

**ABANT İZZET BAYSAL UNIVERSITY
THE GRADUATE SCHOOL OF NATURAL AND APPLIED
SCIENCES
DEPARTMENT OF PHYSICS**



**DYNAMICS OF QUANTUM CORRELATIONS
UNDER NON-LINEAR NON-MARKOVIAN
STOCHASTIC DISTURBANCES**

DOCTOR OF PHILOSOPHY

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BOLU, MARCH 2017

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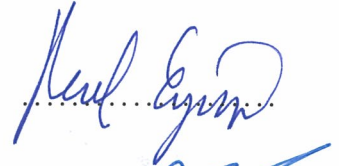
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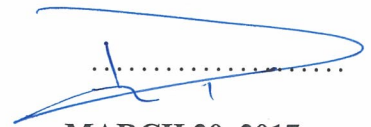
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Arzu KURT



ABSTRACT

DYNAMICS OF QUANTUM CORRELATIONS UNDER NON-LINEAR NON-MARKOVIAN STOCHASTIC DISTURBANCES

PHD THESIS

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**ABANT İZZET BAYSAL UNIVERSITY GRADUATE SCHOOL OF
NATURAL AND APPLIED SCIENCES**

**DEPARTMENT OF PHYSICS
(SUPERVISOR: PROF. DR. RESUL ERYİĞİT)
BOLU, MARCH 2017**

The main aim of this thesis is to obtain exact analytical solutions for three different problems which have relevance in quantum computer and quantum information science research areas. The first problem deals with the dynamics of two interacting qubits which are driven by a periodical external field. Solution of this problem is important for the control of qubits in quantum computers. The problem is split into three parts; in the first step, we investigated the dynamics of a single qubit driven by a harmonic field and have shown that Magnus expansion can be used to derive recursion relations for the exact dynamics of the system. As the second step, we have used Bellomo method to obtain analytical probability amplitudes for two non-interacting qubits driven by an harmonic field. We have derived a set of third order ordinary differential equations for the dynamics of driven two interacting qubits that is the last part of the problem. The second problem deals with the fourth-order corrections to the master equation of a charged harmonic oscillator in a black-body radiation field by using Krylov averaging method. Quantum master equations which determine the dynamics of the density matrix of the system are, almost always, second-order in the interaction parameter and determining the validity range of such an approximation for the relevant system parameters is crucial for assessing the accuracy of the dynamics obtained by solving such an equation. We have shown that the third- and the fourth-order contributions are, mostly, opposite in sign and cancel each other for the charged harmonic oscillator + black-body field system. The third problem that we have studied is related to the dynamics of spin-boson model which has a wide-range of applications in physics, chemistry and biology. As the first part of this problem, we have considered the dynamics of spin-boson model with a structured spectral density which is used to describe the environment in both superconducting qubits and biological light harvesting systems. We were able to obtain exact analytical expressions for the environmental correlation functions and derived exact expressions for the population dynamics of such a system. As the second part of the study on the dynamics of the spin-boson model, we have considered the effect of dichotomous noise in qubit transition frequency on the population and coherence of a qubit in a thermal bath. At high and low temperature limits, we have obtained analytical expressions for the transition rates and decay of coherence by using a non-Markovian master equation approach in the polaron transformation formulation. An interesting result of the study is that the dichotomous noise plays a constructive role on the transition rate in case of low temperature, overdamped and both intermediate and strong coupling.

KEYWORDS: Decoherence, Dichotomous noise, Non-Markovian systems, Structured environment.



