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非馬可夫系統的雙時關聯函數：精確解與微擾近似的研究
Non-Markovian two-time correlation functions: exact result v.s
perturbation approach



蔡宇安

Tsai, Yu-An

指導教授：管希聖博士

Advisor: Goan, Hsi-Sheng, Ph.D.

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究

Non-Markovian two-time correlation functions: exact result
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本論文係蔡宇安君(R98222007)在國立臺灣大學物理學 所完成
之碩士學位論文，於民國 100 年 06 月 28 日承下列考試委員審查通過
及口試及格，特此證明

口試委員：

官希聖

(簽名)

(指導教授)

孫中

陳柏中

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中文摘要

我們研究了一個二能級自發性輻射到真空場的模型，也就是透過 Jaynes-Cummings 模型在旋轉波近似下的研究。當我們考慮最多只有一個激發態時，這個模型能夠被精確的解出。這裡我們使用兩種方法得出非馬可夫開放系統的雙時關聯函數：第一個是直接精確的計算系統運算子隨時間的演化。另一種方法是透過微擾的非馬可夫主宰方程進行計算，我們將非馬可夫主宰方程寫成對時間是非卷積的形式。為了計算雙時關聯函數，我們將系統-環境之間的交互作用，用微擾的方式展開到第四階的近似。我們將以上兩種方法做了一些比較。在這邊，我們同時找到了一種使微擾近似更準確的方法，我們是把加速數列收斂的技術應用在微擾展開的主宰方程上。我們的結果發現，非馬可夫主宰方程比下面幾種方法都還要更接近精確解：馬可夫微擾、非馬可夫微擾與精確解並使用量子退化定理。這樣的結果表示，我們找到的非馬可夫主宰方程是相當有用同時正確的。另外，我們也研究了自發性輻射的光譜，我們發現它在強耦合與弱耦合下有非常不相同的行為。

Abstract

A two-level system which decays spontaneously into field vacuum is studied through the Jaynes-Cummings model in the rotating wave approximation (RWA). When at most one excitation is considered, this model is exactly solvable. Here we evaluate the non-Markovian two-time correlation functions (CF's) of system operators for this model in two ways: one by directly solving the system-environment evolution, and the other by using the perturbative time-convolutionless non-Markovian master equation approach. We derive valid to fourth order in system-bath coupling strength a non-Markovian evolution equation for the two-time CF's of system operators. We use the derived evolution equation to calculate a two-time CF for the two-level model and compare it with the exact result obtained by direct evaluation. Another numerical series acceleration method is applied to the calculation of the perturbation decay, and this method is found to improve the accuracy of the evolution equation. The result obtained by the derived perturbative two-time evolution equation is much better than those by the perturbative Markovian Quantum regression theorem (QRT), the non-Markovian QRT and exact QRT as it agrees more closely with the exact result even when the model is in the regime where the bath correlation time is comparable to the system relaxation time. This demonstrates the validity and usefulness of our derived non-Markovian two-time evolution equation. The exact spontaneous emission spectrum is also calculated, and it has very different behaviours in the strong coupling and the weak coupling regions.

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Chapter 1

Introduction

A realistic quantum systems is always hard to isolate from its environments. The environments that usually contain infinite number of degrees of freedom, interact with the quantum systems. The infinite environment degrees of freedom contain many information that we do not want to know, and the relevant information is from the quantum system of interest. One of the quantities is reduced density matrix of the system, it describes the state of the reduce system. Thus the system-environment interaction will lead to quantum dissipative, quantum decoherence, and spontaneous emission phenomena of the system when the environment degrees of freedom are traced over.

The evolution of reduced density matrix is governed by a equation called the quantum master equation. There are two kind of master equations Nakajima-Zwanzig time-convolution and the time-convolutionless ones. In this thesis, we focus mainly on the time-convolutionless approach.

The two-time correlation functions are important quantities [1–10], they can provide some important information of the system, whereas the single-time expectation values can not. There are some physical quantities that require the calculation of two-time correlation functions, like the spontaneous emission spectrum of an atom, the photon bunching and anti-bunching phenomena, and the current noise in nanos-structure devices.

In the Markovain open quantum systems, an useful method to calculate two-time correlations functions by master equation is the quantum regression theorem (QRT).

It states that the two(multi)-time evolution equation of the system operator correlation functions same as the one-time evolution equation for expectation values. The quantum regression theorem could be valid in Markovian open quantum systems, but may not be valid in non-Markovian open quantum systems [1, 2, 4, 10, 11]. The Markovian approximation assumes that the bath correlation time is very short as compared to typical system response time, and the bath correlation function is treated as delta-correlated in time, this is not true for non-Markovian systems. Therefore, the quantum regression theorem is not applicable when the bath correlation time is large and comparable to system response time.

In Ref.[2], a time-convolutionless equation for two-time correlation function of system operators to second system-environment interaction Hamiltonian has been derived. In this thesis, we extend the derivation of the general time-convolutionless evolution equation for two-time correlation functions in higher order in perturbation expansion. Furthermore, we study a two-level system coupled to zero-temperature bath through the Jaynes-Cummings model. When at most one excitation is considered, this model is exactly solved. That is, all single-time expectation values of this model can be calculated. We also find that a particular two-time correlation function can be evaluated exactly. We then use the derived evolution equation up to fourth order to calculate the two-time correlation function for the two-level model and compare it with the exact result. Since the higher order perturbation result become very complicated and hard to calculate, we employ series acceleration method to improve the perturbation result.

The thesis is organized as follows. In Chapter 2, we derive the general time-convolutionless evolution equation for two-time correlation functions up to fourth order in the system-environment interaction Hamiltonian. In Chapter 3, we evaluate an exact two-time correlation function of system operators for Jaynes-Cummings model with one excitation. In Chapter 4, we use the evolution equation derived in Chapter 2 to obtain a perturbation evolution equation to fourth order, and then deduce an exact evolution equation based on the result obtained in Chapter 3. We

also introduce the series acceleration method in this chapter. In Chapter 5, we show the difference between the perturbation and the exact results in different conditions. We also calculate the exact spontaneous emission spectrum in the chapter. Finally, a short conclusion is given in Chapter 6.



Chapter 2

General master equation of two-time correlation functions for reduced effective density matrix

An open quantum system is usually described by a system Hamiltonian belonging to Hilbert space \mathbb{S} , an environment Hamiltonian belonging to Hilbert space \mathbb{B} and an interaction Hamiltonian between system and environment belonging to Hilbert space $\mathbb{S} \otimes \mathbb{B}$. The total Hamiltonian can be written in the form

$$H = H_s + H_B + H_I = H_0 + H_I, \quad (2.1)$$

where H_s and H_B are system and environment Hamiltonians, which describe the uncoupled state evolutions of system and environment respectively, and H_I is the interaction Hamiltonian which describes the interaction between the system and environment.

To describe the open quantum systems, we use the density matrix approach which is helpful for dealing with mixed states. The density matrix contains the information of not only the probability but also the coherence. The state vector $|\phi\rangle$ is always a pure state. The relation between state vector and density matrix is defined as $\rho = |\phi\rangle\langle\phi|$.

The time-evolution of density matrix is described by the von Neumann equation in the Schrödinger picture.

$$\frac{\partial}{\partial t}\rho(t) = -i[H, \rho(t)]. \quad (2.2)$$

In interaction picture, the von Neumann equation becomes

$$\frac{\partial}{\partial t}\tilde{\rho}(t) = -i[\tilde{H}_I(t), \tilde{\rho}(t)] \equiv \mathcal{L}(t)\rho(t), \quad (2.3)$$

where $\tilde{H}_I(t) = e^{-iH_0t}H_Ie^{iH_0t}$, and $\tilde{\rho}(t) = e^{iH_0t}\rho(t)e^{-iH_0t}$.

The two-time correlation functions can be written in the Heisenberg picture as:

$$\langle A(t_1)B(t_2) \rangle \equiv \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{U^\dagger(t_1)AU(t_1)U^\dagger(t_2)BU(t_2)\rho(0)\} \quad (2.4)$$

$$= \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{AU(t_1 - t_2)B\rho(t_2)U^\dagger(t_1 - t_2)\} \quad (2.5)$$

$$= \text{Tr}_{\mathbb{S}} \{A\text{Tr}_{\mathbb{B}}\{X(t_1)\}\} \quad (2.6)$$

$$= \text{Tr}_{\mathbb{S}} \{e^{-iH_0t_1}Ae^{-iH_0t_1}\text{Tr}_{\mathbb{B}}\{\tilde{\chi}(t_1)\}\}, \quad (2.7)$$

where A, B is any operator which belong to \mathbb{S} , and we have transformed from the Heisenberg picture of Eq.(2.4) to the interaction picture of Eq.(2.7).

$$\chi(t_1) = U(t_1 - t_2)B\rho(t_2)U^\dagger(t_1 - t_2) \quad (2.8)$$

$$\tilde{\chi}(t_1) = e^{iH_0t_1}\chi(t_1)e^{-iH_0t_1}, \quad (2.9)$$

$\chi(t_1)$ and $\tilde{\chi}(t_1)$ are effective density matrix in the Schrödinger and interaction pictures, respectively.

To solve for $\tilde{\chi}(t_1)$, we require that $\tilde{\chi}(t_1)$ satisfies the following conditions

Table 2.1: The requirement of problem

1. The effect density matrix satisfy $\frac{\partial}{\partial t_1}\text{Tr}_{\mathbb{B}}\{\tilde{\chi}(t_1)\} = \text{Tr}_{\mathbb{B}}\{-i[\tilde{H}_I(t_1), \tilde{\chi}(t_1)]\}$
2. The initial condition in the time t_2 $\tilde{\chi}(t_2) = \tilde{B}(t_2)\tilde{\rho}(t_2)$ should be know.
3. The state $\rho(0)$ can be factorized in time $t = 0$, i.e. $\rho(0) = \rho_s(0) \otimes \rho_B$, where $\rho_s \in \mathbb{S}$, $\rho_B \in \mathbb{B}$

All of the following discussion had assume above three conditions be satisfied.

2.1 Projection operator

Most often, the relevant information we want to know is reduce effect density matrix, so we need a technique to separate system and environment. One of widely

used method ([10, 12–16]) is the Nakajima-Zwanzig projection operator \mathcal{P} ([17, 18]). It is defined as follows:

$$\mathcal{P}\tilde{W}(t) \equiv \text{Tr}_{\mathbb{B}}\{\tilde{W}(t)\} \otimes \rho_B = \tilde{W}_s(t) \otimes \rho_B \quad (2.10)$$

Here we do not require $W(t)$ must be a genuine density matrix, and it could be an effective density matrix. ρ_B is some known state of environment called the reference state. We assume that ρ_B would not change with time evolution. The projection operator helps us to separate the relevant part and irrelevant part. It also helps us make calculation easier. In other words, with projection operator \mathcal{P} , we do not need to treat the evolution of environment state. With completeness relation, we define operator $\mathcal{Q} = I - \mathcal{P}$. $\mathcal{Q}W(t)$ contains the information about system-environment entanglement, about whether ρ_B is in an equilibrium or ρ_B change with time. Operator \mathcal{P} and \mathcal{Q} satisfy the following properties.

$$\mathcal{Q} + \mathcal{P} = I, \quad (2.11)$$

$$\mathcal{Q}\mathcal{Q} = \mathcal{Q}, \quad (2.12)$$

$$\mathcal{P}\mathcal{P} = \mathcal{P}, \quad (2.13)$$

$$\mathcal{P}\mathcal{Q} = \mathcal{Q}\mathcal{P} = 0, \quad (2.14)$$

Here, we make an addition assumption. We assume the reference state ρ_B of the environment is a Gaussian state. It implies that we can apply Wick's rule to express the high order moment as a combination of the second order moments, and the interaction Hamiltonian satisfy $\text{Tr}_{\mathbb{B}}\{[\rho_B, H_I]\} = 0$. Using above properties we obtain the following equation.

$$\mathcal{P}\mathcal{L}(t_1)\mathcal{P} = 0, \quad (2.15)$$

$$\mathcal{P}\mathcal{L}(t_1)\mathcal{L}(t_2)\mathcal{L}(t_3)\mathcal{P} = 0, \quad (2.16)$$

⋮

2.2 Cumulant expansion

Cumulant expansion provides an elegant and easy method to obtain exact time-convolutionless master equation or evolution equation for two-time correlation functions. The method introduced by Kubo [19] and Van Kampen [20, 21], can apply to many physical problems. The connection of cumulant is through the moment generating function. Suppose we have an operator χ and reference state ρ_B , which satisfy $\chi \in \mathbb{S} \otimes \mathbb{B}$, and $\text{Tr}_{\mathbb{B}}\{\rho_B\} = 1$. The first and second moments are defined by

$$\langle X \rangle_B = \text{Tr}_{\mathbb{B}}\{X\rho_B\} = \mu_1, \quad (2.17)$$

$$\langle X^2 \rangle_B = \mu_2. \quad (2.18)$$

The moment generating function is defined as

$$M(\xi) = \langle e^{X\xi} \rangle_B = \sum_{i=0}^{\infty} \langle X^i \rangle_B \frac{\xi^i}{i!} = \sum_{n=0}^{\infty} \mu_n \frac{\xi^n}{n!}, \quad (2.19)$$

where ξ is the expansion parameter. The cumulants generating function $K(\xi)$ is

$$K(\xi) = \ln(M(\xi)) = \sum_{n=1}^{\infty} K_n \frac{\xi^n}{n!} \equiv \sum_{n=1}^{\infty} \langle X^n \rangle_c \frac{\xi^n}{n!}, \quad (2.20)$$

$$M(\xi) = \exp(K(\xi)) = \sum_{n=0}^{\infty} \langle X^n \rangle_B \frac{\xi^n}{n!} = \exp\left(\sum_{n=1}^{\infty} \langle X^n \rangle_c \frac{\xi^n}{n!}\right), \quad (2.21)$$

The cumulants K_n are the coefficient of expansion of generating function $K(\xi)$. The cumulant average $\langle X^n \rangle_c$ is not usual average, it is the coefficient in Eq.(2.20) with order ξ^i . It is obviously that $\mu_0 = 1$ makes $K_0 = 0$. The notions for average used in this thesis are summarized below. The notation $\langle \dots \rangle_B$ means to trace the quantity over the environment reference state ρ_B , $\langle \dots \rangle_c$ denotes the cumulant average, the average without any subscript means to trace over all degrees of freedom. For examples, the moment $\langle X^n \rangle_B$ means trace over reference state with operator X^n , $\langle X^n \rangle_c$ is n-th cumulant.

