



Theory of non-Markovian dynamics in Fluorescence spectra

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Theory of non-Markovian dynamics in Fluorescence spectra

by

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Abstract

Robust quantum coherence is an important prerequisite for any system that may be used to perform quantum information processing tasks. For systems that can be probed optically, the fluorescence spectrum may provide indirect evidence of coherence times when supplemented with an appropriate model of the decay process. A common approach is to assume a Markovian system, resulting in exponential decay of correlation functions and Lorentzian features in the associated spectrum.

For physical systems with strongly history-dependent (non-Markovian) dynamics, there is currently limited systematic theoretical approach to establish the associated spectrum.

In this thesis, we present a detailed theoretical method to obtain the fluorescence spectrum for a general system undergoing non-Markovian dynamics. This procedure can be used to systematically account for features in fluorescence spectra due to genuine non-Markovian dynamics. We apply this method to study the non-Markovian behaviour in the side peak of the Mollow-triplet and absorption spectrum of coherent population trapping systems.

Fræðileg athugun á ó-Markóvskum eiginleikum flúrljómunar

Abhishek Kumar

Desember 2014

Útdráttur

Til að nýta megi kerfi til skammtafræðilegra reikninga þarf það að viðhalda skammtafræðilegri samfösun nægjanlega lengi. Fyrir kerfi sem skoðuð eru með ljósi getur flúrljómun veitt upplýsingar um samfösunartíma ef tilheyrandi líkan fyrir dempun kerfisins er til. Hefðbundna aðferðin er að gera ráð fyrir Markóvskri hegðun kerfisins sem leiðir til veldisvísislækkunar fylgnifalla og Lorentz-lögunar tilheyrandi rófs.

Í kerfum þar sem eiginleikar kerfis við fyrri tíma skiptir máli (ó-Markóvsk) til að lýsa hreyfifræði eru til fáar kerfisbundar aðferðir til að lýsa tilheyrandi rófi. Í þessari ritgerð er kynnt kennileg aðferð til að lýsa kerfum sem sýna ó-Markóvska hegðun. Aðferðin nýtist til að finna á kerfibundinn hátt áhrif ó-Markóvskra eiginleika í flúrljómunarrófum. Við beitum þessari aðferð á hliðartoppa Mollow-þrístigsins og einnig á ísogsróf kerfa með samfasa fjöldafestingu.

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Contents

1	Introduction	1
1.1	Two-level system	1
1.1.1	Interaction of a two-level system with a classical monochromatic laser field	2
1.1.2	Two-level cavity-QED system	4
1.2	Coherent population trapping	6
1.3	Pure dephasing	8
1.4	Outline	8
2	Non-Markovian Fluorescence Spectrum	11
2.1	Introduction	11
2.2	Model Hamiltonian	11
2.3	Fluorescence spectra	12
2.4	Linear response theory	14
2.5	Fluorescence spectrum in the stationary regime	18
2.6	Nakajima-Zwanzig generalized master equation	20
2.7	Fluorescence spectrum of a driven two-level system	24
2.8	Fluorescence spectrum	24
2.8.1	Hamiltonian	25
2.8.2	Rotating frame	25
2.8.3	Fluorescence spectrum in the rotating frame	26
2.8.4	Dressed states	27
2.8.5	Fluorescence spectrum in the dressed-state basis	28
2.8.6	Markov approximation	32
2.8.7	Properties of the three-peak fluorescence spectrum	35

3	Non-Markovian Phonon-induced Dephasing in a Cavity-QED System	43
3.1	Introduction	43
3.2	Model Hamiltonian	44
3.2.1	Rotating frame	44
3.2.2	Dressed states	46
3.2.3	Polaron transformation	47
3.2.4	Fluorescence Spectrum in the polaron frame	48
3.3	Fluorescence Spectrum within the Secular Approximation	49
3.4	Simplified Fluorescence Spectrum	53
3.4.1	Side-peak spectrum for finite detuning: non-Markov versus Markov	57
3.5	Conclusion	60
4	Coherent population trapping and the influence of phonon-induced de- phasing on the absorption spectrum of a lambda system	61
4.1	Introduction	61
4.2	Theory	62
4.3	Generalized master equation	65
5	Summary	73
A	Self-energy superoperator and its matrix elements	75
A.1	Self-energy expansion	75
A.2	Matrix elements of superoperators	76
A.3	Matrix elements of reduced self-energy for photon coupling	80
A.3.1	Transition self-energy	83
A.3.2	Dephasing self-energy	83
A.4	Matrix elements of reduced self-energy for phonon coupling	84
A.4.1	Transition self-energy	84
A.4.2	Dephasing self-energy	84
A.5	Detector response function for two-level system in a cavity	86

Chapter 1

Introduction

In this thesis, we will discuss the non-Markovian dynamics of open quantum systems and the influence of the non-Markovian dephasing on the fluorescence associated with such a system. We will derive a general formula that can be used to study the fluorescence spectrum of a multi-level system interacting with an arbitrary quantum environment. To relate the fluorescence spectrum to system dynamics, we will be interested in evaluating the dynamics of a system observable $\mathcal{O}_S(t)$ in contact with an environment. The observable $\mathcal{O}_S$ could be, for example, the population or polarization of an atomic state, the longitudinal or transverse component of spin of an electron, or the electric and magnetic fields generated by the motion of spins or charges.

1.1 Two-level system

The laws of quantum mechanics allow for quantum computers, which are known to be significantly more powerful than classical computers. In a quantum computer, information is stored in quantum bits (qubits), rather than classical bits. Each bit represents either a zero or a one. Similarly a single qubit represents a zero or a one and is a two-level system with two energy levels which can be used to store and process the information. An example of a two-level system, frequently used in quantum optics, is composed of the ground and excited states of an atom. It is therefore important to understand the dynamics of two-level systems and their interaction with an environment. In chapters 2 and 3 we will consider a two-level system formed, e.g., by the

excited and ground states of a single atom to quantitatively study atomic dynamics under spontaneous emission and dephasing. In chapter 4, we will discuss the coherent population trapping absorption spectrum under the influence of dephasing.

1.1.1 Interaction of a two-level system with a classical monochromatic laser field

We consider a two-level system driven by a classical monochromatic laser field of frequency ω as shown in fig. (1.1). The two-level system is represented by an excited state $|a\rangle$ and a ground state $|b\rangle$ where $\omega_{ab} = \omega_a - \omega_b$ is the transition frequency. The Hamiltonian of the entire system can be written in terms of the free Hamiltonian H_0 and the interaction with the laser field $V_{\text{laser}}(t)$ as

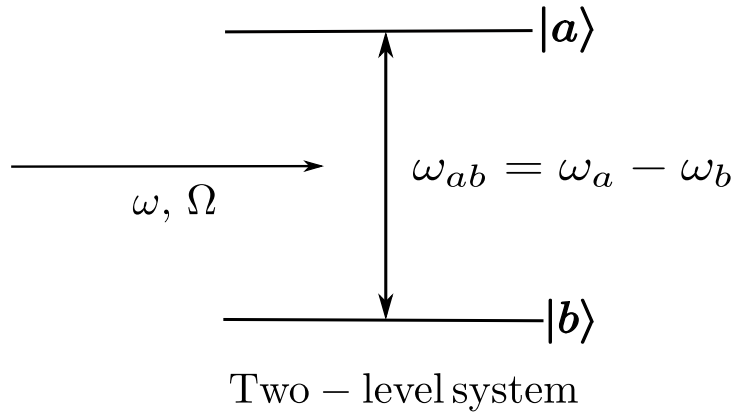


Figure 1.1: A two-level system, with a transition frequency ω_{ab} , driven by a classical monochromatic laser field of frequency ω with drive amplitude Ω .

$$H(t) = H_0 + V_{\text{laser}}(t)$$

$$H_0 = \omega_a |a\rangle\langle b| + \omega_b |b\rangle\langle a| \quad (1.1)$$

$$V_{\text{laser}}(t) = \frac{\Omega}{2} (|a\rangle\langle b| + |b\rangle\langle a|) (e^{-i\omega t} + e^{i\omega t}) \quad (1.2)$$

where Ω is the amplitude of the drive and we have chosen $\hbar = 1$ for the simplicity of units. The wave function of the two-level system at any time $t > 0$ is

$$|\psi(t)\rangle = c_a(t)|a\rangle e^{-i\omega_a t} + c_b(t)|b\rangle e^{-i\omega_b t} \quad (1.3)$$

and satisfies the time-depended Schrödinger equation

$$|\dot{\psi}(t)\rangle = -iH(t)|\psi(t)\rangle. \quad (1.4)$$

Substituting for $H(t)$ and $|\psi(t)\rangle$ in the above equation we obtain coupled equations for the probability amplitudes $c_a(t)$ and $c_b(t)$,

$$\dot{c}_a(t) = -i\frac{\Omega}{2}(e^{i(\omega_{ab}-\omega)t} + e^{i(\omega_{ab}+\omega)t})c_b(t) \quad (1.5)$$

$$\dot{c}_b(t) = -i\frac{\Omega}{2}(e^{-i(\omega_{ab}-\omega)t} + e^{-i(\omega_{ab}+\omega)t})c_a(t). \quad (1.6)$$

In the rotating wave approximation (RWA), we neglect the counter-rotating terms, $\sim e^{\pm i(\omega_{ab}+\omega)t}$ which are rapidly oscillating compared to the terms $\sim e^{\pm i(\omega_{ab}-\omega)t}$ when $\omega_{ab} + \omega \gg \omega_{ab} - \omega$. We write the above coupled equations in terms of the detuning $\Delta = \omega_{ab} - \omega$ as

$$\dot{c}_a(t) = -i\frac{\Omega}{2}e^{i\Delta t}c_b(t) \quad (1.7)$$

$$\dot{c}_b(t) = -i\frac{\Omega}{2}e^{-i\Delta t}c_a(t). \quad (1.8)$$

We assume that the atom is initially in the ground state $|b\rangle$, giving $c_a(0) = 0$ and $c_b(0) = 1$. Solving the above equation by Laplace transform, which is defined through $f(s) = \int_0^\infty dt e^{-st} f(t)$, we obtain

$$c_a(s) = -i\frac{\Omega}{2}c_b(s - i\Delta) \quad (1.9)$$

$$c_b(s) = \frac{1}{s} \left(1 - i\frac{\Omega}{2}c_a(s + i\Delta) \right). \quad (1.10)$$

Here, we have used the Laplace transform of the derivative $\dot{f}(t) \rightarrow sf(s) - f(0)$. Solving for $c_a(s)$ and $c_b(s)$ and going back to the time domain, we then have

$$c_a(t) = e^{-i\frac{\Delta}{2}t} \left[\frac{-i\Omega}{\Omega_R} \sin\left(\frac{\Omega_R}{2}t\right) \right] \quad (1.11)$$

$$c_b(t) = e^{i\frac{\Delta}{2}t} \left[\cos\left(\frac{\Omega_R}{2}t\right) - \frac{i\Delta}{\Omega_R} \sin\left(\frac{\Omega_R}{2}t\right) \right], \quad (1.12)$$

where Ω_R is the generalized Rabi frequency, given by

$$\Omega_R = \sqrt{\Omega^2 + \Delta^2}. \quad (1.13)$$

The populations are given by the expressions

$$|c_a(t)|^2 = \left(\frac{\Omega}{\Omega_R}\right)^2 \sin^2\left(\frac{\Omega_R}{2}t\right) \quad (1.14)$$

$$|c_b(t)|^2 = \cos^2\left(\frac{\Omega_R}{2}t\right) + \left(\frac{\Delta}{\Omega_R}\right)^2 \sin^2\left(\frac{\Omega_R}{2}t\right). \quad (1.15)$$

Defining the inversion $W(t) = |c_b(t)|^2 - |c_a(t)|^2$ and looking at the driving frequency exactly at resonance, i.e., when $\Delta = 0$, we obtain the expression for the inversion

$$W(t) = \cos(\Omega t) \quad (1.16)$$

which oscillates between -1 and 1 with frequency Ω , see fig. (1.2).

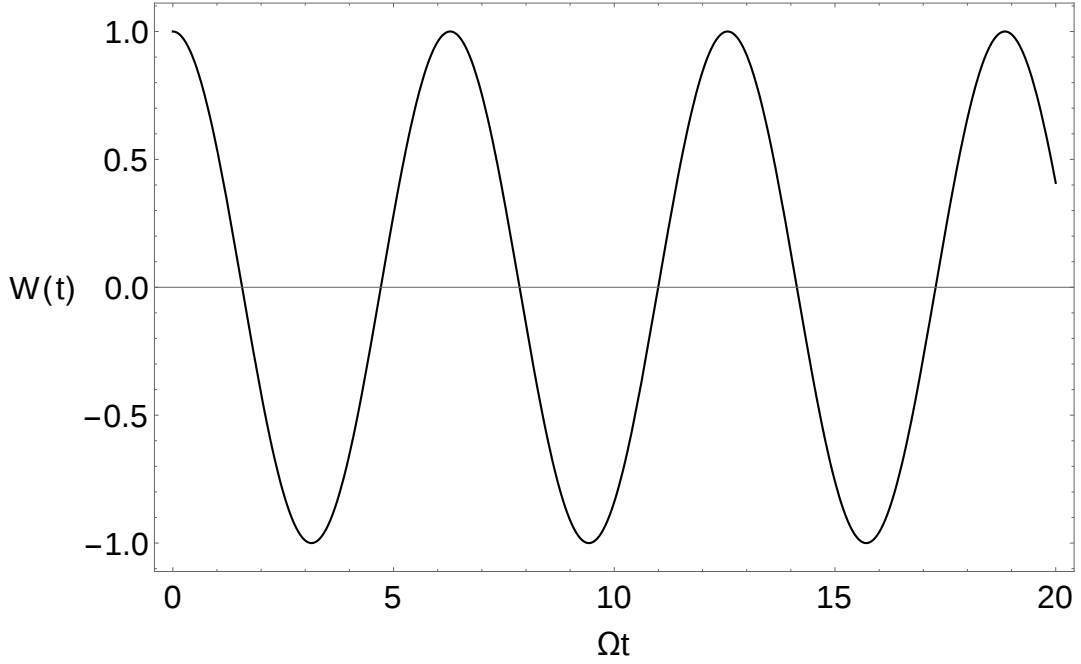


Figure 1.2: Oscillation of the inversion $W(t)$ at zero detuning given by Eq. (1.16) as a function of time .

The two-level system oscillates between the upper level $|a\rangle$ and lower level $|b\rangle$ with Rabi frequency Ω due to the interaction of a classical monochromatic laser field analogous to the Rabi spin- $\frac{1}{2}$ problem [1] under the influence of an oscillating transverse magnetic field.

1.1.2 Two-level cavity-QED system

Cavity quantum electrodynamics (cavity-QED) is the study of the emission properties of atoms, embedded inside a reflective cavity, due to interaction of the atoms with the electric-field modes of the cavity.

In the previous section, we discussed the interaction of a two-level system with a classical monochromatic laser field which gives rise to Rabi oscillations in the two-level system. In this section we will discuss spontaneous emission, giving rise to a transition from the atomic excited to ground state due to interaction of the two-level system with the radiation modes of free space. Moreover, it has been shown that the spontaneous emission rate can be enhanced [2] if the two-level atom is embedded inside a resonant

cavity, i.e., when $\omega_{ab} = \omega_c$, where ω_c is the center frequency of a cavity coupling to the atom.

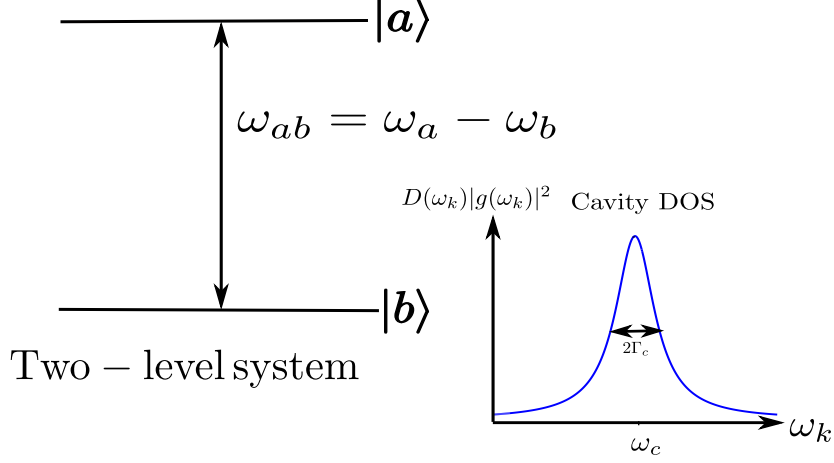


Figure 1.3: A cavity coupled to a two-level system, with a transition frequency ω_{ab} . The central frequency of the cavity modes ω_c is resonant with the transition frequency of the two-level atom. The cavity mode couples to a broad continuum, resulting in a Lorentzian spectrum of radiation modes, described by an effective density of states $D(\omega_k)$.

The Hamiltonian of the cavity-coupled two-level system can be written in the form of the standard Jaynes-Cummings Hamiltonian, within the RWA and after neglecting energy non-conserving terms, as

$$H = \frac{\omega_{ab}}{2}(|a\rangle\langle a| - |b\rangle\langle b|) + \sum_k \omega_k a_k^\dagger a_k + \sum_k g_k(|a\rangle\langle b|a_k + |b\rangle\langle a|a_k^\dagger). \quad (1.17)$$

Here, radiation modes of the cavity are represented by a bosonic field of frequency ω_k with annihilation and creation field operators a_k and $a_k^\dagger$, respectively. The coupling to the radiation modes is given by the coupling constant g_k to a mode of frequency ω_k and is related to the electric-dipole transition matrix element $|\rho_{ab}|$ through [3],

$$g_k = \frac{|\rho_{ab}|\varepsilon_k}{\hbar}. \quad (1.18)$$

The system operators are denoted by $\sigma_{ij} = |i\rangle\langle j|$ where $i, j \in \{a, b\}$ and $\sigma_z = \sigma_{aa} - \sigma_{bb}$ and the quantity $\varepsilon_k = \sqrt{\frac{\hbar\omega_k}{2\epsilon_0 V}}$ is the electric field per photon and V is the effective volume of a cubic cavity resonator.

We assume that at time $t = 0$ the atom is in the excited state $|a\rangle$ and the radiation modes are in the vacuum state $|0_k\rangle$. The state vector of the entire system at a later time $t > 0$ is then

$$|\psi(t)\rangle = c_a(t)|a, 0\rangle + \sum_k c_{b,k}(t)|b, 1_k\rangle e^{-i\omega_k t}, \quad (1.19)$$

where the initial conditions are given as $c_a(0) = 1$ and $c_{b,k}(0) = 0$. We solve for the probability amplitude $c_a(t)$ using the time-dependent Schrödinger equation

$$|\dot{\psi}(t)\rangle = -iH|\psi(t)\rangle. \quad (1.20)$$

Within the standard Wigner-Weiskoff theory and assuming a continuum of modes where $\sum_k \rightarrow \int d\omega D(\omega)$ and $g_k \rightarrow g(\omega_k)$, we obtain an expression for the excited-state population as

$$|c_a(t)|^2 = e^{-\Gamma t} \quad (1.21)$$

where the decay rate Γ is given by

$$\Gamma = 2\pi D(\omega_{ab})|g(\omega_{ab})|^2. \quad (1.22)$$

Here, $D(\omega_{ab})$ is the density of states at the atomic transition frequency ω_{ab} and is assumed to be given by the Lorentzian

$$D(\omega_k)|g(\omega_k)|^2 = \frac{1}{\pi} \frac{g^2 \Gamma_c}{(\omega_k - \omega_c)^2 + \Gamma_c^2} \quad (1.23)$$

where Γ_c is half of the cavity bandwidth, as shown in fig. (1.3). The density of states for the cavity is different from the mode density of free space. The spontaneous emission rate in the presence of the cavity is significantly modified and enhanced relative to the spontaneous emission rate in free space [2]. When a two-level system is embedded in a cavity, its energy levels are broadened when the cavity is resonant with the two-level transition frequency, hence there is an increase in the spontaneous emission rate.

1.2 Coherent population trapping

Coherent population trapping (CPT) [4] refers to the trapping of population in a coherent superposition of two ground states which are coupled by optical fields to an intermediate state in a three-level system. This is a so-called lambda configuration shown in fig. (1.4). We consider a lambda system driven by two classical laser fields of frequencies ω_1 and ω_2 . The Hamiltonian of the entire system within the usual rotating wave approximation (RWA), for $\hbar = 1$, is

$$H(t) = \omega_a |a\rangle\langle a| + \omega_b |b\rangle\langle b| + \omega_c |c\rangle\langle c| + \frac{\Omega_1}{2} (|a\rangle\langle b| e^{-i\omega_1 t} + |b\rangle\langle a| e^{i\omega_1 t}) + \frac{\Omega_2 e^{i\phi}}{2} (|a\rangle\langle c| e^{-i\omega_2 t} + |c\rangle\langle a| e^{i\omega_2 t}), \quad (1.24)$$

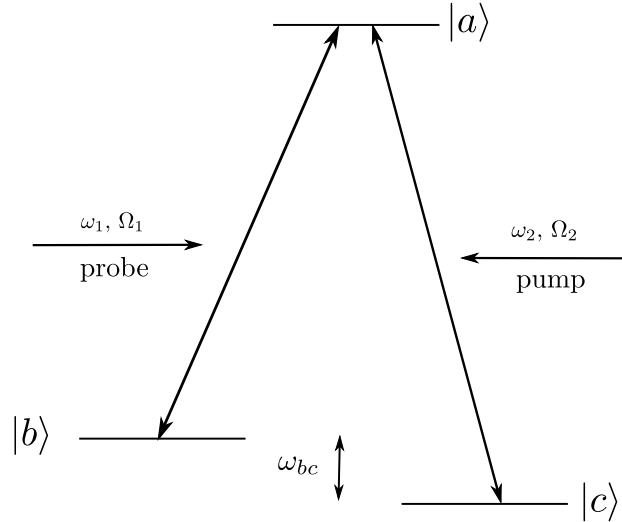


Figure 1.4: A driven lambda system where the two low energy states $|b\rangle$ and $|c\rangle$ are coupled to an intermediate state $|a\rangle$. The lambda system is driven by two classical monochromatic laser fields of frequencies ω_1 and ω_2 and with amplitudes Ω_1 and Ω_2 , respectively. The transitions frequencies of the levels $|a\rangle$ and $|b\rangle$ and between levels $|a\rangle$ and $|c\rangle$ are resonant with the laser probe and pump frequencies ω_1 and ω_2 , respectively, i.e. $\omega_{ab} = \omega_1$ and $\omega_{ac} = \omega_2$. We have assumed that the transition between $|b\rangle \rightarrow |c\rangle$ is not dipole allowed.

where Ω_1 and Ω_2 are assumed real. The wave function of the lambda system at any time $t > 0$ is given by

$$|\psi(t)\rangle = c_a(t)|a\rangle e^{-i\omega_a t} + c_b(t)|b\rangle e^{-i\omega_b t} + c_c(t)|c\rangle e^{-i\omega_c t}, \quad (1.25)$$

and satisfies the time-dependent Schrödinger equation

$$|\dot{\psi}(t)\rangle = -iH(t)|\psi(t)\rangle. \quad (1.26)$$

After substituting the expressions for $H(t)$ and $|\psi(t)\rangle$ in the above equation we obtain the coupled equations-of-motions for the probability amplitudes $c_a(t)$, $c_b(t)$ and $c_c(t)$, for the resonant conditions $\omega_1 = \omega_{ab}$ and $\omega_2 = \omega_{ac}$, given as

$$\dot{c}_a(t) = \frac{i}{2}(\Omega_1 c_b(t) + \Omega_2 e^{i\phi} c_c(t)) \quad (1.27)$$

$$\dot{c}_b(t) = i\frac{\Omega_1}{2}c_c(t) \quad (1.28)$$

$$\dot{c}_c(t) = i\frac{\Omega_2 e^{i\phi}}{2}c_a(t). \quad (1.29)$$

We solve for the probability amplitude $c_a(t)$ by going into Laplace space and using the initial condition $c_a(0) = 0$. This gives the expression for $c_a(s)$,

$$c_a(s) = -\frac{i}{2} \frac{\Omega_1 c_b(0) + \Omega_2 e^{i\phi} c_c(0)}{s^2 + \frac{1}{4}(\Omega_1^2 + e^{2i\phi}\Omega_2^2)}. \quad (1.30)$$

It can be seen from the above expression that we have population trapped in the lower states for the condition

$$\Omega_1 c_b(0) = -\Omega_2 e^{i\phi} c_c(0). \quad (1.31)$$

Under this condition the population is trapped in the lower states and the system is transparent to the incident fields and therefore there is no absorption. Following the notation given in Ref. [5], the dark state is given as a superposition of states $|b\rangle$ and $|c\rangle$

$$|D\rangle = \frac{\Omega_2 e^{i\phi}|b\rangle - \Omega_1|c\rangle}{\sqrt{\Omega_1^2 + \Omega_2^2}}, \quad (1.32)$$

and it implies that when the atom is in this dark state the atom will stay in this state indefinitely. This phenomenon is known as coherent population trapping (CPT).

1.3 Pure dephasing

Due to coupling of a quantum system to the environment degrees of freedom, the system and environment become entangled, giving rise to a rapid decay of the off-diagonal element of the reduced density matrix of the system, when the diagonal elements of the density matrix are preserved, this phenomenon is called pure dephasing. Pure dephasing is a process that destroys quantum coherence for any two-level system that may represent a qubit.

It is therefore important to understand the mechanism behind pure-dephasing due to the interaction of quantum system with its environment. Several excellent experimental [6, 7, 8] and theoretical studies [9, 10] have been performed to understand pure-dephasing mechanisms in cavity-QED systems. In this thesis, we will mainly focus on understanding pure dephasing in a two-level cavity-QED system and the influence of pure dephasing on an associated fluorescence spectrum. Furthermore, we will discuss the pure-dephasing mechanism in a CPT lambda-system and the effect of pure-dephasing on its absorption spectrum.

1.4 Outline

This thesis is structured as follows: Chapter 2 discusses a general formula for the fluorescence spectrum, accounting for non-Markovian effects, of a multi level system interacting with radiation modes of the electromagnetic field which gives rise to spontaneous emission. Chapter 3 presents a detailed analysis of phonon-induced non-Markovian dephasing in a driven two-level cavity-QED system. Chapter 4 describes coherent population trapping in a so-called lambda system and the effect of phonon-induced non-Markovian dephasing on its absorption spectrum.

Chapter 2

Non-Markovian Fluorescence Spectrum

2.1 Introduction

In this chapter we derive a general formula for the non-Markovian fluorescence spectrum of a multi-level system interacting with a bosonic environment. To this end, we apply linear-response theory [11] to describe the dynamics of a detector monitoring the emission spectrum following the method used in Ref. [3]. The associated emission spectrum can be directly related to a correlation function for system observables, which we evaluate beyond the Markov approximation using a Nakajima-Zwanzig Generalized Master Equation (GME).

2.2 Model Hamiltonian

We start with the Hamiltonian of a general multi-level system interacting with radiation modes of the electromagnetic field, which can be written as system (H_S), field (H_R), and interaction (H_{SR}) in terms of the standard Jaynes-Cummings model within a rotating-wave approximation (see fig. 2.1):

$$H = H_0 + H_{SR} \tag{2.1}$$

$$H_0 = H_S + H_R \quad (2.2)$$

$$H_R = \sum_k \hbar \omega_k a_k^\dagger a_k \quad (2.3)$$

$$H_{SR} = \sum_k \hbar g_k (\sigma_+ a_k + \sigma_- a_k^\dagger), \quad (2.4)$$

where $\sigma_+ = |a\rangle\langle b|$ and $\sigma_- = |b\rangle\langle a|$ are the raising and lowering operators between a selected excited state $|a\rangle$ and ground state $|b\rangle$ in the Hilbert space of the system and $a_k, a_k^\dagger$ are the annihilation and creation operators in the Hilbert space of a set of electromagnetic modes coupled to the system. The coupling to the radiation modes is given by the coupling constant g_k to a mode of frequency ω_k . The coupling constant g_k is related to the electric-dipole transition matrix element $|\rho_{ab}|$ through Eq. (1.18).

2.3 Fluorescence spectra

We theoretically describe a realistic detector with the well-known gedanken spectrum analyzer [3, 12] which measures the scattered fluorescence light from the emitting system (see fig. 2.1).