ÖZET

**DOĞRUSAL OLMAYAN NON-MARKOVIAN GÜRÜLTÜNÜN
KUANTUM KORELASYONLARIN DİNAMİĞİNE ETKİSİ
DOKTORA TEZİ
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(TEZ DANIŞMANI: PROF. DR. RESUL ERYİĞİT)
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Bu tezin ana amacı, kuantum bilgisayar ve kuantum bilgi bilimi araştırma konularında gerekli olan üç farklı problem için tam analitik çözümlerinin elde edilmesidir. Birinci problem, periyodik dış alan tarafından sürülen iki etkileşen kübitin dinamiğiyle ilgilidir. Bu problemin çözümü, kuantum bilgisayarlarını oluşturan kübitlerin kontrolü için önemlidir. Problem üç kısımdan oluşmaktadır; birinci adımda, bir harmonik alan tarafından sürülen tek bir kübitin dinamiğini araştırdık ve Magnus açılımının, sistemin tam dinamiği için yineleme bağlantılarını türetmek üzere kullanılabilir olduğunu gösterdik. İkinci adım olarak, bir harmonik alan tarafından sürülen iki etkileşmeyen kübit için analitik olası genliklerini elde etmek üzere Bellomo metodunu kullandık. Sürülen etkileşen iki kübitin dinamiği için üçüncü mertebeden diferansiyel denklemler elde ettik ki bu problemin son kısmını oluşturmaktadır. İkinci problem, Krylov ortalama metodunu kullanarak siyah cisim ışıması alanı altında bir yüklü harmonik salıncığının ana denklemi için dördüncü mertebeden terimlerinin doğrulaması ile ilgilidir. Bu sistemin yoğunluk matrisinin dinamiğini tanımlayan kuantum ana denklemi sıklıkla etkileşme parametresinin ikinci mertebesinde yazılır ve ilgili sistem parametreleri için böyle bir yaklaşımın geçerlilik aralığını belirlemek, bu denklemi çözerek elde edilen dinamiğinin doğruluğunu değerlendirmek için çok önemlidir. Üçüncü ve dördüncü dereceden katkıların yoğunlukla zıt işaretli olduğunu ve yüklü harmonik salıncı + siyah cisim ışıması alan sistemi için birbirlerini yok ettiklerini gösterdik. Üçüncü çalışma fizik, kimya ve biyoloji alanlarında birçok olguyu modellemekte kullanılan spin-bozon modelinin dinamiğiyle ilgilidir. Bu problemin birinci kısmı olarak, hem süperiletken kübit hem de biyolojik ışık depolama sistemlerindeki çevreyi tarif etmek için kullanılan bir yapısal spektral yoğunluklu spin-bozon sistemlerin dinamiğini inceledik. Çevresel korelasyon fonksiyonları için tam analitik ifadeleri bulduk ve böyle bir sistemin populasyon dinamiği için tam ifadelerini türettik. Spin-bozon modelin dinamiği konusundaki çalışmanın ikinci kısmı olarak, bir termal çevre içindeki bir kübitin populasyon ve eşyumluluğu üzerine kübit geçiş frekansındaki iki değerlikli gürültünün etkisini inceledik. Yüksek ve düşük sıcaklık limitlerinde, polaron dönüşüm formüllemesi içindeki Markovian olmayan ana denklem yaklaşımını kullanarak, eşyumluluk azalması ve geçiş oranlarının analitik ifadelerini elde ettik. Ara ve güçlü etkileşim, düşük sıcaklık ve aşırı sönüm durumunda, gürültünün geçiş oranı üzerinde yapıcı bir rolünün olması bu çalışmanın dikkat çekici bir sonucudur.

ANAHTAR KELİMELEER: Eşyumlu olamama, İki değerlikli gürültü, Yapısal çevre, Non-Markovian sistemler.

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1. INTRODUCTION

With the advent of quantum computation and information science in the last two decades, the dynamics of quantum systems and the relation between that dynamics and the interaction between the quantum system and its macroscopic environment have become the main problems that needs to be solved for practical realization of quantum technologies. This interaction leads to phenomena such as decoherence, dissipation, and dephasing which destroys the quantum nature of the system which is expected to provide the advantage of quantum information systems in computational tasks compared to the classical ones. So, investigating the environmental noise and its effect on the dynamics of the quantum system would help designing new protocols that allineate the detrimental effects of the environment.

Quantum master equations are important for the non-unitary and irreversible dynamics that provide a most general description for the open quantum systems. Moreover, they preserve quantum mechanical properties, which are preserving of trace, Hermiticity, and positivity of density matrix through the entire evolution of the system if these master equations are in the Lindblad form. Master equation in Markovian as well as non-Markovian, generally environment contain linear interaction terms between the system and the environment and is taken into account up to second-order (Redfield, 1957; Zwanzig, 1964; Nakajima, 1958). There have been many attempt to include higher-order interaction terms into the master equations because second order equations have a limitation in the weak coupling condition. Higher-order terms, especially in the intermediate coupling regime, are expected to provide a better description of the effect of environment on the system dynamics. The following works that examined how the effect of the fourth-order term is on the system-bath coupling are (Budimir and Skinner, 1987; Laird and Skinner, 1991; Jang et al., 2002).

