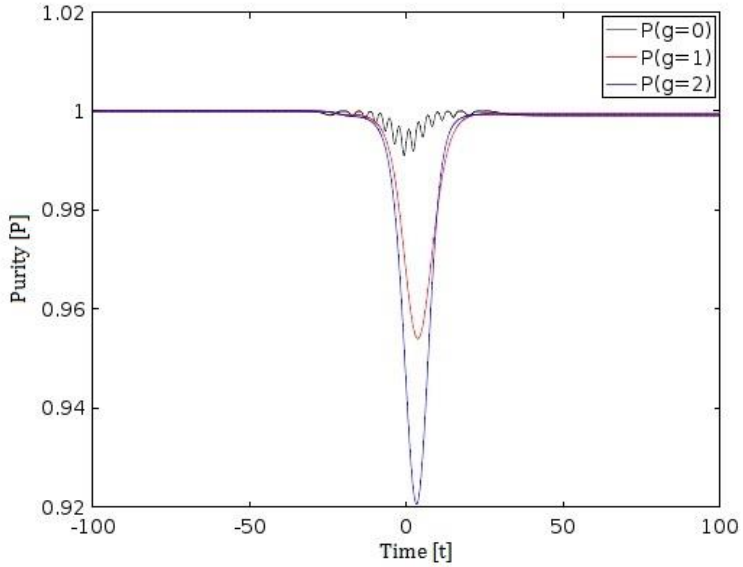

Figure 13: Purity, for different values of γ (denoted g in the insets), for case 3 as a function of time. Once again one can see how STIRAP is hardly affected by the decoherence coming from case 3. The state is starts and ends completely pure only being slightly mixed in the middle.

Figure 13 shows the behavior of case 3 which greatly resembles case 1 (figure 12a). One can clearly see, despite a coupling to the environment at different strengths, how the state starts and ends completely pure and only being slightly mixed in the middle. This shows, yet again, that the environment has little effect for case 3 and case 2, as previously seen in figure 8,9,10 and 11.

4 Conclusions

The results above show the effects of decoherence on the open STIRAP problem. The thesis was aimed at looking at a new approach to the problem, by parametrizing the density operator. The Lindblad master equation (Eq.(2.75)) could be rewritten as a first order linear differential matrix equation. Different types of matrices (Eqs.(3.7), (3.9) and Eq.(3.10)), depending on the actual Lindblad operators studied, in the dimension of 8×8 come as a result. The study showed that under a closed system only two out of eight eigenstates of these matrices showed to be real physical states. That is, the corresponding state has to be a proper density operator. Taking an open system and constant pulses ($G_1 = G_2 = 1$), one could see how more and more physical states emerged as γ was increased, and at the same time the imaginary eigenvalues decayed in pairs. Eventually, for case 2, all eight eigenvalues become real. As a further property of the matrix M the eigenvalues and eigenstates come in pairs if they are complex, this is expected, as the trace of matrix M is real. It is interesting to note that, a previous study [12] making a similar parametrization and writing a corresponding matrix never took this fact that only some of the states are physical into consideration in their study of *Berry phases*.

The system parameters effects on population transfer showed some expected results of STIRAP, that is, the success rate of STIRAP quickly deteriorates when coupling to an environment in the natural case, case 2. Case 1 showed to have no significant effect on STIRAP and greatly resembled case 3, which can never be found in nature itself. Even with a strong coupling to an environment, STIRAP behaved approximately as in the closed system for both cases. One conclusion to draw from this artificial case which preserves coherence is that, STIRAP is very sensitive for dephasing. Case 1 gives dissipation from state $|2\rangle$, but at the same time gives dephasing of the lower states, while case 3 only gives dissipation.

STIRAP relies widely on adibaticity, in order to make the process adiabatic the pulses need to have the correct width and be on the right position relative to each other and make a slight overlap. Setting the pulses relative position fixed, we introduced the adiabaticity parameter k to study the effects of the pulses width on population transfer. The study showed that with γ applied one can accomplish a greater success in population transfer when manipulating the pulses to a more narrow pulse width. Moreover, in case 3 and case 1, one can accomplish a more successful population transfer in an open system ($\gamma > 0$) than in a closed system ($\gamma = 0$), if the pulses are narrow enough. Hence, in that case, STIRAP works more efficiently with a coupling to the environment than without. Adiabaticity favors long time scales while a system interacting with an environment favors a short time scale. The study confirms that in an open quantum system the population transfer is more successful if the pulses are narrower, thus, a faster process. However, for case 3 and case 1, a coupling to the environment leads to just as good or greater success, for almost all values of k even when the process is slow, than for a closed system. Nevertheless, case 3 is not a natural occurring case but case 1 is.