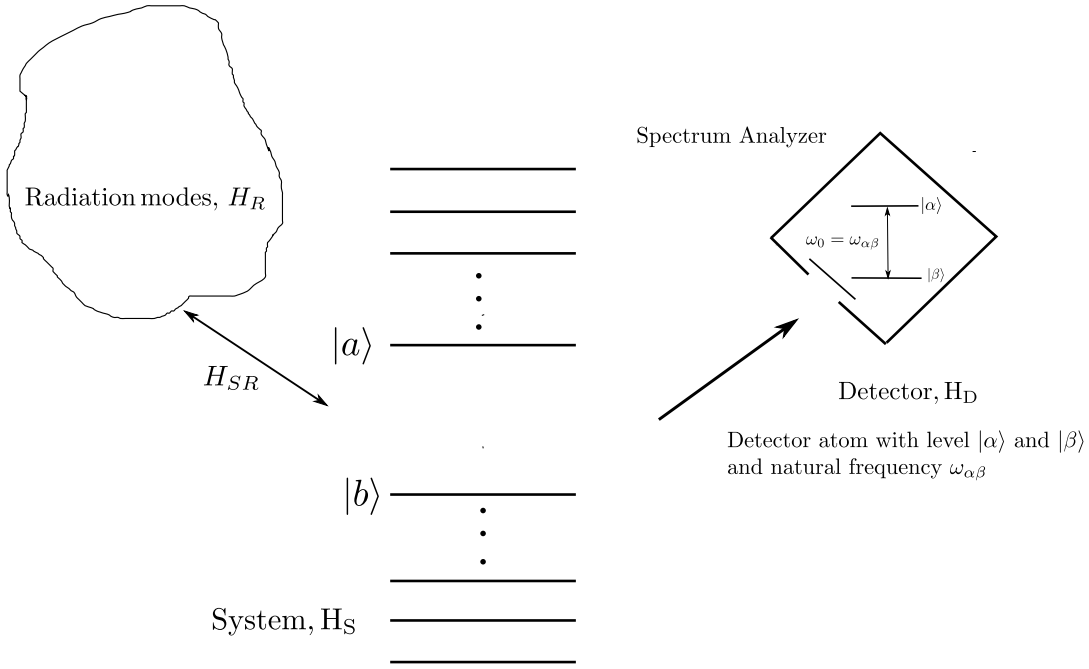


Figure 2.1: Schematic diagram of a multi-level system coupled to the radiation modes of the electromagnetic field. The coupling constant between the system and field mode k is given by g_k , where field modes are represented by a bosonic field of frequency ω_k . The selected two-level system is described by an excited state $|a\rangle$ and a ground state $|b\rangle$ with energies ω_a and ω_b , respectively. The emission spectrum of light scattered from this cavity-QED system is measured by a detector incorporated inside a spectrum analyzer.

We assume that the radiation field emitted by the system is detected by a two-level (detector) atom, with transition frequency $\omega_\alpha - \omega_\beta = \omega_0$.

The detector atom has sharp levels $|\alpha\rangle$ and $|\beta\rangle$ separated by energy ω_0 and is initially prepared in the ground state $|\beta\rangle$ and is placed inside a shutter, that only opens during a certain observation time T , during which it receives the emitted radiation and may be excited to the upper level $|\alpha\rangle$. The excitation rate of the detector sets the detector response profile, centered at frequency ω_0 with bandwidth $\Delta\omega_B$. The detector bandwidth is assumed to be small compared to the typical feature size in the fluorescence spectrum (see fig. (2.2)).

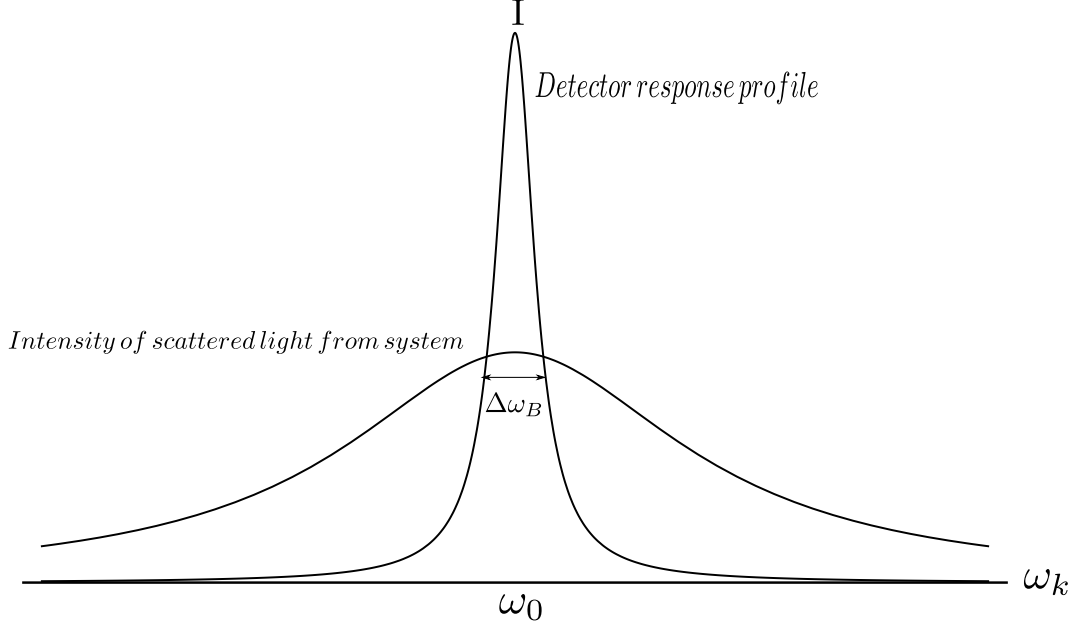


Figure 2.2: Incident light of frequency ω_k is scattered by the system. The detector has a response profile centered at frequency ω_0 with decay time $\tau_{\text{cd}} = \frac{1}{\Delta\omega_B}$.

The detector atom has a Hamiltonian of the form,

$$H_D = \frac{\hbar\omega_0}{2} (|\alpha\rangle\langle\alpha| - |\beta\rangle\langle\beta|). \quad (2.5)$$

The total Hamiltonian (for system, field and detector) has the form,

$$H_T = H + H_D + H_{DR}. \quad (2.6)$$

The coupling between detector atom H_D and the radiation field, included in the Hamiltonian H , is given by the Hamiltonian

$$H_{DR} = \sum_k \hbar g_k^D \left(|\alpha\rangle\langle\beta| a_k + |\beta\rangle\langle\alpha| a_k^\dagger + |\alpha\rangle\langle\beta| a_k^\dagger + |\beta\rangle\langle\alpha| a_k \right), \quad (2.7)$$

where g_k^D is the coupling of the detector to field mode k and the detector coupling Hamiltonian H_{DR} , in the interaction picture with respect to the Hamiltonian $H'_0 = H + H_D$, is given by

$$H_{DR}^I(t) = e^{iH'_0 t} H_{DR} e^{-iH'_0 t}, \quad (2.8)$$

we adopt the rotating wave approximation which is equivalent to dropping the energy non-conserving terms, to write

$$H_{DR}^I(t) = \sum_k \hbar g_k^D \left(\sigma_{\alpha\beta} a_k(t) e^{i\omega_0 t} + \sigma_{\beta\alpha} a_k^\dagger(t) e^{-i\omega_0 t} \right), \quad (2.9)$$

Here, $\sigma_{\alpha\beta} = |\alpha\rangle\langle\beta|$, $\sigma_{\beta\alpha} = |\beta\rangle\langle\alpha|$ and $a_k(t)$ is in the interaction picture with H'_0 , and we assume that the electric field is linearly polarized along the x -axis. The positive-frequency part of the electric field is defined by

$$E^+(t) = \sum_k \varepsilon_k a_k(t), \quad (2.10)$$

the negative-frequency part of the electric field is $E^-(t) = [E^+(t)]^\dagger$ and the detector coupling g_k^D is related to the electric-dipole transition matrix element $|\rho_{\alpha\beta}^k|$ through [3],

$$g_k^D = \frac{|\rho_{\alpha\beta}^k| \varepsilon_k}{\hbar}. \quad (2.11)$$

In the long-wavelength limit, we take the dipole element $\rho_{\alpha\beta} \approx \rho_{\alpha\beta}^k$ (independent of k). From Eqs. (2.9) and (2.11), we find the detector coupling Hamiltonian

$$H_{DR}^I(t) = \rho_{\alpha\beta} \sigma_{\alpha\beta} E^+(t) e^{i\omega_0 t} + \rho_{\alpha\beta} \sigma_{\beta\alpha} E^-(t) e^{-i\omega_0 t}, \quad (2.12)$$

where $\rho_{\alpha\beta} = e\langle\alpha|x|\beta\rangle = \rho_{\alpha\beta}^*$.

2.4 Linear response theory

Linear response theory [11] states that the response to a weak external perturbation is proportional to the perturbation, and therefore all we need to understand is the proportionality constant. We consider that the detector is weakly coupled to the scattered emitted field, which allows us to do the linear response on the detector coupling. With this setup, the idea is to calculate the spectrum of light emitted by the system in the presence of a detector coupled to the emitted field at some point in time $t > 0$. This spectrum is defined in terms of the probability to excite the detector atom, $P(\omega_0, t)$ at time t . The probability of exciting the detector atom to excited level $|\alpha\rangle$ is found

by calculating the expectation value of the projection operator $|\alpha\rangle\langle\alpha|$, which can be evaluated as

$$P(\omega_0, t) = \langle\psi_I(t)|\alpha\rangle\langle\alpha|\psi_I(t)\rangle, \quad (2.13)$$

where $|\psi(t)\rangle$ is the interaction-picture state, and is given by

$$|\psi_I(T)\rangle = U_I(T)|\psi_I(0)\rangle \quad (2.14)$$

$U_I(T)$ is the time-evolution operator given by

$$U_I(T) = \mathcal{T} \exp\left\{-i \int_0^T dt H_{DR}^I(t)\right\} \quad (2.15)$$

$$= 1 - i \int_0^T dt_1 H_{DR}^I(t_1) - \frac{1}{2} \mathcal{T} \int_0^T dt_1 \int_0^T dt_2 H_{DR}^I(t_1) H_{DR}^I(t_2) + \dots \quad (2.16)$$

We assume that the detector atom is initially in the ground state $|\beta\rangle$ and that the initial state of the system, radiation modes, and detector is given by the product state $|\psi_{SR}(0)\rangle \otimes |\beta\rangle$, where $|\psi_{SR}(0)\rangle$ is an eigenstate of the Hamiltonian H , including the coupling between system and the radiation modes. Suppose now that at some time $t=0$, an external perturbation is applied, driving the system out of equilibrium. The perturbation is described by the term $H_{DR}^I(t)$. Now we wish to find the expectation value of the operator $|\alpha\rangle\langle\alpha|$ at time $T > 0$. In order to do so, we must find the time evolution of the state $|\psi_I(T)\rangle$ in the interaction picture with the interaction picture Hamiltonian $H_{DR}^I(t)$. To linear order in H_{DR}^I , we obtain an expression for the state in the interaction picture up to first order in the perturbation

$$|\psi_I(T)\rangle \approx \left[1 - i \int_0^T dt_1 H_{DR}^I(t_1)\right] |\psi_{SR}(0)\rangle. \quad (2.17)$$

Going back to the Schrödinger picture, we have

$$|\psi(T)\rangle = e^{-iH_0 T} \left[1 - i \int_0^T dt_1 H_{DR}^I(t_1)\right] |\psi_{SR}(0)\rangle. \quad (2.18)$$

Therefore the excitation probability, in the Schrödinger picture, takes the form

$$\begin{aligned} P(\omega_0, t = T) &= \langle \psi(0) | U_I^\dagger(T) e^{iH'_0 T} | \alpha \rangle \langle \alpha | e^{-iH'_0 T} U_I(T) | \psi(0) \rangle \\ &= \langle \psi(0) | U_I^\dagger(T) | \alpha \rangle \langle \alpha | U_I(T) | \psi(0) \rangle, \end{aligned} \quad (2.19)$$

since $[H'_0, |\alpha\rangle\langle\alpha|] = 0$.

By substituting the values of $H_{DR}^I(t_1)$ and then $|\psi(T)\rangle$ in equations (2.17) and (2.13), respectively, the resulting expression for the excitation probability in the interaction picture is calculated as,

$$P(\omega_0, T) = |\rho_{\alpha\beta}|^2 \int_0^T dt_1 \int_0^T dt_2 \langle E^-(t_1) E^+(t_2) \rangle e^{-i\omega_0(t_1-t_2)}. \quad (2.20)$$

Here, we have generalized to a mixed-state initial condition and the average $\langle \dots \rangle = \text{Tr}\{\dots\rho(0)\}$, where $\rho(0)$ is the total density matrix of the scattering system plus detector and the radiation field at time $T = 0$ is not a product state.

From Eq. (2.20), it follows that the excitation probability, $P(\omega_0, T)$, of the detector atom is proportional to the two-time correlation function of the field operators, i.e., $\langle E^-(t_1) E^+(t_2) \rangle$.

We write the probability of excitation in terms of observables of the emitting system. We start with the Heisenberg equation of motion for $a_k(t)$ with the Hamiltonian H given by Eq. (2.1), i.e.,

$$\frac{da_k(t)}{dt} = -i[a_k(t), H], \quad (2.21)$$

we obtain an equation for $a_k(t)$

$$\dot{a}_k(t) = -i\left(\omega_k a_k(t) + g_k \sigma_-(t)\right). \quad (2.22)$$

After integrating the above equation starting from time $t = t_0 < 0$, we obtain the expression

$$a_k(t) = a_k(0) e^{-i\omega_k t} - i \int_0^t d\tau g_k \sigma_-(\tau) e^{-i\omega_k(t-\tau)}. \quad (2.23)$$

We insert the above expression for $a_k(t)$ into the positive-frequency part of the electric field operator, giving

$$E^+(t) = \sum_k \varepsilon_k a_k(0) e^{-i\omega_k t} - i \sum_k \varepsilon_k g_k \times \int_0^t d\tau \sigma_-(\tau) e^{-i\omega_k(t-\tau)}. \quad (2.24)$$

Similarly, the negative frequency part of the electric field operator is

$$E^-(t) = \sum_k \varepsilon_k a_k^\dagger(0) e^{i\omega_k t} + i \sum_k \varepsilon_k g_k \times \int_0^t d\tau \sigma_+(\tau) e^{i\omega_k(t-\tau)}. \quad (2.25)$$

Replacing equations (2.24) and (2.25) in eqn. (2.20), we get (neglecting terms $\sim \langle a_k a_k^\dagger \rangle$, $\langle a_k \sigma_+ \rangle$ and $\sigma_- a_k^\dagger$)

$$P(\omega_0, T) = \int_0^T dt_1 \int_0^T dt_2 e^{-i\omega_0(t_1-t_2)} \times \left\{ \int_0^{t_1} d\tau' \int_0^{t_2} d\tau S^*(t_1-\tau') S(t_2-\tau) \langle \sigma_+(\tau') \sigma_-(\tau) \rangle \right\}, \quad (2.26)$$

where $S(t-\tau)$ is the detector response function

$$S(t-\tau) = \sum_k g_k g_k^D e^{-i\omega_k(t-\tau)} \quad (2.27)$$

$$= \alpha \sum_k g_k^2 e^{-i\omega_k(t-\tau)} \quad (2.28)$$

here we have used $g_k^D = |\rho_{\alpha\beta}| \varepsilon_k$ and $\alpha = \frac{g_k^D}{g_k}$ is the ratio of the detector and system coupling strengths.

The detector response function $S(t)$ typically decays rapidly with a decay time τ_{cd} , which is given by the inverse of the detector bandwidth, $\Delta\omega_B$ see Eq. (2.2)

$$\tau_{\text{cd}} = \frac{1}{\Delta\omega_B} \quad (2.29)$$

whereas $\langle \sigma_+(\tau') \sigma_-(\tau) \rangle$ evolves on a typical time scale T_{sys} , where, by assumption, $T_{\text{sys}} \gg \tau_{\text{cd}}$. We apply change of variables in the Eq. (2.26) as $\tau' = t_1 - \tau'$ and

$\tau = t_2 - \tau$ to write

$$P(\omega_0, T) = \int_0^T dt_1 \int_0^T dt_2 e^{-i\omega_0(t_1-t_2)} \times \left\{ \int_0^{t_1} d\tau' \int_0^{t_2} d\tau S^*(\tau') S(\tau) \langle \sigma_+(t_1-\tau') \sigma_-(t_2-\tau) \rangle \right\}. \quad (2.30)$$

When $\langle \sigma_+(t_1 - \tau') \sigma_-(t_2 - \tau) \rangle$ is a slow varying function where $S(\tau)$ is finite, we approximate: $\langle \sigma_+(t_1 - \tau') \sigma_-(t_2 - \tau) \rangle \approx \langle \sigma_+(t_1) \sigma_-(t_2) \rangle$ and extend the upper limit of integrations to $t_{1,2} \rightarrow \infty$ for $t_{1,2} \gg \tau_{cd}$, we obtain

$$P(\omega_0, T) \approx |\bar{I}|^2 \int_0^T dt_1 \int_0^T dt_2 e^{-i\omega_0(t_1-t_2)} \times \left\{ \langle \sigma_+(t_1) \sigma_-(t_2) \rangle \right\}, \quad (2.31)$$

where

$$\bar{I} = \int_0^\infty d\tau S(\tau) \propto g g^D \tau_{cd}. \quad (2.32)$$

Rewriting Eq. (2.31)

$$P(\omega_0, T) \approx |\bar{I}|^2 \int_0^T dt_1 \int_0^T dt_2 e^{-i\omega_0(t_1-t_2)} \times \left\{ \text{Tr} \{ \sigma_-(t_2 - t_1) \rho(t_1) \sigma_+ \} \right\}, \quad (2.33)$$

here we have used the cyclic property of trace.

We apply change of variables in the above equation, $x = t_2 - t_1$, to write

$$P(\omega_0, T) \approx |\bar{I}|^2 \int_0^T dt_1 \int_{-t_1}^{T-t_1} dx e^{i\omega_0 x} \times \left\{ \text{Tr} \{ \sigma_-(x) \rho(t_1) \sigma_+ \} \right\}. \quad (2.34)$$

Here, we will work in the regime where the observation time T is much larger than the system evolution time T_{sys} . In this limit, we take $T - t_1 \rightarrow \infty$ and $-t_1 \rightarrow -\infty$, therefore

$$P(\omega_0, T) \approx |\bar{I}|^2 \int_0^T dt_1 \int_{-\infty}^\infty dx e^{i\omega_0 x} \text{Tr} \left\{ \sigma_-(x) \rho(t_1) \sigma_+ \right\}. \quad (2.35)$$

The contribution of the x-integration from the boundaries is small in the parameter $\frac{T_{\text{sys}}}{T} \rightarrow 0$

$$\begin{aligned} P(\omega_0, T) &\approx |\bar{I}|^2 \int_{-\infty}^\infty dx e^{i\omega_0 x} \text{Tr} \left\{ \sigma_-(x) \int_0^T dt_1 \rho(t_1) \sigma_+ \right\} \\ &= T |\bar{I}|^2 \int_{-\infty}^\infty dx e^{i\omega_0 x} \text{Tr} \left\{ \sigma_-(x) \rho_T \sigma_+ \right\}. \end{aligned} \quad (2.36)$$

where the time-averaged density matrix is

$$\rho_T = \frac{1}{T} \int_0^T dt_1 \rho(t_1). \quad (2.37)$$

2.5 Fluorescence spectrum in the stationary regime

The fluorescence spectrum $F(\omega_0)$ in terms of the two-time correlation function of system observables and in the stationary limit has the form

$$\begin{aligned} F(\omega_0) &= \lim_{T \rightarrow \infty} \frac{1}{T} P(\omega_0, T) \\ &= \lim_{T \rightarrow \infty} \frac{1}{T} |\bar{I}^*|^2 \int_{-\infty}^{\infty} dt e^{i\omega_0 t} \text{Tr} \left\{ \sigma_-(t) T \bar{\rho} \sigma_+ \right\} \\ &= |S(s=0)|^2 \int_{-\infty}^{\infty} dt e^{i\omega_0 t} \langle \sigma_-(t) \sigma_+ \rangle \end{aligned} \quad (2.38)$$

where $\bar{\rho} = \lim_{T \rightarrow \infty} \rho_T$. Taking the complex conjugate of the expression $F(\omega_0)$ to obtain

$$F^*(\omega_0) = |S(0)|^2 \int_{-\infty}^{\infty} dx e^{-i\omega_0 x} \langle (\sigma_-(x) \sigma_+)^{\dagger} \rangle, \quad (2.39)$$

where

$$(\sigma_-(x) \sigma_+)^{\dagger} = (\sigma_- \sigma_+(x)), \quad (2.40)$$

therefore,

$$F^*(\omega_0) = |S(s=0)|^2 \int_{-\infty}^{\infty} dx e^{-i\omega_0 x} \langle \sigma_- \sigma_+(x) \rangle. \quad (2.41)$$

Performing a change of variables $x = -t$ and for stationary conditions, we have

$$F^*(\omega_0) = |S(s=0)|^2 \int_{-\infty}^{\infty} dt e^{i\omega_0 t} \langle \sigma_-(t) \sigma_+ \rangle. \quad (2.42)$$

By comparing the equations (2.42) and (2.38) one can see that $F^*(\omega_0) = F(\omega_0)$, which means that $F(\omega_0)$ is a real function. The Fourier transform appearing in Eq. (2.38) can

then be written as a Laplace transform

$$\begin{aligned} F(\omega_0) &= |S^*(s=0)|^2 2\text{Re} \int_0^\infty dt e^{i\omega_0 t} \text{Tr}\{\sigma_-(t) \bar{\rho} \sigma_+\} \\ &= |S^*(s=0)|^2 2\text{Re} \int_0^\infty dt e^{i\omega_0 t} \text{Tr}\{\sigma_- \Omega(t)\}, \end{aligned} \quad (2.43)$$

where we used the cyclicity of trace in the last step, a factor of two comes from the change in the interval of integration and the operator $\Omega(t)$ is given by the expression¹

$$\Omega(t) = e^{-iHt} \bar{\rho} \sigma_+ e^{iHt}. \quad (2.44)$$

The above operator contains all the information needed for the fluorescence spectrum but it is defined in the Hilbert space of the entire world, i.e. the system and the reservoir. In a sense it contains too much information so we would like to find a new operator defined only in the system Hilbert space. For this purpose, we introduce a projection method which will be discussed in the next chapter.

2.6 Nakajima-Zwanzig generalized master equation

In order to compute the spectrum given by equation (2.43), we write the equation for the dynamics of the operator $\Omega(t)$ and find its Laplace transform. Equation (2.43) is analogous to the expression for the single time expectation value,

$$\langle \sigma_-(t) \rangle = \text{Tr}\{\sigma_- \rho(t)\}, \quad (2.45)$$

with $\rho(t)$ replaced by $\Omega(t)$ [13]. Thus, we first write an equation of motion for the dynamics of the reduced density matrix, then the equation for the dynamics of $\Omega(t)$ can be derived in the same way. The radiation field and system are decoupled for times $t < t_0$ and are prepared independently in the states described by density matrices $\rho_R(0)$ and $\rho_S(0)$, respectively. At time $t = t_0$, when the radiation field and system are brought into contact, the state of the entire system is described by the full density

¹ Since σ_+ and σ_- are operators in the system Hilbert space and $[H_D, H] = 0$, the evolution of $\Omega(t)$ is determined by the Hamiltonian of the emitting system and radiation field, H , in the absence of the detector.

matrix $\rho(t_0)$:

$$\rho(t_0) = \rho_S(t_0) \otimes \rho_R(t_0). \quad (2.46)$$

To evaluate the dynamics of the reduced density operator, we introduce a projection superoperator P , defined by its action on an arbitrary operator $\mathcal{O}$: $P\mathcal{O} = \rho_R(t_0)\text{Tr}_R\mathcal{O}$. P is chosen to preserve all system expectation values:

$$\begin{aligned} \langle \mathcal{O}_S \rangle(t) &= \text{Tr}\{\mathcal{O}_S \rho(t)\} \\ &= \text{Tr}\{\mathcal{O}_S P\rho(t)\} \end{aligned} \quad (2.47)$$

$$\begin{aligned} &= \text{Tr}_S \text{Tr}_R \{\mathcal{O}_S \otimes \mathbb{1}_R\} \rho_R(t_0) \text{Tr}_R \rho(t) \\ &= \text{Tr}_S \mathcal{O}_S \rho_S(t) \text{Tr}_R \mathbb{1}_R \otimes \rho_R(t_0) \\ &= \text{Tr}_S \{\mathcal{O}_S \rho_S(t)\} \end{aligned} \quad (2.48)$$

and satisfies $P^2 = P$. For factorized initial conditions [Eq. (2.46)], $P\rho(t_0) = \rho(t_0)$, which is a sufficient condition to rewrite the von-Neumann equation

$$\dot{\rho}(t) = -i[H, \rho(t)] = -iL\rho(t) \quad (2.49)$$

in the form of the exact Nakajima-Zwanzig generalized master equation (GME)[14], where L is the full Liouvillian, defined as $L_\alpha \mathcal{O} = [H_\alpha, \mathcal{O}]$ and $\alpha = S, R, 0, SR$. Multiplying equation (2.49) by P on both sides, we get

$$P\dot{\rho}(t) = -iPL\rho(t). \quad (2.50)$$

Introducing the complement of P : $Q = \mathbb{1} - P$ and using $P + Q = \mathbb{1}$ we obtain

$$P\dot{\rho}(t) = -iPLP\rho(t) - iPLQ\rho(t). \quad (2.51)$$

To write the above equation in terms of $P\rho(t)$ alone, we multiply equation (2.49) by Q , to get

$$Q\dot{\rho}(t) = -iQLP\rho(t) - iQLQ\rho(t). \quad (2.52)$$

We solve the above equation for $Q\rho(t)$ using the separation of variables method

$$Q\dot{\rho}(t) + iQLQ\rho(t) = -iQLP\rho(t). \quad (2.53)$$

Multiplying the above equation by e^{iQLt} on both sides gives

$$\begin{aligned} e^{iQLt} Q \dot{\rho}(t) + i e^{iQLt} Q L Q \rho(t) &= -i e^{iQLt} Q L P \rho(t) \\ \frac{d}{dt} e^{iQLt} Q \rho(t) &= -i e^{iQLt} Q L P \rho(t). \end{aligned} \quad (2.54)$$

After integrating the above equation and assuming $Q\rho(t_0) = 0$, i.e., assuming that the so-called irrelevant part of $\rho(t_0)$ is zero, we obtain

$$Q\rho(t) = -i \int_{t_0}^t dt' e^{-iQL(t-t')} Q L P \rho(t'). \quad (2.55)$$

Substituting equation (2.55) into equation (2.51), we obtain the standard form of the Nakajima-Zwanzig generalized master equation [14]

$$P \dot{\rho}(t) = -i P L P \rho(t) - i \int_{t_0}^t dt' \Sigma(t-t') P \rho(t'), \quad (2.56)$$

$$\Sigma(t) = -i P L Q e^{-iQLt} Q L P, \quad (2.57)$$

where $\Sigma(t)$ is the self-energy superoperator.

We can derive an equation of motion for $\Omega(t)$ analogous to the equation for $\rho(t)$ [Eq. (2.56)]. However, an additional term appears because $Q\Omega(0) = Q\bar{\rho}\sigma_+ \neq 0$, see Eq. (2.55). The resulting GME for $P\Omega(t)$ is then

$$P \dot{\Omega}(t) = -i P L P \Omega(t) - i \int_0^t dt' \Sigma(t-t') \Omega(t') - i P L Q e^{-iQLt} Q \Omega(0), \quad (2.58)$$

where $\Sigma(t)$ is defined in Eq. (2.57) and the last term in the above equation contains $Q\Omega(0)$, i.e. the irrelevant part of $\Omega(0)$ is non-zero. This last term accounts for conditions that accumulate between the system and radiation modes in the time interval $t \in [t_0, 0]$ for $t_0 < 0$. The long-time average value, is defined as

$$\bar{\rho} = \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T dt \rho(t) = \lim_{s \rightarrow 0} s \rho(s). \quad (2.59)$$

Here, the Laplace transform is defined as $F(s) = \int_{t_0}^{\infty} dt e^{-st} f(t)$. Inserting this definition into Eq. (2.55), we find (assuming that $\frac{1}{0^+ + iQL}$ exists),

$$Q\bar{\rho} = -i \frac{1}{0^+ + iQL} QLP\bar{\rho}. \quad (2.60)$$

Here, 0^+ is a positive infinitesimal. We want to write $Q\Omega(0) = Q\bar{\rho}\sigma_+$ in terms of $P\bar{\rho}\sigma_+$. To this end, substituting for $Q\bar{\rho}$ in equation (2.58), we have

$$P\dot{\Omega}(t) = -iPLP\Omega(t) - i \int_0^t dt' \Sigma(t-t')\Omega(t') - PLQe^{-iQLt} \left(\frac{1}{0^+ + iQL} \right) QLP\bar{\rho}\sigma_+. \quad (2.61)$$

The above equation is identical to equation (2.56) except for the last part, the so-called irrelevant part, which is expressed as

$$\Phi(t) = -PLQe^{-iQLt} \left(\frac{1}{0^+ + iQL} \right) QLP\bar{\rho}\sigma_+. \quad (2.62)$$

The expression for the fluorescence spectrum given in equation (2.43) is written in terms of the trace of the system operator σ_- as $\text{Tr}\{\sigma_- \Omega(t)\}$.