The spin-boson model is the simplest model to analyze the dynamics of a two level system (TLS) interacting with its surrounding bath, which is widely used to characterize charge as well as energy transfer in various systems, such as photosynthetic com-

plexes, tunneling in amorph metals and electron transfer reactions. The environment in spin-boson model is considered to be a collection of harmonic oscillators which are in thermal equilibrium at a given temperature T . In the absence of the environment, the dynamics of the TLS is unitary, *i.e.*, the system would oscillates between its two possible states with a time constant determined by the tunneling matrix element. One would expect that interaction of the TLS with a thermal environment would introduce decoherence which degrades the quantumness of the TLS as well as dissipation which leads to thermalization of the TLS with its environment. Under certain conditions accounting for the effect of the environment by using thermal means might not be adequate. For example, magnetic fluctuations of the environment might modulate the internal parameters of the two-state system which cannot be described by using a thermal bath. The general approach to deal with such situations is to model the internal parameters of the TLS as time-dependent term which describes a classical noise process.

There exists no analytical solution of the spin boson model, which would express the elements of the density matrix of the TLS as function of time. However, various approximate methods have been developed to investigate the system-environment interaction problems over the last fifty years. Among them the most prominent ones are: the path-integral (Hu et al., 1992; Escher and Ankerhold, 2011; Evans and Coalson, 1995), Nakajima Zwanzig projector operator (Zwanzig, 1960; Nakajima, 1958), non-interacting blip approximation (NIBA) (Goychuk et al., 1997, 1995a), Monte Carlo and hierarchical methods (Iles-Smith et al., 2014; Tanaka and Tanimura, 2009; Tanimura, 2012; Liu et al., 2014). However, none of the approximate techniques give a good description of the dynamics for all parameters of the problem. The inadequacy partially stems from the fact that the problem contains many competing time scales, such as the cutoff frequency and the thermal energy of the environment, TLS-environment interaction strength and the internal dynamical times of the TLS (such as the tunneling and the level splitting energies). Presence of the dichotomous noise introduces two new time scales, namely the frequency and the strength of the noise.

As the quantum mechanical system-environment type problems are becoming more and more important in quantum information problems, a more realistic description of such systems and analytical solution of their dynamics are gaining more attention. In such problems, the most important quantity that describes the effect of the coupling to the environment of the two level system is the bath spectral density which accounts for

the interaction strength as well as the frequency distribution of the harmonic oscillator that form the environment.



2. AIM AND SCOPE OF THE STUDY

In this thesis, we have studied three problems that deal with the quantum dynamics of low dimensional systems. Our approach is to start from the dynamics of a simple idealized system of two two-level system, *i. e.*, two qubits which are driven by an external field and include a thermal environment as the second problem and modify the qubit system plus environment with a noise on the qubit in the last problem. We introduce the problems in more detail below.

First problem considered is the analytical unitary dynamics of two interacting qubits that are driven by a periodic external field. The single qubit version of this problem has been investigated thoroughly in the last 80 years starting from the Landau-Zener model and shows interesting features, such as coherent destruction of tunneling which refers to entrapment of the qubit in one of the states at particular values of the driving field frequency and the amplitude. Interestingly, the exact analytical expression for the dynamics of a sinusoidally driven single qubit was obtained only recently (Qiongtao and Wenhua, 2010). In that study, the dynamics of probability amplitudes of a qubit coupled to a monochromatic periodical field are found to be expressible as combination of the Heun confluent and double confluent special functions. The Hamiltonian for the system we have studied can be written as:

$$H = \sum_{i=1,2} \frac{f^i(t)}{2} \sigma_z^i + \frac{v^i}{2} \sigma_x^i + \varepsilon \sigma \cdot \sigma \quad (2.1)$$

where $f(t) = f_0^i + f_1^i \sin(\omega_i t)$ is a sinusoidal driving field on i^{th} qubit. Note that f_0 can be thought of as a static field that determines the splitting frequency of the qubit, while $f_1^i \sin(\omega_i t)$ term modulates the energy levels of the qubit with frequency ω_i . $\sigma = \{\sigma_x, \sigma_y, \sigma_z\}$ and $\sigma_{x,y,z}^i$ are the Pauli spin matrices for the i^{th} qubit. v^i denotes the tunneling splitting of each qubit and ε is the strength of the spin-spin interaction which is assumed to be along the x, y, z -direction. We have found analytically probability amplitudes $a_i(t)$ of the entangled pure state for a periodically driven two qubits system by

using Bellomo's method (Bellomo et al., 2008) and the results of Ref. (Qiongtao and Wenhua, 2010) in absence of interaction between these qubits.