Appendix

A The Gell-Mann matrices

In a three level system one can use the eight Gell-Mann matrices to parametrize the density operator, when doing so one ends up with the following expression

$$\rho(t) = \frac{1}{3}\left[\hat{I} + \sqrt{3}\sum_{i=1}^{8} c_i(t)\lambda_i\right]. \tag{A.1}$$

Here, λ_i are the Gell-Mann matrices. The Gell-Mann matrices are eight different matrices named after Murray Gell-Mann; they are traceless and Hermitian and play the same role for three-level systems as Pauli matrices play for two-level systems. The matrices are [13]

$$\lambda_1 = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \lambda_2 = \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \lambda_3 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

$$\lambda_4 = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \quad \lambda_5 = \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix}, \quad \lambda_6 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \tag{A.2}$$

$$\lambda_7 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix}, \quad \lambda_8 = \frac{1}{\sqrt{3}}\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}.$$

B Detailed calculation of matrix M

The calculation of the matrix M is messy but straightforward and will be presented here in more detail. Some parts are not written out into its full extent but are easy to verify as they are straightforward and simple use of basic linear algebra and the bracket notations.

B.1 Calculation of the commutator

Let us start by calculating the commutator representing the regular Hamiltonian dynamics in the master equation (Eq.(2.75))

$$-i[H_s(t), \rho(t)] = -i(H_s(t)\rho(t) - \rho(t)H_s(t)). \tag{B.1}$$

For the ease of the calculation the time-dependence of the density operator and the Hamiltonian will not be written out in the remaining calculation of the appendix. Let us start by taking the STIRAP Hamiltonian, as seen in Eq.(2.39), and writing it in bracket notation.

$$H_s = G_1(|1\rangle\langle 2| + |2\rangle\langle 1|) + \Delta|2\rangle\langle 2| + G_2(|2\rangle\langle 3| + |3\rangle\langle 2|). \tag{B.2}$$

Here the kets can be represented as

$$|1\rangle = \begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix}, \tag{B.3}$$

$$|2\rangle = \begin{bmatrix} 0 \\ 1 \\ 0 \end{bmatrix}, \tag{B.4}$$

$$|3\rangle = \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix}. \tag{B.5}$$

And the bras as the transpose of these vectors. The first part of the commutator can then be written, by the use of the parametrization of the density operator (Eq.(3.3)), as

$$\begin{aligned} H_s\rho = \frac{1}{3}\Bigg[& H_s + \sqrt{3}\sum_{i=1}^{8} c_i G_1(|1\rangle\langle 2| + |2\rangle\langle 1|)\lambda_i + \Delta c_i |2\rangle\langle 2|\lambda_i \\ & + G_2 c_i(|2\rangle\langle 3| + |3\rangle\langle 2|)\lambda_i \Bigg]. \end{aligned} \tag{B.6}$$

Here c_i are time-dependent coefficients and λ_i are the eight Gell-Mann matrices. The sums follows

$$\begin{aligned} \sqrt{3}\sum_{i=1}^{8} c_i|1\rangle\langle 2|\lambda_i = \sqrt{3}[& c_1|1\rangle\langle 1| + c_2 i|1\rangle\langle 1| - c_3|1\rangle\langle 2| + c_6|1\rangle\langle 3| - c_7 i|1\rangle\langle 3| \\ & + \frac{1}{\sqrt{3}} c_8|1\rangle\langle 2|], \end{aligned} \tag{B.7}$$

$$\begin{aligned} \sqrt{3}\sum_{i=1}^{8} c_i|2\rangle\langle 1|\lambda_i = \sqrt{3}[& c_1|2\rangle\langle 2| - c_2 i|2\rangle\langle 2| + c_3|2\rangle\langle 1| + c_4|2\rangle\langle 3| - \\ & - c_5 i|2\rangle\langle 3| + \frac{1}{\sqrt{3}} c_8|2\rangle\langle 1|], \end{aligned} \tag{B.8}$$

$$\begin{aligned} \sqrt{3}\sum_{i=1}^{8} c_i|2\rangle\langle 3|\lambda_i = \sqrt{3}[& c_4|2\rangle\langle 1| + c_5 i|2\rangle\langle 1| + c_6|2\rangle\langle 2| + \\ & + c_7 i|2\rangle\langle 2| - \frac{2}{\sqrt{3}} c_8|2\rangle\langle 3|], \end{aligned} \tag{B.9}$$

$$\begin{aligned} \sqrt{3}\sum_{i=1}^{8} c_i|3\rangle\langle 2|\lambda_i = \sqrt{3}[& c_1|3\rangle\langle 1| + c_2 i|3\rangle\langle 1| - c_3|3\rangle\langle 2| + c_6|3\rangle\langle 3| - \\ & - c_7 i|3\rangle\langle 3| + \frac{1}{\sqrt{3}} c_8|3\rangle\langle 3|]. \end{aligned} \tag{B.10}$$