Using the properties of the projection superoperator, we have

$$\text{Tr}\{\sigma_- \Omega(t)\} = \text{Tr}\{\sigma_- P\Omega(t)\} = \text{Tr}_S\{\sigma_- \Omega_S(t)\}, \quad (2.63)$$

where in the last step we have defined

$$\Omega_S(t) = \text{Tr}_R\{\Omega(t)\}. \quad (2.64)$$

Therefore, the expression for the fluorescence spectrum given in Eq. (2.43) takes the form

$$F(\omega_0) = 2|S^*(0)|^2 \text{Re} \int_0^{\infty} dt e^{i\omega_0 t} \text{Tr}_S\{\sigma_- \Omega_S(t)\}. \quad (2.65)$$

For completeness, we substitute the expression for the Laplace transform $P\Omega(s)$ in the equation for the fluorescence spectrum to obtain an expression for the fluorescence spectrum

$$F(\omega_0) = 2|S^*(0)|^2 \text{Re} \left[\text{Tr} \left\{ \sigma_- \frac{1}{s + iPLP + i\Sigma(s)} (\mathbb{1} + \Phi(s)) P\bar{\rho}\sigma_+ \right\} \right]_{s=-i\omega_0} \quad (2.66)$$

where the Laplace transforms of the self-energy and the irrelevant part are given, respectively, below

$$\Sigma(s = -i\omega_0) = -iPLQ \frac{1}{-i\omega_0 + iQL} QLP \quad (2.67)$$

$$\Phi(s = -i\omega_0) = -PLQ \left(\frac{1}{-i\omega_0 + iQL} \right) \left(\frac{1}{0^+ + iQL} \right) QLP, \quad (2.68)$$

$$\bar{\rho} = \lim_{s \rightarrow 0} s \frac{1}{s + iPLP + i\Sigma(s)} P\rho(t_0). \quad (2.69)$$

2.7 Fluorescence spectrum of a driven two-level system

We have derived a formula which is valid for studying the fluorescence spectrum of a general system undergoing non-Markovian dynamics. In this section, as an example, we shall apply this theory to study the Markovian resonance fluorescence spectrum of a laser-driven two-level atom and recover the well-known Mollow triplet spectrum. Figure (2.3) describes a laser-driven two-level system which is kept inside a cavity and interacts with the radiation modes of the cavity.

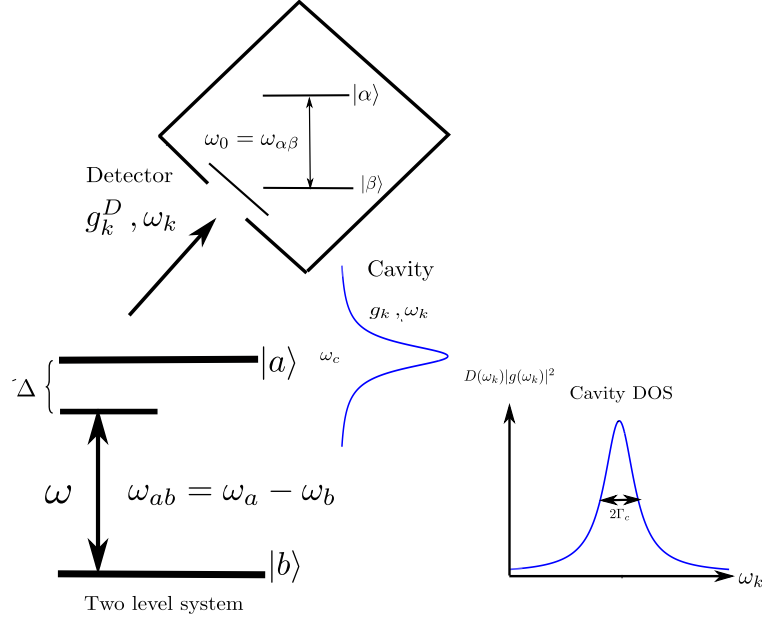


Figure 2.3: A two-level system inside a cavity with natural frequency ω_{ab} is driven by a laser pump frequency ω and the detuning $\Delta = \omega_{ab} - \omega$, is detuning of the transition frequency from the laser. The role of the spectrum analyser is to admit frequencies which are resonant with the transition frequency of the detector. The photonic density of states of the cavity is described by a Lorentzian density of states, where ω_c is the central frequency of the cavity mode and Γ_c is half of the cavity bandwidth.

2.8 Fluorescence spectrum

We consider a two-level system with ground state $|b\rangle$ and excited state $|a\rangle$, separated by an energy difference ω_{ab} . The two-level system is placed inside a cavity and driven by an external monochromatic classical laser field of frequency ω and the two-level system-laser coupling is given by the Rabi frequency Ω . The two-level system interacts with the radiation field of the cavity, which is represented by a collection of modes with frequencies ω_k . It is assumed here that the vacuum state, defined by $|0_k\rangle$, is the initial state of the radiation modes at a time $t_0 < 0$ in the distant past, see Fig. (2.3).

2.8.1 Hamiltonian

The Hamiltonian for the total system described above can be written as

$$H = \frac{\omega_{ab}}{2}\sigma_z + \frac{\Omega}{2}(\sigma_{ab} + \sigma_{ba})(e^{i\omega t} + e^{-i\omega t}) + \sum_k \omega_k a_k^\dagger a_k + \sum_k g_k(\sigma_{ab} + \sigma_{ba})(a_k + a_k^\dagger). \quad (2.70)$$

Here, $\sigma_{ij} = |i\rangle\langle j|$ are the system operators in the bare state basis $\{|a\rangle, |b\rangle\}$ and $\sigma_z = \sigma_{aa} - \sigma_{bb}$.

2.8.2 Rotating frame

In order to evaluate the dynamics of the operator $\Omega(t)$, we want to write an equation of motion for $\Omega(t)$ in the form of the Nakajima-Zwanzig GME, which is derived for a time-independent Hamiltonian. Hence, we would like to remove the explicit time dependence from the above Hamiltonian, Eq. (2.70). This is achieved by transforming the above Hamiltonian to a rotating frame

$$\begin{aligned} e^{i\xi t}(H - \xi)e^{-i\xi t} &= \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}(\sigma_{ab}e^{i\omega t} + \sigma_{ba}e^{-i\omega t})(e^{i\omega t} + e^{-i\omega t}) \\ &\quad + \sum_k \Delta_k a_k^\dagger a_k + \sum_k g_k(\sigma_{ab}e^{i\omega t} + \sigma_{ba}e^{-i\omega t})(a_k e^{-i\omega t} + a_k^\dagger e^{i\omega t}) \\ &= \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}(\sigma_{ab} + \sigma_{ba}) + \sum_k g_k(\sigma_{ab}a_k + \sigma_{ba}a_k^\dagger) + \sum_k \Delta_k a_k^\dagger a_k \\ &\quad + \left(\frac{\Omega}{2}\sigma_{ab}e^{2i\omega t} + \sum_k g_k\sigma_{ab}a_k^\dagger e^{2i\omega t} + \text{h.c.} \right) \end{aligned} \quad (2.71)$$

where $\xi = \frac{\omega}{2}\sigma_z + \sum_k \omega_k a_k^\dagger a_k$. In the rotating wave approximation (RWA) the energy non-conserving terms, i.e., the terms with explicit time dependence, are neglected [12], resulting in

$$\tilde{H} = \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}(\sigma_{ab} + \sigma_{ba}) + \sum_k \Delta_k a_k^\dagger a_k + \sum_k g_k(\sigma_{ab}a_k + \sigma_{ba}a_k^\dagger), \quad (2.72)$$

where, are detunings of the transition frequency ω_{ab} and the radiation mode frequency ω_k from the laser frequency ω ,

$$\Delta = \omega_{ab} - \omega \quad (2.73)$$

$$\Delta_k = \omega_k - \omega \quad (2.74)$$

Following the above discussion, we want to write the von-Neumann equation of motion for $\Omega(t)$ in the form of the Nakajima-Zwanzig GME with a time-independent Hamiltonian. To this end, we introduce $\tilde{\Omega}(t) = e^{i\xi t}\Omega(t)e^{-i\xi t}$ to obtain an equation of motion in the rotating frame,

$$\dot{\tilde{\Omega}}(t) = -i[\tilde{H}, \tilde{\Omega}(t)], \quad (2.75)$$

where $\tilde{H}$ is given by equation (2.72). After finding $\Omega(t)$ in the rotating frame the operator in the lab frame can be obtained using the relation

$$\Omega(t) = e^{-i\xi t}\tilde{\Omega}(t)e^{i\xi t}, \quad (2.76)$$

this relation also holds for any other operator that needs to be transformed back to the lab frame.

2.8.3 Fluorescence spectrum in the rotating frame

After transforming the operators in Eq. (2.43) according to the relation given by equation (2.76), the fluorescence spectrum becomes

$$F(\omega_0) = |S(0)|^2 2\text{Re} \int_0^\infty dt \text{Tr}\{\sigma_{ba}e^{-i\xi t}\tilde{\Omega}(t)e^{i\xi t}\}e^{i\omega_0 t}. \quad (2.77)$$

We use the cyclic property of the trace and the relation $e^{i\xi t}\sigma_{ba}e^{-i\xi t} = \sigma_{ba}e^{-i\omega t}$ to write the fluorescence spectrum in the rotated frame, in the Laplace domain, as

$$F(\Delta_0) = |S^*(0)|^2 2\text{Re} \left[\text{Tr}\{\sigma_{ba}\tilde{\Omega}(-i\Delta_0)\} \right], \quad (2.78)$$

where $\Delta_0 = \omega_0 - \omega$, which is simply a frequency shift resulting from the rotating-wave approximation.

2.8.4 Dressed states

The Hamiltonian given in equation (2.72) can be written in terms of unperturbed and perturbed parts as

$$\tilde{H} = \tilde{H}_0 + \tilde{H}_{SR} \quad (2.79)$$

$$\tilde{H}_0 = \tilde{H}_S + \tilde{H}_R \quad (2.80)$$

$$\tilde{H}_S = \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}(\sigma_{ab} + \sigma_{ba}) \quad (2.81)$$

$$\tilde{H}_R = \sum_k \Delta_k a_k^\dagger a_k \quad (2.82)$$

$$\tilde{H}_{SR} = \sum_k g_k (\sigma_{ab} a_k + \sigma_{ba} a_k^\dagger). \quad (2.83)$$

The above Hamiltonian is written in the bare-state basis $\{|a\rangle, |b\rangle\}$. Due to the strong laser-field interaction, the levels $|a\rangle$ and $|b\rangle$ are strongly coupled. The free Hamiltonian, $\tilde{H}_S$, is no longer diagonal in the bare basis. By introducing the dressed states $|+\rangle$ and $|-\rangle$, which are the eigenstates of the Hamiltonian $\tilde{H}_S$, the free Hamiltonian becomes diagonal. The dressed states are defined in terms of the bare states as

$$|+\rangle = \cos \theta |a\rangle + \sin \theta |b\rangle \quad (2.84)$$

$$|-\rangle = -\sin \theta |a\rangle + \cos \theta |b\rangle, \quad (2.85)$$

where the mixing angles are

$$\cos \theta = \sqrt{\frac{\Omega_R + \Delta}{2\Omega_R}} \quad (2.86)$$

$$\sin \theta = \sqrt{\frac{\Omega_R - \Delta}{2\Omega_R}}. \quad (2.87)$$

and the eigenvalue equation for dressed states is

$$\tilde{H}_S |\pm\rangle = \pm \frac{1}{2} \Omega_R |\pm\rangle, \quad (2.88)$$

where $\Omega_R = \sqrt{\Omega^2 + \Delta^2}$ is the dressed Rabi frequency.

The relation between the dressed atomic operators and the bare atomic operators is the following:

$$\sigma_{ab} = \cos^2 \theta \sigma_{+-} - \sin^2 \theta \sigma_{-+} + \cos \theta \sin \theta \sigma_3 \quad (2.89)$$

$$\sigma_{ba} = -\sin^2 \theta \sigma_{+-} + \cos^2 \theta \sigma_{-+} + \cos \theta \sin \theta \sigma_3 \quad (2.90)$$

$$\sigma_z = (\cos^2 \theta - \sin^2 \theta) \sigma_3 - 2 \cos \theta \sin \theta (\sigma_{+-} + \sigma_{-+}), \quad (2.91)$$

and $\sigma_{ij} = |i\rangle\langle j|$ are the system operators in the dressed state basis $\{|+\rangle, |-\rangle\}$ and $\sigma_3 = \sigma_{++} - \sigma_{--}$ as used in Ref. [15].

Therefore, the Hamiltonian in the dressed state basis can be written in terms of unperturbed and perturbed parts as

$$\tilde{H} = \tilde{H}_0 + \tilde{H}_V \quad (2.92)$$

$$\tilde{H}_0 = \tilde{H}_S + \tilde{H}_R + \tilde{H}_x \quad (2.93)$$

$$\tilde{H}_S = \frac{\Omega_R}{2} \sigma_3 \quad (2.94)$$

$$\tilde{H}_R = \sum_k \Delta_k a_k^\dagger a_k \quad (2.95)$$

$$\tilde{H}_x = \sin \theta \cos \theta \sigma_3 \sum_k g_k (a_k + a_k^\dagger) \quad (2.96)$$

$$\tilde{H}_V = \sum_k g_k \left[(\cos^2 \theta a_k - \sin^2 \theta a_k^\dagger) \sigma_{+-} + (\cos^2 \theta a_k^\dagger - \sin^2 \theta a_k) \sigma_{-+} \right], \quad (2.97)$$

The unperturbed part of the Hamiltonian, $\tilde{H}_0$, consists of three terms: the two-level Hamiltonian $\tilde{H}_S$ the free-field Hamiltonian $\tilde{H}_R$, and the pure-dephasing Hamiltonian $\tilde{H}_x$. In contrast, the perturbed part of the Hamiltonian, $\tilde{H}_V$, gives rise to relaxation in the system due to coupling to the cavity photons.

2.8.5 Fluorescence spectrum in the dressed-state basis

The expression for the fluorescence spectrum given previously given in equation (2.78) is

$$F(\Delta_0) = 2|S^*(0)|^2 \text{Re} \underbrace{\text{Tr}_S \{ \sigma_{ba} \tilde{\Omega}_S(-i\Delta_0) \}} \quad (2.98)$$

where the reduced operator $\tilde{\Omega}_S(-i\Delta_0) = \text{Tr}_R\{\tilde{\Omega}(-i\Delta_0)\}$. Using the completeness relation $|+\rangle\langle+| + |-\rangle\langle-| = 1$, the quantity under the brace can be written as

$$\begin{aligned} \text{Tr}_S\{\sigma_{ba}\tilde{\Omega}_S(-i\Delta_0)\} = & \langle+|\tilde{\Omega}_S(-i\Delta_0)|+\rangle\langle a|+\rangle\langle+|b\rangle + \langle-|\tilde{\Omega}_S(-i\Delta_0)|-\rangle\langle a|-\rangle\langle-|b\rangle \\ & + \langle-|\tilde{\Omega}_S(-i\Delta_0)|+\rangle\langle a|+\rangle\langle-|b\rangle + \langle+|\tilde{\Omega}_S(-i\Delta_0)|-\rangle\langle a|-\rangle\langle+|b\rangle \end{aligned} \quad (2.99)$$

We use the relations between dressed state operators and bare state operators and substitute the above expression in the expression for the fluorescence spectrum to write

$$\begin{aligned} F(\Delta_0) = & |S^*(0)|^2 2\text{Re} \left[\cos\theta \sin\theta \tilde{\Omega}_z(-i\Delta_0) \right. \\ & \left. + \cos^2\theta \tilde{\Omega}_{+-}(-i\Delta_0) - \sin^2\theta \tilde{\Omega}_{-+}(-i\Delta_0) \right], \end{aligned} \quad (2.100)$$

where

$$\tilde{\Omega}_z(-i\Delta_0) = \tilde{\Omega}_{++}(-i\Delta_0) - \tilde{\Omega}_{--}(-i\Delta_0) \quad (2.101)$$

and we have also used the identity $\tilde{\Omega}_{ij}(\Delta_0) = \text{Tr}_S\{|j\rangle\langle i|\tilde{\Omega}_S(t)\} = \langle i|\tilde{\Omega}_S(t)|j\rangle$. In addition, the steady-state density matrix is given by analogy to Eq. (2.59),

$$\bar{\rho}_S = \lim_{T \rightarrow \infty} \frac{1}{T} \int_{t_0}^{t_0+T} dt \tilde{\rho}_S(t), \quad (2.102)$$

$\bar{\rho}_S$ and σ_{ab} can be written in the dressed-state representation as

$$\bar{\rho}_S = \bar{\rho}_{++}|+\rangle\langle+| + \bar{\rho}_{--}|-\rangle\langle-| + \bar{\rho}_{+-}|+\rangle\langle-| + \bar{\rho}_{-+}|-\rangle\langle+| \quad (2.103)$$

$$\sigma_{ab} = \cos\theta \sin\theta |+\rangle\langle+| - \cos\theta \sin\theta |-\rangle\langle-| + \cos^2\theta |+\rangle\langle-| - \sin^2\theta |-\rangle\langle+|, \quad (2.104)$$

respectively. The initial value of the operator $\tilde{\Omega}_S(t)$ can be written in the dressed-state representation as

$$\begin{aligned}\tilde{\Omega}_S(0) = \tilde{\rho}_S \sigma_{ab} = & \left(\cos \theta \sin \theta \tilde{\rho}_{++} - \sin^2 \theta \tilde{\rho}_{+-} \right) |+\rangle\langle +| \\ & + \left(-\cos \theta \sin \theta \tilde{\rho}_{--} + \cos^2 \theta \tilde{\rho}_{-+} \right) |-\rangle\langle -| \\ & + \left(\cos^2 \theta \tilde{\rho}_{++} - \cos \theta \sin \theta \tilde{\rho}_{+-} \right) |+\rangle\langle -| \\ & + \left(-\sin^2 \theta \tilde{\rho}_{--} + \cos \theta \sin \theta \tilde{\rho}_{-+} \right) |-\rangle\langle +|. \quad (2.105)\end{aligned}$$

To evaluate the dynamics of the reduced operator $\tilde{\Omega}_S(t)$, we derive the Nakajima-Zwanzig generalized master equation, analogous to Eq. (2.61), for $P\tilde{\Omega}(t)$ using the dressed-state Hamiltonian $\tilde{H} = \tilde{H}_0 + \tilde{H}_V$ given by Eq. (2.92)

$$P\dot{\tilde{\Omega}}(t) = -iPLP\tilde{\Omega}(t) - i \int_0^t dt' \Sigma(t-t')\tilde{\Omega}(t') - iPLQe^{-iQLt}Q\tilde{\Omega}(0). \quad (2.106)$$

Here, the tilde denotes that the operators are in the rotating frame, the self-energy superoperator $\Sigma(-i\Delta_0)$ is expressed as

$$\Sigma(-i\Delta_0) = -iPLQ \frac{1}{-i\Delta_0 + iLQ} QLP, \quad (2.107)$$

and $L = L_0 + L_V$ is the full Liouvillian superoperator, where L_m ($m = 0, S, R, x, V$) is defined by its action on an arbitrary operator as $L_m \mathcal{O} = [\tilde{H}_m, \mathcal{O}]$. The irrelevant part $Q\tilde{\Omega}(t_0)$ can be written as

$$\begin{aligned}Q\tilde{\Omega}(t_0) &= Q\tilde{\rho}\sigma_{ab} = \tilde{\rho}\sigma_{ab} - P(\tilde{\rho}\sigma_{ab}) \\ &= (\tilde{\rho} - \rho_R(t_0)\tilde{\rho}_S)\sigma_{ab}. \quad (2.108)\end{aligned}$$

We have assumed as before, that the system and radiation modes are initially in a product state, $Q\tilde{\Omega}(t_0) = 0$. If we neglect the term $Q\tilde{\Omega}(t_0)$, then Eq. (2.106), in the Laplace domain, takes the form

$$P\tilde{\Omega}(-i\Delta_0) = \frac{1}{-i\Delta_0 + iPLP + i\Sigma(-i\Delta_0)} P\tilde{\Omega}(t_0). \quad (2.109)$$

The projection superoperator P obeys the following important identities:

$$PLP = L_S P, \quad (2.110)$$

$$PL_V P = 0. \quad (2.111)$$

We apply the identities in (2.110) and (2.111), and perform the trace in equation (2.109) over the radiation modes and use

$$PLQ = P(L_x + L_V), \quad (2.112)$$

$$QPL = (L_x + L_V)P. \quad (2.113)$$

$$(2.114)$$

We then obtain the expressions

$$\tilde{\Omega}_S(-i\Delta_0) = \frac{1}{-i\Delta_0 + L_S + i\Sigma_S(-i\Delta_0)} \tilde{\Omega}_S(t_0), \quad (2.115)$$

$$\Sigma_S(-i\Delta_0) = -i\text{Tr}_R(L_x + L_V) \frac{1}{-i\Delta_0 + L_Q} (L_x + L_V) \rho_R, \quad (2.116)$$

where $L_S O = [\tilde{H}_S, O]$ and $\Sigma_S(-i\Delta_0)$ is the reduced self-energy superoperator. $\tilde{\Omega}_S(-i\Delta_0) = \text{Tr}_R\{\tilde{\Omega}(-i\Delta_0)\}$ is the system reduced operator $\tilde{\Omega}_S(t_0) = \bar{\rho}_S \sigma_{ab}$, and $\bar{\rho}_S$ is the stationary density matrix. After using the identities (2.110) and (2.111) and performing the trace over equation (2.59) over the variables of radiation modes

$$\bar{\rho}_S = \lim_{s \rightarrow 0} s \frac{1}{s + iL_S + i\Sigma_S(s)} \tilde{\rho}_S(t_0). \quad (2.117)$$

Defining

$$[M(\Delta_0)] = -i\Delta_0 \otimes \mathbb{1} + i[L_S] + i[\Sigma_S(-i\Delta_0)], \quad (2.118)$$

here square brackets represents a matrix and the equation (2.115) can be written as

$$M(-i\Delta_0) \tilde{\Omega}_S(-i\Delta_0) = \tilde{\Omega}_S(t_0). \quad (2.119)$$

The self-energy superoperator can be expanded in terms of the perturbation Liouvillian $L_V = L_{SR} + L_{SP}$,

$$\Sigma_S(-i\Delta_0) = \Sigma_S^{(2)}(-i\Delta_0) + \Sigma_S^{(4)}(-i\Delta_0) + \dots \quad (2.120)$$

since only even powers of L_V contribute in the final trace over the radiation modes. In the Born approximation, the self-energy superoperator is replaced by its lowest order $\Sigma_S(-i\Delta_0) \rightarrow \Sigma_S^{(2)}(-i\Delta_0)$ (see Appendix A for detail). Similarly, the stationary density matrix and the propagator $M(\Delta_0)$ in Born approximation take the form

$$\bar{\rho}_S \approx \lim_{s \rightarrow 0} s \frac{1}{s + iL_S + i\Sigma_S^{(2)}(s)} \tilde{\rho}_S(t_0) \quad (2.121)$$

$$[M(-i\Delta_0)] \approx -i\Delta_0 \otimes \mathbb{1} + i[L_S] + i[\Sigma_S^{(2)}(-i\Delta_0)]. \quad (2.122)$$

The matrix for L_S (see Appendix A), in the $\{+, -\}$ basis, takes the form

$$[L_S] = \begin{matrix} & \begin{matrix} ++ & -- & +- & -+ \end{matrix} \\ \begin{matrix} ++ \\ -- \\ +- \\ -+ \end{matrix} & \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & \Omega_R & 0 \\ 0 & 0 & 0 & -\Omega_R \end{pmatrix} \end{matrix}.$$

Similarly, the Matrix for the reduced self-energy superoperator $\Sigma_S^{(2)}(-i\Delta_0)$, in the $\{+, -\}$ basis, takes the form

$$[\Sigma_S^{(2)}(-i\Delta_0)] = \begin{matrix} & \begin{matrix} ++ & -- & +- & -+ \end{matrix} \\ \begin{matrix} ++ \\ -- \\ +- \\ -+ \end{matrix} & \begin{pmatrix} \Sigma_{++,++}^{(2)}(-i\Delta_0) & \Sigma_{++,--}^{(2)}(-i\Delta_0) & \Sigma_{++,+-}^{(2)}(-i\Delta_0) & \Sigma_{++, -+}^{(2)}(-i\Delta_0) \\ \Sigma_{--,++}^{(2)}(-i\Delta_0) & \Sigma_{--,--}^{(2)}(-i\Delta_0) & \Sigma_{--,+-}^{(2)}(-i\Delta_0) & \Sigma_{--, -+}^{(2)}(-i\Delta_0) \\ \Sigma_{+-,++}^{(2)}(-i\Delta_0) & \Sigma_{+-,--}^{(2)}(-i\Delta_0) & \Sigma_{+-,+-}^{(2)}(-i\Delta_0) & \Sigma_{+-, -+}^{(2)}(-i\Delta_0) \\ \Sigma_{-+,++}^{(2)}(-i\Delta_0) & \Sigma_{-+,-}^{(2)}(-i\Delta_0) & \Sigma_{-+,+-}^{(2)}(-i\Delta_0) & \Sigma_{-+, -+}^{(2)}(-i\Delta_0) \end{pmatrix} \end{matrix}.$$

2.8.6 Markov approximation

In the secular approximation, [12] the equations for the populations $\dot{\tilde{\Omega}}_z(t)$ and coherences $\dot{\tilde{\Omega}}_{+-}(t)$ are decoupled and take the form

$$\dot{\tilde{\Omega}}_z(t) = N_z(t) - i \int_0^t dt' \Sigma_z^{(2)}(t-t') \tilde{\Omega}_z(t'), \quad (2.123)$$

where $N_z(s) = -\frac{i}{s}(\Sigma_{++,++}^{(2)}(s) + \Sigma_{++,--}^{(2)}(s))$ and

$$\dot{\tilde{\Omega}}_{+-}(t) = -i\Omega_R \tilde{\Omega}_{+-}(t) - i \int_0^t dt' \Sigma_{+-,+-}^{(2)}(t-t') \tilde{\Omega}_{+-}(t'), \quad (2.124)$$

$$\dot{\tilde{\Omega}}_{-+}(t) = i\Omega_R \tilde{\Omega}_{-+}(t) - i \int_0^t dt' \Sigma_{-+, -+}^{(2)}(t-t') \tilde{\Omega}_{-+}(t'). \quad (2.125)$$

The reduced self-energy superoperator matrix elements which couple populations $\tilde{\Omega}_z(t)$ and the coherence $\tilde{\Omega}_{+-}(t)$ are separated by an energy difference given by the Rabi frequency Ω_R . Since Ω_R is one of the largest frequencies in the problem, the terms which couple $\tilde{\Omega}_z(t)$ and $\tilde{\Omega}_{+-}(t)$ are weakly coupled or suppressed by Ω_R compared to the other terms in the equation-of-motion. Following the discussion given in Ref. [12], in the secular approximation we neglect the terms which couple populations and the coherences in the equation-of-motion.