The cumulants expansion can be extended to multi-observable as,

$$M(\xi_1, \dots, \xi_N) = \sum_{v_1=0}^{\infty} \cdots \sum_{v_N=0}^{\infty} \prod_j \frac{\xi_j^{v_j}}{v_j!} \langle X_1^{v_1} \cdots X_N^{v_N} \rangle_B, \quad (2.22)$$

$$M(\vec{\xi}) = \exp(K(\vec{\xi})) = \exp\left(\sum_{v_1=0}^{\infty} \cdots \sum_{v_N=0}^{\infty} \prod_j \frac{\xi_j^{v_j}}{v_j!} \langle X_1^{v_1} \cdots X_N^{v_N} \rangle_c\right), \quad (2.23)$$

and the cumulant $\langle 1 \rangle_c = 0$. The results of cumulants with multi-observable can be found in Ref.[19]. For example

$$\langle X_i \rangle_c = \langle X_i \rangle_B, \quad (2.24)$$

$$\langle X_i X_j \rangle_c = \langle X_i X_j \rangle_B - \langle X_i \rangle_B \langle X_j \rangle_B, \quad (2.25)$$

$$\begin{aligned} \langle X_i X_j X_k \rangle_c &= \langle X_i X_j X_k \rangle_B + 2\langle X_i \rangle_B \langle X_j \rangle_B \langle X_k \rangle_B \\ &\quad - \{\langle X_i \rangle_B \langle X_j X_k \rangle_B + \langle X_j \rangle_B \langle X_i X_k \rangle_B + \langle X_k \rangle_B \langle X_i X_j \rangle_B\}, \end{aligned} \quad (2.26)$$

The most important result to us is the relation of the time-ordering function and cumulant. The relation was found in Ref. [19–21]

$$\begin{aligned} &\langle T \exp\left(\int_{t_0}^t d\tau X(\tau)\right) \rangle_B \\ &= \sum_{n=0}^{\infty} \int_{t_0}^t d\tau_1 \int_{t_0}^{\tau_1} d\tau_2 \cdots \int_{t_0}^{\tau_{n-1}} d\tau_n \langle X(\tau_1) X(\tau_2) \cdots X(\tau_n) \rangle_B \end{aligned} \quad (2.27)$$

$$= \exp\left(\sum_{n=1}^{\infty} \int_{t_0}^t d\tau_1 \int_{t_0}^{\tau_1} d\tau_2 \cdots \int_{t_0}^{\tau_{n-1}} d\tau_n \langle X(\tau_1) X(\tau_2) \cdots X(\tau_n) \rangle_c\right) \quad (2.28)$$

$$= \exp\left(\sum_{n=1}^{\infty} K'_n(t)\right) \equiv \exp(K'(t)), \quad (2.29)$$

where T denote as time-ordering operator and

$$K'(t) = \sum_{n=1}^{\infty} K'_n(t) \quad (2.30)$$

$$= \sum_{n=1}^{\infty} \int_{t_0}^t d\tau_1 \int_{t_0}^{\tau_1} d\tau_2 \cdots \int_{t_0}^{\tau_{n-1}} d\tau_n \langle X(\tau_1) X(\tau_2) \cdots X(\tau_n) \rangle_c, \quad (2.31)$$

$$K'_n(t) = \int_{t_0}^t d\tau_1 \int_{t_0}^{\tau_1} d\tau_2 \cdots \int_{t_0}^{\tau_{n-1}} d\tau_n \langle X(\tau_1) X(\tau_2) \cdots X(\tau_n) \rangle_c. \quad (2.32)$$

2.3 Cumulant expansion to homogenous master equation

First, we give a connection between the moments and the projection operator.

We can write an alternate form of average through the projection operator.

$$\mathcal{P}X\mathcal{P} = \text{Tr}_{\mathbb{B}}\{X\rho_B\}\text{Tr}_{\mathbb{B}}\{\cdots \otimes \rho_B\} = \langle X \rangle_B \mathcal{P}, \quad (2.33)$$

$$\langle X_i X_j \rangle_c \mathcal{P} = \mathcal{P}X_i X_j \mathcal{P} - \mathcal{P}X_i \mathcal{P} \mathcal{P}X_j \mathcal{P}, \quad (2.34)$$

$$\langle X_i \cdots X_j \rangle \cdots \langle X_l \cdots X_n \rangle \mathcal{P} = \mathcal{P}X_i \cdots X_j \mathcal{P} \cdots \mathcal{P}X_l \cdots X_n \mathcal{P}, \quad (2.35)$$

The moment followed by projection operator is equal to projection operators in both side of operator. Our main goal is to get an exact time-convolutionless evolution equation for two-time correlation functions or equivalently the time-convolutionless for the effective density matrix $\tilde{\chi}_s(t)$. The requirement of the master equation for $\tilde{\chi}_s(t)$ was listed in Table 2.1. We will solve the following equation with the requirement,

$$\frac{\partial}{\partial t_1} \tilde{\chi}(t_1) = -i[\tilde{H}_I(t_1), \tilde{\chi}(t_1)] = \mathcal{L}(t_1)\tilde{\chi}(t_1), \quad (2.36)$$

where \mathcal{L} is Liouville operator defined in Eq.(2.3). The formal solution is

$$\tilde{\chi}(t_1) = T \exp\left(\int_{t_2}^{t_1} d\tau \mathcal{L}(\tau)\right)\tilde{\chi}(t_2), \quad (2.37)$$

$$\mathcal{P}\tilde{\chi}(t_1) = \mathcal{P}T \exp\left(\int_{t_2}^{t_1} d\tau \mathcal{L}(\tau)\right)(\mathcal{P} + \mathcal{Q})\tilde{\chi}(t_2), \quad (2.38)$$

If we apply the projection operator on $\tilde{\chi}(t)$, we obtain

$$\begin{aligned} \mathcal{P}\tilde{\chi}(t_1) &= \langle T \exp\left(\int_{t_0}^t d\tau \mathcal{L}(\tau)\right) \rangle_B \mathcal{P}\tilde{\chi}(t_2) \\ &\quad + \mathcal{P}T \exp\left(\int_{t_1}^t d\tau \mathcal{L}(\tau)\right)\mathcal{Q}\tilde{\chi}(t_2), \end{aligned} \quad (2.39)$$

Using Eq.(2.29) - Eq.(2.31), we obtain Eq.(2.41)

$$\mathcal{P}\tilde{\chi}(t_1) = \exp\left(\sum_{n=1}^{\infty} K'_n(t_1, t_2)\right)\tilde{\chi}(t_2) + \mathcal{P}T \exp\left(\int_{t_1}^t d\tau \mathcal{L}(\tau)\right)\mathcal{Q}\tilde{\chi}(t_2), \quad (2.40)$$

Differential Eq.(2.40) with respect to t_1 , we obtain

$$\begin{aligned} \frac{\partial}{\partial t_1} \mathcal{P} \tilde{\chi}(t_1) &= \sum_{n=1}^{\infty} K_n(t_1, t_2) \mathcal{P} \exp\left(\sum_{n=1}^{\infty} K'_n(t_1, t_2)\right) \mathcal{P} \tilde{\chi}(t_2) \\ &+ \mathcal{P} \mathcal{L}(t_1) T \exp\left(\int_{t_2}^{t_1} d\tau \mathcal{L}(\tau)\right) \mathcal{Q} \tilde{\chi}(t_2), \end{aligned} \quad (2.41)$$

where

$$K_n(t) = \frac{\partial}{\partial t} K'_n(t). \quad (2.42)$$

Using the relation obtained from Eq.(2.40)

$$\exp\left(\sum_{n=1}^{\infty} K_n(t, t_2)\right) \tilde{\chi}(t_2) = \mathcal{P} \tilde{\chi}(t_1) - \mathcal{P} T \exp\left(\int_{t_2}^{t_1} d\tau \mathcal{L}(\tau)\right) \mathcal{Q} \tilde{\chi}(t_1), \quad (2.43)$$

and insert Eq.(2.43) to Eq.(2.41), we can get

$$\begin{aligned} \frac{\partial}{\partial t_1} \mathcal{P} \tilde{\chi}(t_1) &= \sum_{n=1}^{\infty} K_n(t_1, t_2) \mathcal{P} \tilde{\chi}(t_1) \\ &+ \mathcal{P} \left\{ \mathcal{L}(t_1) - \sum_{n=1}^{\infty} K_n(t, t_2) \mathcal{P} \right\} T \exp\left(\int_{t_2}^{t_1} d\tau \mathcal{L}(\tau)\right) \mathcal{Q} \tilde{\chi}(t_2) \end{aligned} \quad (2.44)$$

$$\equiv \sum_{n=1}^{\infty} K_n(t_1, t_2) \mathcal{P} \tilde{\chi}(t_1) + R_n(t_1, t_2) \mathcal{Q} \tilde{\chi}(t_2), \quad (2.45)$$

The lowest four cumulants K_n in terms of moments are written in the following

$$K_1(t_1, t_2) = \langle \mathcal{L}(t_1) \rangle_c = 0, \quad (2.46)$$

$$K_2(t_1, t_2) = \int_{t_2}^{t_1} d\tau_1 \langle \mathcal{L}(t_1) \mathcal{L}(\tau_1) \rangle_c = \int_{t_2}^{t_1} d\tau_1 \langle \mathcal{L}(t_1) \mathcal{L}(\tau_1) \rangle_B, \quad (2.47)$$

$$K_3(t_1, t_2) = \int_{t_2}^{t_1} d\tau_1 \int_{t_2}^{\tau_1} d\tau_2 \langle \mathcal{L}(t_1) \mathcal{L}(\tau_1) \mathcal{L}(\tau_2) \rangle_c = 0, \quad (2.48)$$

$$\begin{aligned} K_4(t_1, t_2) &= \int_{t_2}^{t_1} d\tau_1 \int_{t_2}^{\tau_1} d\tau_2 \int_{t_2}^{\tau_2} d\tau_3 \langle \mathcal{L}(t_1) \mathcal{L}(\tau_1) \mathcal{L}(\tau_2) \mathcal{L}(\tau_3) \rangle_c, \\ &= \int_{t_2}^{t_1} d\tau_1 \int_{t_2}^{\tau_1} d\tau_2 \int_{t_2}^{\tau_2} d\tau_3 \langle \mathcal{L}(t_1) \mathcal{L}(\tau_1) \mathcal{L}(\tau_2) \mathcal{L}(\tau_3) \rangle_B \\ &- \langle \mathcal{L}(t_1) \mathcal{L}(\tau_1) \rangle_B \langle \mathcal{L}(\tau_2) \mathcal{L}(\tau_3) \rangle_B - \langle \mathcal{L}(t_1) \mathcal{L}(\tau_2) \rangle_B \langle \mathcal{L}(\tau_1) \mathcal{L}(\tau_3) \rangle_B \\ &- \langle \mathcal{L}(t_1) \mathcal{L}(\tau_3) \rangle_B \langle \mathcal{L}(\tau_1) \mathcal{L}(\tau_2) \rangle_B \end{aligned} \quad (2.49)$$

The relation between cumulants and moments of higher order are not showing here.

The expressions of the lowest three $R_n(t_1, t_2)$ shown in the following

$$R_1(t_1, t_2) = \mathcal{P}\mathcal{L}(t_1)\mathcal{Q}, \quad (2.50)$$

$$R_2(t_1, t_2) = \int_{t_2}^{t_1} d\tau_1 \mathcal{P}\mathcal{L}(t_1)\mathcal{L}(\tau_1)\mathcal{Q}, \quad (2.51)$$

$$R_3(t_1, t_2) = \int_{t_2}^{t_1} d\tau_1 \int_{t_2}^{\tau_1} d\tau_2 \mathcal{P}\mathcal{L}(t_1)\mathcal{L}(\tau_1)\mathcal{L}(\tau_2)\mathcal{Q} \\ - \mathcal{P}\mathcal{L}(t_1)\mathcal{L}(\tau_1)\mathcal{P}\mathcal{L}(\tau_2)\mathcal{Q} - \mathcal{P}\mathcal{L}(t_1)\mathcal{L}(\tau_2)\mathcal{P}\mathcal{L}(\tau_1)\mathcal{Q}. \quad (2.52)$$

Here, we have used the relation $\mathcal{P}\mathcal{Q} = 0$.

2.4 Inhomogenous term

The second term in the r.h.s. of Eq. (2.41) can not be evaluated directly because we do not know about $\mathcal{Q}\tilde{\chi}(t_2)$, that contains the information of the environment degrees of freedom. An alternative way is to use the requirement 3 in Table 2.1 such that $\rho(0) = \mathcal{P}\rho(0)$. The first step is to separate $\mathcal{Q}\tilde{\chi}(t_2)$ as $\mathcal{Q}\tilde{\chi}(t_2) = \tilde{B}(t_2)\mathcal{Q}\rho(t_2)$ since $\tilde{B}(t_2)$ is a system operator, Then the problem becomes how to derive $\mathcal{Q}\rho(t_2)$ in term of $\mathcal{P}\rho(t_2)$.