Plugging Eq.(B.7), (B.8), (B.9) and (B.10) into Eq.(B.6) and one obtains

$$
\begin{aligned}
H_s\rho = \frac{1}{3}\Big[&\sqrt{3}(G_1c_1 + G_1c_2i)|1\rangle\langle 1| + \big(G_1 - \sqrt{3}G_1c_3 + +G_1c_8\big)|1\rangle\langle 2| \\
&+ \sqrt{3}(G_1c_6 - G_1c_7i)|1\rangle\langle 3| \\
&+ \Big(G_1 + G_1c_8 + \sqrt{3}(\Delta c_1 + \Delta c_2 i + G_2c_4 + G_2c_5i + G_1c_3)\Big)|2\rangle\langle 1| \\
&+ \Big(\Delta + \Delta c_8 + \sqrt{3}(G_1c_1 + G_1c_2i - \Delta c_3 + G_2c_6 + G_2c_7i)\Big)|2\rangle\langle 2| \\
&+ \Big(G_2 - 2G_2c_8 + \sqrt{3}(G_1c_4 - G_1c_5i + \Delta c_6 - \Delta c_7 i)\Big)|2\rangle\langle 3| \\
&+ \sqrt{3}(G_2c_1 + G_2c_2i)|3\rangle\langle 1| + \big(G_2 - \sqrt{3}G_2c_3 + G_2c_8\big)|3\rangle\langle 2| \\
&+ \sqrt{3}(G_2c_6 - G_2c_7i)|3\rangle\langle 3|\Big].
\end{aligned}
\tag{B.11}
$$

Doing the same with the second part of the commutator in Eq.(B.1)

$$
\begin{aligned}
\rho H_s = \frac{1}{3}\Bigg[&H_s + \sqrt{3}\sum_{i=1}^{8} G_1c_i\lambda_i(|1\rangle\langle 2| + |2\rangle\langle 1|) + \Delta c_i\lambda_i|2\rangle\langle 2| \\
&+ G_2c_i\lambda_i(|2\rangle\langle 3| + |3\rangle\langle 2|)\Bigg],
\end{aligned}
\tag{B.12}
$$

where Eq.(B.12) can be rewritten as

$$
\begin{aligned}
\rho H_s = \frac{1}{3}\Big[&\sqrt{3}(G_1c_1 - G_1c_2i)|1\rangle\langle 1| \\
&+ \Big(G_1 + G_1c_8 + \sqrt{3}(G_1c_3 + \Delta c_1 - \Delta c_2 i + G_2c_4 - G_2c_5i)\Big)|1\rangle\langle 2| \\
&+ \sqrt{3}(G_2c_1 - G_2c_2i)|1\rangle\langle 3| + \big(G_1 + G_1c_8 - \sqrt{3}G_1c_3\big)|2\rangle\langle 1| \\
&+ \Big(\Delta + \Delta c_8 + \sqrt{3}(G_1c_1 + G_1c_2i - \Delta c_3 + G_2c_6 - G_2c_7i)\Big)|2\rangle\langle 2| \\
&+ \big(G_2 - \sqrt{3}G_2c_3 + G_2c_8\big)|2\rangle\langle 3| + \sqrt{3}(G_1c_6 + G_1c_7i)|3\rangle\langle 1| \\
&+ \Big(G_2 - 2G_2c_8 + \sqrt{3}(G_1c_4 + G_1c_5i + \Delta c_6 + \Delta c_7 i)\Big)|3\rangle\langle 2| \\
&+ \sqrt{3}(G_2c_6 + G_2c_7i)|3\rangle\langle 3|\Big].
\end{aligned}
\tag{B.13}
$$