We carry out the Markov approximation on equation (2.123). To do this, we change variables, replacing t' by $t-t'$ in the integrand, and changing the interval of integration accordingly, which results in

$$\dot{\tilde{\Omega}}_z(t) = N_z(t) - i \int_0^t dt' \Sigma_z^{(2)}(t') \tilde{\Omega}_z(t-t'), \quad (2.126)$$

Next, we approximate $\tilde{\Omega}_z(t-t')$ by $\tilde{\Omega}_z(t)$ and extend the upper limit of integration up to infinity, to obtain

$$\dot{\tilde{\Omega}}_z(t) \approx N_z(t) - i \int_0^\infty dt' \Sigma_z^{(2)}(t') \tilde{\Omega}_z(t). \quad (2.127)$$

This approximation (the Markov approximation) is justified when the typical time scale for evolution of $\tilde{\Omega}_z(t-t')$ is large compared to the decay time of the self-energy matrix element $\Sigma_z^{(2)}(t')$. In the Laplace domain

$$\tilde{\Omega}_z(s) = \frac{\tilde{\Omega}_z(0) + N_z(s)}{s + \kappa}, \quad (2.128)$$

where $\text{Re}[\kappa]$ is the spontaneous emission rate, given explicitly by the expression

$$\kappa = i \int_0^\infty dt \Sigma_z^{(2)}(t) = i \Sigma_z^{(2)}(s=0). \quad (2.129)$$

When $N_z(s)$ can be neglected, the condition for the validity of the Markov approximation carried out in order to write equation (2.128) is given by [14]

$$|\kappa| \tau_c \ll 1, \quad (2.130)$$

where

$$\tau_c = \frac{1}{|\kappa|} \int_0^\infty |\psi(t)| dt \quad (2.131)$$

$$\psi(t) = i \int_t^\infty \Sigma_z^{(2)}(t') dt'. \quad (2.132)$$

Using the above relations, the condition for the validity of the Markov approximation is

$$\int_0^\infty \left(\left| i \int_t^\infty \Sigma_z^{(2)}(t') dt' \right| \right) dt \ll 1, \quad (2.133)$$

which leads to the condition, in terms of the parameters involved in the problem

$$\frac{g}{\Gamma_c} \ll 1. \quad (2.134)$$

Here, we take $g_k \approx g$ to give the typical system coupling and Γ_c is the half of cavity bandwidth, (see Fig. 1.3). We now introduce

$$\Omega'_{+-}(t) = e^{i\Omega_R t} \tilde{\Omega}_{+-}(t), \quad (2.135)$$

or

$$\tilde{\Omega}_{+-}(t) = e^{-i\Omega_R t} \Omega'_{+-}(t). \quad (2.136)$$

We take the derivative of the above equation to write the equation of motion for $\Omega'_{+-}(t)$

$$\dot{\Omega}'_{+-}(t) = -i \int_0^t dt' e^{i\Omega_R(t-t')} \Sigma_{+-,+-}^{(2)}(t-t') \Omega'_{+-}(t'). \quad (2.137)$$

Following the same argument given above, we carry out the Markov approximation to write the equation for $\Omega'_{+-}(t)$

$$\dot{\Omega}'_{+-}(t) \approx -i \int_0^\infty dt' e^{i\Omega_R t'} \Sigma_{+-,+-}^{(2)}(t') \Omega'_{+-}(t). \quad (2.138)$$

The integral kernel $e^{i\Omega_R t'} \Sigma_{+-,+-}^{(2)}(t')$ in the above equation is complex valued, therefore the integral can be decomposed into real and imaginary parts as

$$\Delta\omega = -\text{Im} \left[i \int_0^\infty dt \Sigma_{+-,+-}^{(2)}(t) e^{i\Omega_R t} \right], \quad (2.139)$$

$$\Gamma = \text{Re} \left[i \int_0^\infty dt \Sigma_{+-,+-}^{(2)}(t) e^{i\Omega_R t} \right], \quad (2.140)$$

where $\Delta\omega$ is the Lamb shift and Γ is the decay rate. In the Laplace domain,

$$\tilde{\Omega}_{+-}(s) = \frac{1}{s + i\Omega_R - i\Delta\omega + \Gamma} \tilde{\Omega}_{+-}(0). \quad (2.141)$$

Similarly, the condition for the validity of the Markov approximation carried out to write equation (2.141) is [14]

$$|i \int_0^\infty \Sigma_{+-,+-}^{(2)}(t') e^{i\Omega_R t'} dt'| \tau_c \ll 1. \quad (2.142)$$

From the definition of τ_c , the above inequality can be written as

$$\frac{\int_0^\infty \left(|i \int_t^\infty \Sigma_{+-,+-}^{(2)}(t') e^{i\Omega_R t'} dt'| \right) dt}{|i \int_0^\infty \Sigma_{+-,+-}^{(2)}(t') e^{i\Omega_R t'} dt'|} \ll 1. \quad (2.143)$$

This leads to the same condition for the validity of the Markov approximation,

$$\frac{g}{\Gamma_c} \ll 1. \quad (2.144)$$

We substitute the expressions for $\tilde{\Omega}_z(s)$ and $\tilde{\Omega}_{+-}(s)$ from equation (2.128) and equation (2.141), respectively into equation (2.100), to obtain the fluorescence spectrum in the Markov limit

$$\begin{aligned} F_{\text{mar}}(\Delta_0) = & |S^*(s=0)|^2 2\text{Re} \left[\frac{\cos^2 \theta \sin^2 \theta (\bar{\rho}_{++} + \bar{\rho}_{--})}{-i\Delta_0 + \kappa} \right. \\ & - \frac{i \cos^2 \theta \sin^2 \theta (\bar{\rho}_{++} - \bar{\rho}_{--}) (\Sigma_{++}^{(2)}(-i\Delta_0) + \Sigma_{+-}^{(2)}(-i\Delta_0))}{-i\Delta_0 (-i\Delta_0 + \kappa)} \\ & \left. + \frac{\cos^4 \theta \bar{\rho}_{++}}{-i\Delta_0 + i\Omega_R - i\Delta\omega + \Gamma} + \frac{\sin^4 \theta \bar{\rho}_{--}}{-i\Delta_0 - i\Omega_R + i\Delta\omega + \Gamma} \right]. \quad (2.145) \end{aligned}$$

This spectrum is demonstrated by three peaks at $\Delta_0 = 0, \pm\Omega_R$ and describes the well-known Mollow-triplet.

2.8.7 Properties of the three-peak fluorescence spectrum

In this section we will discuss and determine the widths of the three lines [16] of the fluorescence spectrum, given by equation (2.145). The spontaneous emission rate is

given by

$$\kappa = i\Sigma_z(s=0), \quad (2.146)$$

where

$$\Sigma_z(s) = \Sigma_{++}^{(2)}(s) - \Sigma_{+-}^{(2)}(s). \quad (2.147)$$

The matrix elements of the reduced self-energy are expressed as (see Appendix A.3 for details),

$$\Sigma_{++}^{(2)}(s) = -i \cos^4 \theta g^2 \left(\frac{1}{s - i(\Delta_c - \Omega_R) + \Gamma_c} + \frac{1}{s + i(\Delta_c - \Omega_R) + \Gamma_c} \right), \quad (2.148)$$

$$\Sigma_{+-}^{(2)}(s) = i \sin^4 \theta g^2 \left(\frac{1}{s - i(\Delta_c + \Omega_R) + \Gamma_c} + \frac{1}{s + i(\Delta_c + \Omega_R) + \Gamma_c} \right). \quad (2.149)$$

here, $\Delta_c = \omega_c - \omega$ is the detuning of the central frequency of the cavity from the laser pump frequency. Using the above expression, the spontaneous emission rate, κ , is calculated as

$$\kappa = 2g^2\Gamma_c \left(\frac{\cos^4 \theta}{\Gamma_c^2 + (\Delta_c - \Omega_R)^2} + \frac{\sin^4 \theta}{\Gamma_c^2 + (\Delta_c + \Omega_R)^2} \right). \quad (2.150)$$

The frequency shift, $\Delta\omega$, is expressed as

$$\begin{aligned} \Delta\omega &= -\text{Im} \left[i\Sigma_{+-}^{(2)}(s) \right] \\ &= -\text{Im} \left[i\Sigma_{+-}^{SR}(s) + i\Sigma_{+-}^x(s) \right], \end{aligned} \quad (2.151)$$

where matrix elements of the reduced self-energy are given by

$$\Sigma_{+-}^{SR}(s) = -ig^2 \left(\frac{\cos^4 \theta}{s - i\Delta_c + \Gamma_c} + \frac{\sin^4 \theta}{s + i\Delta_c + \Gamma_c} \right) \quad (2.152)$$

$$\Sigma_{+-}^x(s) = -2i \sin^2 \theta \cos^2 \theta g^2 \left(\frac{1}{s - i\Delta_c + \Gamma_c} + \frac{1}{s + i\Delta_c + \Gamma_c} \right). \quad (2.153)$$

Using the above expressions, we have

$$\Delta\omega = -g^2 \left(\frac{\cos^4 \theta (\Delta_c + \Omega_R)}{\Gamma_c^2 + (\Delta_c + \Omega_R)^2} - \frac{\sin^4 \theta (\Delta_c - \Omega_R)}{\Gamma_c^2 + (\Delta_c - \Omega_R)^2} \right). \quad (2.154)$$

The decay rate, Γ , is

$$\begin{aligned}\Gamma &= \text{Re} \left[i \Sigma_{+-,+-}^{(2)}(-i\Omega_R) \right] \\ &= \text{Re} \left[i \Sigma_{+-,+-}^{SR}(-i\Omega_R) + i \Sigma_{+-,+-}^x(-i\Omega_R) \right]\end{aligned}\quad (2.155)$$

$$= g^2 \Gamma_c \left(\frac{\cos^4 \theta}{\Gamma_c^2 + (\Delta_c + \Omega_R)^2} + \frac{\sin^4 \theta}{\Gamma_c^2 + (\Delta_c - \Omega_R)^2} + \frac{4 \sin^2 \theta \cos^2 \theta}{\Gamma_c^2 + \Delta_c^2} \right). \quad (2.156)$$

In order to discuss the spectrum in the Markov approximation, we consider a simple case when the laser frequency is exactly on resonance with the system's natural frequency and central frequency, i.e.,

$$\Delta = \Delta_c = 0. \quad (2.157)$$

Therefore, the second term in equation (2.145) on resonance is

$$\begin{aligned}\Sigma_{++,++}^{(2)}(s) + \Sigma_{++,--}^{(2)}(s)|_{s=-i\Delta_0} &= -i \frac{g^2}{4} \left(\frac{1}{s + i\Omega + \Gamma_c} + \frac{1}{s - i\Omega + \Gamma_c} \right. \\ &\quad \left. - \frac{1}{s - i\Omega + \Gamma_c} - \frac{1}{s + i\Omega + \Gamma_c} \right)|_{s=-i\Delta_0} = 0.\end{aligned}\quad (2.158)$$

At resonance, the fluorescence spectrum given in equation (2.145) takes the form

$$F_{\text{mar}}(\Delta_0) = \frac{\alpha^2 g^4}{2\Gamma_c^2} \left(\frac{\kappa}{\Delta_0^2 + \kappa^2} + \frac{\Gamma \bar{\rho}_{++}}{(\Delta_0 - \Omega + \Delta\omega)^2 + \Gamma^2} + \frac{\Gamma \bar{\rho}_{--}}{(\Delta_0 + \Omega - \Delta\omega)^2 + \Gamma^2} \right). \quad (2.159)$$

We can rewrite the above equation as

$$F_{\text{mar}}(\Delta_0) = \frac{\alpha^2 g^4}{2\Omega \Gamma_c^2} \left(\frac{\left(\frac{\kappa}{\Omega}\right)}{\left(\frac{\Delta_0}{\Omega}\right)^2 + \left(\frac{\kappa}{\Omega}\right)^2} + \frac{\left(\frac{\Gamma}{\Omega}\right) \bar{\rho}_{++}}{\left(\frac{\Delta_0 - \Omega + \Delta\omega}{\Omega}\right)^2 + \left(\frac{\Gamma}{\Omega}\right)^2} + \frac{\left(\frac{\Gamma}{\Omega}\right) \bar{\rho}_{--}}{\left(\frac{\Delta_0 + \Omega - \Delta\omega}{\Omega}\right)^2 + \left(\frac{\Gamma}{\Omega}\right)^2} \right). \quad (2.160)$$

The spontaneous emission rate in the Markov approximation, when $\Gamma_c \gg \Omega$, and on resonance takes the form

$$\kappa = \frac{g^2 \Gamma_c}{\Gamma_c^2 + \Omega^2} \approx \frac{g^2}{\Gamma_c}. \quad (2.161)$$

Similarly, the frequency shift is given by

$$\Delta\omega = -\frac{g^2}{4} \left(\frac{\Omega}{\Gamma_c^2 + \Omega^2} + \frac{\Omega}{\Gamma_c^2 + \Omega^2} \right) \approx \frac{-g^2\Omega}{2\Gamma_c^2} \quad (2.162)$$

and the decay rate is

$$\Gamma = \frac{g^2\Gamma_c}{2(\Gamma_c^2 + \Omega^2)} + \frac{g^2\Gamma_c}{\Gamma_c^2} \approx \frac{3g^2}{2\Gamma_c}. \quad (2.163)$$

Therefore, the fluorescence spectrum on resonance and in the Markov approximation takes the form,

$$F_{\text{mar}}(\Delta_0) = \frac{\alpha^2 g^4}{2\Omega\Gamma_c^2} \left(\frac{\left(\frac{\kappa(\bar{\rho}_{++} + \bar{\rho}_{--})}{\Omega} \right)}{\left(\frac{\Delta_0}{\Omega} \right)^2 + \left(\frac{\kappa}{\Omega} \right)^2} + \frac{\left(\frac{3\kappa}{2\Omega} \right) \bar{\rho}_{++}}{\left(\frac{\Delta_0 - \Omega + \Delta\omega}{\Omega} \right)^2 + \left(\frac{3\kappa}{2\Omega} \right)^2} + \frac{\left(\frac{3\kappa}{2\Omega} \right) \bar{\rho}_{--}}{\left(\frac{\Delta_0 + \Omega - \Delta\omega}{\Omega} \right)^2 + \left(\frac{3\kappa}{2\Omega} \right)^2} \right) \quad (2.164)$$

with $\kappa = \frac{g^2}{\Gamma_c}$.

The stationary density matrix is expressed as

$$\bar{\rho}_{++} = \lim_{s \rightarrow 0} s \rho_{++}(s) \quad (2.165)$$

where the matrix element $\rho_{++}(s)$ of the density matrix can be found using equation-of-motion given by equation (2.56)

$$\rho_{++}(s) = \frac{1}{s + i\Sigma_z^{(2)}(s)} \left(\rho_{++}(t_0) - \frac{i\Sigma_{++,-}^{(2)}(s)}{s(s + i\Sigma_z^{(2)}(s))} \right). \quad (2.166)$$

Exactly on resonance, we find

$$\bar{\rho}_{++} = \frac{-i\Sigma_{++,-}^{(2)}(0)}{i\Sigma_z^{(2)}(0)} = \frac{1}{2}, \quad (2.167)$$

and

$$\bar{\rho}_{--} = 1 - \bar{\rho}_{++} = \frac{1}{2}. \quad (2.168)$$

Therefore, in the strong driving limit, Ω is much greater than $\Delta = \omega_{ab} - \omega$ and $\kappa = \frac{g^2}{\Gamma_c}$, the fluorescence spectrum reduces to the expression

$$F_{\text{mar}}(\Delta_0) = \frac{\alpha^2 g^4}{2\Omega \Gamma_c^2} \left(\frac{\left(\frac{\kappa}{\Omega}\right)}{\left(\frac{\Delta_0}{\Omega}\right)^2 + \left(\frac{\kappa}{\Omega}\right)^2} + \frac{\left(\frac{3\kappa}{4\Omega}\right)}{\left(\frac{\Delta_0 - \Omega + \Delta\omega}{\Omega}\right)^2 + \left(\frac{3\kappa}{2\Omega}\right)^2} + \frac{\left(\frac{3\kappa}{4\Omega}\right)}{\left(\frac{\Delta_0 + \Omega - \Delta\omega}{\Omega}\right)^2 + \left(\frac{3\kappa}{2\Omega}\right)^2} \right) \quad (2.169)$$

The physical interpretation of the above expression can be explained in the dressed-state representation, see Figure (2.4). The above expression corresponds to the sum of three Lorentzian lines centered at $\Delta_0 + \Omega - \Delta\omega$, Δ_0 and $\Delta_0 - \Omega + \Delta\omega$. These lines arise from the transition between upper $|-\rangle$ to the lower $|+\rangle$ state, from the upper $|+\rangle$ state to the lower $|+\rangle$ state and from the upper $|+\rangle$ state to the lower $|-\rangle$ state, respectively.

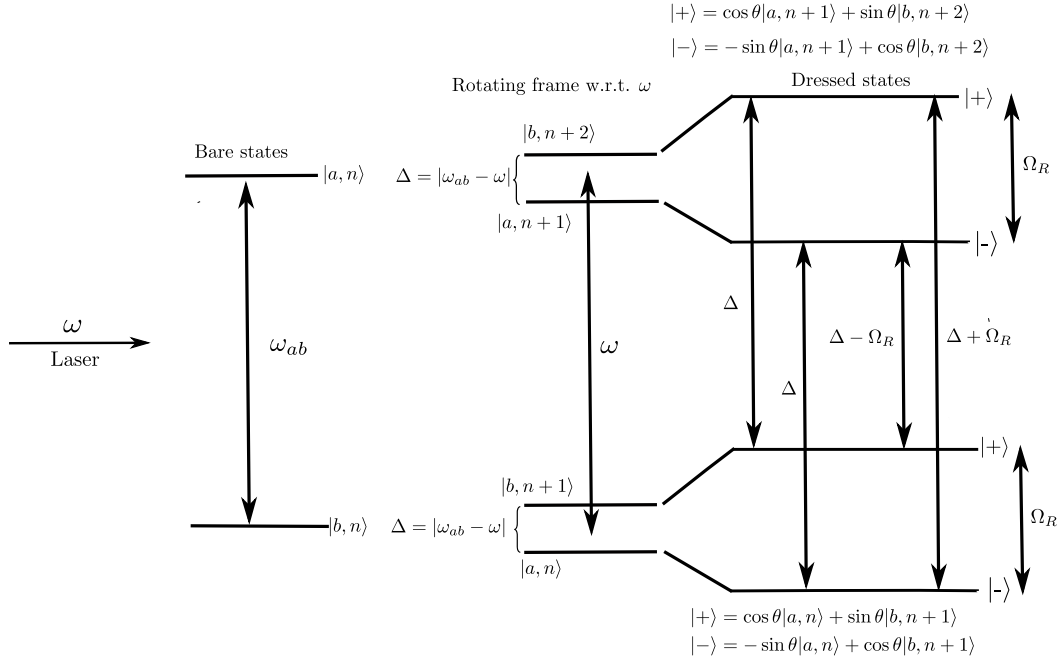


Figure 2.4: Allowed transitions between dressed states in the strong-coupling limit.

As an alternative explanation, we will explain the allowed transitions between the dressed-states fig. (2.4) using the Floquet quasi-energy levels, since the two-level problem is no longer the standard Jaynes-Cummings problem. The Hamiltonian for

the laser-driven two-level system,

$$H(t) = \frac{\omega_{ab}}{2}(|a\rangle\langle a| - |b\rangle\langle b|) + \frac{\Omega}{2}(e^{i\omega t} + e^{-i\omega t})(|a\rangle\langle b| + |b\rangle\langle a|) \quad (2.170)$$

where ω is the frequency of a drive laser with amplitude Ω and the wave function for the two-level system is at any time t :

$$|\psi(t)\rangle = c_a(t)e^{-i\omega_a t}|a\rangle + c_b(t)e^{-i\omega_b t}|b\rangle. \quad (2.171)$$

We have quasi-energy states which are an infinite number of resonant and off-resonant states coupled through the interaction part of the Hamiltonian.

Introducing

$$\psi_\alpha(t) = e^{-i\varepsilon_\alpha t}\Phi_\alpha(t), \quad (2.172)$$

where $\Phi_\alpha(t) = \Phi_\alpha(t + \frac{2\pi}{\omega})$ is periodic and determined by

$$\left(H(t) - i\frac{\partial}{\partial t}\right)\Phi_\alpha(t) = \varepsilon_\alpha\Phi_\alpha(t), \quad (2.173)$$

also

$$\Phi_\alpha(t) = \sum_{n=-\infty}^{\infty} \Phi_{\alpha,n} e^{i\omega_n t}. \quad (2.174)$$

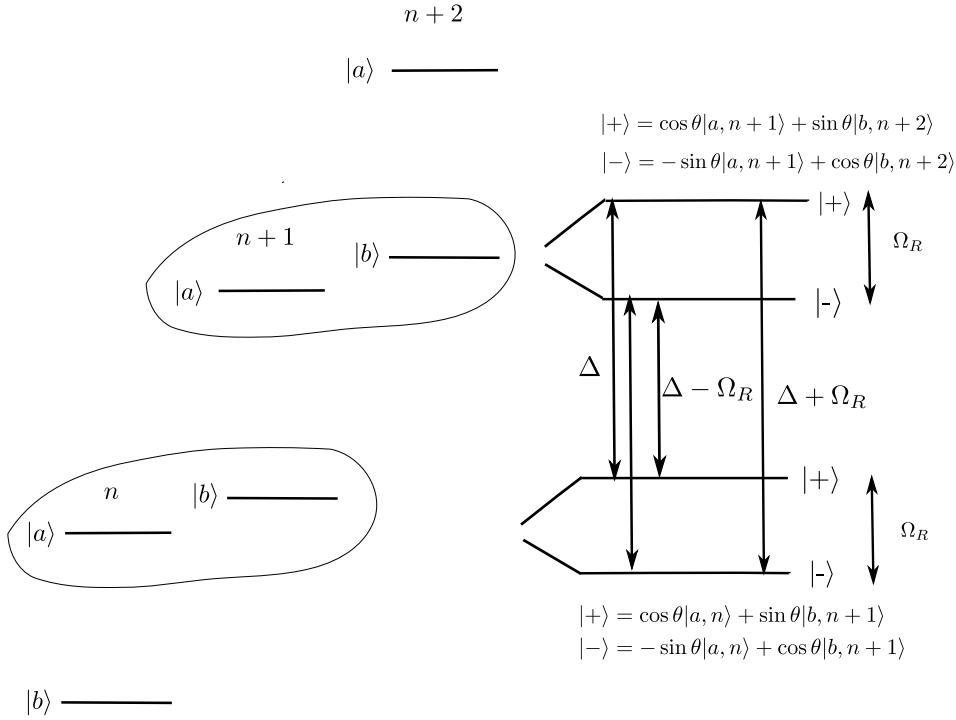


Figure 2.5: The allowed transitions in dressed states explained as floquet quasistates representation.

We will explain the allowed transitions in the dressed state Fig. (2.5) using the Floquet quasienergy levels. The laser interaction part of the Hamiltonian H couples pairs of quasi-energy states $\Phi_{\alpha,n}(t)$. In Fig. (2.5), the state $|a, n + 1\rangle$ is coupled to the state $|b, n + 2\rangle$ through the interaction part of the Hamiltonian. Similarly, the state $|a, n\rangle$ is coupled by the interaction Hamiltonian to the state $|b, n + 1\rangle$. The interaction Hamiltonian also introduces non-resonant couplings, which are separated by energy difference $\pm 2\omega$ and larger. These transitions are ignored in the rotating wave approximation.

The result obtained in equation (2.169) demonstrates the basic physical properties of Mollow-triplet spectrum, like heights, widths etc.

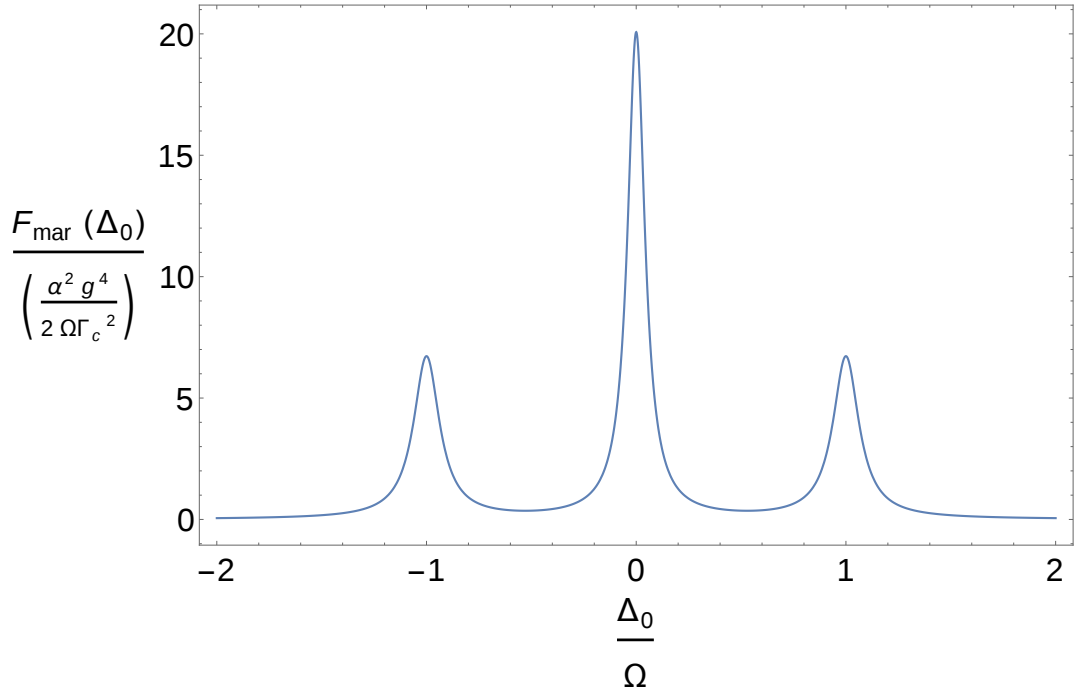


Figure 2.6: Fluorescence spectrum of a two-level atom inside a cavity, given by equation (2.169), in the units of $\frac{\alpha^2 g^4}{2\Omega\Gamma_c^2}$ for $\Omega = 20\kappa$.

The widths of the peaks in Fig. (2.6), centered at $\Delta_0 = -(\Omega - \Delta\omega)$, $\Delta_0 = 0$ and $\Delta_0 = (\Omega - \Delta\omega)$ are $\frac{3g^2}{\Gamma_c}$, $\frac{2g^2}{\Gamma_c}$ and $\frac{3g^2}{\Gamma_c}$, respectively. The relative heights of the three peaks in the limit, when $\Gamma_c \gg \Omega$, is in the ratio 1 : 3 : 1 and the integrated intensities in the peaks are in the ratio of 1 : 2 : 1.

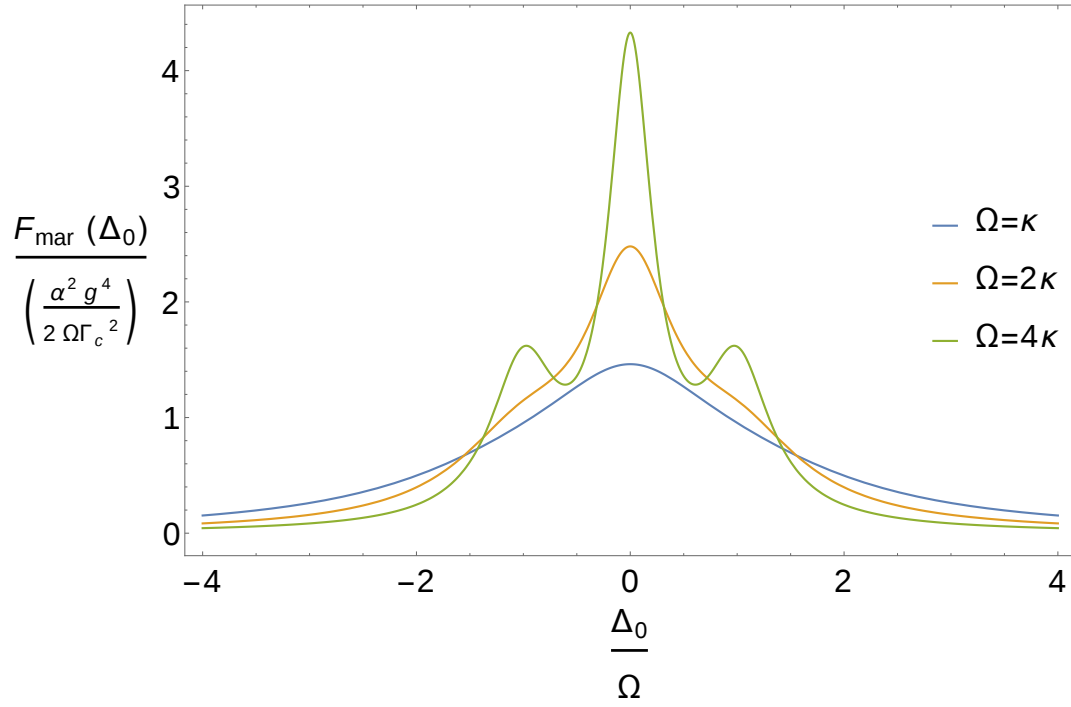


Figure 2.7: Fluorescence spectrum of a two-level atom inside a cavity $F(\Delta_0)$, given by equation (2.169), in the units of $\frac{g^4}{2\Omega\Gamma_c^2}$ for different values of Rabi frequency Ω .