Starting from Eq.(2.2) and requirement 3 in Table 2.1, we can obtain

$$\frac{\partial}{\partial t_2} \tilde{\rho}(t_2) = \mathcal{L}(t_2)\tilde{\rho}(t_2), \quad (2.53)$$

$$\tilde{\rho}(t_2) = \{I + \sum_{n=1}^{\infty} \int_0^{t_2} d\tau_1 \cdots \int_0^{\tau_{n-1}} d\tau_n \mathcal{L}(\tau_1) \cdots \mathcal{L}(\tau_n)\} \mathcal{P}\rho(0) \\ \equiv \{I + M\} \mathcal{P}\rho(0), \quad (2.54)$$

where

$$M = \sum_{n=1}^{\infty} \int_0^{t_2} d\tau_1 \cdots \int_0^{\tau_{n-1}} d\tau_n \mathcal{L}(\tau_1) \cdots \mathcal{L}(\tau_n). \quad (2.55)$$

We apply \mathcal{P} and \mathcal{Q} to Eq.(2.54)

$$\mathcal{Q}\tilde{\rho}(t_2) = \mathcal{Q}(I + M)\mathcal{P}\rho(0), \quad (2.56)$$

$$\mathcal{P}\tilde{\rho}(t_2) = \mathcal{P}(I + M)\mathcal{P}\rho(0), \quad (2.57)$$

We invert $(I + \mathcal{P}M\mathcal{P})$ in Eq.(2.57) to obtain

$$\mathcal{P}\rho(0) = \mathcal{P}\{I + \mathcal{P}M\mathcal{P}\}^{-1}\mathcal{P}\tilde{\rho}(t_2), \quad (2.58)$$

Substituting Eq.(2.58) in Eq.(2.56), we obtain the relation between $\mathcal{Q}\tilde{\rho}(t_2)$ and $\mathcal{P}\tilde{\rho}(t_2)$

$$\mathcal{Q}\tilde{\rho}(t_2) = \mathcal{Q}(I + M)\mathcal{P}\{I + \mathcal{P}M\mathcal{P}\}^{-1}\mathcal{P}\tilde{\rho}(t_2). \quad (2.59)$$

Then we expand $\mathcal{P}\{I + \mathcal{P}M\mathcal{P}\}^{-1}$ to a geometric series to obtain

$$\begin{aligned} \mathcal{Q}\tilde{\rho}(t_2) &= \mathcal{Q}M\mathcal{P}\{I - \mathcal{P}M\mathcal{P} + \{\mathcal{P}M\mathcal{P}\}^2 - \{\mathcal{P}M\mathcal{P}\}^3 + \cdots\}\mathcal{P}\tilde{\rho}(t_2) \\ &\equiv \mathcal{Q}\sum_{n=1}^{\infty} J_n(t_2, 0)\mathcal{P}\tilde{\rho}(t_2). \end{aligned} \quad (2.60)$$

The first few $J_n(t_2, 0)$ are given as follows.

$$J_1(t_2, 0) = \int_0^{t_2} d\tau_1 \mathcal{Q}\mathcal{L}(\tau_1)\mathcal{P}, \quad (2.61)$$

$$J_2(t_2, 0) = \int_0^{t_2} d\tau_1 \int_0^{\tau_1} d\tau_2 \mathcal{Q}\mathcal{L}(\tau_1)\mathcal{L}(\tau_2)\mathcal{P}, \quad (2.62)$$

$$\begin{aligned} J_3(t_2, 0) &= \mathcal{Q} \int_0^{t_2} d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \\ &\quad \mathcal{L}(\tau_1)\mathcal{L}(\tau_2)\mathcal{L}(\tau_3)\mathcal{P} - \mathcal{L}(\tau_1)\mathcal{P}\mathcal{L}(\tau_2)\mathcal{L}(\tau_3)\mathcal{P} \\ &\quad - \mathcal{L}(\tau_2)\mathcal{P}\mathcal{L}(\tau_1)\mathcal{L}(\tau_3)\mathcal{P} - \mathcal{L}(\tau_3)\mathcal{P}\mathcal{L}(\tau_1)\mathcal{L}(\tau_2)\mathcal{P}, \end{aligned} \quad (2.63)$$

We can combine Eqs.(2.50)-(2.52) and (2.61)-(2.63) to get the final results of the master equation for the effective density matrix from Eq.(2.45)

$$\frac{\partial}{\partial t_1} \mathcal{P}\tilde{\chi}(t_1) = \sum_{n=1}^{\infty} K_n(t_1, t_2)\mathcal{P}\tilde{\chi}(t_1) + \sum_{n=1}^{\infty} I_n(t_1, t_2, 0, B)\mathcal{P}\tilde{\rho}(t_2), \quad (2.64)$$

where the first few inhomogeneous terms are

$$I_1(t_1, t_2, 0, B) = 0, \quad (2.65)$$

$$I_3(t_1, t_2, 0, B) = 0, \quad (2.66)$$

$$\begin{aligned} I_2(t_1, t_2, 0, B) &= R_1(t_1, t_2) \tilde{B}(t_2) J_1(t_2, 0) \\ &= \int_0^{t_2} d\tau_1 \mathcal{P} \mathcal{L}(t_1) \tilde{B}(t_2) \mathcal{L}(\tau_1) \mathcal{P}, \end{aligned} \quad (2.67)$$

$$\begin{aligned} I_4(t_1, t_2, 0, B) &= R_1(t_1, t_2) \tilde{B}(t_2) J_3(t_2, 0) \\ &\quad + R_2(t_1, t_2) \tilde{B}(t_2) J_2(t_2, 0) + R_3(t_1, t_2) \tilde{B}(t_2) J_1(t_2, 0) \quad (2.68) \\ &= \int_0^{t_2} d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \mathcal{P} \mathcal{L}(t_1) \tilde{B}(t_2) \mathcal{L}(\tau_1) \mathcal{L}(\tau_2) \mathcal{L}(\tau_3) \mathcal{P} \\ &\quad - \mathcal{P} \mathcal{L}(t_1) \tilde{B}(t_2) \mathcal{L}(\tau_1) \mathcal{P} \mathcal{L}(\tau_2) \mathcal{L}(\tau_3) \mathcal{P} - \mathcal{P} \mathcal{L}(t_1) \tilde{B}(t_2) \mathcal{L}(\tau_2) \mathcal{P} \mathcal{L}(\tau_1) \mathcal{L}(\tau_3) \mathcal{P} \\ &\quad - \mathcal{P} \mathcal{L}(t_1) \tilde{B}(t_2) \mathcal{L}(\tau_3) \mathcal{L}(\tau_1) \mathcal{P} \mathcal{L}(\tau_2) \mathcal{P} \\ &\quad + \int_{t_2}^{t_1} d\tau_1 \int_0^{t_2} d\tau_2 \int_0^{\tau_2} d\tau_3 \mathcal{P} \mathcal{L}(t_1) \mathcal{L}(\tau_1) \tilde{B}(t_2) \mathcal{L}(\tau_2) \mathcal{L}(\tau_3) \mathcal{P} \\ &\quad - \mathcal{P} \mathcal{L}(t_1) \mathcal{L}(\tau_1) \mathcal{P} \tilde{B}(t_2) \mathcal{L}(\tau_2) \mathcal{L}(\tau_3) \mathcal{P} \\ &\quad + \int_{t_2}^{t_1} d\tau_1 \int_{t_2}^{\tau_1} d\tau_2 \int_0^{t_2} d\tau_3 \mathcal{P} \mathcal{L}(t_1) \mathcal{L}(\tau_1) \mathcal{L}(\tau_2) \tilde{B}(t_2) \mathcal{L}(\tau_3) \mathcal{P} \\ &\quad - \mathcal{P} \mathcal{L}(t_1) \mathcal{L}(\tau_1) \mathcal{P} \mathcal{L}(\tau_2) \tilde{B}(t_2) \mathcal{L}(\tau_3) \mathcal{P} - \mathcal{P} \mathcal{L}(t_1) \mathcal{L}(\tau_2) \mathcal{P} \mathcal{L}(\tau_1) \tilde{B}(t_2) \mathcal{L}(\tau_3) \mathcal{P}. \end{aligned} \quad (2.69)$$

Equation (2.64) is the exact master equation for the effective density matrix if the series converges. However, in some parameter region the series may diverge. This may happen in the strong coupling region. When the series of Eq.(2.64) diverges, the effect density matrix can not be obtained from Eq.(2.64). The same result of Eq.(2.64) was also found in [10].

2.5 Insert the interaction Hamiltonian to master equation

We consider the interaction Hamiltonian in the following form;

$$H_I = \sum_k g_k a_k L^\dagger + g_k a_k^\dagger L \equiv \sum_k \sum_{i=0}^1 g_k a_k^i L^i, \quad (2.70)$$

where $a_k^0 \equiv a_k, a_k^1 \equiv a_k^\dagger; L^0 \equiv L^\dagger, L^1 \equiv L$, a_k^i is the bath annihilation operator acting on bath mode k , L^i is the system operator, g_k is the coupling strength between bath mode k and system.

We assume the reference state ρ_R to be Gaussian. Therefore we can use Wick's rule to express the fourth order moment to second order moment as following.

$$\mathcal{P} a^i a^j a^k a^l \mathcal{P} = \mathcal{P} a^i a^j \mathcal{P} a^k a^l \mathcal{P} + \mathcal{P} a^i a^k \mathcal{P} a^j a^l \mathcal{P} + \mathcal{P} a^i a^l \mathcal{P} a^j a^k \mathcal{P}, \quad (2.71)$$

define the bath correlation functions as

$$\langle a^i(t_1) a^j(\tau_1) \rangle \equiv \sum_{m,n} \text{Tr}_{\mathbb{B}} \{ g_m g_n a_m^i(t_1) a_n^j(\tau_1) \rho_B \} \quad (2.72)$$

where $a_m^j(t)$ are annihilation operator for the bath mode m in the interaction picture. Then we can express the first few terms in the series of Eq.(2.64) in terms of the second order moment as follows.

$$\begin{aligned} K_2(t_1, t_2) \mathcal{P} \tilde{\chi}(t_1) &= \int_{t_2}^{t_1} d\tau_1 \mathcal{P} \mathcal{L}(t_1) \mathcal{L}(\tau_1) \mathcal{P} \tilde{\chi}(t_1) \\ &= \int_{t_2}^{t_1} d\tau_1 \sum_{i,j=0}^1 \langle a^j(\tau_1) a^i(t_1) \rangle [L^i(t_1), \tilde{\chi}_s(t_1) L^j(\tau_1)] \\ &\quad - \langle a^i(t_1) a^j(\tau_1) \rangle [L^i(t_1), L^j(\tau_1) \tilde{\chi}_s(t_1)], \end{aligned} \quad (2.73)$$

$$\begin{aligned}
K_4(t_1, t_2) \mathcal{P} \tilde{\chi}(t_1) &= \int_{t_2}^{t_1} d\tau_1 \int_{t_2}^{\tau_1} d\tau_2 \int_{t_2}^{\tau_2} d\tau_3 \langle \mathcal{L}(t_1) \mathcal{L}(\tau_1) \mathcal{L}(\tau_2) \mathcal{L}(\tau_3) \rangle_c \\
&= \int_{t_2}^{t_1} d\tau_1 \int_{t_2}^{\tau_1} d\tau_2 \int_{t_2}^{\tau_2} d\tau_3 \sum_{i,j,k,l=0}^1 \{ \\
&\quad \langle a^i(t_1) a^k(\tau_2) \rangle \langle a^j(\tau_1) a^l(\tau_3) \rangle [L^i(t_1), [L^j(\tau_1), L^k(\tau_2)] L^l(\tau_3) \tilde{\chi}_s(t_1)] \\
&\quad - \langle a^i(t_1) a^k(\tau_2) \rangle \langle a^l(\tau_3) a^j(\tau_1) \rangle [L^i(t_1), [L^j(\tau_1), L^k(\tau_2)] \tilde{\chi}_s(t_1) L^l(\tau_3)] \\
&\quad - \langle a^k(\tau_2) a^i(t_1) \rangle \langle a^j(\tau_1) a^l(\tau_3) \rangle [L^i(t_1), L^l(\tau_3) \tilde{\chi}_s(t_1) [L^j(\tau_1), L^k(\tau_2)]] \\
&\quad + \langle a^k(\tau_2) a^i(t_1) \rangle \langle a^l(\tau_3) a^j(\tau_1) \rangle [L^i(t_1), \tilde{\chi}_s(t_1) L^l(\tau_3) [L^j(\tau_1), L^k(\tau_2)]] \\
&\quad + \langle a^i(t_1) a^l(\tau_3) \rangle \langle a^j(\tau_1) a^k(\tau_2) \rangle ([L^i(t_1), [L^l(\tau_3), L^k(\tau_2)] \tilde{\chi}_s(t_1) L^j(\tau_1)] \\
&\quad \quad + [L^i(t_1), [L^j(\tau_1) L^k(\tau_2), L^l(\tau_3)] \tilde{\chi}_s(t_1)]) \\
&\quad + \langle a^l(\tau_3) a^i(t_1) \rangle \langle a^k(\tau_2) a^j(\tau_1) \rangle ([L^i(t_1), L^j(\tau_1) \tilde{\chi}_s(t_1) [L^l(\tau_3), L^k(\tau_2)]] \\
&\quad \quad + [L^i(t_1), \tilde{\chi}_s(t_1) [L^k(\tau_2) L^j(\tau_1), L^l(\tau_3)]]) \\
&\quad - \langle a^i(t_1) a^l(\tau_3) \rangle \langle a^k(\tau_2) a^j(\tau_1) \rangle ([L^i(t_1), [L^j(\tau_1), L^l(\tau_3)] \tilde{\chi}_s(t_1) L^k(\tau_2)] \\
&\quad - \langle a^l(\tau_3) a^i(t_1) \rangle \langle a^j(\tau_1) a^k(\tau_2) \rangle ([L^i(t_1), L^k(\tau_2) \tilde{\chi}_s(t_1) [L^j(\tau_1), L^l(\tau_3)]]) \}, \quad (2.74)
\end{aligned}$$

$$\begin{aligned}
I_2(t_1, t_2, 0, B) \mathcal{P} \tilde{\rho}(t_2) &= \int_0^{t_2} d\tau_1 \mathcal{P} \mathcal{L}(t_1) \tilde{B}(t_2) \mathcal{L}(\tau_1) \mathcal{P} \tilde{\chi}(t_1) \\
&= \int_0^{t_2} d\tau_1 \sum_{i,j=0}^1 \langle a^j(\tau_1) a^i(t_1) \rangle [L^i(t_1), \tilde{B}(t_2) \tilde{\rho}_s(t_2) L^j(\tau_1)] \\
&\quad - \langle a^i(t_1) a^j(\tau_1) \rangle [L^i(t_1), \tilde{B}(t_2) L^j(\tau_1) \tilde{\rho}_s(t_2)], \quad (2.75)
\end{aligned}$$