By the use of Eqs.(B.11) and (B.13) one can calculate the commutator (Eq.(B.1)) and write it as an ordinary matrix

$$
-i[H,\rho] = -\frac{i}{3}\begin{pmatrix}
2\sqrt{3}G_1c_2i & \begin{matrix}-\sqrt{3}2G_1c_3 + \\ +\sqrt{3}\Delta(-c_1 + c_2i) + \\ +\sqrt{3}G_2(-c_4 + c_5i)\end{matrix} & \begin{matrix}\sqrt{3}G_1(c_6 - c_7i) + \\ +\sqrt{3}G_2(-c_1 + c_2i)\end{matrix} \\
\begin{matrix}2\sqrt{3}G_1c_3 + \\ +\Delta\sqrt{3}(c_1 + c_2i) + \\ +G_2\sqrt{3}(c_4 + c_5i)\end{matrix} & \begin{matrix}-2\sqrt{3}G_1c_2i + \\ +2\sqrt{3}G_2c_7i\end{matrix} & \begin{matrix}\sqrt{3}G_1(c_4 - c_5i) + \\ +\sqrt{3}\Delta(c_6 - c_7i) + \\ +G_2\big(\sqrt{3}c_3 - 3c_8\big)\end{matrix} \\
\begin{matrix}\\ G_1\sqrt{3}(-c_6 - c_7i) + \\ +G_2\sqrt{3}(c_1 + c_2i)\end{matrix} & \begin{matrix}\sqrt{3}G_1(-c_4 - c_5i) + \\ +\sqrt{3}\Delta(-c_6 - c_7i) + \\ +G_2\big(-c_3\sqrt{3} + 3c_8\big)\end{matrix} & -2\sqrt{3}G_2c_7i
\end{pmatrix}.
\tag{B.14}
$$

B.2 Calculation of case 1

In case 1, the Lindblad jump operators are

$$A_1 = |1\rangle\langle 2|, \tag{B.15a}$$

$$A_2 = |3\rangle\langle 2|. \tag{B.15b}$$

The corresponding master equation then reads

$$\begin{aligned}\dot{\rho} = -i[H_S, \rho] + \gamma(A_1\rho A_1^\dagger - \frac{1}{2}\left(A_1^\dagger A_1\rho + \rho A_1^\dagger A\right) + \\ +\gamma(A_2\rho A_2^\dagger - \frac{1}{2}\left(A_2^\dagger A_2\rho + \rho A_2^\dagger A\right).\end{aligned} \tag{B.16}$$

The calculation is again straightforward and follows by the use of the parametrization of the density operator (Eq.(3.3)) as before

$$A_1\rho A_1^\dagger = \frac{1}{3}(|1\rangle\langle 2| + \sqrt{3}\sum_{i=1}^{8} c_i|1\rangle\langle 2|\lambda_i|2\rangle\langle 1|), \tag{B.17}$$

$$A_1^\dagger A_1\rho = \frac{1}{3}(|2\rangle\langle 2| + \sqrt{3}\sum_{i=1}^{8} c_i|2\rangle\langle 2|\lambda_i), \tag{B.18}$$

$$\rho A_1^\dagger A = \frac{1}{3}(|2\rangle\langle 2| + \sqrt{3}\sum_{i=1}^{8} c_i\lambda_i|2\rangle\langle 2|). \tag{B.19}$$

Here the sums are

$$\begin{aligned}\sqrt{3}\sum_{i=1}^{8} c_i|2\rangle\langle 2|\lambda_i = \sqrt{3}(c_1|2\rangle\langle 1| + c_2 i|2\rangle\langle 1| - c_3|2\rangle\langle 2| + c_6|2\rangle\langle 3| - \\ -c_7 i|3\rangle\langle 2| + \frac{c_8}{\sqrt{3}}|2\rangle\langle 2|),\end{aligned} \tag{B.20}$$

$$\begin{aligned}\sqrt{3}\sum_{i=1}^{8} c_i\lambda_i|2\rangle\langle 2| = \sqrt{3}[c_1|1\rangle\langle 2| - c_2 i|1\rangle\langle 2| - c_3|2\rangle\langle 2| + c_6|3\rangle\langle 2| + \\ +c_7 i\,|3\rangle\langle 2| + \frac{1}{\sqrt{3}}c_8|2\rangle\langle 2|],\end{aligned} \tag{B.21}$$

$$\sqrt{3}\sum_{i=1}^{8} c_i|1\rangle\langle 2|\lambda_i|2\rangle\langle 1| = -\sqrt{3}c_3|1\rangle\langle 1| + c_8|1\rangle\langle 1|. \tag{B.22}$$