Fig. (2.7) shows the fluorescence spectrum $F_{\text{mar}}(\Delta_0)$ in the units of $\frac{g^4}{2\Omega\Gamma_c^2}$, given by equation (2.169), for different values of the Rabi frequency $\Omega = \kappa$, $\Omega = 2\kappa$ and $\Omega = 4\kappa$.

Chapter 3

Non-Markovian Phonon-induced Dephasing in a Cavity-QED System

3.1 Introduction

In this chapter, we present a detailed theoretical analysis of a non-Markovian pure-dephasing process in a two-level cavity-QED system and its effect on the corresponding fluorescence spectrum. In particular, we will discuss the properties of one of the side bands of the Mollow-triplet spectrum. A number of experimental [6, 7, 8] and theoretical studies [9, 10] have been performed to understand pure-dephasing mechanisms in cavity-QED systems. In recent experiments [6] a distinct narrowing of one of the triplet sidebands has been observed with increasing detuning of a drive laser from an atomic transition frequency. This behaviour appears unexpected experimentally and does not agree with the recent theoretical model presented in [10] where broadening of the sideband is predicted as a result of excitation induced dephasing (EID) when either the drive power or detuning are increased.

In chapter 2, we have derived a formula to study the fluorescence spectrum of a general system undergoing non-Markovian dynamics. We shall apply this theory to the fluorescence spectrum of a two-level system interacting with radiation modes of a cavity, where the two-level system is driven by a classical monochromatic laser field. We consider a two-level system with an excited state $|a\rangle$, representing, e.g., a single exciton, and ground state $|b\rangle$, which represents, e.g., the absence of an exciton. The

excited state $|a\rangle$ is coupled to a phonon reservoir and can be modelled with the usual independent boson model (IBM) [17].

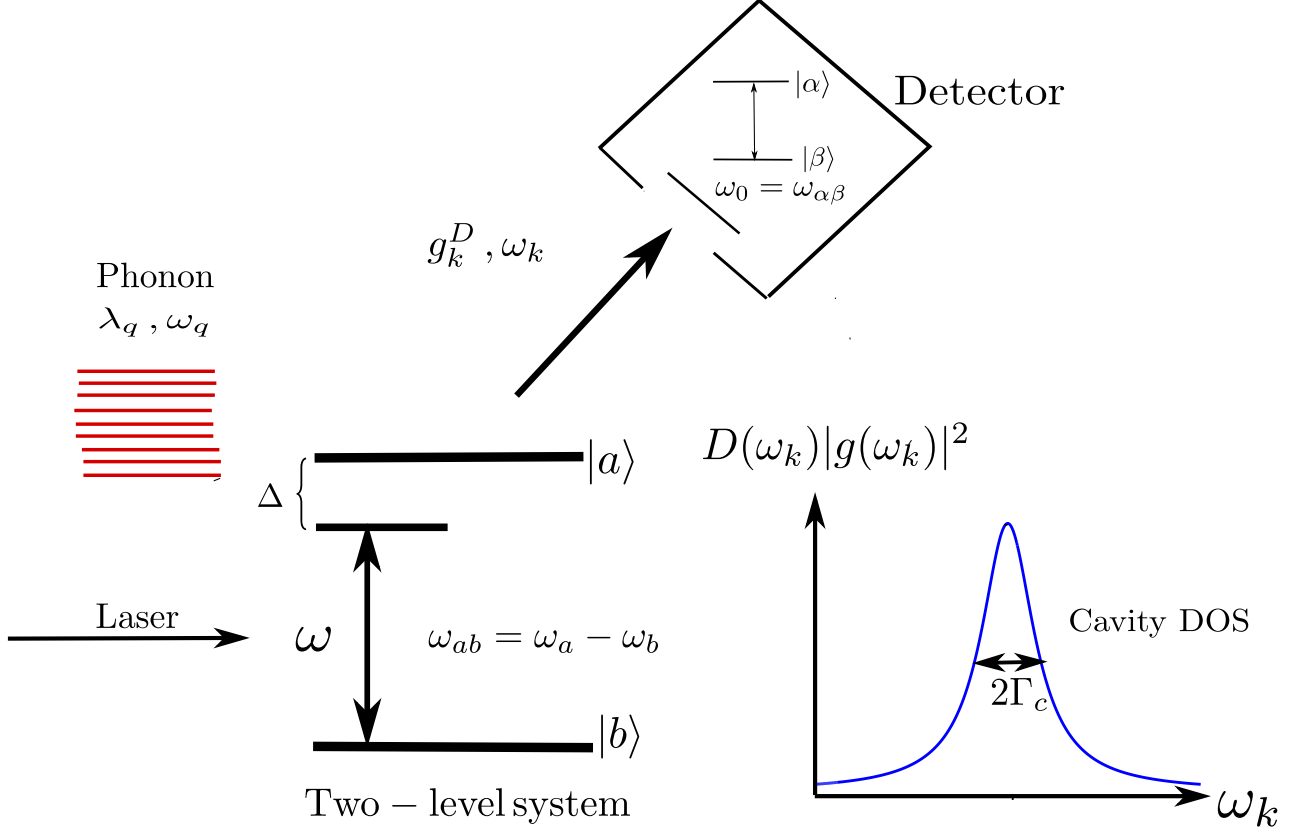


Figure 3.1: Schematic diagram of a cavity coupled to a two-level system driven by a classical monochromatic laser field of frequency ω and amplitude Ω . The two-level system couples to radiation mode k with strength g_k . The two-level system is described by an excited state $|a\rangle$ and a ground state $|b\rangle$ with energies ω_a and ω_b , respectively. A phonon reservoir is coupled to the excited state of the two level system with the coupling constant λ_q to the phonon mode with wavevector q and is represented by a bosonic field of frequency ω_q . The emission spectrum of light scattered from this cavity-QED system is measured by a detector placed behind a spectrum analyzer.

3.2 Model Hamiltonian

We consider a driven two-level cavity-QED system interacting with cavity photons and a phonon reservoir which is coupled to the excited state of the two-level system. The

Hamiltonian for the total system reads,

$$\begin{aligned}
 H(t) = & \frac{\omega_{ab}}{2}\sigma_z + \frac{\Omega}{2}(\sigma_{ab} + \sigma_{ba})(e^{i\omega t} + e^{-i\omega t}) + \sum_k \omega_k a_k^\dagger a_k + \sum_k g_k(\sigma_{ab} + \sigma_{ba})(a_k + a_k^\dagger) \\
 & + \sum_k \omega_q b_q^\dagger b_q + \sum_q \lambda_q \sigma_{aa}(b_q + b_q^\dagger)
 \end{aligned} \tag{3.1}$$

where ω_{ab} is the transition frequency of the two-level system and ω is the frequency of the laser field. The photon (phonon) modes are represented by bosonic fields with frequencies ω_k (ω_q) with creation and annihilation operators $a_k^\dagger$ ($b_q^\dagger$) and a_k (b_q), respectively. The system-photon (system-phonon) coupling strength is given by g_k (λ_q) and Ω is the coupling between the two-level system and laser field. The system operators are denoted by $\sigma_{ij} = |i\rangle\langle j|$ where $i, j \in \{a, b\}$ and $\sigma_z = \sigma_{aa} - \sigma_{bb}$. This physical model is illustrated schematically in Fig. (3.1).

3.2.1 Rotating frame

The explicit time dependence in H can be removed by going to a rotating frame and applying a rotating-wave approximation as discussed in Section 2.8.2. The Hamiltonian in the rotating frame is then

$$\begin{aligned}
 e^{i\xi t}(H - \xi)e^{-i\xi t} = & \frac{\omega_{ab}}{2}\sigma_z - \frac{\omega}{2}\sigma_z + \sum_k \omega_k a_k^\dagger a_k - \omega \sum_k a_k^\dagger a_k \\
 & + \sum_q \omega_q b_q^\dagger b_q + \sum_q \lambda_q \sigma_{aa}(b_q + b_q^\dagger) + \frac{\Omega}{2}(\sigma_{ab}e^{i\omega t} + \sigma_{ba}e^{-i\omega t})(e^{i\omega t} + e^{-i\omega t}) \\
 & + \sum_k g_k(\sigma_{ab}e^{i\omega t} + \sigma_{ba}e^{-i\omega t})(a_k e^{-i\omega t} + a_k^\dagger e^{i\omega t})
 \end{aligned} \tag{3.2}$$

where $\xi = \frac{\omega}{2}\sigma_z + \omega \sum_k a_k^\dagger a_k$.

We now perform the rotating-wave approximation (RWA) on both the driving term and the system-photon coupling term, in which we neglect the rapidly-oscillating terms, keeping only the time-independent part of Eq. (3.2). The Hamiltonian, within the

range of validity of the RWA, is then

$$\begin{aligned}\tilde{H} \approx & \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}(\sigma_{ab} + \sigma_{ba}) + \sum_k \Delta_k a_k^\dagger a_k \\ & + \sum_k g_k(\sigma_{ab}a_k + \sigma_{ba}a_k^\dagger) \\ & + \sum_k \omega_q b_q^\dagger b_q + \sum_k \lambda_q \sigma_{aa}(b_q + b_q^\dagger)\end{aligned}\quad (3.3)$$

where $\Delta = \omega_{ab} - \omega$ and $\Delta_k = \omega_k - \omega$ are the detunings of the atomic and cavity frequencies from the laser pump frequency ω . The RWA adopted here is justified when the detuning Δ and Rabi frequency Ω are small compared to the laser frequency ω , i.e.

$$|\omega| > |\Delta|, |\Omega|. \quad (3.4)$$

In general, an operator in the rest frame is transformed to the rotating frame according to:

$$\tilde{\mathcal{O}}(t) = e^{i\xi t} \mathcal{O}(t) e^{-i\xi t}. \quad (3.5)$$

Thus, the fluorescence spectrum derived in Chapter 2 [Eq. (2.43)] can be written (transforming to the interaction picture) as

$$F(\Delta_0) = |S^*(0)|^2 2\text{Re} \left[\text{Tr}_S \{ \sigma_{ba} \tilde{\Omega}_S(-i\Delta_0) \} \right], \quad (3.6)$$

where $\Delta_0 = \omega_0 - \omega$ and ω_0 is the probe frequency.

3.2.2 Dressed states

We write the RWA Hamiltonian, $\tilde{H}$, in a simpler form by transforming to basis of dressed states in the presence of the intense laser field. In terms of the bare atomic basis, the dressed states are

$$|+\rangle = \cos \theta |a\rangle + \sin \theta |b\rangle \quad (3.7)$$

$$|-\rangle = -\sin \theta |a\rangle + \cos \theta |b\rangle \quad (3.8)$$

where $\Omega_R = \sqrt{\Omega^2 + \Delta^2}$ is the dressed Rabi frequency and the mixing angle θ is given by

$$\cos \theta = \sqrt{\frac{\Omega_R + \Delta}{2\Omega_R}} \quad (3.9)$$

$$\sin \theta = \sqrt{\frac{\Omega_R - \Delta}{2\Omega_R}}. \quad (3.10)$$

The dressed states $|+\rangle$ and $|-\rangle$ are eigenstates of Hamiltonian

$$\tilde{H}_S = \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}(\sigma_{ab} + \sigma_{ba}) \quad (3.11)$$

with energy eigenvalues $E_{\pm} = \pm \frac{1}{2}\Omega_R$. The Hamiltonian, in terms of the dressed-state basis, is then

$$\begin{aligned} \tilde{H} = & \frac{\Omega_R}{2}\sigma_3 + \cos \theta \sin \theta \sum_k g_k (a_k + a_k^\dagger) + \sum_k \Delta_k a_k^\dagger a_k + \sum_q \omega_q b_q^\dagger b_q \\ & + \sum_k g_k \left[(\cos^2 \theta a_k - \sin^2 \theta a_k^\dagger) \sigma_{+-} + (\cos^2 \theta a_k^\dagger - \sin^2 \theta a_k) \sigma_{-+} \right] \\ & + \frac{1}{2} (\mathbb{1} + (\cos^2 \theta - \sin^2 \theta) \sigma_3) \sum_q \lambda_q (b_q + b_q^\dagger) \\ & - \sin \theta \cos \theta \sum_q \lambda_q (\sigma_{+-} + \sigma_{-+}) (b_q + b_q^\dagger) \end{aligned} \quad (3.12)$$

where $\sigma_{ij} = |i\rangle\langle j|$, $i, j \in \{+, -\}$ and $\sigma_3 = |+\rangle\langle+| - |-\rangle\langle-|$.

Following the discussion in Section 2.8.5, the expression for the fluorescence spectrum in the basis of dressed states [Eq. (2.100)] can be written as

$$\begin{aligned} F(\Delta_0) = & |S^*(0)|^2 2\text{Re} \left[\cos \theta \sin \theta \tilde{\Omega}_z(-i\Delta_0) \right. \\ & \left. + \cos^2 \theta \tilde{\Omega}_{+-}(-i\Delta_0) - \sin^2 \theta \tilde{\Omega}_{-+}(-i\Delta_0) \right], \end{aligned} \quad (3.13)$$

where

$$\tilde{\Omega}_z(-i\Delta_0) = \tilde{\Omega}_{++}(-i\Delta_0) - \tilde{\Omega}_{--}(-i\Delta_0). \quad (3.14)$$

3.2.3 Polaron transformation

We write the RWA Hamiltonian in the polaron frame by using a canonical transformation

$$H' = e^S \tilde{H} e^{-S} \quad (3.15)$$

where $S = \frac{1}{2} \mathbb{1} \otimes \sum_q \frac{\lambda_q}{\omega_q} (b_q^\dagger - b_q)$ and $S^\dagger = -S$. The result of the transformation is to shift the phonon ladder operators $b_q \rightarrow b_q + \frac{\lambda_q}{\omega_q}$, and the Hamiltonian H' resulting from the polaron transformation, is then

$$H' = H'_0 + H_V \quad (3.16)$$

$$H'_0 = H'_S + H'_x + H'_R + H'_y + H'_P \quad (3.17)$$

$$H'_V = H'_{SR} + H'_{SP}. \quad (3.18)$$

The individual terms appearing in Eqs. (3.17) and (3.18) are the unperturbed two-level system, cavity and phonon Hamiltonians

$$H'_S = \left(\frac{\Omega_R - \Delta_P}{2} \right) \sigma_3 \quad (3.19)$$

$$H'_R = \sum_k \Delta_k a_k^\dagger a_k \quad (3.20)$$

$$H'_P = \sum_q \omega_q b_q^\dagger b_q \quad (3.21)$$

where $\Delta_P = \sum_q \frac{\lambda_q^2}{\omega_q}$ is the polaron shift and coupling terms describing pure dephasing due to cavity and phonons, respectively,

$$H'_x = \cos \theta \sin \theta \sum_k g_k \sigma_3 (a_k + a_k^\dagger) \quad (3.22)$$

$$H'_y = \left(\frac{\cos^2 \theta - \sin^2 \theta}{2} \right) \sum_q \lambda_q \sigma_3 (b_q + b_q^\dagger) \quad (3.23)$$

and finally two terms that account for transitions due to the cavity, and due to phonons, respectively

$$H'_{SR} = \sum_k g_k \left[(\cos^2 \theta a_k - \sin^2 \theta a_k^\dagger) \sigma_{+-} + (\cos^2 \theta a_k^\dagger - \sin^2 \theta a_k) \sigma_{-+} \right] \quad (3.24)$$

$$H'_{SP} = -\sin \theta \cos \theta \sum_q \lambda_q (\sigma_{+-} + \sigma_{-+}) (b_q + b_q^\dagger) \quad (3.25)$$

and $H'_V = H'_{SR} + H'_{SP}$. The separation into pure-dephasing and transition terms is determined by the form of the system coupling, i.e. the dephasing terms contain σ_3 and the transition terms contain σ_{+-} and σ_{-+} .

3.2.4 Fluorescence Spectrum in the polaron frame

We recall the expression for the fluorescence spectrum, equation (3.6)

$$F(\Delta_0) = |S^*(0)|^2 2\text{Re} \int_0^\infty dt e^{i\Delta_0 t} \text{Tr} \{ \sigma_{ba} \tilde{\Omega}(t) \}. \quad (3.26)$$

The operator $\tilde{\Omega}(t) = e^{-i\tilde{H}t} \tilde{\rho} \sigma_{ab} e^{i\tilde{H}t}$ describes the dynamics of the system in the RWA. Inserting this into the above equation

$$F(\Delta_0) = |S^*(0)|^2 2\text{Re} \int_0^\infty dt e^{i\Delta_0 t} \text{Tr} \{ \sigma_{ba} e^{-i\tilde{H}t} \tilde{\rho} \sigma_{ab} e^{i\tilde{H}t} \}. \quad (3.27)$$

After performing the polaron transformation on the Hamiltonian the accompanying transformation needs to be done on the operators. Using the relation $\tilde{H} = e^{-S} H' e^S$, we can write

$$e^{i\tilde{H}t} = e^{ie^{-S} H' e^S t} = e^{-S} e^{iH't} e^S \quad (3.28)$$

the expression for the fluorescence spectrum takes the form

$$F(\Delta_0) = |S^*(0)|^2 2\text{Re} \int_0^\infty dt e^{i\Delta_0 t} \text{Tr} \{ \sigma_{ba} \underbrace{e^{-iH't} e^S \tilde{\rho} \sigma_{ab} e^{-S}}_{\text{polaron transformed density matrix}} e^{iH't} \}. \quad (3.29)$$

where we have used that $e^S \sigma_{ba} e^{-S} = \sigma_{ba}$ and the fluorescence spectrum can therefore be described in terms of dynamics in the polaron frame

$$F(\Delta_0) = |S^*(0)|^2 2\text{Re Tr}\{\sigma_{ba}\Omega'(\Delta_0)\} \quad (3.30)$$

$$= |S^*(0)|^2 2\text{Re Tr}_S\{\sigma_{ba}\Omega'_S(\Delta_0)\} \quad (3.31)$$

where $\Omega'_S(\Delta'_0) = \text{Tr}_R \text{Tr}_P\{\Omega'(\Delta'_0)\}$ and here the prime denotes that operators are in the polaron frame with a modified initial condition

$$\Omega'(0) = e^S \bar{\rho} \sigma_{ab} e^{-S}, \quad (3.32)$$

which we will discuss in Appendix A.4.2.

3.3 Fluorescence Spectrum within the Secular Approximation

In this section, the matrix elements, $\Omega'_z(t) = \Omega'_{++}(t) - \Omega'_{--}(t)$ and $\Omega'_{+-}(t)$, of the operator $\Omega'_S(t)$ will be evaluated in the polaron frame. Primes, however, will be omitted for convenience from the expectation values. We introduce the projection superoperator P , defined by its action on an operator $\mathcal{O}$

$$P\mathcal{O} = \rho_R(t_0)\rho_P(t_0)\text{Tr}_R \text{Tr}_P\mathcal{O} \quad (3.33)$$

where $\rho_R(t_0)$ and $\rho_P(t_0)$ are the initial density matrices for radiation and phonon modes and Tr_R and Tr_P are the partial traces over radiation and phonon modes, respectively. The generalized master equation for the operator $\Omega(t)$ is, similar to Eq. (2.58) and assuming a vanishing irrelevant part,

$$P\dot{\Omega}(t) = iPLP\Omega(t) - i \int_0^t dt' \Sigma(t-t')P\Omega(t') \quad (3.34)$$

The radiation modes are assumed to be initially in the vacuum state given by the density matrix $\rho_R(t_0) = \prod_k |0_k\rangle\langle 0_k|$ and the phonon modes density matrix is given by $\rho_P(t_0) = \frac{e^{-\beta H_P}}{\text{Tr}\{e^{-\beta H_P}\}}$ and $\beta = \frac{1}{k_B T}$. When the initial radiation modes and phonon modes

are of the form described above, P obeys the following important identities

$$PLP = L_S P \quad (3.35)$$

$$PL_V P = 0 \quad (3.36)$$

where $L = L_0 + L_V$ is the full Liouvillian superoperator associated with the Hamiltonians given in Eqs. (3.19)-(3.25), and is defined by its action on an arbitrary operator as $L\mathcal{O} = [H', \mathcal{O}]$. Using the above identities, we obtain a similar equation to Eq. (2.115) for the reduced operator $\Omega_S(\Delta_0)$ as

$$M(\Delta_0)\Omega_S(-i\Delta_0) = \Omega_S(0), \quad (3.37)$$

where

$$\Omega_S(-i\Delta_0) = \text{Tr}_R \text{Tr}_P \{\Omega(-i\Delta_0)\}. \quad (3.38)$$

The initial condition is

$$\Omega_S(0) = \text{Tr}_R \text{Tr}_P \{\Omega(0)\}, \quad (3.39)$$

and $\tilde{\rho}_S = \text{Tr}_R \text{Tr}_P \{\tilde{\rho}\}$ is the stationary reduced system density matrix given by expression Eq.(2.59)

$$\tilde{\rho}_S = \lim_{s \rightarrow 0} s \tilde{\rho}_S(s), \quad (3.40)$$

and the reduced density matrix in Laplace space, is

$$\tilde{\rho}_S(s) = \frac{1}{s + iL_S + i\Sigma_S(s)} \tilde{\rho}_S(t_0). \quad (3.41)$$

The propagator, $M(\Delta_0)$, is defined as [Eq. (2.118)]

$$[M(\Delta_0)]^{-1} = \frac{1}{-i\Delta_0 \otimes \mathbb{1} + i[L_S] + i[\Sigma_S(-i\Delta_0)]} \quad (3.42)$$

$$\Sigma_S(s) = \text{Tr}_R \text{Tr}_P \left[LQ \frac{1}{s + iQL} QL \rho_R(t_0) e^s \rho_P(t_0) e^{-s} \right]. \quad (3.43)$$

The self-energy superoperator can be expanded in powers of the transition part of the perturbation Liouvillian $L_V = L_{SR} + L_{SP}$ but to all orders in the dephasing part of the perturbation Liouvillian (See Appendix A for detail)

$$\Sigma_S(\Delta_0) = \Sigma_S^{(2)}(\Delta_0) + \Sigma_S^{(4)}(\Delta_0) + \dots \quad (3.44)$$

since only even powers of L_V contribute in the final trace over the radiation/phonon modes. In the Born approximation, we replace the relaxation part of the self-energy superoperator by its lowest order in terms of L_{SR} , L_{SP} ($\Sigma_S(\Delta_0) \rightarrow \Sigma_S^{(2)}(\Delta_0)$), but treat dephasing exactly. In Born approximation, the stationary density matrix and propagator are

$$\bar{\rho}_S = \lim_{s \rightarrow 0} s \frac{1}{s + iL_S + i\Sigma_S^{(2)}(s)} \rho_S(0) \quad (3.45)$$

$$[M(\Delta_0)]^{-1} \approx \frac{1}{-i\Delta_0 \otimes \mathbb{1} + i[L_S] + i[\Sigma_S^{(2)}(-i\Delta_0)]}. \quad (3.46)$$

The matrix for L_S is

$$[L_S]_{\alpha\beta,\gamma\delta} = \text{Tr} \left\{ |\delta\rangle \langle \gamma| L_S |\alpha\rangle \langle \beta| \right\} \quad (3.47)$$

where α, β, γ and $\delta \in \{+, -\}$,

$$[L_S] = \begin{matrix} & \begin{matrix} ++ & -- & +- & -+ \end{matrix} \\ \begin{matrix} ++ \\ -- \\ +- \\ -+ \end{matrix} & \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & (\Omega_R - \Delta_P) & 0 \\ 0 & 0 & 0 & -(\Omega_R - \Delta_P) \end{pmatrix} \end{matrix}.$$

We now set

$$\Delta'_0 = \Delta_0 - (\Omega_R - \Delta_P), \quad (3.48)$$

where $|\Delta'_0| \ll \Omega_R$ and $\Delta_0 = \Delta'_0 + (\Omega_R - \Delta_P)$. Substituting for Δ_0 in equation (2.118), we obtain

$$[M'(\Delta'_0)]^{-1} = \frac{1}{-i[\Delta'_0 + (\Omega_R - \Delta_P)] \otimes \mathbb{1} + i[L_S] + i[\Sigma_S^{(2)}(\Delta'_0)]}. \quad (3.49)$$

Within the secular approximation, $|\Omega_R - \Delta_P| \gg |\Sigma_S^{(2)}(\Delta'_0)|$, we neglect off-diagonal corrections to the propagator, giving

$$\begin{aligned} [M'(\Delta'_0)]_{+-,+-}^{-1} &= \frac{1}{-i[\Delta'_0 + (\Omega_R - \Delta_P)] + i(\Omega_R - \Delta_P) + i[\Sigma_{+-,+-}^{(2)}(\Delta'_0)]} \\ &= \frac{1}{-i\Delta'_0 + i\Sigma_{+-,+-}^{(2)}(\Delta'_0)}. \end{aligned} \quad (3.50)$$

From equations (3.37) and (3.50), within the secular approximation, the equation for the matrix element $\Omega_{+-}(\Delta'_0)$ takes the form

$$\Omega_{+-}(\Delta'_0) \approx \frac{1}{-i\Delta'_0 + i\Sigma'^{(2)}_{+-,+-}(\Delta'_0)} \Omega_{+-}(0). \quad (3.51)$$

The reduced self-energy matrix element, $\Sigma'^{(2)}_{+-,+-}(\Delta'_0)$, consists of four terms

$$\Sigma'^{(2)}_{+-,+-}(\Delta'_0) = \Sigma'^x_{+-,+-}(\Delta'_0) + \Sigma'^y_{+-,+-}(\Delta'_0) + \Sigma'^{SR}_{+-,+-}(\Delta'_0) + \Sigma'^{SP}_{+-,+-}(\Delta'_0) \quad (3.52)$$

where the first two terms

$$\Sigma'^x_{+-,+-}(\Delta'_0) = -i\text{Tr}_R L_x^+ \frac{1}{s + i(L_S + L_R + L_x^+)} L_x^+ \rho_R(0) \quad (3.53)$$

$$\Sigma'^y_{+-,+-}(\Delta'_0) = -i\text{Tr}_P L_y^+ \frac{1}{s + i(L_S + L_P + L_y^+)} L_y^+ \rho_P(0) \quad (3.54)$$

in the above equation give rise to the pure dephasing (T_2^* process) due to system-photon coupling and system-phonon couplings, respectively, where $L_{x(y)}^+ \mathcal{O} = \{H'_{x(y)}, \mathcal{O}\}$ and braces indicate the anti-commutator. Whereas the last two terms

$$\Sigma'^{SR}_{+-,+-}(\Delta'_0) = -i\text{Tr}_R L_{SR} \frac{1}{s + i(L_S + L_R + L_x^+)} L_{SR} \rho_R(0) \quad (3.55)$$

$$\Sigma'^{SP}_{+-,+-}(\Delta'_0) = -i\text{Tr}_P L_{SP} \frac{1}{s + i(L_S + L_P + L_y^+)} L_{SP} \rho_P(0) \quad (3.56)$$

in the above equation give rise to the relaxation (T_1 process) due to system-photon coupling and system-phonon couplings, respectively.

We neglect the contributions $\Sigma'^{SR}_{+-,+-}(\Delta'_0)$, $\Sigma'^{SP}_{+-,+-}(\Delta'_0)$, giving rise to relaxation, arguing that they are small relative to the dephasing parts of self-energy since they are suppressed by a large frequency $\Omega_R - \Delta_P$, Eq. (3.51) takes the form

$$\Omega_{+-}(\Delta'_0) \approx \frac{1}{-i\Delta'_0 + i\Sigma'^x_{+-,+-}(\Delta'_0) + i\Sigma'^y_{+-,+-}(\Delta'_0)} \Omega_{+-}(0). \quad (3.57)$$

3.4 Simplified Fluorescence Spectrum

As shown in Chapter 2, the fluorescence spectrum for a strongly-driven two-level system generally leads to three peaks, describing the Mollow triplet. Here, we will derive a simplified expression for one of the side-peaks in the Mollow triplet spectrum. We will analyze the lineshape and frequency shift of this side-peak in both the conventional Markovian regime as well as the non-Markovian regime. The expression for the three-peak (Mollow-triplet) spectrum is given by Eq. (3.13), in terms of matrix elements of $\Omega_S(\Delta_0)$

$$F(\Delta_0) = |S^*(0)|^2 2\text{Re} \left[\cos \theta \sin \theta \Omega_z(-i\Delta_0) + \cos^2 \theta \Omega_{+-}(-i\Delta_0) - \sin^2 \theta \Omega_{-+}(-i\Delta_0) \right]. \quad (3.58)$$

Setting

$$\Delta'_0 = \Delta_0 - (\Omega_R - \Delta_P), \quad (3.59)$$

the above expression can be written as

$$F'(\Delta'_0) = |S^*(0)|^2 2\text{Re} \left[\cos \theta \sin \theta \Omega'_z(-i\Delta'_0) + \cos^2 \theta \Omega'_{+-}(-i\Delta'_0) - \sin^2 \theta \Omega'_{-+}(-i\Delta'_0) \right] \quad (3.60)$$

where $F'(\Delta'_0) = F(\Delta'_0 + \Omega_R - \Delta_P)$.