The first term in the fourth order inhomogeneous term $I_4(t_1, t_2, 0, B)$ can be

written as

$$\begin{aligned}
R_1(t_1, t_2) \tilde{B}(t_2) J_3(t_2, 0) \mathcal{P} \tilde{\rho}(t_2) &= \int_0^{t_2} d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \sum_{i,j,k,l=0}^1 \{ \\
&\langle a^i(t_1) a^k(\tau_2) \rangle \langle a^j(\tau_1) a^l(\tau_3) \rangle [L^i(t_1), \tilde{B}(t_2) [L^j(\tau_1), L^k(\tau_2)] L^l(\tau_3) \tilde{\rho}_s(t_2)] \\
&- \langle a^i(t_1) a^k(\tau_2) \rangle \langle a^l(\tau_3) a^j(\tau_1) \rangle [L^i(t_1), \tilde{B}(t_2) [L^j(\tau_1), L^k(\tau_2)] \tilde{\rho}_s(t_2) L^l(\tau_3)] \\
&- \langle a^k(\tau_2) a^i(t_1) \rangle \langle a^j(\tau_1) a^l(\tau_3) \rangle [L^i(t_1), \tilde{B}(t_2) L^l(\tau_3) \tilde{\rho}_s(t_2) [L^j(\tau_1), L^k(\tau_2)]] \\
&+ \langle a^k(\tau_2) a^i(t_1) \rangle \langle a^l(\tau_3) a^j(\tau_1) \rangle [L^i(t_1), \tilde{B}(t_2) \tilde{\rho}_s(t_2) L^l(\tau_3) [L^j(\tau_1), L^k(\tau_2)]] \\
&+ \langle a^i(t_1) a^l(\tau_3) \rangle \langle a^j(\tau_1) a^k(\tau_2) \rangle ([L^i(t_1), \tilde{B}(t_2) [L^l(\tau_3), L^k(\tau_2)] \tilde{\rho}_s(t_2) L^j(\tau_1)] \\
&\quad + [L^i(t_1), \tilde{B}(t_2) [L^j(\tau_1) L^k(\tau_2), L^l(\tau_3)] \tilde{\rho}_s(t_2)]) \\
&+ \langle a^l(\tau_3) a^i(t_1) \rangle \langle a^k(\tau_2) a^j(\tau_1) \rangle ([L^i(t_1), \tilde{B}(t_2) L^j(\tau_1) \tilde{\rho}_s(t_2) [L^l(\tau_3), L^k(\tau_2)]] \\
&\quad + [L^i(t_1), \tilde{B}(t_2) \tilde{\rho}_s(t_2) [L^k(\tau_2) L^j(\tau_1), L^l(\tau_3)]]) \\
&- \langle a^i(t_1) a^l(\tau_3) \rangle \langle a^k(\tau_2) a^j(\tau_1) \rangle ([L^i(t_1), \tilde{B}(t_2) [L^j(\tau_1), L^l(\tau_3)] \tilde{\rho}_s(t_2) L^k(\tau_2)] \\
&- \langle a^l(\tau_3) a^i(t_1) \rangle \langle a^j(\tau_1) a^k(\tau_2) \rangle ([L^i(t_1), \tilde{B}(t_2) L^k(\tau_2) \tilde{\rho}_s(t_2) [L^j(\tau_1), L^l(\tau_3)]]), \\
\end{aligned} \tag{2.76}$$

The second term in Eq.(2.68) is

$$\begin{aligned}
R_2(t_1, t_2) \tilde{B}(t_2) J_2(t_2, 0) \mathcal{P} \tilde{\rho}(t_2) &= \int_{t_2}^{t_1} d\tau_1 \int_0^{t_2} d\tau_2 \int_0^{\tau_2} d\tau_3 \sum_{i,j,k,l=0}^1 \{ \\
&(\langle a^i(t_1) a^k(\tau_2) \rangle \langle a^j(\tau_1) a^l(\tau_3) \rangle + \langle a^i(t_1) a^l(\tau_3) \rangle \langle a^j(\tau_1) a^k(\tau_2) \rangle) \\
&\quad [L^i(t_1), [L^j(\tau_1), \tilde{B}(t_2) L^k(\tau_2) L^l(\tau_3) \tilde{\rho}_s(t_2)]] \\
&+ (\langle a^l(\tau_3) a^i(t_1) \rangle \langle a^k(\tau_2) a^j(\tau_1) \rangle + \langle a^k(\tau_2) a^i(t_1) \rangle \langle a^l(\tau_3) a^j(\tau_1) \rangle) \\
&\quad [L^i(t_1), [L^j(\tau_1), \tilde{B}(t_2) \tilde{\rho}_s(t_2) L^l(\tau_3) L^k(\tau_2)]] \\
&- (\langle a^l(\tau_3) a^i(t_1) \rangle \langle a^j(\tau_1) a^k(\tau_2) \rangle + \langle a^i(t_1) a^k(\tau_2) \rangle \langle a^l(\tau_3) a^j(\tau_1) \rangle) \\
&\quad [L^i(t_1), [L^j(\tau_1), \tilde{B}(t_2) L^k(\tau_2) \tilde{\rho}_s(t_2) L^l(\tau_3)]] \\
&- (\langle a^i(t_1) a^l(\tau_3) \rangle \langle a^k(\tau_2) a^j(\tau_1) \rangle + \langle a^k(\tau_2) a^i(t_1) \rangle \langle a^j(\tau_1) a^l(\tau_3) \rangle) \\
&\quad [L^i(t_1), [L^j(\tau_1), \tilde{B}(t_2) L^l(\tau_3) \tilde{\rho}_s(t_2) L^k(\tau_2)]]), \\
\end{aligned} \tag{2.77}$$

The third term in Eq.(2.68) is

$$\begin{aligned}
R_3(t_1, t_2) \tilde{B}(t_2) J_1(t_2, 0) \mathcal{P} \tilde{\rho}(t_2) &= \int_{t_2}^{t_1} d\tau_1 \int_{t_2}^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \sum_{i,j,k,l=0}^1 \{ \\
&\langle a^i(t_1) a^k(\tau_2) \rangle \langle a^j(\tau_1) a^l(\tau_3) \rangle [L^i(t_1), [L^j(\tau_1), L^k(\tau_2)] \tilde{B}(t_2) L^l(\tau_3) \tilde{\rho}_s(t_2)] \\
&- \langle a^i(t_1) a^k(\tau_2) \rangle \langle a^l(\tau_3) a^j(\tau_1) \rangle [L^i(t_1), [L^j(\tau_1), L^k(\tau_2)] \tilde{B}(t_2) \tilde{\rho}_s(t_2) L^l(\tau_3)] \\
&+ \langle a^k(\tau_2) a^i(t_1) \rangle \langle a^l(\tau_3) a^j(\tau_1) \rangle [L^i(t_1), \tilde{B}(t_2) \tilde{\rho}_s(t_2) L^l(\tau_3) [L^j(\tau_1), L^k(\tau_2)]] \\
&- \langle a^k(\tau_2) a^i(t_1) \rangle \langle a^j(\tau_1) a^l(\tau_3) \rangle [L^i(t_1), \tilde{B}(t_2) L^l(\tau_3) \tilde{\rho}_s(t_2) [L^j(\tau_1), L^k(\tau_2)]] \\
&+ \langle a^i(t_1) a^l(\tau_3) \rangle \langle a^j(\tau_1) a^k(\tau_2) \rangle [L^i(t_1), [L^j(\tau_1), L^k(\tau_2)] \tilde{B}(t_2) L^l(\tau_3) \tilde{\rho}_s(t_2)] \\
&- \langle a^i(t_1) a^l(\tau_3) \rangle \langle a^k(\tau_2) a^j(\tau_1) \rangle [L^i(t_1), [L^j(\tau_1), \tilde{B}(t_2) L^l(\tau_3) \tilde{\rho}_s(t_2) L^k(\tau_2)]] \\
&+ \langle a^l(\tau_3) a^i(t_1) \rangle \langle a^k(\tau_2) a^j(\tau_1) \rangle [L^i(t_1), [L^j(\tau_1), \tilde{B}(t_2) \tilde{\rho}_s(t_2) L^l(\tau_3) L^k(\tau_2)]] \\
&- \langle a^l(\tau_3) a^i(t_1) \rangle \langle a^j(\tau_1) a^k(\tau_2) \rangle [L^i(t_1), [L^j(\tau_1), L^k(\tau_2)] \tilde{B}(t_2) \tilde{\rho}_s(t_2) L^l(\tau_3)]] \}.
\end{aligned} \tag{2.78}$$

Chapter 3

The exact two time correlation function

3.1 Hamiltonian

We consider Jaynes-Cummings (JC) model[22] with a Lorentz spectral density. This model describes a two-level systems interaction with a bath represented by a collection of harmonic oscillators. This model is exactly solvable in zero temperature when at most one excitations considered, or when the bath is replaced with a single mode of an EM field. Many researches use this model to demonstrate their new method or ideas by analysing some properties in this model. ([15, 23–25]).

The Hamiltonian of JC model can be written as

$$H = H_0 + H_I, \quad (3.1)$$

$$H_0 = \omega_0 \sigma_+ \sigma_- + \sum_{k=1}^{\infty} \omega_k a_k^\dagger a_k, \quad (3.2)$$

$$H_I = \sum_{k=1}^{\infty} g_k \{ \sigma_- a_k^\dagger + \sigma_+ a_k \}, \quad (3.3)$$

where H_I is the interaction between the system and the bath, σ_+ (σ_-) is the raising (lowering) operator of two-level system, and a_k^\dagger (a_k) is the raising (lowering) operator of the harmonic oscillator in mode k . We can define the total excitation numbers operator which measure the excitation numbers in the total system as

$$N = \sigma_+ \sigma_- + \sum_{k=1}^{\infty} a_k^\dagger a_k$$

In the JC model it is obvious that $[H, N] = 0$. Thus the total excitation number is conserved during the total system evolution. Because of this property, we can block-diagonalize the total system space into the subspace with the eigenvalue of $N = 0, 1, 2, \dots$ respectively.

3.2 Exact two time-correlation function

Because the propagator is block-diagonal with eigenvalue N , we will classify the basis with different excitation numbers. The elements of the propagator will be calculated in each subspace of eigenvalue N . The subspace can be classified by different excitation numbers as follows.

1. Zero excitation :

$$|0\rangle = |0\rangle_s \otimes \sum_{k=1}^n |0\rangle_k, \quad (3.4)$$

where the subscript of k means Bath mode with frequency ω_k , the subscript of s means system's state.

2. One excitation :

$$|1\rangle = |1\rangle_s \otimes \sum_{k=1}^n |0\rangle_k, \quad (3.5a)$$

$$|m\rangle = |0\rangle_s \otimes \sum_{k \neq m}^n |0\rangle_k \otimes |1\rangle_m, \quad (3.5b)$$

3. Two excitations :

$$|1m\rangle = |1\rangle_s \otimes \sum_{k \neq m}^{\infty} |0\rangle_k \otimes |1\rangle_m, \quad (3.6a)$$

$$|2m\rangle = |0\rangle_s \otimes \sum_{k \neq m}^{\infty} |0\rangle_k \otimes |2\rangle_m, \quad (3.6b)$$

$$|m_1, m_2\rangle = |0\rangle_s \otimes \sum_{k \neq m_1, m_2}^{\infty} |0\rangle_k \otimes |1\rangle_{m_1} \otimes |1\rangle_{m_2}, \quad (3.6c)$$

and so on.

We consider the bath initially in a vacuum state, i.e. all of the bath modes are in the ground state. In other words, it is a zero-temperature environment. We will also consider the case that at most one excitation happen in the total system. The one-excitation problem was solved in Ref.[23, 26]. The two-excitation problem was solved in Ref. [27], but we do not consider the two-excitation problem. When at most one excitation is consider, the general initial state of the total system can be written as

$$|\Phi(0)\rangle = C_0(0)|0\rangle + C_1(0)|1\rangle, \quad (3.7)$$

The state evolving to $|\Phi(t)\rangle$ in time t can be written as

$$\begin{aligned} |\Phi(t)\rangle &\equiv U(t, 0)|\Phi(0)\rangle \\ &= C_0(0)|0\rangle + C_1(t)|1\rangle + \sum_k C_k(t)|k\rangle, \end{aligned} \quad (3.8)$$

where $U(t, 0) = e^{-iHt}$ is propagator, $C_k(t) \equiv C_1(0)\langle k|U(t, 0)|1\rangle$, $C_1(t) \equiv C_1(0)\langle 1|U(t, 0)|1\rangle$ and $\langle 0|U(t, 0)|0\rangle = I$. The coefficients of $|\Phi(t)\rangle$ will be determined by the Schrödinger equation.