Let us define L_1 as

$$L_1 = \gamma\left(A_1\rho A_1^\dagger - \frac{1}{2}\left(A_1^\dagger A_1\rho + \rho A_1^\dagger A\right)\right), \tag{B.23}$$

and it follows after plugging Eqs.(B.17), (B.18) and (B.19) into Eq.(B.23) and using the calculated sums

$$L_1 = \frac{\gamma}{3}[\left(1 - \sqrt{3}c_3 + c_8\right)|1\rangle\langle 1| - \frac{\sqrt{3}}{2}(c_1 - c_2 i)|1\rangle\langle 2| -$$
$$-\frac{\sqrt{3}}{2}(c_1 + c_2 i)|2\rangle\langle 1| + \left(-1 + \sqrt{3}c_3 - c_8\right)|2\rangle\langle 2| - \quad (B.24)$$
$$-\frac{\sqrt{3}}{2}(c_6 - c_7 i)|2\rangle\langle 3| - \frac{\sqrt{3}}{2}(c_6 + c_7 i)|3\rangle\langle 2|]\,.$$

Doing the same for A_2

$$A_2 \rho A_2^\dagger = \frac{1}{3}(|3\rangle\langle 3| + \sqrt{3}\sum_{i=1}^{8} c_i |3\rangle\langle 2|\lambda_i|2\rangle\langle 3|), \quad (B.25)$$

$$A_2^\dagger A\rho = \frac{1}{3}(|2\rangle\langle 2| + \sqrt{3}\sum_{i=1}^{8} c_i |2\rangle\langle 2|\lambda_i), \quad (B.26)$$

$$\rho A_2^\dagger A_2 = \frac{1}{3}(|2\rangle\langle 2| + \sqrt{3}\sum_{i=1}^{8} c_i \lambda_i |2\rangle\langle 2|). \quad (B.27)$$

With the corresponding sums sums

$$\sqrt{3}\sum_{i=1}^{8} c_i |3\rangle\langle 2|\lambda_i|2\rangle\langle 3| = -\sqrt{3}c_3|3\rangle\langle 3| + c_8|3\rangle\langle 3|, \quad (B.28)$$

$$\sqrt{3}\sum_{i=1}^{8} c_i |2\rangle\langle 2|\lambda_i = \sqrt{3}(c_1|2\rangle\langle 1| + ic_2|2\rangle\langle 1| - c_3|2\rangle\langle 2| + c_6|2\rangle\langle 3| - \quad (B.29)$$
$$-c_7 i|2\rangle\langle 3| - \frac{1}{\sqrt{3}}c_8|2\rangle\langle 2|),$$

$$\sqrt{3}\sum_{i=1}^{8} c_i \lambda_i |2\rangle\langle 2|) = \sqrt{3}(c_1|1\rangle\langle 2| - c_2 i|1\rangle\langle 2| - c_3|2\rangle\langle 2| + c_6|3\rangle\langle 2| + \quad (B.30)$$
$$+c_7 i|3\rangle\langle 2| - \frac{1}{\sqrt{3}}c_8|3\rangle\langle 3|).$$

And again let us define L_2 as

$$L_2 = \gamma\left(A_2 \rho A_2^\dagger - \frac{1}{2}\left(A_2^\dagger A\rho + \rho A_2^\dagger A_2\right)\right), \quad (B.31)$$

and we arrive at

$$L_2 = \frac{\gamma}{3}[(c_1 - c_2 i)|1\rangle\langle 2| - \frac{\sqrt{3}}{2}(c_1 + c_2 i)|2\rangle\langle 1| + \left(-1 + \sqrt{3}c_3 - c_8\right)|2\rangle\langle 2|$$
$$-\frac{\sqrt{3}}{2}(c_6 - c_7 i)|2\rangle\langle 3| - \frac{\sqrt{3}}{2}(c_6 + c_7 i)|3\rangle\langle 2| \quad (B.32)$$
$$+\left(1 - \sqrt{3}c_3 + c_8\right)|3\rangle\langle 3|].$$

Combining Eq.(B.32) with Eq.(B.23) and plugging them into Eq.(B.16) together with the commutator Eq.(B.14) and one will find a 3×3 matrix. This matrix can then be used to identify each time derivative of the time-dependent coefficients c_i in $\dot{\rho}$ which has the form

$$\dot{\rho} = \frac{\sqrt{3}}{3}\sum_{i=1}^{8} \dot{c}_i \lambda_i. \tag{B.33}$$