The matrix elements of the operator $\Omega_S(t)$ are given by [Eq. (2.123)-(2.124)]. These equations are written in the time domain. We now perform a Laplace transform on Eqs. (2.123)-(2.124) and evaluate at $s = -i\Delta'_0$ to obtain

$$\Omega'_z(\Delta'_0) = \frac{\Omega'_z(0) + N_z(\Delta'_0)}{-i\Delta'_0 + i(\Omega_R - \Delta_P) + i\Sigma'^{(2)}_z(\Delta'_0)} \quad (3.61)$$

$$\Omega'_{+-}(\Delta'_0) = \frac{1}{-i\Delta'_0 + i\Sigma'^{(2)}_{+-}(\Delta'_0)} \Omega'_{+-}(0) \quad (3.62)$$

$$\Omega'_{-+}(\Delta'_0) = \frac{1}{-i\Delta'_0 + 2i(\Omega_R - \Delta_P) + i\Sigma'^{(2)}_{-+}(\Delta'_0)} \Omega'_{-+}(0). \quad (3.63)$$

We note that $\Omega'_z(0)$, $\Omega'_{+-}(0)$, and $\Omega'_{-+}(0)$ are matrix elements of $\Omega'_S(t)$ evaluated at $t = 0$. It can be seen above that the expressions for $\Omega'_z(\Delta'_0)$ and $\Omega'_{-+}(\Delta'_0)$ are suppressed, in the limit $|\Delta'_0| \ll |\Omega_R - \Delta_P|$, by relatively large frequencies $(\Omega_R - \Delta_P)$ and $2(\Omega_R - \Delta_P)$, respectively compared to the expression for $\Omega'_{+-}(\Delta'_0)$. We argue that, in the limit when

$$|(\Omega_R - \Delta_P)| \gg |\Sigma'_z(2)(\Delta'_0)|, |\Sigma'_{-+,-+}(\Delta'_0)|, \quad (3.64)$$

and for $|\Delta'_0| \ll |\Omega_R - \Delta_P|$ the three-peak spectrum in Eq. (3.60) reduces to a single-peak spectrum centered at $\Delta'_0 = 0$ and takes the form

$$F'(\Delta'_0) \approx 2|S^*(0)|^2 \text{Re} [\cos^2 \theta \Omega'_{+-}(\Delta'_0)] \quad (3.65)$$

$$|S^*(0)|^2 = \frac{\alpha^2 g^4}{\Gamma_c^2 + \Delta_c^2}, \quad (3.66)$$

where $|S^*(0)|^2$ is the detector response function discussed in Appendix A.

Substituting for $\Omega'_{+-}(\Delta'_0)$ from equation (3.57) into equation (3.65), we obtain

$$F'(\Delta'_0) \approx 2|S^*(0)|^2 \text{Re} \left[\cos^2 \theta \frac{1}{-i\Delta'_0 + i\Sigma'_{+-,+-}(\Delta'_0) + i\Sigma'_{+-,+-}(\Delta'_0)} \Omega'_{+-}(0) \right] \quad (3.67)$$

The system-phonon coupling reduced self-energy matrix element can be calculated exactly to all orders in L_y^+ , while the system-photon coupling reduced self-energy matrix element can be calculated within Born approximation (to second order in L_x^+).

The contribution from photon coupling, $\Sigma'_{+-,+-}(\Delta'_0)$, is expressed as (refer to Appendix A for detail)

$$\Sigma'_{+-,+-}(\Delta'_0) = -2ig^2 \sin^2 \theta \cos^2 \theta \left(\frac{1}{-i\Delta'_0 - i\Delta_c + \Gamma_c} + \frac{1}{-i\Delta'_0 + i\Delta_c + \Gamma_c} \right). \quad (3.68)$$

We recall that $\Delta_c = \omega_c - \omega$ is the detuning of the cavity from the laser pump frequency. Equation (3.63) can be written in terms of a dimensionless decay rate $\Gamma_R(\Delta'_0)$ and dimensionless frequency shift $\Delta\omega_R(\Delta'_0)$, each scaled by a common frequency scale

γ_R :

$$\Sigma'_{+-,+-}(\Delta'_0) = -i\gamma_R\Gamma_R(\Delta'_0) + \gamma_R\Delta\omega_R(\Delta'_0), \quad (3.69)$$

$$\gamma_R = \frac{2g^2}{\Gamma_c} \left(\frac{\Omega_R^2 - \Delta^2}{4\Omega_R^2} \right) \quad (3.70)$$

where $\Gamma_R(\Delta'_0)$ and $\Delta\omega_R(\Delta'_0)$ are the detuning-dependent decay and frequency-shift, respectively, due to photon coupling, and are given explicitly as

$$\Gamma_R(\Delta'_0) = \frac{1}{\left(\frac{\Delta'_0 + \Delta_c}{\Gamma_c}\right)^2 + 1} + \frac{1}{\left(\frac{\Delta'_0 - \Delta_c}{\Gamma_c}\right)^2 + 1} \quad (3.71)$$

$$\Delta\omega_R(\Delta'_0) = \frac{\left(\frac{\Delta'_0 + \Delta_c}{\Gamma_c}\right)}{\left(\frac{\Delta'_0 + \Delta_c}{\Gamma_c}\right)^2 + 1} + \frac{\left(\frac{\Delta'_0 - \Delta_c}{\Gamma_c}\right)}{\left(\frac{\Delta'_0 - \Delta_c}{\Gamma_c}\right)^2 + 1}. \quad (3.72)$$

In a similar fashion, the contribution due to phonon coupling can be expressed as (refer to Appendix A for detail)

$$\Sigma'_{+-,+-}(\Delta'_0) = -i\frac{\alpha_P\Delta^2}{\Omega_R^2}\Gamma_P(\Delta'_0) + \frac{\alpha_P\Delta^2}{\Omega_R^2}\Delta\omega_P(\Delta'_0) \quad (3.73)$$

where $\alpha_P = \lambda^2 K_3$ and $\Gamma_P(\Delta'_0)$ and $\Delta\omega_P(\Delta'_0)$ are the frequency-dependent decay rate and frequency-shift, respectively due to phonon coupling and are given as

$$\Gamma_P(\Delta'_0) = \frac{\pi}{2}|\Delta'_0|^3 \coth\left(\frac{\Delta'_0}{2k_B T}\right) e^{\frac{-|\Delta'_0|}{\epsilon_c}} \quad (3.74)$$

$$\Delta\omega_P(\Delta'_0) = \frac{1}{2}P \int_{-\infty}^{\infty} d\epsilon \frac{|\epsilon|^3 \coth\left(\frac{\epsilon}{2k_B T}\right) e^{-\frac{|\epsilon|}{\epsilon_c}}}{\Delta'_0 - \epsilon}. \quad (3.75)$$

Here, 'P' indicates a principal-value integral.

From Eq. (2.105), the initial condition for $\Omega'_{+-}(0) = \cos^2 \theta \tilde{\rho}_{++} - \sin^2 \theta \tilde{\rho}_{+-}$ and noting that

$$\tilde{\rho}_{+-}(s) = \frac{1}{s + i(\Omega_R - \Delta_P) + i\Sigma'_{+-,+-}(s)} \tilde{\rho}_{+-}(0) \quad (3.76)$$

and for the condition $|(\Omega_R - \Delta_P)| \gg |\Sigma'_{+-,+}(s)|$ the matrix element $\tilde{\rho}_{+-}(s)$ does not have a pole at $s = 0$, therefore taking the stationary limit gives

$$\bar{\rho}_{+-} = \lim_{s \rightarrow 0} s \tilde{\rho}_{+-}(s) = 0. \quad (3.77)$$

And the initial condition is then

$$\Omega'_{+-}(0) = \cos^2 \theta \bar{\rho}_{++}, \quad (3.78)$$

where the matrix element of the stationary density matrix $\bar{\rho}_{++}$ is given as (refer to Appendix B)

$$\begin{aligned} \bar{\rho}_{++} &= \lim_{s \rightarrow 0} s \rho'_{++}(s) \\ &= \frac{\frac{(\Omega_R - \Delta)^2}{\Gamma_c^2 + (\Omega_R - \Delta_P + \Delta_c)^2}}{\frac{(\Omega_R + \Delta)^2}{\Gamma_c^2 + (\Omega_R - \Delta_P - \Delta_c)^2} + \frac{(\Omega_R - \Delta)^2}{\Gamma_c^2 + (\Omega_R - \Delta_P + \Delta_c)^2}} \end{aligned} \quad (3.79)$$

$$(3.80)$$

and the mixing angle is

$$\cos \theta = \left(\frac{\Omega_R + \Delta}{2\Omega_R} \right)^{\frac{1}{2}}. \quad (3.81)$$

Substituting for $\cos^2 \theta \bar{\rho}'_{++}$ and $\Sigma'^y_{+-,+}(\Delta'_0)$ from equation (3.73) in the final expression for the one-peak spectrum takes the form

$$F'(\Delta'_0) = X \frac{\Gamma_R(\Delta'_0) + \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Gamma_P(\Delta'_0)}{\left(\frac{\Delta'_0}{\gamma_R} - \Delta \omega_R(\Delta'_0) - \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Delta \omega_P(\Delta'_0) \right)^2 + \left(\Gamma_R(\Delta'_0) + \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Gamma_P(\Delta'_0) \right)^2}, \quad (3.82)$$

where $X = \cos^4 \theta \bar{\rho}'_{++}$ is the Δ'_0 - independent prefactor having dimensions of frequency. This prefactor can be expressed in terms of other parameters as

$$X = \frac{\alpha^2 \left(\frac{g}{\gamma_R} \right)^4 \gamma_R}{2 \left(\frac{\Gamma_c^2 + \Delta_c^2}{\gamma_R^2} \right)} \frac{\frac{(\Omega_R^2 - \Delta^2)^2}{\Omega_R^2 [\Gamma_c^2 + (\Omega_R - \Delta_P + \Delta_c)^2]}}{\frac{(\Omega_R + \Delta)^2}{\Gamma_c^2 + (\Omega_R - \Delta_P - \Delta_c)^2} + \frac{(\Omega_R - \Delta)^2}{\Gamma_c^2 + (\Omega_R - \Delta_P + \Delta_c)^2}}. \quad (3.83)$$

3.4.1 Side-peak spectrum for finite detuning: non-Markov versus Markov

Equation (3.82) is non-Markovian in both photon and phonon terms. We can perform a Markov approximation with respect to the system-photon coupling only. This approximation is justified when the time-dependent self-energy matrix element, $\Sigma'_{+-,+}(t)$ decays rapidly compared to the timescale of evolution for the observable $\Omega'_{+-}(t)$. This condition is satisfied for $\gamma_R \ll \Gamma_c$, in which case we can approximate the fluorescence spectrum by

$$F'(\Delta'_0) \approx X \frac{\Gamma_R(\Delta\omega) + \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Gamma_P(\Delta'_0)}{\left(\frac{\Delta'_0}{\gamma_R} - \Delta\omega_R(\Delta\omega) - \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Delta\omega_P(\Delta'_0)\right)^2 + \left(\Gamma_R(\Delta\omega) + \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Gamma_P(\Delta'_0)\right)^2} \quad (3.84)$$

where $\Delta\omega$ is given implicitly by the equation

$$\Delta\omega - \Delta\omega_R(\Delta\omega) - \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Delta\omega_P(\Delta\omega) = 0. \quad (3.85)$$

Equation (3.84) is Markovian in photon terms but non-Markovian in phonon terms. We want to discuss the spectrum in a regime where spontaneous emission can be treated within a Markov approximation while the phonon-induced dephasing term describes non-Markov dynamics. In order to see the distinction between Markovian and non-Markovian phonon-induced dephasing, we carry out the Markov approximation on equation (3.82) in terms of the phonon coupling to write the spectrum within the Markov approximation for both photon and phonon couplings

$$F'(\Delta'_0) \approx X \frac{\Gamma_R(\Delta\omega) + \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Gamma_P(\Delta\omega)}{\left(\frac{\Delta'_0}{\gamma_R} - \Delta\omega_R(\Delta\omega) - \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Delta\omega_P(\Delta\omega)\right)^2 + \left(\Gamma_R(\Delta\omega) + \frac{\alpha_P \Delta^2}{\gamma_R \Omega_R^2} \Gamma_P(\Delta\omega)\right)^2} \quad (3.86)$$

where $\Delta\omega$ is given implicitly by the Eq. (3.85). Equation (3.86) is Markovian in terms of both photon and phonon couplings and the Markov approximation carried out here is justified when the self-energy matrix element $\Sigma'_{+-,+}(t)$ decays rapidly on the time scale of evolution of the observable $\Omega'_{+-}(t)$.

From the expression given in Eq. (3.86), we can discuss the regime of validity of the Markov approximation, i.e. we can determine when the one-peak spectrum will exhibit a pure Lorentzian or non-Lorentzian lineshape.

For the elevated temperature $k_B T \gg \epsilon_c$ the width of side peak is set by $\Delta'_0 \sim \epsilon_c$ and for large α_P , the condition for the validity of the Markov approximation in terms of phonon coupling can be given by the condition

$$\alpha_P < \frac{\gamma_R \Omega_R^2}{\Delta^2 \epsilon_c^2 k_B T}. \quad (3.87)$$

It is worth noting that for an exact resonance condition $\Delta = 0$, the phonon decay term vanishes and we obtain a Lorentzian lineshape of width given by γ_R , which is a consequence of vanishing phonon density of states at $\Delta = 0$ in the dressed state representation. In other words, the phonon coupling is zero for zero detuning and brings a non-Lorentzian feature in the lineshape for a non-zero detuning.

In Figs. (3.2) and (3.3) we discuss the one side-peak spectra given by Eqs. (3.84) (red curve) and (3.86) (blue curve) for different parameter values in both the Markovian and the non-Markovian regimes. For the condition given by Eq. (3.87), the two curves (red and blue) fall on the top of each other. In other words, in this limit the decay is dominated by the spontaneous decay rate while in the opposite regime the decay is dominated by the frequency-dependent decay and curves deviate from each other. We observe a dip in the red curve at $\Delta'_0 = 0$, and it can be understood in terms of vanishing phonon density of states at $\Delta'_0 = 0$. Noting that the frequency-dependent decay $\Gamma_P(\Delta'_0)$ is identically zero at zero probe frequency i.e. $\Delta'_0 = 0$.

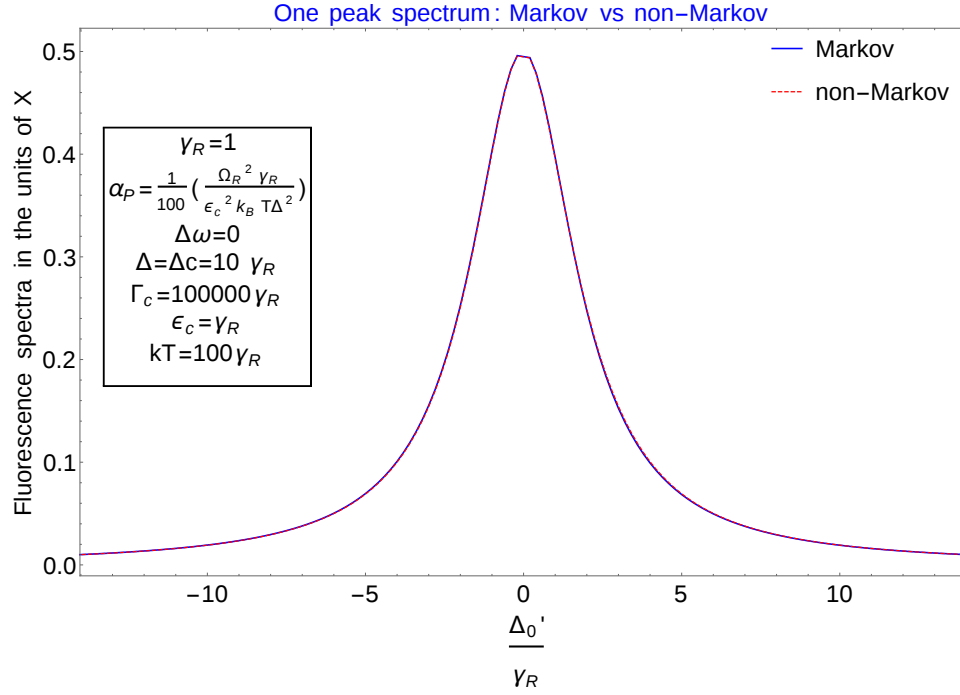


Figure 3.2: non-Markovian and Markovian fluorescence spectra: The red curve represents the non-Markovian spectrum given by Eq. (3.84) and the blue curve represents the Markovian spectrum given by Eq. (3.86). In the Markovian limit given by Eq. (3.87) both curves, non-Markovian (red) and Markovian (blue), fall on the top of each other.

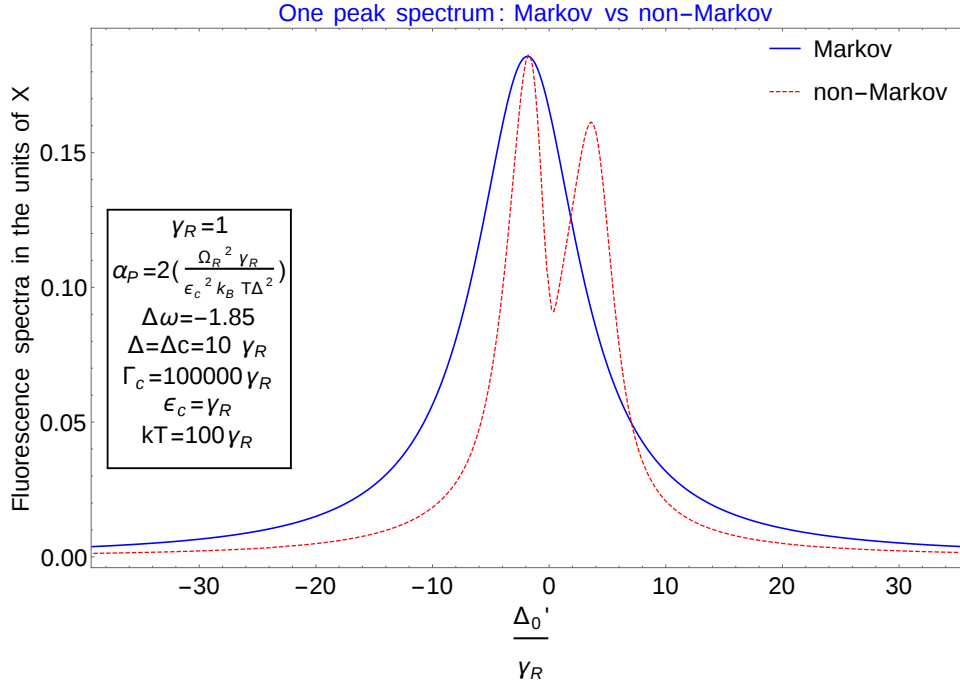


Figure 3.3: Full non-Markovian fluorescence spectrum alongside the Markovian fluorescence spectrum: The red curve represents the non-Markovian spectrum given by Eq. (3.84) and the blue curve represents the Markovian spectrum given by Eq. (3.86). In the non-Markovian limit both curves, non-Markovian (red) and Markovian (blue), deviate from each other and a dip is observed in the red curve at $\Delta'_0 = 0$.

3.5 Conclusion

In this chapter, we discussed the fluorescence spectrum of a two-level cavity-QED system with phonon-induced non-Markovian dephasing. We have illustrated the phonon-induced dephasing in the dressed state representation accounting for an off-resonant drive laser. We have derived a simplified formula for the Mollow-triplet spectrum, which clearly describes the phonon-induced dephasing in the side-peak spectrum. We have shown the differences between the Markovian and non-Markovian spectrum with phonon-induced non-Markovian dephasing. By changing the phonon parameters like α_P and kT we can switch from Markov regime to non-Markov regime in terms of phonons. In the fully non-Markovian regime we see a deviation in the lineshape which is a signature of the non-Markovian effects due to phonon coupling. In the fully non-Markov regime with a non-zero detuning between the two-level atomic frequency and laser drive, we observe a dip in the side-peak spectrum at $\Delta'_0 = 0$. This dip is a result of vanishing density of states of phonons at $\Delta'_0 = 0$.

Chapter 4

Coherent population trapping and the influence of phonon-induced dephasing on the absorption spectrum of a lambda system

4.1 Introduction

Coherent population trapping (CPT) [4] refers to the trapping of population in a coherent superposition of two ground states which are coupled by optical fields to an intermediate state in a three-level system. This is a so-called lambda configuration as shown in fig. (4.2). The laws of quantum mechanics allow for powerful computational tasks to be performed with quantum systems through, e.g., a quantum computer. In quantum computers, the information is stored and processed in qubits rather than classical bits. Storage and processing of quantum information requires long-lived quantum states. As discussed in chapter 1, an interesting demonstration of quantum coherence is achieved through the creation of a long-lived dark state through coherent population trapping. Due to a coupling to uncontrolled environmental degrees of freedom, a quantum system may undergo pure dephasing, resulting in the loss of quantum coherence. To this end, it is necessary to understand the interaction of a quantum system with its environment which gives rise to dephasing in the system.

Recently, a number of experimental studies [18, 19] have been performed to realise and understand the coherent population trapping in nitrogen vacancy centers in diamond and semiconductor quantum dots. The decoherence process due to the interaction between a quantum system and a quantum environment is typically non-Markovian in nature for, e.g., an electron spin interacting with a nuclear spin environment [20]. However, the experimental results presented in refs. [18, 19] have been analyzed assuming a purely Markovian environment.

In this chapter, we present a preliminary analysis of the absorption spectrum of a lambda system undergoing a non-Markovian dephasing process due to coupling to a phonon reservoir. In chapter 3, we developed a method to treat the phonon-induced pure dephasing self-energy matrix element exactly. We shall apply this method to study a pure-dephasing mechanism in the absorption spectrum of a lambda system in the regime where it exhibits CPT. We consider a three-level system in the lambda configuration with two ground-state levels, $|b\rangle$ and $|c\rangle$, coupled to a common level $|a\rangle$. One of the ground states $|c\rangle$ is coupled to a phonon reservoir and can be modelled with the usual independent boson model (IBM) [17].

4.2 Theory

We consider a three-level lambda system driven by two laser fields of frequencies ω_1 and ω_2 with amplitudes given by Rabi frequencies Ω_1 and Ω_2 . In this lambda configuration the two lower levels $|b\rangle$ and $|c\rangle$ are coupled to a single upper level $|a\rangle$. The splittings between states $|a\rangle$ and $|b\rangle$ and between $|a\rangle$ and $|c\rangle$ are given by the frequencies ω_{ab} and ω_{ac} . The detunings of the transition frequencies ω_{ab} and ω_{ac} from the laser frequencies ω_1 and ω_2 are defined by $\Delta_1 = \omega_{ab} - \omega_1$ and $\Delta_2 = \omega_{ac} - \omega_2$, respectively. We have assumed that only transitions $|a\rangle \rightarrow |b\rangle$ and $|a\rangle \rightarrow |c\rangle$ are dipole allowed, i.e. there is no direct coupling between $|b\rangle$ and $|c\rangle$.

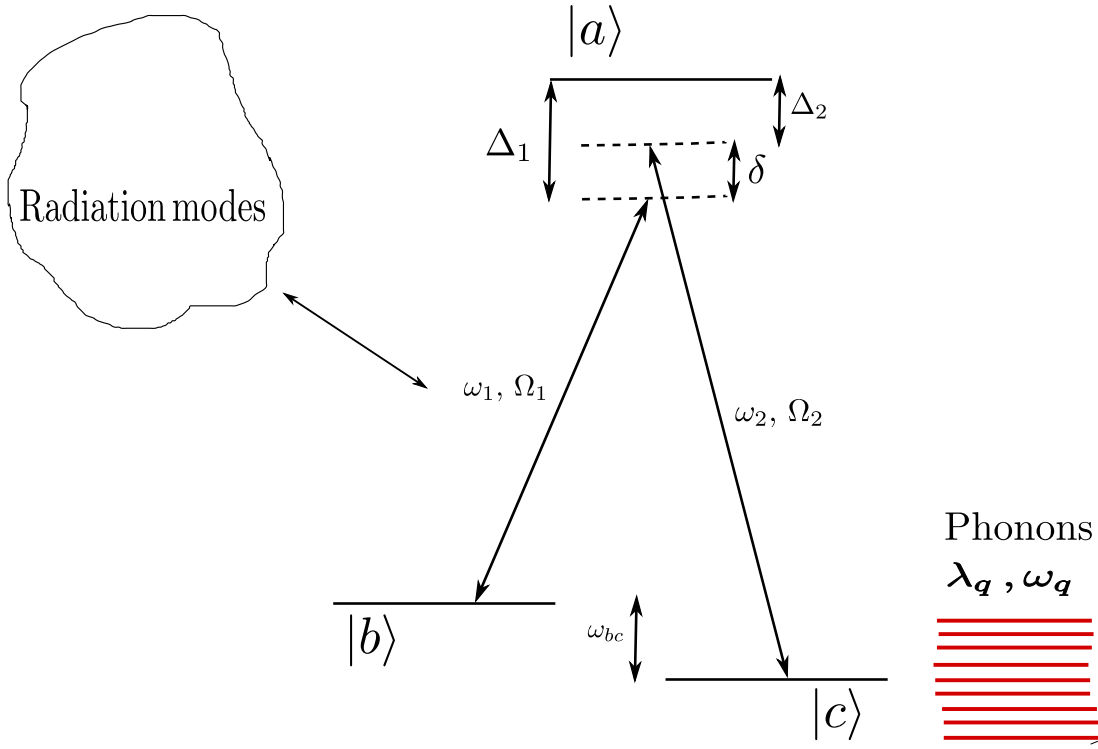


Figure 4.1: Schematic representation of a driven lambda system. The two ground states $|b\rangle$ and $|c\rangle$ are coupled by two laser fields of frequencies ω_1 and ω_2 to a common upper state $|a\rangle$ and a phonon reservoir is coupled to one of the ground states $|c\rangle$.