3.3 Exact two-time correlation function and exact QRT result

The possible two-time correlation functions of system operator for a two-level system are $\langle \sigma_{\pm}(t_1)\sigma_{\mp}(t_2)\rangle$, $\langle \sigma_{\pm}(t_1)\sigma_z(t_2)\rangle$ and $\langle \sigma_z(t_1)\sigma_{\pm}(t_2)\rangle$. In this thesis, we focus on $\langle \sigma_+(t_1)\sigma_-(t_2)\rangle$ which can be exactly evaluated by calculating the propagator with at most one excitation. Furthermore, this two-time correlation function is also important and required to calculate the spontaneous emission spectrum of the two-level system. The two time correlation functions can be evaluated as follows.

$$\begin{aligned}
\langle \sigma_+(t_1)\sigma_-(t_2) \rangle &= \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{ U^\dagger(t_1)\sigma_+U(t_1)U^\dagger(t_2)\sigma_-U(t_2)\rho(0) \} \\
&= \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{ \sigma_+U(t_1 - t_2)\sigma_-U(t_2)|\Phi(0)\rangle\langle\Phi(0)|U^\dagger(t_1) \} \\
&= \text{Tr}_{\mathbb{S}} \{ \sigma_+ \text{Tr}_{\mathbb{B}} \{ U(t_1 - t_2)\sigma_-|\Phi(t_2)\rangle\langle\Phi(t_1)| \} \}. \tag{3.9}
\end{aligned}$$

Equation (3.9) can be written as

$$\langle \sigma_+(t_1)\sigma_-(t_2) \rangle = \text{Tr}_{\mathbb{S}} \{ \sigma_+ \chi_e(t_1, t_2) \} \tag{3.10}$$

where $\chi_e(t_1, t_2) = \text{Tr}_{\mathbb{B}} \{ U(t_1 - t_2)\sigma_-|\Phi(t_2)\rangle\langle\Phi(t_1)| \}$ is the exact effective density matrix, and will be used latter. To evaluate Eq.(3.9), we note that $\sigma_-|\Phi(t)\rangle = C_1(t)|0\rangle$, $U(t_1 - t_2)|0\rangle = |0\rangle$ and then $\sigma_+|0\rangle = |1\rangle$. Then

$$\langle \sigma_+(t_1)\sigma_-(t_2) \rangle = \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{ C_1(t_2)|1\rangle\langle\Phi(t_1)| \} = C_1(t_2)C_1^*(t_1) \tag{3.11}$$

Equation (3.11) is the exact two-time correlation function of $\langle \sigma_+(t_1)\sigma_-(t_2) \rangle$. To compare the two-time correlation function with QRT, we also calculate the two-time correlation function by the QRT. The QRT says that the two-time evolution is as same as one time evolution. We denote the form of the two-time correlation function by the QRT as

$$\langle A_1(t_1)A_2(t_2) \rangle_{QRT} \simeq \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{ A_1U(t_1 - t_2)\text{Tr}_{\mathbb{B}} \{ A_2\rho(t_2) \} \otimes \rho_B U^\dagger(t_1 - t_2) \}. \tag{3.12}$$

In other words, the QRT neglects the bath correlation between $t < t_2$ and $t > t_2$ so one can trace over the bath degrees of freedom in time $0 < t < t_2$ first. But this bath correlation will in general affect the system dynamic. In Markovian systems, the system affected by bath operators between $t < t_2$ and $t > t_2$ are not correlated, and therefore QRT is a useful method to calculate the multi-time correlation functions. Using Eq.(3.12), we obtain the two-time correlation function of $\langle \sigma_+(t_1)\sigma_-(t_2) \rangle$ as

$$\begin{aligned}
\langle \sigma_+(t_1)\sigma_-(t_2) \rangle_{QRT} &= \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{ \sigma_+(t_1)\sigma_-(t_2)\rho(0) \} \\
&= \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{ \sigma_+ U(t_1 - t_2) \text{Tr}_{\mathbb{B}} \{ \sigma_- \rho(t_2) \} \otimes (|0\rangle\langle 0|) U^\dagger(t_1 - t_2) \} \\
&= \text{Tr}_{\mathbb{S} \otimes \mathbb{B}} \{ \sigma_+ U(t_1 - t_2) [|C_1(t_2)|^2 |0\rangle\langle 1| + C_1(t_2)C_0^*(0)|0\rangle\langle 0|] U^\dagger(t_1 - t_2) \} \\
&= |C_1(t_2)|^2 C_1^*(t_1 - t_2), \tag{3.13}
\end{aligned}$$

In this thesis, we will call the result of Eq.(3.13) as the exactQRT result. We can see that the exact two-time correlation function Eq.(3.11) and the exact QRT Eq.(3.13) have very different structure.

3.4 Evaluation of $C_1(t)$

To evaluate $C_1(t)$, we solve Schrödinger equation in interaction picture

$$i \frac{d|\tilde{\Phi}(t)\rangle}{dt} = \tilde{H}_I(t)|\tilde{\Phi}(t)\rangle, \tag{3.14}$$

$$|\tilde{\Phi}(t)\rangle = e^{iH_0 t} |\Phi(t)\rangle, \tilde{H}_I(t) = e^{-iH_0 t} H_I e^{iH_0 t} \tag{3.15}$$

Inserting the Eq(3.8) into the Eq.(3.14) and then separating the vector differential equation to a set of coupled differential equations, we obtain

$$\frac{d\tilde{C}_1(t)}{dt} = -i \sum_{k=1}^{\infty} g_k \tilde{C}_k(t) e^{i(\omega_0 - \omega_k)t} \tag{3.16}$$

$$\frac{d\tilde{C}_k(t)}{dt} = -i g_k \tilde{C}_1(t) e^{-i(\omega_0 - \omega_k)t} \tag{3.17}$$

We directly integrate of Eq.(3.17) and use the initial environment state being in the vacuum state ($\tilde{C}_k(0) = 0$) which we required is Eq.(3.7), we obtain the formal solution of $\tilde{C}_k(t)$ as

$$\tilde{C}_k(t) = -i g_k \int_0^t d\tau \tilde{C}_k(\tau) e^{-i(\omega_0 - \omega_k)\tau}, \tag{3.18}$$

Then insert Eq. (3.18) to the Eq.(3.16), we can get an uncoupled equation of motion for $\tilde{C}_1(t)$.

$$\frac{d\tilde{C}_1(t)}{dt} = - \sum_{k=1}^{\infty} |g_k|^2 \int_0^t d\tau \tilde{C}_1(\tau) e^{i(\omega_0 - \omega_k)(t-\tau)}, \quad (3.19)$$

Defining $f(t - \tau) = \sum_k |g_k|^2 e^{i(\omega_0 - \omega_k)(t-\tau)}$ then Eq.(3.19) become

$$\frac{d\tilde{C}_1(t)}{dt} = - \int_0^t d\tau f(t - \tau) \tilde{C}_1(\tau), \quad (3.20)$$

The summation over k in $f(t - \tau)$ can be changed to a continuum integral over ω through the spectral density $J(\omega) = \sum_k |g_k|^2 \delta(\omega - \omega_k)$. Thus we have

$$f(t - \tau) = \sum_{k=1}^{\infty} |g_k|^2 e^{i(\omega_0 - \omega_k)(t-\tau)} = \int_0^{\infty} d\omega J(\omega) e^{i(\omega_0 - \omega)(t-\tau)} \quad (3.21)$$

We consider the bath spectral density is Lorentz form

$$J(\omega) = \frac{\gamma_0}{2\pi} \frac{\lambda^2}{(\omega - \omega_0)^2 + \lambda^2}, \quad (3.22)$$

where γ_0 is decay rate (in Markovian case), λ is cutoff frequency. The cutoff frequency λ is an important parameter in the non-Markovian system, because the bath correlation time τ_B is about $\tau_B \simeq \lambda^{-1}$. When the bath correlation time $\tau_B \rightarrow 0$, one has a Markovian system. Otherwise, one has a non-Markovian system. We assume system frequency ω_0 is larger than spectral width λ ($\omega_0 \gg \lambda$), then we can extend the lower limit of the integral to infinity (i.e., $\int_{-\omega_0}^{\infty} \rightarrow \int_{-\infty}^{\infty}$) to obtain $f(t - \tau)$. Substituting Eq.(3.22) into Eq.(3.21) we obtain

$$\begin{aligned} f(t - \tau) &= \int_{-\omega_0}^{\infty} d\omega \frac{e^{-i\omega(t-\tau)}}{\omega^2 + \lambda^2} = \frac{\gamma\lambda^2}{2\pi} \int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega(t-\tau)}}{\omega^2 + \lambda^2} \\ &= \frac{\gamma\lambda}{2} e^{-\lambda(t-\tau)} \end{aligned} \quad (3.23)$$

Inserting Eq.(3.23) to Eq.(3.17) and differentiating the resultant integro-differential equation again, we can obtain a second order ordinary differential equation

$$\ddot{\tilde{C}}_1(t) + \lambda \dot{\tilde{C}}_1(t) + \frac{\gamma_0\lambda}{2} \tilde{C}_1(t) = 0, \quad (3.24)$$

with initial conditions

$$\tilde{C}_1(0) = C_1(0), \dot{\tilde{C}}_1(0) = 0. \quad (3.25)$$

Solving the above equation, we obtain

$$C_1(t) = C_1(0)e^{-\lambda t/2}(\cosh(dt/2) + \frac{\lambda}{d} \sinh(dt/2))e^{-i\omega_0 t}, \quad (3.26)$$

for $\lambda > 2\gamma_0$ where $d = \sqrt{\lambda^2 - 2\lambda\gamma_0}$.

Inserting Eq.(3.26) to Eq.(3.11), we obtain for $\lambda > 2\gamma_0$ the exact two-time correlation function as

$$\begin{aligned} \langle \sigma_+(t_1)\sigma_-(t_2) \rangle &= |C_1(0)|^2 e^{i\omega_0(t_1-t_2) - \frac{\lambda}{2}(t_1+t_2)} \\ &* (\cosh(\frac{dt_1}{2}) + \frac{\lambda}{d} \sinh(\frac{dt_1}{2})) (\cosh(\frac{dt_2}{2}) + \frac{\lambda}{d} \sinh(\frac{dt_2}{2})). \end{aligned} \quad (3.27)$$

Similarly for $\lambda < 2\gamma_0$ case, the two-time correlation function can be obtained as

$$\begin{aligned} \langle \sigma_+(t_1)\sigma_-(t_2) \rangle &= |C_1(0)|^2 e^{i\omega_0(t_1-t_2) - \frac{\lambda}{2}(t_1+t_2)} \\ &* (\cos(\frac{dt_1}{2}) + \frac{\lambda}{d} \sin(\frac{dt_1}{2})) (\cos(\frac{dt_2}{2}) + \frac{\lambda}{d} \sin(\frac{dt_2}{2})), \end{aligned} \quad (3.28)$$

where $d = \sqrt{2\lambda\gamma_0 - \lambda^2}$.

Chapter 4

The exact master equation of the reduced effective density matrix in the JC model

4.1 Master equation of the JC model

In this section, we insert the Jaynes-Cumming model (3.1) to the general master equation (2.73 - 2.78) with a zero temperature bath reference state.

The Jaynes-Cumming model's Hamiltonian in the interaction picture is

$$\tilde{H}_I(t) = \sum_k g_k \{ \sigma_- a_k^\dagger e^{-i(\omega_0 - \omega_k)t} + \sigma_+ a_k e^{-i(\omega_0 - \omega_k)t} \}. \quad (4.1)$$

Comparing above equation with Eq.(2.70), we can find the system and bath operator $L^i(t)$ and $a_k^j(t)$ as following

$$L^0(t) = \sigma_- e^{-i\omega_0 t}, L^1(t) = \sigma_+ e^{i\omega_0 t}; \quad (4.2)$$

$$a_k^0(t) = a_k^\dagger e^{i\omega_k t}, a_k^1(t) = a_k e^{-i\omega_k t}, \quad (4.3)$$

The reference state ρ_B was set to be a zero-temperature vacuum state, i.e., $\rho_B = \sum_{kq} (|0\rangle_{kq} \langle 0|)$ which means all the bath modes are in the ground state. The bath spectral $J(\omega)$ is in a Lorentz form in (3.22). The bath correlation functions Eq. (2.72) can be calculated.

$$\begin{aligned}
\langle a(t_1)a^\dagger(\tau_1) \rangle &= \sum_{m,n} \text{Tr}_{\mathbb{B}} \{ g_m g_n a_m(t_1) a_n^\dagger(\tau_1) \sum_{kq} (|0\rangle_{kq} \langle 0|) \} \\
&= \int_0^\infty d\omega J(\omega) e^{-i\omega(t_1-\tau_1)} = \frac{\gamma_0 \lambda}{2} \exp(-\lambda|t_1 - \tau_1|) e^{i\omega_0(t_1-\tau_1)}, \quad (4.4)
\end{aligned}$$

Here, to obtain the final result of Eq.(4.4) we have also extended the integral limits from $(0, \infty)$ to $(-\infty, \infty)$ by assuming $\omega_0 \gg \lambda$. Setting $L = \sigma_-$ and substituting the bath correlation function Eq.(4.4) into Eqs.(2.73) - (2.78), we then obtain from Eq.(2.64) the perturbation master equation of the reduced density matrix up to fourth order in the JC model with

$$K_2(t_1, t_2) \tilde{\chi}_s(t_1) = \Delta\gamma_2(t_1 - t_2) \{ \sigma_- \tilde{\chi}_s(t_1) \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \tilde{\chi}_s(t_1) \}_+ \}, \quad (4.5)$$

$$K_4(t_1, t_2) \tilde{\chi}_s(t_1) = \Delta\gamma_4(t_1 - t_2) \{ \sigma_- \tilde{\chi}_s(t_1) \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \tilde{\chi}_s(t_1) \}_+ \}, \quad (4.6)$$

$$I_2(t_1, t_2, 0, \sigma_-) \tilde{\rho}_s(t_2) = \Delta i_2(t_1, t_2) \frac{-1}{2} (\tilde{\sigma}_-(t_2) \tilde{\rho}_s(t_2)) \sigma_+ \sigma_-, \quad (4.7)$$

$$I_4(t_1, t_2, 0, \sigma_-) \tilde{\rho}_s(t_2) = \Delta i_4(t_1, t_2) \frac{-1}{2} (\tilde{\sigma}_-(t_2) \tilde{\rho}_s(t_2)) \sigma_+ \sigma_-, \quad (4.8)$$

where

$$\Delta\gamma_2(t_1 - t_2) = \gamma_0 (1 - e^{-\lambda(t_1-t_2)}) \quad (4.9)$$

$$\Delta\gamma_4(t_1 - t_2) = \frac{\gamma_0^2}{\lambda} e^{-\lambda(t_1-t_2)} (\sinh(\lambda(t_1 - t_2)) - \lambda(t_1 - t_2)) \quad (4.10)$$

are decay rates coming from homogeneous terms in 2nd order and 4th order, respectively, and.

$$\Delta i_2(t_1, t_2) = \gamma_0 e^{-\lambda(t_1-t_2)} (1 - e^{-\lambda t_2}) \quad (4.11)$$

$$\begin{aligned}
\Delta i_4(t_1, t_2) &= \frac{\gamma_0^2}{\lambda} \{ e^{-\lambda t_1} [\sinh(\lambda t_2) - \lambda t_2] \\
&\quad + e^{-\frac{3}{2}\lambda(t_1-t_2)} [1 - e^{-\lambda t_2}] [\frac{\lambda}{2}(t_1 - t_2) - \sinh(\frac{\lambda}{2}(t_1 - t_2))] \} \quad (4.12)
\end{aligned}$$

are effective decay rates coming from inhomogeneous terms. The effective decay rates are strongly depend on memory effect.