The final equation then takes the form

$$\dot{\vec{R}} = M_1 \vec{R} + \overrightarrow{b_1}, \tag{B.34}$$

where the matrix M_1 is

$$M_1 = \begin{pmatrix} -\gamma & \Delta & 0 & 0 & G_2 & 0 & 0 & 0 \\ -\Delta & -\gamma & -2G_1 & -G_2 & 0 & 0 & 0 & 0 \\ 0 & 2G_1 & -3\gamma/2 & 0 & 0 & 0 & -G_2 & \sqrt{3}\gamma/2 \\ 0 & G_2 & 0 & 0 & 0 & 0 & -G_1 & 0 \\ -G_2 & 0 & 0 & 0 & 0 & G_1 & 0 & 0 \\ 0 & 0 & 0 & 0 & -G_1 & -\gamma & -\Delta & 0 \\ 0 & 0 & G_2 & G_1 & 0 & \Delta & -\gamma & -\sqrt{3}G_2 \\ 0 & 0 & \sqrt{3}\gamma/2 & 0 & 0 & 0 & \sqrt{3}G_2 & -\gamma/2 \end{pmatrix}, \tag{B.35}$$

and $\vec{b}_1$ and $\vec{R}$ are

$$\overrightarrow{b_1} = \begin{bmatrix} 0 \\ 0 \\ \sqrt{3}\gamma/2 \\ 0 \\ 0 \\ 0 \\ 0 \\ -\gamma/2 \end{bmatrix}, \qquad \vec{R} = \begin{bmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \\ c_5 \\ c_6 \\ c_7 \\ c_8 \end{bmatrix}. \tag{B.36}$$

B.3 Calculation of case 2

In case 2 the Lindblad jump operator is

$$B = |1\rangle\langle 1| - |3\rangle\langle 3|. \tag{B.37}$$

The calculations follows as in the previous section appendix A.2, starting by identifying each term in

$$L_3 = \gamma\left(B\rho B^\dagger - \frac{1}{2}\left(B^\dagger B\rho + \rho B^\dagger B\right)\right). \tag{B.38}$$

Having the same approach as before, using Eq.(3.3) and Eq.(B.37) one can calculate each part of Eq.(B.38). One derives the following

$$B\rho B^{\dagger} = \frac{1}{3}\left(|1\rangle\langle 1| + |3\rangle\langle 3| + \sqrt{3}\sum_{i=1}^{8} c_i[(|1\rangle\langle 1| - |3\rangle\langle 3|)\lambda_i(|1\rangle\langle 1| - |3\rangle\langle 3|)] \right), \tag{B.39}$$

$$B^{\dagger}B\rho = \frac{1}{3}\left(|1\rangle\langle 1| + |3\rangle\langle 3| + \sqrt{3}\sum_{i=1}^{8} c_i[(|1\rangle\langle 1| + |3\rangle\langle 3|)\lambda_i] \right), \tag{B.40}$$

$$\rho B^{\dagger}B = \frac{1}{3}\left(|1\rangle\langle 1| + |3\rangle\langle 3| + \sqrt{3}\sum_{i=1}^{8} c_i\lambda_i(|1\rangle\langle 1| - |3\rangle\langle 3|) \right). \tag{B.41}$$

Identifying each summation term

$$\begin{aligned} &\sqrt{3}\sum_{i=1}^{8} c_i[(|1\rangle\langle 1| - |3\rangle\langle 3|)\lambda_i(|1\rangle\langle 1| - |3\rangle\langle 3|)] = \\ &= \sqrt{3}\big(c_3|1\rangle\langle 1| - c_4(|1\rangle\langle 3| + |3\rangle\langle 1|) + ic_5(|1\rangle\langle 3| - |3\rangle\langle 1|)\big) + \\ &\quad + c_8(|1\rangle\langle 1| - 2|3\rangle\langle 3|), \end{aligned} \tag{B.42}$$

$$\begin{aligned} \sqrt{3}\sum_{i=1}^{8} c_i|1\rangle\langle 1|\lambda_i \; &= \sqrt{3}(c_1|1\rangle\langle 2| - c_2 i|1\rangle\langle 2| + c_3|1\rangle\langle 1| + c_4|1\rangle\langle 3| - c_5 i|1\rangle\langle 3|) + \\ &\quad + c_8|1\rangle\langle 1|, \end{aligned} \tag{B.43}$$

$$\sqrt{3}\sum_{i=1}^{8} c_i|3\rangle\langle 3|\lambda_i = \sqrt{3}(c_4|3\rangle\langle 1| + c_5 i|3\rangle\langle 1| + c_6|3\rangle\langle 2| + c_7 i|3\rangle\langle 2|) - 2c_8|3\rangle\langle 3| \tag{B.44}$$

$$\begin{aligned} \sqrt{3}\sum_{i=1}^{8} c_i\lambda_i|1\rangle\langle 1| = \sqrt{3}(&c_1|2\rangle\langle 1| + c_2 i|2\rangle\langle 1| + c_{\#}|1\rangle\langle 1| + c_4|3\rangle\langle 1| + c_5 i|3\rangle\langle 1|) \\ &+ c_8|1\rangle\langle 1|, \end{aligned} \tag{B.45}$$

$$\sqrt{3}\sum_{i=1}^{8} c_i\lambda_i|3\rangle\langle 3| = \sqrt{3}(c_4|1\rangle\langle 3| - c_5 i|1\rangle\langle 3| + c_6|2\rangle\langle 3| - c_7 i|2\rangle\langle 3|) - 2c_8|3\rangle\langle 3| \tag{B.46}$$