Second problem is related with dynamics of a charged harmonic oscillator weakly coupled to an electromagnetic field which has a form of velocity-coupling for the system-bath interaction including diamagnetic term which is generally ignored. Our aim was to investigate the effect of the fourth-order contribution to master equation on this system. We have applied the von Neumann equation to obtain the fourth-order master equation of the reduced density matrix in case of weak interaction by using Krylov averaging method. The Hamiltonian of the system will be described by

$$H = H_S + H_R + \varepsilon H_I, \quad (2.2)$$

where $H_S = \frac{p^2}{2m} + \frac{1}{2}Kx^2$, $H_R = \sum_j \left(\frac{p_j^2}{2m_j} + \frac{1}{2}m_j\omega_j^2 q_j^2 \right)$, and $H_I = \frac{pA}{m} + \frac{A^2}{2m}$ are the Hamiltonian of the system, the bath Hamiltonian, and the interaction Hamiltonian between the system and the bath respectively. The term A is the field operator that is described by $A = \sum_j m_j \omega_j q_j$ and ε is the weak coupling constant. In fact, the problem was examined in (Ford et al., 1996) and they have derived a master equation for an oscillator weakly coupled to an electromagnetic field by using Krylov averaging method which can be improved by including higher-order interaction terms in a systematical way. The aim of this work is to extend the previous work by adding the fourth order term in the master equation of the oscillator subject to a electromagnetic field. Our main findings in this context can be summarized as: The single most important parameter in the dynamics of the charge oscillator is found to be the cutoff frequency of the electromagnetic bath which is considered to be infinite in previous studies. The effect of the black-body radiation is found to be in renormalizing the electron mass and the anomalous as well the normal diffusion constants of the electron. We were able to obtain exact analytical expressions for those normalizations at second, third and the fourth orders and prove that including higher than third order terms would not change the commutative structure of the master equation. Also, we have shown that the second order master equation provides a satisfactory description of the system when the cutoff frequency of the environmental oscillators times the electron time is lower than a threshold.

We have analyzed such a system which can be described as a two-state system whose energy splitting is modulated by a two-state discrete stochastic process. The stochastic process is the so called dichotomous Markovian noise which jumps between

its two possible values with a certain frequency. Our aim is to examine the effect of the discrete noise frequency and the amplitude on the dynamics of the dissipative TLS. As one can consider the interaction of the system with the thermal bath as a Gaussian noise, competition and collaboration of the extra noise source with the unavoidable environmental noise is expected to show interesting dynamical features.

To investigate the effect of the thermal environment as well as the dichotomous noise on the dynamics of the TLS, we have adopted and improved the method developed by Goychuk and coworkers who have obtained a set of integro-differential equations for the population difference of the dissipative TLS driven by a telegraph noise. The main findings of this section of the thesis study can be summarized as follows: At very high noise intensity, the two level system would remain in its initial state at all times as expected, because very strong noise would mean a two level system with very large transition energy which is not available. On the other hand, at weak and intermediate noise strengths, contrary to what one expects, the effect of the dichotomous was found to be in opposition to that of the thermal noise as well as the coupling between the TLS and its environment for some parameters of the problem. Such opposition is found in situations where dichotomous noise leads to destruction of tunneling. One can find also the opposite case where dynamics without noise is no-tunneling type because of the strong TLS-bath interaction, but including a weak dichotomous noise leads to tunneling type dynamics. Also, in-line with expectations, we have found that intermediate strength noise would transform the coherent decay dynamics of the TLS to the non-coherent type. Overall, we have found that a dichotomous type noise might improve the transfer process for a restricted range of the problem parameters.

As the final part of the thesis research, we have considered a structured spectral density that has relevance in practical quantum computer implementations and investigated the bath correlation functions and the dynamics of such a system in non-interacting blip approximation (NIBA). We were able to obtain an exact analytical expression for the bath correlation function for the considered spectral density. Also, for a restricted range of parameters, a Markovian form of the NIBA equations were found to be amenable to an analytical solution.