The exact master equation of the system density matrix, which multiple by a system operator and the traced over the system states lead to the evolution equation of the single-time had found in Ref.[10, 23, 28, 29].

In the following, we derive an exact master equation for effective density matrix $\chi_e(t)$. The exact master equation can be derived from the exact operator method. First, we can evaluate the exact effective density matrix $\chi_e(t_1, t_2)$ from Eq.(3.10) following the similar procedure to evaluate the exact two-time correlation function of Eq.(3.11). We then obtain

$$\begin{aligned}\tilde{\chi}_e(t_1, t_2) &= Tr_{\mathbb{B}}\{\tilde{U}(t_1 - t_2)\tilde{\sigma}_-(t_2)\tilde{U}(t_2)(|\Phi(0)\rangle\langle\Phi(0)|)\tilde{U}^\dagger(t_1)\} \\ &= \begin{bmatrix} 0 & 0 \\ \tilde{C}_1(t_1)\tilde{C}_1(t_2) & \tilde{C}_1(t_2)C_0 \end{bmatrix} e^{-i\omega_0 t_2}.\end{aligned}\quad (4.13)$$

Taking a derivative of Eq.(4.13) with respect to time and observing the operator forms of Eqs.(4.5)-(4.8), we find that the exact effective density matrix $\tilde{X}_e(t_1, t_2)$ satisfies the following master equation

$$\begin{aligned}\frac{\partial}{\partial t_1}\tilde{\chi}_e(t_1, t_2) &= \gamma(t_1 - t_2)\{\sigma_- \tilde{\chi}_e \sigma_+ - \frac{1}{2}\{\sigma_+ \sigma_-, \tilde{\chi}_e\}_+\} \\ &\quad + i(t_1, t_2)\frac{-1}{2}\tilde{\sigma}_-(t_2)\tilde{\rho}_s(t_2)\sigma_+ \sigma_-, \end{aligned}\quad (4.14)$$

where

$$\gamma(t) = \frac{2\gamma_0\lambda \sinh(dt/2)}{d \cosh(dt/2) + \lambda \sinh(dt/2)}\quad (4.15)$$

$$i(t_1, t_2) = (\gamma(t_1) - \gamma(t_1 - t_2))\frac{\tilde{C}_1(t_1)}{\tilde{C}_1(t_2)}\quad (4.16)$$

are the exact decay rate and effective decay rate.

Multiplied by the operator $\tilde{\sigma}_+(t_1)$ and trace over the system state, Eq.(4.14) then leads to the exact evolution equation for the two-time correlation function $\langle\sigma_+(t_1)\sigma_-(t_2)\rangle$. The structure of the exact master equation is the same as the perturbation result of Eqs.(4.5)-(4.8). The only difference is the time-dependent coefficient. The time dependent coefficient $\Delta\gamma_2(t)$ and $\Delta\gamma_4(t)$ are the first two terms of the Taylor expansion of $\gamma(t, \gamma_0)$ in power of γ_0 . $\Delta i_2(t_1, t_2)$ and $\Delta i_4(t_1, t_2)$ are also the first two terms of the Taylor expansion of $i(t_1, t_2, \gamma_0)$ in power of γ_0 .

4.2 Aitken's δ^2 method

In previous chapter, we had seen the perturbation of higher order need calculate many terms. We can see from Eqs.(2.73)-(2.78) that the second order expansion of the master equation for effective density matrix $\tilde{\chi}_s$ contains near 30 terms, and the fourth order expansion of master equation contains near 600 terms. We can expect that the sixth order contains at least thousands of terms, and the calculation would become very tedious and complicated. Therefore, we provide an easier method, which can improve the accuracy of the perturbation time-dependent decay rates without really going to calculate the higher order contributes of K_n and I_n .

The method is Aitken's delta-squared method. It is a numerical method used for accelerate the rate of convergence of the sum of a series. Aitken's delta-squared method can be described as follows. Suppose $S_n = \sum_{i=0}^n X_i$ is a partial sum of X_i to the n th term of a slowly convergent sequence where exact result is achieved when $n \rightarrow \infty$. The new sequence S'_n transformed by Aitken's δ^2 method will converges faster or closer to the exact result than S_n does. The expression of the new sequence is form from S_n and previous two sequence S_{n-1} and S_{n-2} as

$$S'_n = S_n - \frac{(S_n - S_{n-1})^2}{S_n - 2S_{n-1} + S_{n-2}}. \quad (4.17)$$

In one case, we have calculate the decay rate up to fourth order to obtain $\gamma_4(t) = \Delta\gamma_2(t) + \Delta\gamma_4(t)$. If we set the 0th and 2nd order decay rates to be $\gamma_0(t) = 0$ and $\gamma_2(t) = \Delta\gamma_2(t)$, we may apply Aitken's δ^2 method to find a new decay rate as

$$\gamma'_4(t) = \gamma_4(t) - \frac{(\gamma_4(t) - \gamma_2(t))^2}{\gamma_4(t) - 2\gamma_2(t) + 0}. \quad (4.18)$$

Similarly, the effective 0th, 2nd and 4th order decay rates $i_0(t_1, t_2) = 0$, $i_2(t_1, t_2) = \Delta i_2(t_1, t_2)$ and $i_4(t_1, t_2) = i_2(t_1, t_2) + \Delta i_4(t_1, t_2)$. One can also apply Aitken's δ^2 method to find a new effective decay rate

$$i'_4(t_1, t_2) = i_4(t_1, t_2) - \frac{(i_4(t_1, t_2) - i_2(t_1, t_2))^2}{i_4(t_1, t_2) - 2i_2(t_1, t_2) + 0}. \quad (4.19)$$

In Fig. 4.1(a), we show the decay rates calculated by different methods for $\lambda = 2.001\gamma_0$ (in the weak-coupling region). The black solid line is exact decay obtained from Eq. (4.15), the blue dotted line is the 2nd-order decay rate $\gamma_2(t)$, the green dot-dashed line is the 4th order decay rate $\gamma_4(t)$, and the red dashed line is decay rate $\gamma'_4(t)$ obtained by Aitken's δ^2 method. It is obvious that the 4th order perturbation result is better than the 2nd order one, and the Aitken's δ^2 method can improve the decay rate as the result obtained from it is closer to the exact result than the 4th order perturbation. In section 5.3, we apply Aitken's δ^2 method to perturbative master equation up to 4th order and then to obtain the two-time correlation function.

Figures 4.1(b) - 4.1(d) show the effective decay rates with different value of t_2 . One can see that the decay time of the effective decay rate is about $\tau_B \sim \lambda^{-1}$ which is the bath correlation time. The strength of the effective decay rate dependent strongly on t_2 . When t_2 is small, the strength of effective decay rate is also small. When t_2 increases, the strength of the effective decay rate also increase, but it would reach a steady state and will not increase any more at largest t_2 .

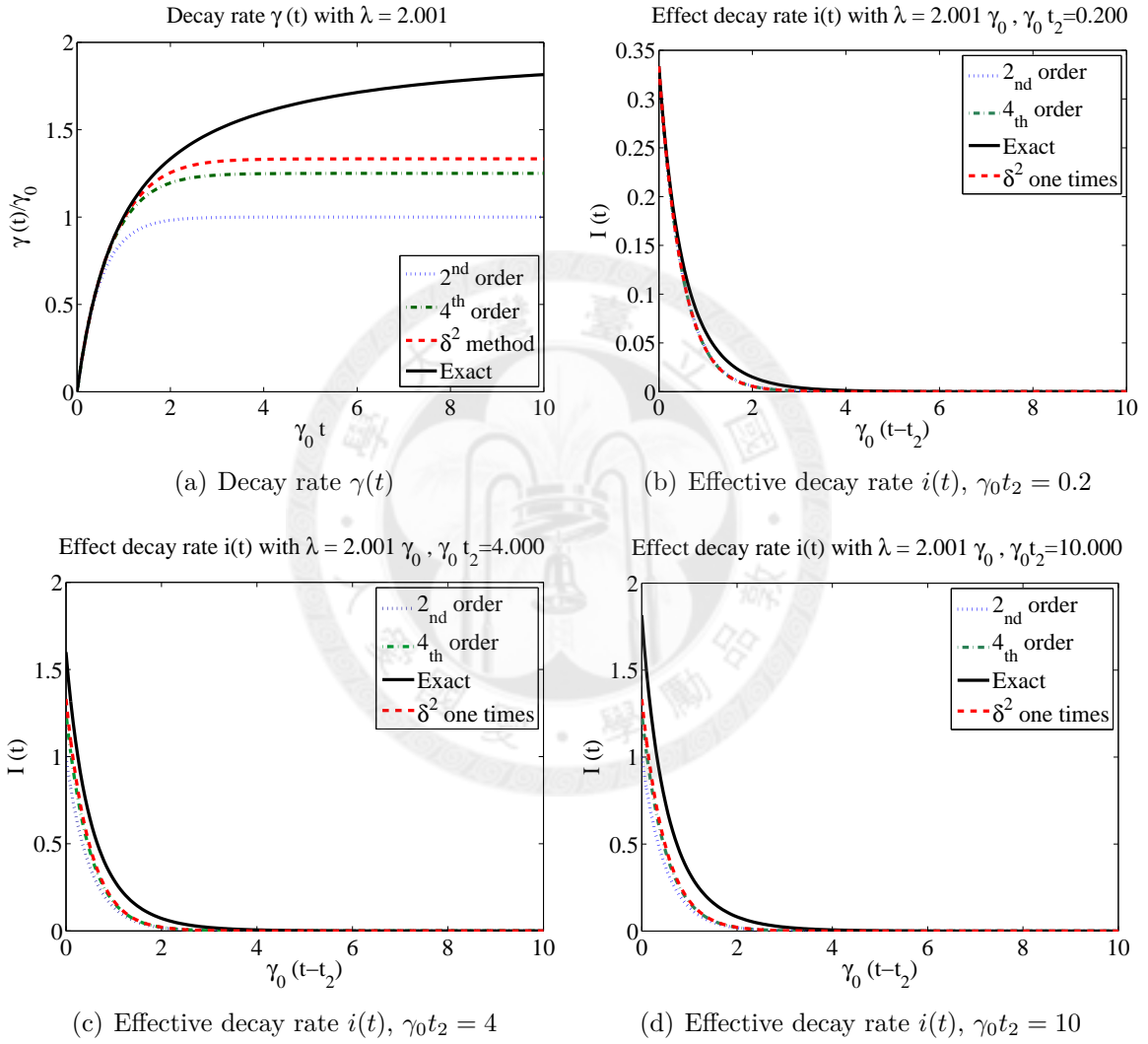


Figure 4.1: Time-dependent decay rates and effective rates obtained by different methods.

Chapter 5

Comparison between the exact result and the perturbation results

With the various master equation for the reduced density matrix \tilde{X}_s obtained, we can find the solution of $\tilde{\chi}_s$ then trace the product of $\tilde{\sigma}_+\tilde{\chi}_s(t_1, t_2)$ over the system state to obtain the two-time correlation function $\langle\sigma_+(t_1)\sigma_-(t_2)\rangle$. In this chapter, we will show the time evolution of two-time correlation function $\langle\sigma_+(t_1)\sigma_-(t_2)\rangle$ obtained using different methods.

To eliminate the oscillating factor of $e^{i\omega_0(t_1-t_2)}$ and to make the time evolution behaviors clearly the absolute value of two-time correlation function ($|\langle\sigma_+(t_1)\sigma_-(t_2)\rangle|$) illustrated, we plot in all the figures shown in this Chapter.