Plugging Eqs.(B.39), (B.40) and Eq.(B.41) into Eq.(B.38) with the corresponding calculated sums, and one will find

$$\begin{aligned} L_3 = \frac{\gamma}{3}\Bigg[&-\frac{\sqrt{3}}{2}(c_1 - c_2 i)|1\rangle\langle 2| + 2\sqrt{3}(-c_4 + ic_5)|1\rangle\langle 3| - \frac{\sqrt{3}}{2}(c_1 + c_2 i)|2\rangle\langle 1| \\ &- \frac{\sqrt{3}}{2}(c_6 - c_7 i)|2\rangle\langle 3| + 2\sqrt{3}(-c_4 - ic_5)|3\rangle\langle 1| \\ &- \frac{\sqrt{3}}{2}(c_6 + c_7 i)|3\rangle\langle 2| \Bigg] \end{aligned} \tag{B.47}$$

Combining Eq.(B.47) with the commutator (Eq.(B.14)) and identifying the time derivative of the time-dependent coefficients as done in section A.2. One will then get an equation on the form

$$\dot{\vec{R}} = M_2 \vec{R}, \tag{B.48}$$

where the matrix M_2 is

$$M_2 = \begin{pmatrix} -\gamma/2 & \Delta & 0 & 0 & G_2 & 0 & 0 & 0 \\ -\Delta & -\gamma/2 & -2G_1 & -G_2 & 0 & 0 & 0 & 0 \\ 0 & 2G_1 & 0 & 0 & 0 & 0 & -G_2 & 0 \\ 0 & G_2 & 0 & -2\gamma & 0 & 0 & -G_1 & 0 \\ -G_2 & 0 & 0 & 0 & -2\gamma & G_1 & 0 & 0 \\ 0 & 0 & 0 & 0 & -G_1 & -\gamma/2 & -\Delta & 0 \\ 0 & 0 & G_2 & G_1 & 0 & \Delta & -\gamma/2 & -\sqrt{3}G_2 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{3}G_2 & 0 \end{pmatrix}, \tag{B.49}$$

B.4 Calculation of case 3

For the final case 3, the Lindblad jump operator is

$$C = |1\rangle\langle 2| + |3\rangle\langle 2| \tag{B.50}$$

Doing the same approach as previously done in this appendix and start identifying each term

$$L_4 = \gamma\left(C\rho C^\dagger - \frac{1}{2}\left(C^\dagger C\rho + \rho C^\dagger C\right)\right), \tag{B.51}$$

where

$$\begin{aligned} C\rho C^\dagger = \frac{1}{3}\Bigg[& |1\rangle\langle 1| + |1\rangle\langle 3| + |3\rangle\langle 1| + |3\rangle\langle 3| \\ & + \sqrt{3}\sum_{i=1}^{8} c_i(|1\rangle\langle 2|\lambda_i|2\rangle\langle 1| + |1\rangle\langle 2|\lambda_i|2\rangle\langle 3| + |3\rangle\langle 2|\lambda_i|2\rangle\langle 1| \\ & + |3\rangle\langle 2|\lambda_i|2\rangle\langle 3|)\Bigg]. \end{aligned} \tag{B.52}$$

Here the terms within the sum are easy to verify as they just pick the middle matrix element from the Gell-Mann matrices. Thus, Eq.(B.52) becomes

$$\begin{aligned} C\rho C^\dagger = \frac{1}{3}\Bigg[& |1\rangle\langle 1| + |1\rangle\langle 3| + |3\rangle\langle 1| + |3\rangle\langle 3| + \sqrt{3}(-c_3|1\rangle\langle 1| + \frac{c_8}{\sqrt{3}}|1\rangle\langle 1| \\ & - c_3|1\rangle\langle 3| + \frac{c_8}{\sqrt{3}}|1\rangle\langle 3| - c_3|3\rangle\langle 1| + \frac{c_8}{\sqrt{3}}|3\rangle\langle 1| - c_3|3\rangle\langle 3| \\ & + \frac{c_8}{\sqrt{3}}|3\rangle\langle 3|)\Bigg]. \end{aligned} \tag{B.53}$$