The Hamiltonian for the entire system consists of a lambda system plus radiation modes, phonon modes, and a diagonal pure dephasing part all included in the unperturbed Hamiltonian, H_0 , the laser part $H_{\text{laser}}(t)$, and the coupling to the radiation modes part H_V Hamiltonians. Here and in what follows we take $\hbar = 1$. Within the rotating-wave approximation (RWA), in which counter-rotating terms are neglected,

the Hamiltonian is:

$$H(t) = H_0 + H_{\text{laser}}(t) + H_V$$

$$H_0 = H_S + H_R + H_P + H_{dp}$$

$$H_S = \omega_a |a\rangle\langle a| + \omega_b |b\rangle\langle b| + \omega_c |c\rangle\langle c| \quad (4.1)$$

$$H_R = \sum_k \omega_k a_k^\dagger a_k + \sum_{k'} \omega_{k'} a_{k'}^\dagger a_{k'} \quad (4.2)$$

$$H_P = \sum_q \omega_q b_q^\dagger b_q \quad (4.3)$$

$$H_{dp} = |c\rangle\langle c| \sum_q \lambda_q (b_q + b_q^\dagger) \quad (4.4)$$

$$H_{\text{laser}}(t) = \frac{\Omega_1}{2} (|a\rangle\langle b| e^{-i\omega_1 t} + |b\rangle\langle a| e^{i\omega_1 t}) + \frac{\Omega_2}{2} (|a\rangle\langle c| e^{-i\omega_2 t} + |c\rangle\langle a| e^{i\omega_2 t}) \quad (4.5)$$

$$H_V = \sum_k g_k^b \left(|a\rangle\langle b| a_k + |b\rangle\langle a| a_k^\dagger \right) + \sum_{k'} g_{k'}^c \left(|a\rangle\langle c| a_{k'} + |c\rangle\langle a| a_{k'}^\dagger \right). \quad (4.6)$$

We assume states $|b\rangle$ and $|c\rangle$ are separated in energy by more than the radiative linewidth. Under this condition, we adopt a model in which $|b\rangle$ and $|c\rangle$ are coupled separately to independent sets of radiation modes, labelled by k and k' , respectively. In order to remove the explicit time dependence from the above Hamiltonian we follow the method given in Ref. [21]. Namely, we go to a rotating frame with respect to the laser frequencies to obtain

$$\begin{aligned} e^{i\xi t} (H(t) - \xi) e^{-i\xi t} = & \frac{1}{2} (\Delta_1 + \Delta_2) |a\rangle\langle a| - \frac{1}{2} \delta (|b\rangle\langle b| - |c\rangle\langle c|) + \sum_{k \in k_{ab}} \Delta_k a_k^\dagger a_k \\ & + \sum_{k' \in k_{ac}} \Delta_{k'} a_{k'}^\dagger a_{k'} + \sum_q \omega_q b_q^\dagger b_q + |c\rangle\langle c| \sum_q \lambda_q (b_q + b_q^\dagger) \\ & + \frac{\Omega_1}{2} (|a\rangle\langle b| + |b\rangle\langle a|) + \frac{\Omega_2}{2} (|a\rangle\langle c| + |c\rangle\langle a|) \\ & + \sum_{k \in k_{ab}} g_k^b \left(|a\rangle\langle b| a_k + |b\rangle\langle a| a_k^\dagger \right) + \sum_{k' \in k_{ac}} g_{k'}^c \left(|a\rangle\langle c| a_{k'} + |c\rangle\langle a| a_{k'}^\dagger \right), \end{aligned} \quad (4.7)$$

here, the sum over k is a sum over radiation modes around the transition frequency ω_{ab} for the $|a\rangle \rightarrow |b\rangle$ transition, and k' runs over modes around the transition frequency ω_{ac} for the $|a\rangle \rightarrow |c\rangle$ transition with $k_{ab} = \frac{\omega_{ab}}{c}$ and $k_{ac} = \frac{\omega_{ac}}{c}$. The detunings are

defined as $\delta = \Delta_1 - \Delta_2$, $\Delta_k = \omega_k - \omega_1$, $\Delta_{k'} = \omega_{k'} - \omega_2$ and

$$\begin{aligned} \xi = & \left[\omega_a - \frac{1}{2}(\Delta_1 + \Delta_2) \right] |a\rangle\langle a| + \left[\omega_b + \frac{1}{2}(\Delta_1 - \Delta_2) \right] |b\rangle\langle b| \\ & + \left[\omega_c - \frac{1}{2}(\Delta_1 - \Delta_2) \right] |c\rangle\langle c| + \omega_1 \sum_{k \in k_{ab}} a_k^\dagger a_k + \omega_2 \sum_{k' \in k_{ac}} a_{k'}^\dagger a_{k'}. \end{aligned} \quad (4.8)$$

The time-independent Hamiltonian in terms of a diagonal part, the laser part and the coupling part can be written as

$$\tilde{H} = \tilde{H}_0 + \tilde{H}_{\text{laser}} + \tilde{H}_V \quad (4.9)$$

$$\tilde{H}_0 = \tilde{H}_S + \tilde{H}_R + H_P + H_{dp} \quad (4.10)$$

$$\tilde{H}_S = \frac{1}{2}(\Delta_1 + \Delta_2)|a\rangle\langle a| - \frac{\delta}{2}(|b\rangle\langle b| - |c\rangle\langle c|) \quad (4.11)$$

$$\tilde{H}_R = \sum_{k \in k_{ab}} \Delta_k a_k^\dagger a_k + \sum_{k' \in k_{ac}} \Delta_{k'} a_{k'}^\dagger a_{k'} \quad (4.12)$$

$$\tilde{H}_{\text{laser}} = \frac{\Omega_1}{2}(|a\rangle\langle b| + |b\rangle\langle a|) + \frac{\Omega_2}{2}(|a\rangle\langle c| + |c\rangle\langle a|) \quad (4.13)$$

$$\tilde{H}_V = \sum_{k \in k_{ab}} g_k^b \left(|a\rangle\langle b| a_k + |b\rangle\langle a| a_k^\dagger \right) + \sum_{k' \in k_{ac}} g_{k'}^c \left(|a\rangle\langle c| a_{k'} + |c\rangle\langle a| a_{k'}^\dagger \right) \quad (4.14)$$

where $\tilde{H}_P \equiv H_P$ and $\tilde{H}_{dp} \equiv H_{dp}$. The expectation value of an operator $\mathcal{O}_S(t)$ in the system Hilbert space is given by

$$\langle \mathcal{O}_S(t) \rangle = \text{Tr}\{\mathcal{O}_S \rho(t)\}, \quad (4.15)$$

and the operators in the lab frame and rotating frame are related by

$$\mathcal{O}_S(t) = e^{-i\xi t} \tilde{\mathcal{O}}_S(t) e^{i\xi t}. \quad (4.16)$$

Using the relation (4.16), we obtain an expression for the expectation value of $\mathcal{O}_S(t)$ in the rotating frame

$$\begin{aligned} \langle \mathcal{O}_S(t) \rangle &= \text{Tr}\{\mathcal{O}_S e^{-i\xi t} \tilde{\rho}(t) e^{i\xi t}\} \\ &= \text{Tr}\{e^{i\xi t} \mathcal{O}_S e^{-i\xi t} \tilde{\rho}(t)\}. \end{aligned} \quad (4.17)$$

4.3 Generalized master equation

The lambda system, radiation modes, and phonon modes are decoupled for times $t < 0$ and prepared separately in the states described by the density matrices $\rho_S(0)$, $\rho_R(0)$ and $\rho_P(0)$, respectively. At time $t = 0$, the lambda system, radiation modes, and phonon modes are brought into contact and the state of the entire system is described by the total density matrix $\rho(0)$ at time $t = 0$, which is given by

$$\rho(0) = \rho_S(0) \otimes \rho_R(0) \otimes \rho_P(0). \quad (4.18)$$

In order to evaluate the dynamics of the reduced density matrix $\tilde{\rho}_S(t) = \text{Tr}_R \text{Tr}_P \{\tilde{\rho}(t)\}$, where Tr_R and Tr_P are partial traces over the radiation and phonon modes, respectively, we introduce a projection superoperator P defined by its action on an arbitrary operator $\mathcal{O}$ as

$$P\mathcal{O} = \rho_R(0)\rho_P(0)\text{Tr}_R \text{Tr}_P \{\mathcal{O}\}. \quad (4.19)$$

The product-state initial condition given in Eq. (4.18) satisfies $P\rho(0) = \rho(0)$, which is a sufficient condition to write the von-Neumann equation of motion

$$\dot{\tilde{\rho}}(t) = -i[\tilde{H}, \tilde{\rho}(t)] \quad (4.20)$$

in form of Nakajima-Zwanzig generalized master equation

$$\begin{aligned} P\dot{\tilde{\rho}}(t) &= -iPLP\tilde{\rho}(t) - i \int_0^t dt' \Sigma(t-t')P\tilde{\rho}(t') \\ \Sigma(t) &= -iPLQe^{-iQLt}QLP \end{aligned} \quad (4.21)$$

where $\Sigma(t)$ is the self-energy superoperator, $Q = 1 - P$ is the complement of P and $L = L_0 + L_V$ is the full Louvillian superoperator, where L_α ($\alpha = 0, V, S, \text{laser}, dp, R, P$) is defined as $L_\alpha \mathcal{O} = [\tilde{H}_\alpha, \mathcal{O}]$.

If we assume the radiation modes are initially described by a vacuum:

$$\rho_R(0) = \prod_k |0_k\rangle\langle 0_k| \quad (4.22)$$

and the phonon modes density matrix is assumed to describe thermal equilibrium

$$\rho_P(0) = \frac{e^{-\beta H_P}}{\text{Tr}\{e^{-\beta H_P}\}} \quad (4.23)$$

where $\beta = \frac{1}{k_B T}$, P then obeys the following important identities

$$PLP = L_S P + L_{\text{laser}} P \quad (4.24)$$

$$PL_V P = 0. \quad (4.25)$$

Applying the above identities and performing the traces on Eq. (4.21) over radiation modes and phonon modes, we obtain

$$\dot{\tilde{\rho}}_S(t) = -iL'_0 \tilde{\rho}_S(t) - i \int_0^t dt' \Sigma_S(t-t') \tilde{\rho}_S(t') \quad (4.26)$$

$$\Sigma_S(t) = -i \text{Tr}_R \text{Tr}_P \left\{ (L_{dp} + L_V) e^{-iQLt} (L_{dp} + L_V) \rho_R(0) \rho_P(0) \right\} \quad (4.27)$$

$L'_0 = L_S + L_{\text{laser}}$ and $\Sigma_S(t)$ is the reduced self-energy superoperator.

The self-energy superoperator in Laplace space

$$\Sigma_S(s) = -i \text{Tr}_R \text{Tr}_P \left\{ (L_{dp} + L_V) \frac{1}{s + iQL} (L_{dp} + L_V) \rho_R(0) \rho_P(0) \right\} \quad (4.28)$$

can be expanded in powers of the perturbation Louivillian L_V by iterating the Dyson identity,

$$\frac{1}{s + iQL} = \sum_k \frac{1}{s + iQL_0} \left(-iQL_V \frac{1}{s + iQL_0} \right)^k. \quad (4.29)$$

In Born approximation, the reduced self-energy superoperator is replaced by the form obtained from the lowest-order self-energy, i.e., $\Sigma_S(s) \rightarrow \Sigma_S^{(2)}(s)$, where the superscript indicates the number of occurrences of L_V , but we include all orders in the dephasing part, L_{dp} . Therefore, at lowest order in L_V but keeping the dephasing Louivillian L_{dp} exactly, the reduced self-energy takes the form

$$\Sigma_S^{(2)}(s) = -i \text{Tr}_R \text{Tr}_P \left\{ (L_{dp} + L_V) \frac{1}{s + iL_0} (L_{dp} + L_V) \rho_R(0) \rho_P(0) \right\} \quad (4.30)$$

In order to find the elements of the reduced density matrix of the system we need to find the matrices for the superoperators $\frac{1}{s+iL_0}$ and L_{dp} , L_V .

We recall the GME in Eq. (4.26)

$$\dot{\tilde{\rho}}_S(t) = -iL'_0\tilde{\rho}_S(t) - i \int_0^t dt' \Sigma_S^{(2)}(t-t')\tilde{\rho}_S(t') \quad (4.31)$$

ignoring the coupling to the environment for the moment and omitting the second term on the right hand side of the above equation, we write

$$\dot{\tilde{\rho}}_S(t) = -iL'_0\tilde{\rho}_S(t). \quad (4.32)$$

The matrix elements of L'_0 are defined by

$$[L'_0]_{\alpha\beta,\gamma\delta} = \text{Tr}\{|\delta\rangle\langle\gamma|L'_0|\alpha\rangle\langle\beta|\} \quad (4.33)$$

where $\alpha, \beta, \gamma \in \{a, b, c\}$

$$[L'_0] = \begin{matrix} & \begin{matrix} aa & bb & cc & ab & ba & ac & ca & bc & ca \end{matrix} \\ \begin{matrix} aa \\ bb \\ cc \\ ab \\ ba \\ ac \\ ca \\ bc \\ cb \end{matrix} & \left(\begin{array}{ccccccccc} 0 & 0 & 0 & \frac{\Omega_1}{2} & -\frac{\Omega_1}{2} & \frac{\Omega_2}{2} & -\frac{\Omega_2}{2} & 0 & 0 \\ 0 & 0 & 0 & -\frac{\Omega_1}{2} & \frac{\Omega_1}{2} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -\frac{\Omega_2}{2} & \frac{\Omega_2}{2} & 0 & 0 \\ \frac{\Omega_1}{2} & -\frac{\Omega_1}{2} & 0 & \frac{\Delta_1+\Delta_2+\delta}{2} & 0 & 0 & 0 & 0 & -\frac{\Omega_2}{2} \\ -\frac{\Omega_1}{2} & \frac{\Omega_1}{2} & 0 & 0 & \frac{-(\Delta_1+\Delta_2+\delta)}{2} & 0 & 0 & \frac{\Omega_2}{2} & 0 \\ \frac{\Omega_2}{2} & 0 & -\frac{\Omega_2}{2} & 0 & 0 & \frac{\Delta_1+\Delta_2-\delta}{2} & 0 & -\frac{\Omega_1}{2} & 0 \\ -\frac{\Omega_2}{2} & 0 & \frac{\Omega_2}{2} & 0 & 0 & 0 & \frac{-(\Delta_1+\Delta_2)+\delta}{2} & 0 & \frac{\Omega_1}{2} \\ 0 & 0 & 0 & 0 & \frac{\Omega_2}{2} & -\frac{\Omega_1}{2} & 0 & \delta & 0 \\ 0 & 0 & 0 & -\frac{\Omega_2}{2} & 0 & 0 & \frac{\Omega_1}{2} & 0 & -\delta \end{array} \right) \end{matrix}$$

The equations-of-motion for the diagonal elements of the reduced density matrix (the populations) take the form

$$\dot{\tilde{\rho}}_{aa}(t) = -i\frac{\Omega_1}{2}(\tilde{\rho}_{ab}(t) - \tilde{\rho}_{ba}(t)) + i\frac{\Omega_2}{2}(\tilde{\rho}_{ac}(t) - \tilde{\rho}_{ca}(t)) \quad (4.34)$$

$$\dot{\tilde{\rho}}_{bb}(t) = i\frac{\Omega_1}{2}(\tilde{\rho}_{ab}(t) - \tilde{\rho}_{ba}(t)) \quad (4.35)$$

$$\dot{\tilde{\rho}}_{cc}(t) = -i\frac{\Omega_2}{2}(\tilde{\rho}_{ac}(t) - \tilde{\rho}_{ca}(t)) \quad (4.36)$$

and the coherences are given by

$$\dot{\tilde{\rho}}_{ab}(t) = -i \left[\frac{1}{2}(\Delta_1 + \Delta_2) + \frac{1}{2}\delta \right] \tilde{\rho}_{ab}(t) - i \frac{\Omega_1}{2} \left(\tilde{\rho}_{aa}(t) - \tilde{\rho}_{bb}(t) \right) + i \frac{\Omega_2}{2} \tilde{\rho}_{cb}(t) \quad (4.37)$$

$$\dot{\tilde{\rho}}_{cb}(t) = -i\delta \tilde{\rho}_{cb}(t) + i \frac{\Omega_2}{2} \tilde{\rho}_{ab}(t) - i \frac{\Omega_1}{2} \tilde{\rho}_{ca}(t) \quad (4.38)$$

$$\dot{\tilde{\rho}}_{ac}(t) = -i \left[\frac{1}{2}(\Delta_1 + \Delta_2) - \frac{1}{2}\delta \right] \tilde{\rho}_{ac}(t) - i \frac{\Omega_2}{2} \left(\tilde{\rho}_{aa}(t) - \tilde{\rho}_{cc}(t) \right) + i \frac{\Omega_1}{2} \tilde{\rho}_{bc}(t). \quad (4.39)$$

We assume that the atom is initially in the ground state $|b\rangle$ which gives the initial condition

$$\tilde{\rho}_{bb}(0) = 1, \tilde{\rho}_{aa}(0) = \tilde{\rho}_{cc}(0) = \tilde{\rho}_{ac}(0) = 0. \quad (4.40)$$

The levels $|a\rangle$ and $|b\rangle$ are coupled to a weak probe laser field of amplitude Ω_1 , therefore we are interested in the absorption between levels $|a\rangle$ and $|b\rangle$ which is given in terms of the coherence $\tilde{\rho}_{ab}(t)$ [3] and want to find the coherence to the first order in Ω_1 . We solve for $\tilde{\rho}_{ab}(t)$ and $\tilde{\rho}_{cb}(t)$ by Laplace transform to the leading order in Ω_1 , we obtain the coupled set of equations

$$\left[s + i \left(\frac{1}{2}(\Delta_1 + \Delta_2) + \frac{1}{2}\delta \right) \right] \tilde{\rho}_{ab}(s) = \tilde{\rho}_{ab}(0) + i \frac{\Omega_1}{2s} + i \frac{\Omega_2}{2} \tilde{\rho}_{cb}(s) \quad (4.41)$$

$$(s + i\delta) \tilde{\rho}_{cb}(s) = \tilde{\rho}_{cb}(0) + i \frac{\Omega_2}{2} \tilde{\rho}_{ab}(s). \quad (4.42)$$

The decay rates of the off-diagonal density matrix elements, $\tilde{\rho}_{ab}(s)$ and $\tilde{\rho}_{cb}(s)$, will be included in the above expressions in terms of reduced self-energy matrix elements from the second term on the right hand side of the Eq. (4.31).

We recall the expression for the reduced self-energy superoperator $\Sigma_S^{(2)}(s)$ given in Eq. (4.30) and write it in terms of pure-dephasing and transition parts as

$$\Sigma_S^{(2)}(s) = \Sigma_S^{ph}(s) + \Sigma_S^{rad}(s) \quad (4.43)$$

where $\Sigma_S^{ph}(s)$ gives rise to the pure-dephasing and $\Sigma_S^{rad}(s)$ gives rise to system-phonon and system-photon couplings, respectively, and are given by the expressions

$$\begin{aligned}\Sigma_S^{ph}(s) &= -i\text{Tr}_R \text{Tr}_P \left\{ L_{dp} \frac{1}{s + iL_0} L_{dp} \rho_R(0) \rho_P(0) \right\} \\ \Sigma_S^{rad}(s) &= -i\text{Tr}_R \text{Tr}_P \left\{ L_V \frac{1}{s + iL_0} L_V \rho_R(0) \rho_P(0) \right\}.\end{aligned}\quad (4.44)$$

Because L_{dp} acts only on the operators in the phonon mode while L_V acts in the operators acting in the radiation modes, the cross terms in Eq. (4.30) will not contribute to the final trace in the expression for the reduced self-energy. To the leading order in the probe amplitude Ω_1 , evolution under L'_0 is closed in the space of $\tilde{\rho}_{ab}(s)$ and $\tilde{\rho}_{cb}(s)$. We therefore focus on evolution in this subspace accounting for pure-dephasing through L_{dP} . We want to find the matrices for L_{dp} and $\frac{1}{s+iL_0}$ in the basis of outer products $\{|a\rangle\langle b|, |c\rangle\langle b|\}$. The matrix for L_{dP}

$$[L_{dp}] = \begin{array}{cc} & \begin{matrix} ab & cb \end{matrix} \\ \begin{matrix} ab \\ cb \end{matrix} & \begin{pmatrix} 0 & 0 \\ 0 & X_P^l \end{pmatrix} \end{array}.$$

where the superoperator for left-multiplication, X_P^l , is defined as

$$X_P^l \mathcal{O}_P = \sum_q \lambda_q (b_q + b_q^\dagger) \mathcal{O}_P \quad (4.45)$$

and $\mathcal{O}_P$ is an operator acting on the phonon modes. The form of the phonon coupling is such that the only non-zero matrix element in the present subspace $\{|a\rangle\langle b|, |c\rangle\langle b|\}$ arising from the phonons is $\Sigma_{cb,cb}^{(2)}(s)$, which can be directly added to the equation of motion in Eq. (4.42). The radiation modes are assumed to be fully Markovian and can thus be replaced by effective rates. Relevant matrix elements of the reduced self-energy matrix element due to phonons are expressed as

$$\Sigma_{cb,cb}^{(2)}(s) = -i\text{Tr}_R \text{Tr}_P \left\{ X_P^l \left[\frac{1}{s + iL_0} \right]_{cb,cb} X_P^l \rho_R(0) \rho_P(0) \right\} \quad (4.46)$$

The above reduced self-energy matrix element will be responsible for the non-Markovian behaviour in the CPT absorption spectrum. We solve the set of equations given by

equations (4.41) and (4.42) by writing them in terms of a 2x2 matrix.

$$M(s)\tilde{\rho}_{coh}(s) = \tilde{\rho}_{coh}(0) + A \quad (4.47)$$

where

$$M(s) = \begin{pmatrix} s + i(\frac{1}{2}(\Delta_1 + \Delta_2) + \frac{1}{2}\delta) + \gamma_{ab} & i\frac{\Omega_2}{2} \\ i\frac{\Omega_2}{2} & s + i\delta + \gamma_{cb} + i\Sigma_{cb,cb}^{(2)}(s) \end{pmatrix}, \quad (4.48)$$

$$\tilde{\rho}_{coh}(s) = \begin{pmatrix} \tilde{\rho}_{ab}(s) \\ \tilde{\rho}_{cb}(s) \end{pmatrix}, \quad (4.49)$$

and

$$A = \begin{pmatrix} \frac{-i\Omega_1}{2s} \\ 0 \end{pmatrix}, \quad (4.50)$$

and we have used that $\Sigma_{ab,cb}^{(2)}(s) = \Sigma_{cb,ab}^{(2)}(s) = 0$ in Eq. (4.48) and the radiative decay rates for the off-diagonal density matrix elements $\tilde{\rho}_{ab}(s)$ and $\tilde{\rho}_{cb}(s)$ are given by γ_{ab} and γ_{cb} , respectively.

In order to clarify the CPT absorption we put $\Sigma_{cb,cb}^{(2)}(s) = 0$ for the moment, we solve for $\tilde{\rho}_{ab}(s)$ to obtain

$$\tilde{\rho}_{ab}(s) = \frac{1}{\left(s + i[\frac{1}{2}(\Delta_1 + \Delta_2) + \frac{1}{2}\delta] + \gamma_{ab}\right)\left(s + i\delta + \gamma_{cb}\right) + \frac{\Omega_2^2}{4}} \times \left[i\frac{\Omega_1(s + i\delta + \gamma_{cb})}{2s} + (s + i\delta + \gamma_{cb})\tilde{\rho}_{ab}(0) - i\frac{\Omega}{2}\tilde{\rho}_{cb}(0) \right], \quad (4.51)$$

We are interested in the absorption between the levels $|a\rangle$ and $|b\rangle$ which are coupled by a weak probe field of amplitude Ω_1 . Following the discussion in Ref. [3], the absorption is given in the terms of the imaginary part of the complex susceptibility χ'' , in the long time limit as

$$\chi'' \propto \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T dt \text{Im}[\tilde{\rho}_{ab}(t)] = \lim_{s \rightarrow 0} \text{Im}[s\tilde{\rho}_{ab}(s)], \quad (4.52)$$

The imaginary part of the complex susceptibility can then be written as

$$\chi'' \propto \text{Im} \left[\frac{\frac{\Omega_1}{2}(i\delta + \gamma_{cb})}{\left(i[\frac{1}{2}(\Delta_1 + \Delta_2) + \frac{1}{2}\delta] + \gamma_{ab}\right)(i\delta + \gamma_{cb}) + \frac{\Omega_2^2}{4}} \right] \quad (4.53)$$

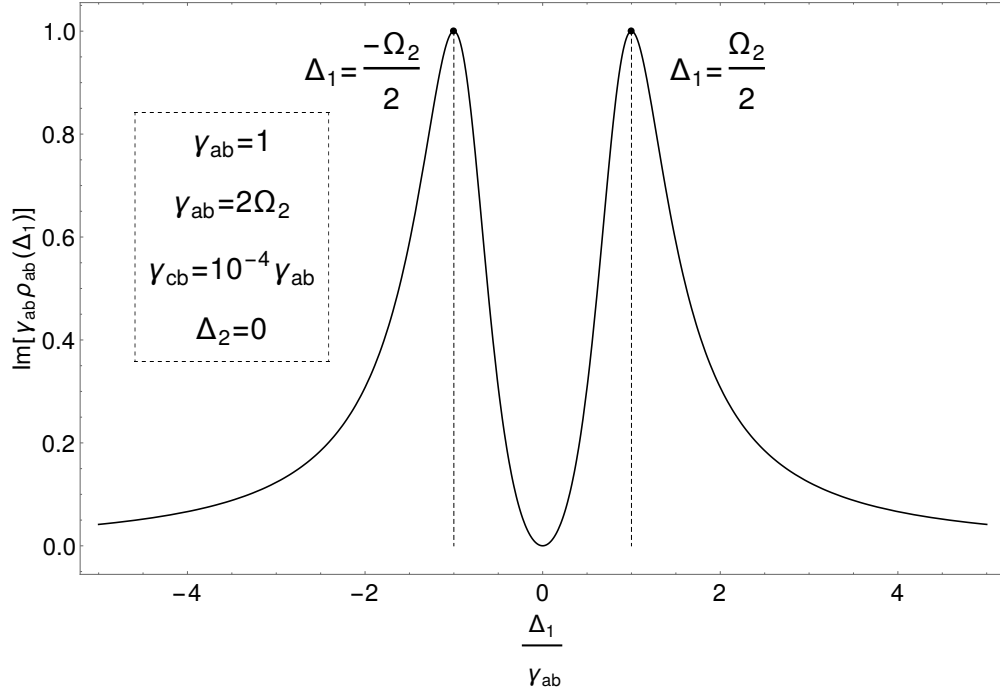


Figure 4.2: Imaginary part of the complex susceptibility versus probe detuning Δ_1 given by Eq. (4.54) in the units of γ_{ab} . The dip in the absorption spectrum results from coherent population trapping.

The imaginary part of the complex susceptibility in Eq. (4.52) can then be expressed in the terms of the probe detuning, for $\Delta_2 = 0$, as

$$\chi'' \propto \frac{\Omega_1}{2} \left[\frac{\Delta_1^2(\gamma_{ab} + \gamma_{cb}) - \gamma_{cb}(\Delta_1^2 - \gamma_{ab}\gamma_{cb} - \frac{\Omega_2^2}{4})}{(\Delta_1^2 - \gamma_{ab}\gamma_{cb} - \frac{\Omega_2^2}{4})^2 + \Delta_1^2(\gamma_{ab} + \gamma_{cb})^2} \right]. \quad (4.54)$$

The above equations describe an absorption peak of width γ_{ab} along with a dip at $\Delta_1 = 0$ due to the coherent superposition of the ground states and the width of the dip is determined by Ω_2 [3]. It can be noted that at $\Delta_1 = 0$ the absorption is proportional

to γ_{cb} where γ_{cb} is the decay rate between the levels which we have assumed dipole forbidden and is small compared to the decay rate γ_{ab} between $|a\rangle$ and $|b\rangle$.

It can be seen from the Eq. (4.54) that, for $(\gamma_{ab}\gamma_{cb}) \ll \Omega_2^2$, the coherence $\rho_{ab}(\Delta_1)$ has a maxima at points satisfying $\Delta_1^2 = \frac{\Omega_2^2}{4}$. We expect that at these points $\Delta_1^2 = \frac{\Omega_2^2}{4}$ the dephasing reduced self-energy matrix element $\Sigma_{cb,cb}^{(2)}(s)$

$$\Sigma_{cb,cb}^{(2)}(s) = \text{Tr}_R \text{Tr}_P \left\{ X_P^l \left[\frac{1}{s + iL_0} \right]_{cb,cb} X_P^l \rho_R(0) \rho_P(0) \right\} \quad (4.55)$$

will affect the absorption spectrum significantly and we expect non-Markovian effects to manifest themselves around these points in the spectrum. Furthermore, in order to see a significant non-Markovian effect in the CPT absorption the dephasing self-energy should be large or comparable to the radiative decay rate γ_{ab} , i.e. $\gamma_{cb} \ll |\Sigma_{cb,cb}^{(2)}(s)| \sim \gamma_{ab}$. The dephasing reduced self-energy matrix element $\Sigma_{cb,cb}^{(2)}(s)$ in Eq. (4.55) can be calculated exactly using the equation-of-motion method discussed in the chapter 3 and Appendix A.4.2 and remains a work in progress.

Chapter 5

Summary

The main goal of this thesis is to study decoherence in the open quantum systems undergoing non-Markovian dynamics. Decoherence in semiconductor quantum dots due to coupling to the environment degrees of freedom has been of prime interest in the field of quantum information technology [22, 23, 24]. In quantum information processing, information is stored and processed in quantum bits (qubits) where a single qubit can be represented as a two-level system with two energy levels which can be used to store and process the information [25]. In order to store and process data in the form qubits it is required to create long-lived quantum states and it is necessary to understand decoherence process of these states due to coupling to its environment.