The different methods and corresponding time evolution were shown in the figures are summarized below. The first method is the perturbative master equation approach for the reduced effective density matrix. The time-evolutions calculated using Eq.(2.64) with different perturbation order are presented. We denote K_2 with I_2 in black dashed line as calculation using Eq.(2.64) with homogeneous and inhomogeneous terms up to 2nd order, K_4 with I_2 in green dotted line as with homogeneous terms up to 4th order and inhomogeneous terms up to 2nd order, K_4 with I_4 in purple solid line as with homogeneous and inhomogeneous terms up to 4th order. We also plot the Markovian time evolution to 2nd order in blue dot-solid line as *Markovian*. The second method is the exact direct evaluation by operator technique. The time evolution obtained by the exact result Eq.(3.11) in red solid line denoted as *Exact*. Another result obtained by Eq.(3.13) that neglects the bath correlation between $t < t_2$

and $t > t_2$ but treats the reduce time evolution from t_2 to t_1 exactly is plotted in pink solid line and demoted as *Exact QRT*

The initial states of the environments is in the zero-temperature vacuum state, $\sum_k |0\rangle_k$, and the initial system state is set to be $|\phi(0)\rangle = \frac{1}{\sqrt{2}}(|0\rangle_s + |1\rangle_s)$

5.1 Numerical result in the weak coupling region of $\lambda > 2\gamma_0$

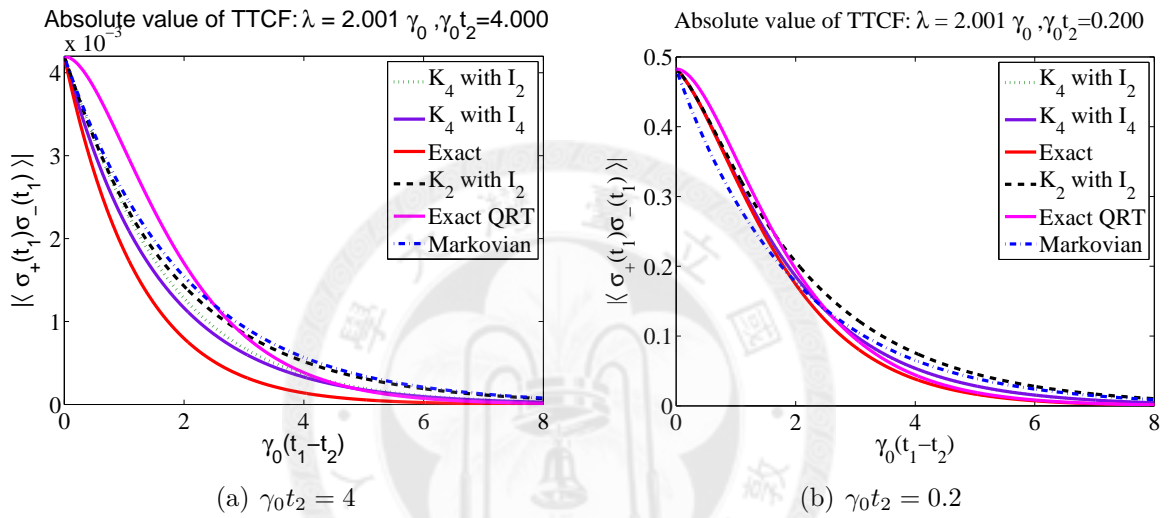


Figure 5.1: Two-time correlation functions of $|\langle \sigma_+(t_1)\sigma_-(t_2) \rangle|$ obtained by different methods with $\lambda = 2.001\gamma_0$ for different value of (a) $t_2 = 4\gamma_0$, (b) $0.1\gamma_0$ respectively. In Fig. (a), when we consider perturbation of homogeneous and inhomogeneous term up to 4th order, the result is better than exact QRT case, even 2nd order perturbation it is also better than exact QRT in short time region. In Fig. (b), if the t_2 is not large enough do not have enough memory about the time before $t < t_2$ (i.e. $t_2 < \lambda^{-1}$), QRT is applicable. The initial condition of $\tilde{X}_s(t_2)$ was obtained by exact operator method, it makes the contribution of inhomogeneous terms clear.

In this section, we consider the region with $\lambda > 2\gamma_0$ (referred to as the weak coupling region). Specifically, we choose the cutoff frequency $\lambda = 2.001\gamma_0$. In this region, the two-time correlation functions will decrease monotonically.

We can see from Fig.5.1(a) that the difference between the exact result and the result by the exact QRT method is obvious. The reason is that the QRT that neglects the bath correlation between $t < t_2$ and $t > t_2$ does not consider the non-Markovian memory effect of the bath comes from $t < t_2$ that may affect the system

dynamics in $t > t_2$.

Next, we compare the perturbation results in Fig.5.1(a). The two-time correlation function obtained by perturbation method with homogeneous and inhomogeneous term up to 4th order is closer to the result by the exact operator evaluation than the exact QRT, which demonstrates clearly the validity of the evolution equation Eq.(2.64).

As expected, the result of K_4 with I_4 is more accurate than the result of K_2 with I_2 . One can also observe that even the second order perturbation result with inhomogeneous contribution is better than the exact QRT in the short time region. After $\gamma_0 t > 3$ the inhomogeneous contribution dies out as shown in Fig.4.1(c), the exact QRT result is then close to the exact result. The Markovian result also seems better than the exact QRT in the short time region. This is because Markovian result assumes a time-dependent decay rate $\gamma_M \sim \gamma_2(t \rightarrow \infty)$, so it has a large decay rate than all other cases in the short time region.

The more high-order terms are considered, the more accuracy the results are. However, to include the higher-order perturbation contribution requires much more tedious calculations. An alternative scheme of Aitken's δ^2 method to improve accuracy introduced in Sec.4.2, will be discussed in Sec.5.3.

The difference between K_4 with I_4 and K_4 with I_2 is that K_4 with I_4 contains the 4th order contribution of the inhomogeneous terms. We can see from Fig.4.1(c) that the contribution of $\Delta i_4(t_1, t_2)$ is very small. Furthermore, the bath correlation time or memory time is about $\tau_B \sim \lambda^{-1}$, so the contribution of the inhomogeneous terms becomes less affected $t = t_1 - t_2 > \lambda^{-1}$

The difference between the Exact result and the Exact QRT result is not significant. For small t_2 , the inhomogeneous contribution from $t < t_2$ is expected to be small. This can be seen from Fig.4.1(b) and Fig.4.1(c) that the magnitude of the effective decay rate $i(t)$ coming from inhomogeneous contribution for $\gamma_0 t = 0.2$ is about 5 times smaller than that for $\gamma_0 t = 4$ case. Thus we conclude that for $t_2 \ll \lambda^{-1}$ and for $\tau_B \sim \lambda^{-1} \ll \gamma_0^{-1}$, the memory effect of the bath coming from

the $t < t_2$ to affect the system dynamics in $t > t_2$ will be small and thus the QRT is valid in this case.

5.2 Numerical result in the Strong coupling region of $\lambda < 2\gamma_0$

In this section, we discuss the strong coupling region, i.e. $\lambda < 2\gamma_0$. Specifically, $\lambda = 0.05\gamma_0$ is chosen.

The exact decay rate in the strong coupling region of $\lambda < 2\gamma_0$ from Eq.(4.15) can be written as

$$\gamma(t) = \frac{2\gamma_0\lambda \sin(dt/2)}{\lambda \sin(dt/2) - d \cos(dt/2)}, \quad (5.1)$$

with $d = \sqrt{2\gamma_0\lambda - \lambda^2}$ would become positive and negative infinity at time near $t = t_{div}$ as

$$t_{div} \forall \lambda \sin(dt_{div}/2) - d \cos(dt_{div}/2) = 0. \quad (5.2)$$

The positive infinity decay rate would make the population of the excite state of two-level system falls into zero suddenly, and then the negative decay rate will cause a sudden birth to the the population of the excite state of the system.

In principle , the perturbation theory described in previous chapter can not be applied to this strong coupling region of $\lambda < 2\gamma_0$ as the decay rate will diverge at time $t = t_{div}$. For the parameter of $\lambda = 0.05\gamma_0$ chosen, the divergent time of the decay rate is about $\gamma_0 t_{div} \simeq 11.08$. Nevertheless, let us plot the two-time correlation function $|\langle \sigma_+(t_1)\sigma_-(t_2) \rangle|$ in Fig.5.2 using the perturbative master equation approach in short time region and compare the results with those by exact evaluation. Bear in mind that the perturbation result is only possibly to be valid before the exact decay rate become very large.

For the correlation function $|\langle \sigma_+(t_1)\sigma_-(t_2) \rangle|$ at $t_2 = 0$, Fig.5.2(a) investigates the single-time expectation value and this can treat the validity of the perturbative mas-

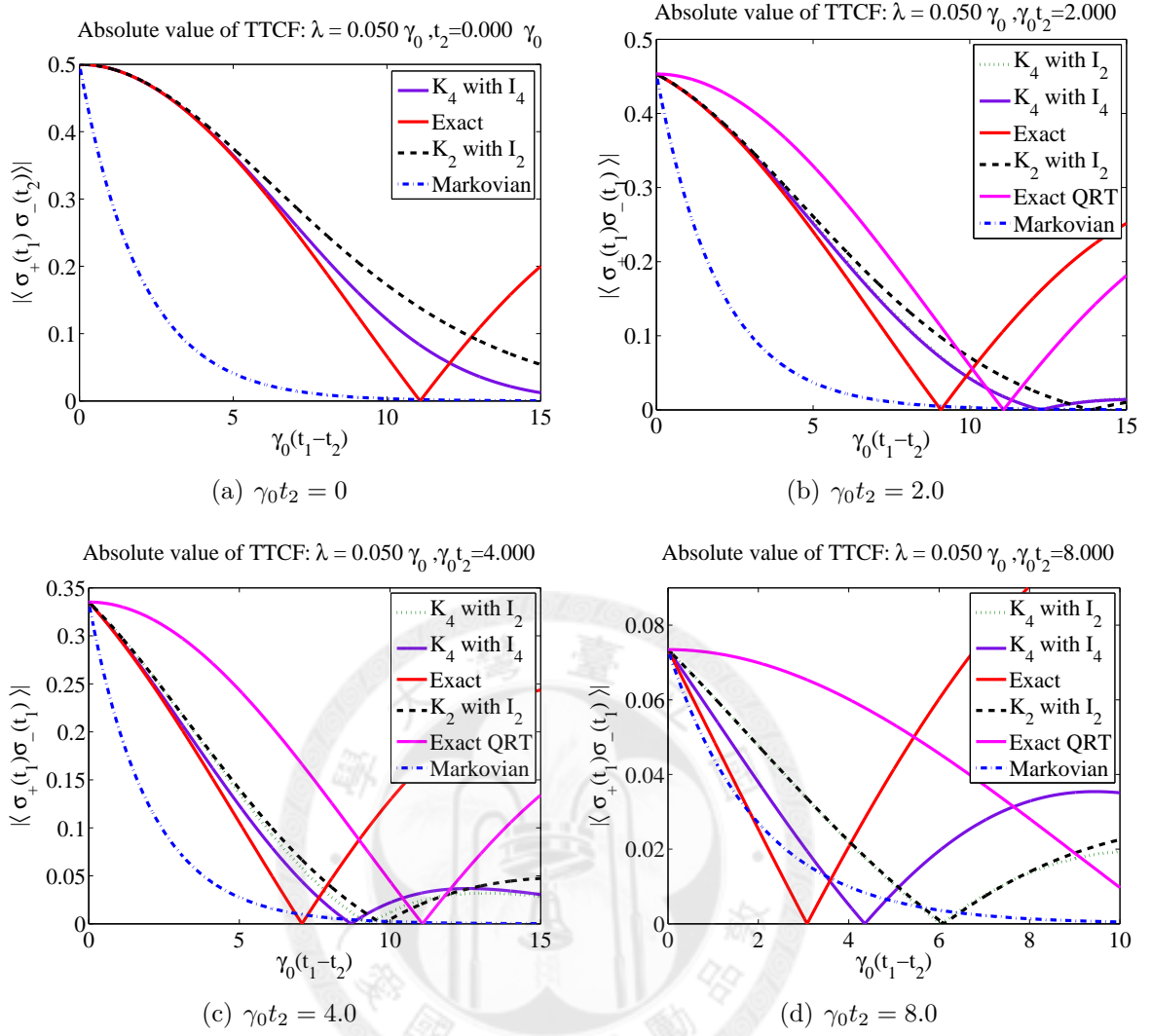


Figure 5.2: Time evolution of the two-time correlation functions for different values of t_2 with $\lambda = 0.05\gamma_0$.

ter equation approach in the short time region since the inhomogeneous contribution are zero for $t_2 = 0$. It is obvious that the result of the Markovain approximation is pretty bad in this strong coupling region. The non-Markovain second order perturbation result seems to close to the exact result for $\gamma_0 t < 5$ and the fourth order seem to be valid for $\gamma_0 t < 8$.

In the previous chapter we had required the series of perturbation master equation (2.64) should be bounded and converged. However, the infinity of decay rate violate the requirement, and therefore the prediction of perturbation method made would be failed. The divergent point of decay rate is at $t_1 = 11.08$.

Next, we investigate the time evolution of the two-time correlation function.

The behaviors of the exact result of the two-time correlation function shown in Figs.5.2(b)-5.2(d). We can also observe that in the short time region all the perturbation with inhomogeneous contributions are much better than those of the exact QRT. As expected, result of K_4 with I_4 is closer to the exact result than the other methods. For large t_2 value in Fig.5.2(c) and Fig.5.2(d), the result of K_2 with I_2 and K_4 with I_2 are closer to each other than to K_4 with I_4 . Thus the homogeneous contribution of K_4 is smaller than the inhomogeneous contribution of I_4 . This again indicates that the non-Markovian memory effect is considerable and important.

5.3 Numerical result of applying Aitken's δ^2 method

The Aitken's delta-squared method was introduced in section 4.2, where we assumed the form of higher order master equation differs only in the by time-dependent coefficients. We applied the Aitken's δ^2 method to the first three partial sums of the decay rates, $\{0, \gamma_2(t), \gamma_4(t) = \gamma_2(t) + \Delta\gamma_4(t)\}$, to obtain a better new decay rate $\gamma'_4(t)$. Similar procedure was applied to the effect decay rate coming from the inhomogeneous contribution.