Doing the same with the rest of Eq.(B.51)

$$C^{\dagger}C\rho = \frac{2}{3}\left(|2\rangle\langle 2| + \sqrt{3}\sum_{i=1}^{8} c_i|2\rangle\langle 2|\lambda_i\right), \tag{B.54}$$

$$\rho C^{\dagger}C = \frac{2}{3}\left(|2\rangle\langle 2| + \sqrt{3}\sum_{i=1}^{8} c_i\lambda_i|2\rangle\langle 2|\right). \tag{B.55}$$

The sums in Eq.(B.54) and (B.55) are

$$\begin{aligned}\sqrt{3}\sum_{i}^{8} c_i|2\rangle\langle 2|\lambda_i = \sqrt{3}(c_1|2\rangle\langle 1| + c_2 i|2\rangle\langle 1| - c_3|2\rangle\langle 2| + c_6|2\rangle\langle 3| - \\ -c_7 i|2\rangle\langle 3| + \frac{c_8}{\sqrt{3}}|2\rangle\langle 2|),\end{aligned} \tag{B.56}$$

$$\begin{aligned}\sqrt{3}\sum_{i}^{8} c_i\lambda_i|2\rangle\langle 2| = \sqrt{3}(c_1|1\rangle\langle 2| - c_2 i|1\rangle\langle 2| - c_3|2\rangle\langle 2| + c_6|3\rangle\langle 2| + \\ +c_7 i|3\rangle\langle 2| + \frac{c_8}{\sqrt{3}}|2\rangle\langle 2|).\end{aligned} \tag{B.57}$$

Combining Eqs.(B.52), (B.53) and Eq.(B.55) and plugging them into Eq.(B.51), one will find

$$\begin{aligned}L_4 = \frac{\gamma}{3}\big[&\left(1 - \sqrt{3}c_3 + c_8\right)|1\rangle\langle 2| + \sqrt{3}(-c_1 + c_2 i)|1\rangle\langle 2| + \left(1 - \sqrt{3}c_3 + c_8\right)|1\rangle\langle 3| \\ &+ \sqrt{3}(-c_1 - c_2 i)|2\rangle\langle 1| + \left(-2 + 2\sqrt{3}c_3 - 2c_8\right)|2\rangle\langle 2| \\ &+ \sqrt{3}(-c_6 + c_7 i)|2\rangle\langle 3| + \left(1 - \sqrt{3}c_3 + c_8\right)|3\rangle\langle 1| \\ &+ \sqrt{3}(-c_6 - c_7 i)|3\rangle\langle 2| + \left(1 - \sqrt{3}c_3 + c_8\right)|3\rangle\langle 3|\big].\end{aligned} \tag{B.58}$$

Finally one can construct the following equation

$$\dot{\vec{R}} = M_3\vec{R} + \vec{b}_3, \tag{B.59}$$

where the matrix M_3 is

$$M_3 = \begin{pmatrix} -\gamma & \Delta & 0 & 0 & G_2 & 0 & 0 & 0 \\ -\Delta & -\gamma & -2G_1 & -G_2 & 0 & 0 & 0 & 0 \\ 0 & 2G_1 & -\frac{3}{2}\gamma & 0 & 0 & 0 & -G_2 & \frac{\sqrt{3}}{2}\gamma \\ 0 & G_2 & -\gamma & 0 & 0 & 0 & -G_1 & \frac{\gamma}{\sqrt{3}} \\ -G_2 & 0 & 0 & 0 & 0 & G_1 & 0 & 0 \\ 0 & 0 & 0 & 0 & -G_1 & -\gamma & -\Delta & 0 \\ 0 & 0 & G_2 & G_1 & 0 & \Delta & -\gamma & -\sqrt{3}G_2 \\ 0 & 0 & \frac{\sqrt{3}}{2}\gamma & 0 & 0 & 0 & \sqrt{3}G_2 & -\frac{\gamma}{2} \end{pmatrix}, \tag{B.60}$$

and the vector $\vec{b}_3$ is

$$\overrightarrow{b_3} = \begin{bmatrix} 0 \\ 0 \\ \frac{\sqrt{3}}{2}\gamma \\ \frac{\gamma}{\sqrt{3}} \\ 0 \\ 0 \\ 0 \\ -\frac{\gamma}{2} \end{bmatrix}. \tag{B.61}$$

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