The thesis starts with discussing the dynamics of a two-level system in chapter 1. We discuss the Rabi problem for a two-level atom interacting with a single mode of a classical monochromatic laser field. We further discuss the spontaneous emission mechanism in a cavity-coupled two-level system using Wigner-Weiskoff approximation. In following sections we discuss the coherent population trapping a so-called lambda system. We discuss the formation of dark state as a superposition of the two ground states in the driven lambda system using the pump-probe technique. In the end of chapter 1, we discuss the pure dephasing mechanism which gives rise to the decay of the off-diagonal elements of the reduced density matrix which reflects the loss of quantum coherence, i.e. dephasing.

In chapter 2, we derive a general formula to study the fluorescence spectrum of a multi-level system coupled to radiation modes of the electromagnetic field. We apply

linear response theory to describe the well known gedanken spectrum analyzer [3, 12] model which theoretically describes the dynamics of a detector monitoring the fluorescence spectrum. As an example, we use this general formula to recover the well known Mollow-triplet spectrum of a two-level system and its properties for a Markovian interaction with the radiation modes of the cavity.

In chapter 3, we focus on the pure dephasing mechanism in a cavity-QED two-level system. We discuss the phonon induced non-Markovian pure dephasing in a cavity-coupled two-level system. We study the effect of phonon-induced pure dephasing on one of the side-peaks of the Mollow-triplet spectrum. We observe a distinct behaviour in the side-peak spectrum due to the non-Markovian effect from coupling to the phonon reservoir, that leads to a splitting of the side-peak.

In the final chapter 4, we discuss the coherent population trapping in a lambda system. We discuss non-Markovian decoherence in the coherent population trapping absorption spectrum due to phonon coupling. We expect to see non-Markovian behaviour in the CPT absorption spectrum due to phonon-induced decoherence which will change the form of the absorption spectrum.

Appendix A

Self-energy superoperator and its matrix elements

In this section, we will calculate the reduced self-energy superoperator matrix elements for both system-photon and system-phonon couplings. The system-photon coupling reduced self-energy matrix elements will be calculated within Born approximation by expanding them in the powers of L_X^+/L_X while the system-phonon coupling reduced self-energy matrix elements will be calculated exactly.

A.1 Self-energy expansion

Reduced self-energy matrix in equation given in (2.118) reads

$$\Sigma_S(-i\Delta_0) = -iTr_E \left(PL_x + PL_{SR} + PL_y + PL_{SP} \right) \frac{1}{-i\Delta_0 + iQL} \left(L_x P + L_{SR} P + L_y P + L_{SP} P \right) \rho_E(0) \quad (\text{A.1})$$

which can be expanded in powers of perturbation $L_V = L_{SR} + L_{SP}$

$$\frac{1}{-i\Delta_0 + iQL} = \sum_k \frac{1}{-i\Delta_0 + iQL_0} \left(-iQL_V \frac{1}{-i\Delta_0 + iQL_0} \right)^k, \quad (\text{A.2})$$

since only even powers of L_V contribute in the final trace on variables of radiation/phonon modes, we have

$$\frac{1}{-i\Delta_0 + iQL} = \sum_k^{\infty} \frac{1}{-i\Delta_0 + iQL_0} \left(-iQL_V \frac{1}{-i\Delta_0 + iQL_0} \right)^{2k}. \quad (\text{A.3})$$

In Born approximation, the reduced self-energy is replaced by the forms obtained from the lowest order self-energy, i.e., $\Sigma_S(-i\Delta_0) \rightarrow \Sigma_S^{(2)}(-i\Delta_0)$, where the superscript indicates the number of occurrences of L_V . Therefore, in the lowest order reduced self-energy takes the form

$$\Sigma_S^{(2)}(-i\Delta_0) = -iTr_E \left(L_x + L_{SR} + L_y + L_{SP} \right) \frac{1}{-i\Delta_0 + iL_0} \left(L_x + L_{SR} + L_y + L_{SP} \right) \rho_R(0). \quad (\text{A.4})$$

A.2 Matrix elements of superoperators

Recalling:

$$\begin{aligned} H'_S &= \left(\frac{\Omega_R - \Delta_P}{2} \right) \sigma_3 \\ L'_S \mathcal{O} &= [H'_S, \mathcal{O}] \\ [L'_S]_{\alpha\beta, \gamma\delta} &= Tr\{ |\beta\rangle \langle \alpha| L'_S |\gamma\rangle \langle \delta| \}. \end{aligned} \quad (\text{A.5})$$

We find the matrix elements for L'_S

$$L'_S |+\rangle \langle +| = \left[\left(\frac{\Omega_R - \Delta_P}{2} \right) \sigma_3, |+\rangle \langle +| \right] \quad (\text{A.6})$$

$$L'_S |+\rangle \langle +| = 0. \quad (\text{A.7})$$

Similarly,

$$L'_S |-\rangle \langle -| = 0. \quad (\text{A.8})$$

$$L'_S |+\rangle \langle -| = |+\rangle \langle -| (\Omega_R - \Delta_P). \quad (\text{A.9})$$

$$L'_S|-\rangle\langle+| = -|-\rangle\langle+|(\Omega_R - \Delta_P). \quad (\text{A.10})$$

Thus, the matrix for L'_S , in $\{+, -\}$ basis, takes the form

$$[L'_S] = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & (\Omega_R - \Delta_P) & 0 \\ 0 & 0 & 0 & -(\Omega_R - \Delta_P) \end{pmatrix}. \quad (\text{A.11})$$

In same manner, recalling:

$$H'_x = \sum_k \tilde{g}_k x_k \sigma_3$$

$$L'_x \mathcal{O} = [H'_x, \mathcal{O}]. \quad (\text{A.12})$$

$$L'_x|+\rangle\langle+|\mathcal{O}_R = [\sum_k \tilde{g}_k x_k \sigma_3, |+\rangle\langle+|\mathcal{O}_R], \quad (\text{A.13})$$

$$L'_x|+\rangle\langle+|\mathcal{O}_R = |+\rangle\langle+|L_X \mathcal{O}_R, \quad (\text{A.14})$$

where

$$L_X \mathcal{O}_R = \sum_k \tilde{g}_k [x_k, \mathcal{O}_R]. \quad (\text{A.15})$$

Similarly,

$$L'_x|-\rangle\langle-|\mathcal{O}_R = -|-\rangle\langle-|L_X \mathcal{O}_R. \quad (\text{A.16})$$

$$L'_x|+\rangle\langle-|\mathcal{O}_R = |+\rangle\langle-|L_X^+ \mathcal{O}_R. \quad (\text{A.17})$$

where

$$L_X^+ \mathcal{O}_R = \sum_k \tilde{g}_k \{x_k, \mathcal{O}_R\}. \quad (\text{A.18})$$

$$L'_x|-\rangle\langle+|\mathcal{O}_R = -|-\rangle\langle+|L_X^+ \mathcal{O}_R. \quad (\text{A.19})$$

Thus, the matrix for L'_x , in $\{+, -\}$ basis, takes the form

$$[L'_x] = \begin{pmatrix} L_X & 0 & 0 & 0 \\ 0 & -L_X & 0 & 0 \\ 0 & 0 & L_X^+ & 0 \\ 0 & 0 & 0 & -L_X^+ \end{pmatrix}. \quad (\text{A.20})$$

Recalling H'_{SR} :

$$\begin{aligned} H'_{SR} &= |-\rangle\langle -| + X^\dagger |-\rangle\langle +| \\ X &= \sum_k g_k \left(\cos^2 \theta a_k - \sin^2 \theta a_k^\dagger \right) \\ X^\dagger &= \sum_k g_k \left(\cos^2 \theta a_k^\dagger - \sin^2 \theta a_k \right) \\ L'_{SR} \mathcal{O} &= [H'_{SR}, \mathcal{O}]. \end{aligned} \quad (\text{A.21})$$

$$\begin{aligned} L'_{SR} |+\rangle\langle +| \mathcal{O}_R &= [X |+\rangle\langle -| + X^\dagger |+\rangle\langle -|, |+\rangle\langle +| \mathcal{O}_R] \\ &= -| \mathcal{O}_R X + |+\rangle\langle -| X^\dagger \mathcal{O}_R \\ &= -X_R \mathcal{O}_R |+\rangle\langle -| + X_L^\dagger \mathcal{O}_R |-\rangle\langle +|. \end{aligned} \quad (\text{A.22})$$

we introduce the superoperator for right and left multiplication

$$X_R \mathcal{O}_R = \mathcal{O}_R \sum_k g_k (\cos^2 \theta a_k - \sin^2 \theta a_k^\dagger) \quad (\text{A.23})$$

$$X_L^\dagger \mathcal{O}_R = \sum_k g_k (\cos^2 \theta a_k^\dagger - \sin^2 \theta a_k) \mathcal{O}_R. \quad (\text{A.24})$$

$$\begin{aligned} L'_{SR} |-\rangle\langle -| \mathcal{O}_R &= [X |+\rangle\langle -| + X^\dagger |-\rangle\langle +|, |-\rangle\langle -| \mathcal{O}_R] \\ &= |+\rangle\langle -| X \mathcal{O}_R - |-\rangle\langle +| \mathcal{O}_R X^\dagger \\ &= X_L \mathcal{O}_R |+\rangle\langle -| - X_R^\dagger \mathcal{O}_R |-\rangle\langle +|. \end{aligned} \quad (\text{A.25})$$

$$X_L \mathcal{O}_R = \sum_k g_k (\cos^2 \theta a_k - \sin^2 \theta a_k^\dagger) \mathcal{O}_R \quad (\text{A.26})$$

$$X_R^\dagger \mathcal{O}_R = \mathcal{O}_R \sum_k g_k (\cos^2 \theta a_k^\dagger - \sin^2 \theta a_k). \quad (\text{A.27})$$

$$\begin{aligned} L'_{SR} |+\rangle \langle -| \mathcal{O}_R &= [X|+\rangle \langle -| + X^\dagger |-\rangle \langle +|, |+\rangle \langle -| \mathcal{O}_R] \\ &= |-\rangle \langle -| X^\dagger \mathcal{O}_R - |+\rangle \langle +| \mathcal{O}_R X^\dagger \\ &= X_L^\dagger \mathcal{O}_R |-\rangle \langle -| - X_R^\dagger \mathcal{O}_R |+\rangle \langle +|. \end{aligned} \quad (\text{A.28})$$

$$\begin{aligned} L'_{SR} |-\rangle \langle +| \mathcal{O}_R &= [X|+\rangle \langle -| + X^\dagger |-\rangle \langle +|, |-\rangle \langle +| \mathcal{O}_R] \\ &= X \mathcal{O}_R - |-\rangle \langle -| \mathcal{O}_R X \\ &= X_L \mathcal{O}_R |+\rangle \langle +| - X_R \mathcal{O}_R |-\rangle \langle -|. \end{aligned} \quad (\text{A.29})$$

Matrix for L'_{SR} in $\{+, -\}$ basis:

$$[L'_{SR}] = \begin{pmatrix} 0 & 0 & -X_R^\dagger & X_L \\ 0 & 0 & X_L^\dagger & -X_R \\ -X_R & X_L & 0 & 0 \\ X_L^\dagger & -X_R^\dagger & 0 & 0 \end{pmatrix}. \quad (\text{A.30})$$

A.3 Matrix elements of reduced self-energy for photon coupling

Recalling:

$$\begin{aligned}
H'_0 &= H'_S + H'_x + H'_R \\
H'_S &= \frac{\Omega_R}{2} \sigma_3 \\
H'_x &= \sum_k \tilde{g}_k x_k \sigma_3 \\
H'_R &= \sum_k \Delta_k a_k^\dagger a_k \\
L'_0 \mathcal{O} &= (L'_S + L'_x + L'_R) \mathcal{O} \\
&= [H'_S + H'_x + H'_R, \mathcal{O}] \\
&= [H'_0, \mathcal{O}].
\end{aligned} \tag{A.31}$$

$$L'_0 |+\rangle \langle +| \mathcal{O}_R = |+\rangle \langle +| (L'_R + L_X) \mathcal{O}_R. \tag{A.32}$$

Similarly,

$$L'_0 |-\rangle \langle -| \mathcal{O}_R = |-\rangle \langle -| (L'_R - L_X) \mathcal{O}_R. \tag{A.33}$$

$$L'_0 |+\rangle \langle -| \mathcal{O}_R = |+\rangle \langle -| (L'_R \mathcal{O}_R + \Omega_R + L_X^+) \mathcal{O}_R. \tag{A.34}$$

$$L'_0 |-\rangle \langle +| \mathcal{O}_R = |-\rangle \langle +| (L'_R \mathcal{O}_R - \Omega_R - L_X^+) \mathcal{O}_R. \tag{A.35}$$

Thus, the matrix for $\frac{1}{s+iL'_0}$, in $\{+, -\}$ basis, takes the form

$$\left[\frac{1}{s + iL'_0} \right] = \begin{pmatrix} \frac{1}{s+i(L'_R+L_X)} & 0 & 0 & 0 \\ 0 & \frac{1}{s+i(L'_R-L_X)} & 0 & 0 \\ 0 & 0 & \frac{1}{s+i(L'_R+\Omega_R+L_X^+)} & 0 \\ 0 & 0 & 0 & \frac{1}{s+i(L'_R-\Omega_R-L_X^+)} \end{pmatrix}. \tag{A.36}$$

Matrix for free propagator $\left[\frac{1}{s+iL'_0}\right]$ is diagonal in dressed state basis and is given in terms of 2×2 blocks by

$$\left[\frac{1}{s+iL'_0}\right] = \begin{pmatrix} G & 0 \\ 0 & G' \end{pmatrix} \quad (\text{A.37})$$

where

$$G = \begin{pmatrix} \frac{1}{s+i(L'_R+L_X)} & 0 \\ 0 & \frac{1}{s+i(L'_R-L_X)} \end{pmatrix} \quad (\text{A.38})$$

$$G' = \begin{pmatrix} \frac{1}{s+i(L'_R+\Omega_R+L_X^+)} & 0 \\ 0 & \frac{1}{s+i(L'_R-\Omega_R-L_X^+)} \end{pmatrix} \quad (\text{A.39})$$

And for L'_{SR}

$$[L'_{SR}] = \begin{pmatrix} 0 & V \\ V' & 0 \end{pmatrix} \quad (\text{A.40})$$

where

$$V = \begin{pmatrix} -X_R^\dagger & X_L \\ X_L^\dagger & -X_R \end{pmatrix} \quad (\text{A.41})$$

$$V' = \begin{pmatrix} -X_R & X_L \\ X_L^\dagger & -X_R^\dagger \end{pmatrix} \quad (\text{A.42})$$

Also,

$$[L'_x] = \begin{pmatrix} Y & 0 \\ 0 & Y' \end{pmatrix} \quad (\text{A.43})$$

$$Y = \begin{pmatrix} L_X & 0 \\ 0 & -L_X \end{pmatrix} \quad (\text{A.44})$$

$$Y' = \begin{pmatrix} L_X^+ & 0 \\ 0 & -L_X^+ \end{pmatrix} \quad (\text{A.45})$$

The reduced self-energy can be written in two parts as

$$\Sigma_S^{(2)}(s) = \Sigma_S^x(s) + \Sigma_S^{SR}(s), \quad (\text{A.46})$$

where

$$\Sigma_S^x(s) = -i\text{Tr}_R \begin{pmatrix} YGY & 0 \\ 0 & Y'G'Y' \end{pmatrix} \rho_R(0) \quad (\text{A.47})$$

$$\Sigma_S^{SR}(s) = -i\text{Tr}_R \begin{pmatrix} VG'V' & 0 \\ 0 & V'GV \end{pmatrix} \rho_R(0). \quad (\text{A.48})$$

The matrix elements we want to calculate in order to compute the fluorescence spectra, the transition reduced self-energy matrix elements

$$\Sigma_{++,+}(s) = -iT r_R \left[X_R^\dagger \frac{1}{s + i(L'_R + \Omega_R + L_X^+)} X_R \rho_R(0) + X_L \frac{1}{s + i(L'_R - \Omega_R - L_X^+)} X_L^\dagger \rho_R(0) \right] \quad (\text{A.49})$$

$$\Sigma_{+,-}(s) = iT r_R \left[X_R^\dagger \frac{1}{s + i(L'_R + \Omega_R + L_X^+)} X_L \rho_R(0) + X_L \frac{1}{s + i(L'_R - \Omega_R - L_X^+)} X_R^\dagger \rho_R(0) \right] \quad (\text{A.50})$$

$$\Sigma_{+-}(s) = -iT r_R \left[X_R \frac{1}{s + i(L'_R + L_X)} X_R^\dagger \rho_R(0) + X_L \frac{1}{s + i(L'_R - L_X)} X_L^\dagger \rho_R(0) \right]. \quad (\text{A.51})$$

and dephasing reduced self-energy matrix elements are given by

$$\Sigma_{++}^x(s) = -iT r_R \left[L_X \frac{1}{s + i(L'_R + L_X)} L_X \rho_R(0) \right] \quad (\text{A.52})$$

$$\Sigma_{--}^x(s) = -iT r_R \left[L_X \frac{1}{s + i(L'_R - L_X)} L_X \rho_R(0) \right] \quad (\text{A.53})$$

$$\Sigma_{+-}^x(s) = -iT r_R \left[L_X^+ \frac{1}{s + i(L'_R + \Omega_R + L_X^+)} L_X^+ \rho_R(0) \right]. \quad (\text{A.54})$$

We will calculate the matrix elements of reduced self-energy for our two level system kept within a cavity. In order to calculate the matrix elements of reduced self-energy, we have to perform sums appearing in above equations. We apply the continuum of modes and for the case of two-level system kept inside a cavity, we replace sum by integration $\sum_k g_k^2 \rightarrow \int d\epsilon D(\epsilon) |g(\epsilon)|^2$ where $D(\epsilon)$ is the photonic density states of the cavity. The standard model for the photonic DOS is given by a Lorentzian

$$D(\epsilon) |g(\epsilon)|^2 = \frac{1}{\pi} \frac{g^2 \Gamma_c}{(\epsilon - \Delta_c)^2 + \Gamma_c^2}, \quad (\text{A.55})$$

where $\Delta_c = \omega_c - \omega$ is the detuning of cavity central frequency (ω_c) from laser frequency (ω) and quantity Γ_c is half of the cavity bandwidth.

A.3.1 Transition self-energy

We want to calculate the reduced self-energy matrix elements due to system-photon coupling in the Born approximation after expanding then in powers of L_X^+/L_X . As discussed in previous section, we apply the continuum of modes to replace sum by integration $\sum_k g_k^2 \rightarrow \int d\epsilon D(\epsilon) |g(\epsilon)|^2$ and using Lorentzian DOS, we solve for reduced self-energy matrix elements appearing in Eqs. (A.49)-(A.51), using contour integration method to obtain

$$\Sigma_{++,,++}^{SR}(s) = -ig^2 \cos^4 \theta \left[\frac{1}{s + i(\Omega_R - \Delta_c) + \Gamma_c} + \frac{1}{s - i(\Omega_R - \Delta_c) + \Gamma_c} \right] \quad (\text{A.56})$$

$$\Sigma_{++,,--}^{SR}(s) = ig^2 \sin^4 \theta \left[\frac{1}{s + i(\Omega_R + \Delta_c) + \Gamma_c} + \frac{1}{s - i(\Omega_R + \Delta_c) + \Gamma_c} \right] \quad (\text{A.57})$$

$$\Sigma_{+-,,+-}^{SR}(s) = -ig^2 \left[\frac{\cos^4 \theta}{s - i\Delta_c + \Gamma_c} + \frac{\sin^4 \theta}{s + i\Delta_c + \Gamma_c} \right]. \quad (\text{A.58})$$

A.3.2 Dephasing self-energy

In the similar function, the dephasing reduced self-energy matrix elements in Eqs. (A.52)-(A.54), within Born approximation, take the form

$$\Sigma_{++,,++}^x(s) = 0 \quad (\text{A.59})$$

$$\Sigma_{--,,--}^x(s) = 0 \quad (\text{A.60})$$

$$\Sigma_{+-,,+-}^x(s) = -2i \cos^2 \theta \sin^2 \theta \left[\frac{1}{s + i(\Omega_R - \Delta_c) + \Gamma_c} + \frac{1}{s + i(\Omega_R + \Delta_c) + \Gamma_c} \right]. \quad (\text{A.61})$$

A.4 Matrix elements of reduced self-energy for phonon coupling

A.4.1 Transition self-energy

The reduced self-energy matrix elements which give rise to transition due to system-phonon coupling are expressed as:

$$\Sigma_{++,++}^{SP}(s) = -i \cos^2 \theta \sin^2 \theta \sum_q \lambda_q^2 \left[\frac{1}{s + i(\Omega_R - \Delta_P - \omega_q)} + \frac{1}{s - i(\Omega_R - \Delta_P - \omega_q)} \right] \quad (\text{A.62})$$

$$\Sigma_{++,-}^{SP}(s) = -i \cos^2 \theta \sin^2 \theta \sum_q \lambda_q^2 \left[\frac{1}{s + i(\Omega_R - \Delta_P + \omega_q)} + \frac{1}{s - i(\Omega_R - \Delta_P + \omega_q)} \right] \quad (\text{A.63})$$

$$\Sigma_{+-,+-}^{SP}(s) = -i \cos^2 \theta \sin^2 \theta \sum_q \lambda_q^2 \left[\frac{1}{s + i\omega_q} + \frac{1}{s - i\omega_q} \right]. \quad (\text{A.64})$$

A.4.2 Dephasing self-energy

The reduced self-energy matrix element which gives rise to pure-dephasing is given in polaron frame as

$$\Sigma_{+-,+-}^y(\Delta_0) = -iTr_P \left[e^{-S} L_Y^+ \frac{1}{-i\Delta_0 + i(\Omega_R - \Delta_P) + i(L_P + L_Y^+)} L_Y^+ e^S \rho_P(0) \right] \quad (\text{A.65})$$

where $S = \frac{1}{2} \sum_q \frac{\lambda_q}{\omega_q} (b_q^\dagger - b_q)$. We want to calculate the reduced self-energy matrix element exactly without expanding it in the powers of L_Y^+ . To this end, we set $\Delta'_0 = \Delta_0 - (\Omega_R - \Delta_P)$ to write

$$\Sigma_{+-,+-}'^y(\Delta'_0) = -iTr_P \left[e^{-S} L_Y^+ \frac{1}{-i\Delta'_0 + i(L_P + L_Y^+)} L_Y^+ e^S \rho_P(0) \right] \quad (\text{A.66})$$

which can be written in time domain as

$$\Sigma_{+-,+-}'^y(t) = -iTr_P \left[e^{-S} \{Y(t), Y(0)\} e^S \rho_P(0) \right] \quad (\text{A.67})$$

where

$$Y(t) = e^{-i(L_Y^+ - L_P)t} Y(0) \quad (\text{A.68})$$

$$Y(0) = \frac{\cos 2\theta}{2} \sum_q \lambda_q (b_q + b_q^\dagger), \quad (\text{A.69})$$

and braces indicate the anti-commutator. Solving the above equations for $Y(t)$, we obtain

$$Y(t) = \frac{\cos 2\theta}{2} \sum_q \left[\lambda_q (b_q e^{-i\omega_q t} + b_q^\dagger e^{i\omega_q t}) + \frac{\lambda_q^2}{\omega_q} - \frac{\lambda_q^2}{\omega_q} (e^{-i\omega_q t} + e^{i\omega_q t}) \right]. \quad (\text{A.70})$$

We apply the polaron transform on $Y(t)$ and $Y(0)$, to obtain

$$e^{-S} Y(t) e^S = \frac{\cos 2\theta}{2} \sum_q \left[\lambda_q (b_q e^{-i\omega_q t} + b_q^\dagger e^{i\omega_q t}) + \frac{\lambda_q^2}{\omega_q} \right] \quad (\text{A.71})$$

$$e^{-S} Y(0) e^S = \frac{\cos 2\theta}{2} \sum_q \left[\lambda_q (b_q + b_q^\dagger) + \frac{2\lambda_q^2}{\omega_q} \right]. \quad (\text{A.72})$$

We replace for $Y(t)$ and $Y(0)$ in the expression for reduced self-energy matrix element to obtain

$$\Sigma'_{+-,+-}(t) = -i \cos^2 2\theta \sum_q \text{Tr}_P \left[\left\{ \lambda_q (b_q e^{-i\omega_q t} + b_q^\dagger e^{i\omega_q t}) + \frac{\lambda_q^2}{\omega_q}, \lambda_q (b_q e^{-i\omega_q t} + b_q^\dagger e^{i\omega_q t}) + \frac{2\lambda_q^2}{\omega_q} \right\} \rho_P(0) \right]. \quad (\text{A.73})$$

After solving the anti-commutator and applying the final trace, obtain

$$\Sigma'_{+-,+-}(t) = -i \cos^2 2\theta \sum_q \lambda_q^2 \coth \left(\frac{\beta \omega_q}{2} \right) \cos(\omega_q t) \quad (\text{A.74})$$

We apply the continuum of limit to replace $\sum_q \lambda_q^2 \rightarrow \int d\epsilon D(\epsilon) |\lambda(\epsilon)|^2$, to obtain

$$\Sigma'_{+-,+-}(t) = -i \cos^2 2\theta \int d\epsilon D(\epsilon) |\lambda(\epsilon)|^2 \coth \left(\frac{\beta \epsilon}{2} \right) \cos(\epsilon t). \quad (\text{A.75})$$

Here, we introduce an exponential cut-off at $\epsilon = \epsilon_c$ for $3 - d$ longitudinal acoustic phonons

$$D(\epsilon) |\lambda(\epsilon)|^2 = \lambda^2 K_3 |\epsilon|^3 e^{-\frac{|\epsilon|}{\epsilon_c}} \quad (\text{A.76})$$

defining $\alpha_P = \lambda^2 K_3$ as the phonon coupling parameter has the dimension of frequency⁻². The pure dephasing self-energy matrix element $\Sigma'_{+-,+-}(\Delta'_0)$ can be written in the terms of the decay and the frequency shift as

$$\Sigma'_{+-,+-}(\Delta'_0) = -i \frac{\alpha_P \Delta'^2}{\Omega_R^2} \Gamma_P(\Delta'_0) + \frac{\alpha_P \Delta'^2}{\Omega_R^2} \Delta\omega_P(\Delta'_0) \quad (\text{A.77})$$

where $\Gamma_P(\Delta'_0)$ and $\Delta\omega_P(\Delta'_0)$ are the frequency-dependent decay and the frequency-shift, respectively due to phonon coupling and given as

$$\Gamma_P(\Delta'_0) = \frac{\pi}{2} |\Delta'_0|^3 \coth\left(\frac{\Delta'_0}{2k_B T}\right) e^{\frac{-|\Delta'_0|}{\epsilon_c}} \left(\theta(\Delta'_0) - \theta(-\Delta'_0) \right) \quad (\text{A.78})$$

where $H(\Delta'_0)$ is the unit step function and given as

$$\theta(\Delta'_0) = \begin{cases} 1 & : \Delta'_0 \geq 0 \\ 0 & : \Delta'_0 < 0 \end{cases}$$

$$\Delta\omega_P(\Delta'_0) = \frac{1}{2} P \int_{-\infty}^{\infty} d\epsilon \frac{|\epsilon|^3 \coth\left(\frac{\epsilon}{2k_B T}\right) e^{-\frac{|\epsilon|}{\epsilon_c}}}{\Delta'_0 - \epsilon}. \quad (\text{A.79})$$

A.5 Detector response function for two-level system in a cavity

In this section, we calculate the detector response function for our two level system kept inside a cavity. Recalling detector response function given in equation (2.28),

$$S(t) = \sum_k g_k g_k^D e^{-i\omega_k(t)} \quad (\text{A.80})$$

$$= \alpha \sum_k g_k^2 e^{-i\omega_k(t)} \quad (\text{A.81})$$

taking the Laplace transform and applying the continuum of modes by replacing $\sum_k \rightarrow \int d\epsilon D(\epsilon)$ and $g_k^2 \rightarrow |g(\epsilon)|^2$, to obtain

$$S(s) = \alpha \int d\epsilon D(\epsilon) |g(\epsilon)|^2 \frac{1}{s + i\epsilon}, \quad (\text{A.82})$$

using equation (A.55), to get

$$S(s) = \alpha \int d\epsilon \frac{1}{\pi} \frac{g^2 \Gamma_c}{(\epsilon - \omega_c)^2 + \Gamma_c^2} \frac{1}{s + i\epsilon}. \quad (\text{A.83})$$

Solving above equation using contour integration method to obtain,

$$S(s) = \frac{\alpha g^2}{s - i\Delta_c + \Gamma_c}. \quad (\text{A.84})$$

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