In Fig.5.3, we compare the results of two-time correlation function obtained by applying the Aitken's δ^2 method with those by K_4 with I_4 and by the exact operator evaluation.

We can see from Fig.5.3(a) with $\lambda = 2.001\gamma_0$ and Fig.5.3(b) that the delta-squared method can slightly improve the accuracy of perturbation method. However, we should be cautious to apply the Aitken's δ^2 method in the strong coupling region of $\lambda < 2\gamma_0$ as in the case of Fig.5.3(b). Since at large time the decay rate may diverge and the Aitken's δ^2 method may not be applicable. Nevertheless, Aitken's δ^2 method is an easy way to obtain a better convergent result in a sequence in the problem or regime for which it is valid

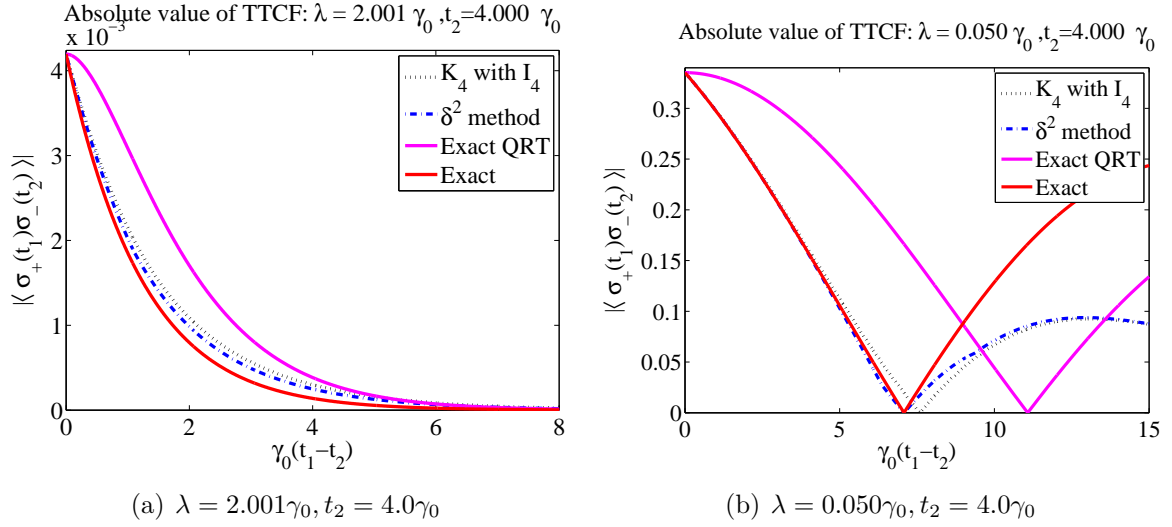


Figure 5.3: Time-evolution of the two-time correlation function $|\langle \sigma_+(t_1)\sigma_-(t_2) \rangle|$ for different value of (a) $\lambda = 2.001\gamma_0$, (b) $\lambda = 0.05\gamma_0$. The value of t_2 is $\gamma_0 t_2 = 4.0$

5.4 Spontaneous emission spectrum

The spectrum of spontaneous emission in the two-level system can be evaluated through the two-time correlations functions $\langle \sigma_+(t_1)\sigma_-(t_2) \rangle$. The spectrum of emission photons is the double Fourier transform of $\langle \sigma_+(t_1)\sigma_-(t_2) \rangle$ [4]

$$S(\omega) \propto \int_0^\infty dt_1 \int_0^\infty dt_2 e^{-i\omega(t_1-t_2)} \langle \sigma_+(t_1)\sigma_-(t_2) \rangle. \quad (5.3)$$

Substituting the exact expression of Eq.(3.27) and Eq.(3.28) into Eq.(5.3), we obtain respectively the spectrum of the Jaynes-Cummings model in the weak coupling region ($\lambda > 2\gamma_0$) as

$$S_w(\omega) \propto \frac{[(\omega - \omega_0)^2 + \lambda^2]}{[(\omega - \omega_0)^2 + (\frac{\lambda-d}{2})^2][(\omega - \omega_0)^2 + (\frac{\lambda+d}{2})^2]}, \quad (5.4)$$

and in strong coupling region ($\lambda < 2\gamma_0$) as

$$S_s(\omega) \propto \frac{[(\omega - \omega_0)^2 + \lambda^2]}{[(\omega - \omega_0 + \frac{d}{2})^2 + (\frac{\lambda}{2})^2][(\omega - \omega_0 - \frac{d}{2})^2 + \frac{\lambda^2}{2}]}. \quad (5.5)$$

In Fig.5.4(a), show the spontaneous emission spectra for different value of λ in the weak coupling region. The peaks of spectra are at the system frequency ω_0 . The widths of the spectra are determined by cut-off frequency λ . Where the λ increase, the width becomes a little bit narrower. When $\lambda \rightarrow \infty$, the spectra reach

the Markovian result $S(\omega) \propto [(\omega - \omega_0)^2 + (\gamma_0/2)^2]^{-1}$ [4]. The only difference between the non-Markovian and the Markovian spectra is the width.

However, the spontaneous emission spectra are more interesting in the strong coupling. Fig.5.4(b), shows the spontaneous emission spectra for different value of λ in the strong coupling region ($\lambda < 2\gamma_0$). Note that the vertical axis in Fig.5.4(b) is in logarithmic scale. When the values of the cutoff frequency decrease, the spectrum from a single-peak structure centered at $\omega = \omega_0$ to a double-peak structure centered at $\omega = \omega_0 \pm \frac{d}{2}$, where $d = \sqrt{2\gamma_0\lambda - \lambda^2}$. There exists a critical cutoff frequency λ_c at which the second derivative of $S(\omega)$ at $\omega = \omega_0$ is zero, i.e., $\frac{d^2S(\omega)}{d\omega^2}|_{\omega_0, \lambda_c} = 0$. When the cutoff frequency is smaller than $\lambda_c \simeq 1.2\gamma_0$, the two-peaks structure starts to develop.

The peak structure of spectrum may be understood from the two-time correlation function. Figure 5.4(c) shows a typical time-evolution of $\text{Re}\langle\sigma_+(t_1)\sigma_-(t_2)\rangle$ oscillating with the frequency ω_0 in the weak coupling region. The monotonically decay of the envelope of the two-time correlation function explains the spontaneous emission spectra in this region at $\omega = \omega_0$. In contrast the envelope of, a typical time-evolution for $\lambda < \lambda_c$ shown in Fig.5.4(d) is modulated by $\cos(\frac{dt}{2})$. As a result, the spectrum exhibits a double-peak structure centered at $\omega = \omega_0 \pm \frac{d}{2}$. Another point is the height of the emission spectrum at $\omega = \omega_0$ remains the same independent of the values of cutoff frequency.

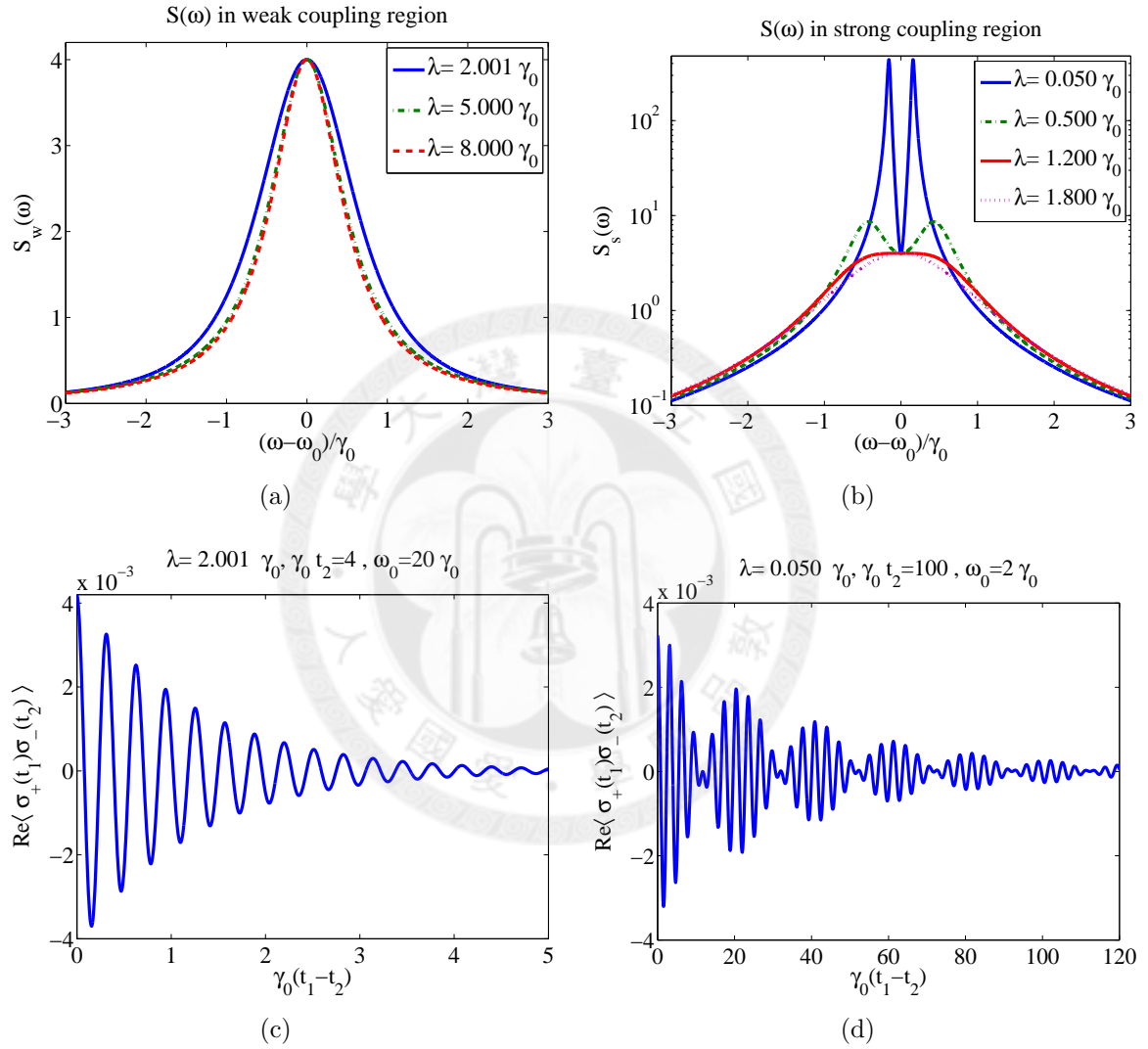


Figure 5.4: spontaneous emission spectra in arbitrary unit for different value of cutoff frequency (a) in the weak coupling region ($\lambda < 2\gamma_0$) (b) in the strong coupling region ($\lambda > 2\gamma_0$), and time evolutions of the two-time correlation function (c) in the weak coupling with $\lambda = 2.001\gamma_0$ (d) in the strong coupling with $\lambda = 0.05\gamma_0$

Chapter 6

Conclusion

We have derived in Chapter 2 the perturbative non-Markovian time-convolutionless master equation for reduced effective density matrix $\tilde{\chi}_s(t)$ through the cumulants expansion. The master equation can be directly applied to calculate the two-time correlation functions. The master equation is only based on a few requirements, (1) the effective density matrix $\chi_s(t)$ satisfies von Neumann equation, (2) initial system-bath is factorized in $t = 0$, (3) knowing the initial condition $\tilde{\chi}(t_2)$ is known and (4) the perturbative expansion series converges. We inserted the general interaction Hamiltonian up to fourth order, it is useful for any kind of problems.

We have calculated in Chapter 3 an exact two-time correlation function for a many-mode Jaynes-Cummings model with a Lorentz spectral density at zero-temperature. The exact two-time correlation function can be used to check the validity and applicable region of the master equation approach developed in Chapter 2. We focus that the exact result of the two-time correlation function guide different from that obtaining the exact QRT method that neglects the non-Markovian bath correlation between $t < t_2$ and $t > t_2$. From the exact result of the two-time correlation function, we were able to find an exact master for the reduced effective density matrix $\tilde{\chi}_s$. This allows us to make direct comparison between the exact two-time correlation with that obtained perturbatively.

We have calculate the two-time correlation function using the perturbative master equation up to fourth order. Here, we have used Aitken's δ^2 method to improve the perturbation master. To go beyond that higher order is a heavy and tedious

task. The perturbation master equation with Aitken's delta-squared method can slightly improve the result of the two-time correlation function.

The perturbation result up to fourth order agrees with the exact result in the weak coupling region. In the strong coupling region, the perturbation method is valid only for $t \ll t_{div}$. The contribution from the inhomogeneous terms depends strongly on the value of t_2 . The smaller the value of t_2 the smaller contribution from the inhomogeneous terms.

Finally, we derived spontaneous emission spectrum analytically. The spectrum shows dramatically different structure in the weak and the strong coupling region. In the weak coupling region, the spectrum has only one peak located at $\omega = \omega_0$ and the spectrum width is determined by the cut-off frequency. In the strong coupling region, there exist a critical cut-off frequency λ_c below which the spectrum goes from a one-peak structure to a two-peak structure with peak centers located at $\omega = \omega_0 \pm d/2$.

In summary, the two-time correlation functions are important physical quantity. They can provide additional information about the system, which the single-time expectation values can not provide. We believe that we are the first group to calculate the exact two-time correlation function and the spontaneous emission spectrum for the many-mode JC model. The calculations provide significant insight into how the non-Markovian memory effect influences the behavior of the two-time correlation functions.

Although it is commendable to calculate the exact two-time correlation function, but not many problems can have the exact solutions. The perturbation master equation approach developed in this thesis can be applied to calculate the two-time correlation functions perturbatively for the non-Markovian open (dissipative) quantum systems. We believe that this master equation approach that generalizes the QRT to the non-Markovian case will find broad applications in many different branches of physics.

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