3. PERIODICALLY DRIVEN TWO QUBITS SYSTEM

3.1 Introduction

In last two decades, with rapid developments in quantum information and computation research, dynamics of such systems become one of the most studied subjects. Because the main building block of any quantum computational system, called qubit, is generally, a two-level system (TLS). The need to control qubit operation requires external time-dependent control fields. It arises in research topics such as Josephson-junction devices (Nakamura et al., 2001), qubit control in quantum computing applications (Poem et al., 2011; Greilich et al., 2009), Bose-Einstein condensates (BEC) related applications (Eckardt et al., 2005; Haroutyunyan and Nienhuis, 2004; Salmond et al., 2002), dynamical localization (Grossmann and Hanggi, 1992; Barata and Wreszinski, 2000), and nuclear magnetic resonance applications (Rau, 1998; Angelo and Wreszinski, 2005). The first two-level system under time dependent external field was studied in early 1930s by the Landau-Majorana-Stückelberg-Zener (Landau, 1932) (LMSZ). The analytical expressions for the probability amplitudes of the system's state were obtained under rotating wave approximation (Shevchenko et al., 2010).

Even the simple system of a periodically driven TLS is hard to solve analytically. Over the years a number of approximate techniques for its solution have been developed. Among them the most widely used ones are the rotating-wave approximation (RWA) (Ashhab et al., 2007; Angelo and Wreszinski, 2005; Miao and Zheng, 2016), the perturbation theory (Zhou and Fang, 2008; Barata and Cortez, 2002), averaging theory (Barata and Wreszinski, 2000; Kayanuma, 1994), reverse-engineering approach (Barnes and Sarma, 2012; Barnes, 2013; Yang and Wu, 2006), iterative approach (Wu and Yang, 2007), Floquet theorem (Gomez Llorente and Plata, 1992; Xie, 2007, 2009; Verdeny et al., 2016), and Magnus expansion method (Grossmann and Hanggi, 1992; Gangopadhyay et al., 2010; Grifoni et al., 1993; Ashhab et al., 2007; Xie, 2009).

Only in recent years there have been reports of an exact analytic solution for a two-level system driven by a time periodic field (Xie and Hai, 2010), by a class of chirped pulses (Jha and Rostovtsev, 2010a) and the unchirped pulse (Jha and Rostovtsev, 2010b). Xie and Hai have examined the dynamics of a single two-level system coupled to a monochromatic magnetic field. They have found that an analytical solution can be expressed in terms of the confluent and the double confluent Heun special functions in absence (presence) of static driving term, respectively (Xie and Hai, 2010). Jha and Rostovtsev have presented an exact analytical solution for the time evolution of a two-level system driven (TLS) by a class of chirped pulses (Jha and Rostovtsev, 2010a) and the unchirped pulse (Jha and Rostovtsev, 2010b) which can be expressed in terms of Heun functions. They have shown that the strength in population transfer can be enhanced up to fourth order with proper choice of chirping parameters.

Barnes has exhibited a simple algorithm to obtain an exact analytical solutions for Schrödinger equation with a time dependent Hamiltonian including only two level system along a family of axis of Bloch sphere by using "reverse-engineering" approach which is especially necessary for application of quantum computation. He has shown that this method is convenient to derive analytic expressions for LMSZ problem (Barnes, 2013). Yang and Wu have developed an efficient approach for finding an exact analytical solution for a periodically driven two-level system and suggested that this approach can be used to seek dynamics of periodically driven multi-level systems (Yang and Wu, 2006).

Zhou and Fang have given an alternative approach to obtain an approximate analytic solution for a periodically driven two-level system in case of weak coupling by adapting a perturbative theory. They have indicated that this solution is consistent with the exact numerical solution in the case of resonance and almost resonance when Rabi frequency is equal and smaller than 0.3 times transition frequency (Zhou and Fang, 2008). Barata and Cortez have described a quite useful method to obtain the time evolution of a two-level system subject to a time-periodic field more accurately for a long time. They have obtained that this methods, having the uniform convergence of the expansion for all time, leads to be more robust numerical calculations for very long times (Barata and Cortez, 2002).

Barata and Wreszinski have presented a novel strong coupling expansion for examining time-evolution of a periodically driven two-atom level. An unexpected finding

for the coherent suppression of tunneling in the case of high frequency and resonance is that coherent destruction in tunneling process is, in deed, a third-order in tunneling frequency (Barata and Wreszinski, 2000).

Wu and Yang have studied the dynamics of periodically driven two level system in the case of strong coupling between the driving external field and the system. For this work they have developed a simple and important iteration approximation that is valid for a general expression of time-periodic field for wide frequency regime. An analytical solution of probability amplitudes of the system is obtained when the transition frequency of the TLS is smaller than the Rabi frequency even if a rather arbitrary form of periodic external field is used (Wu and Yang, 2007).

Ashhab and co-workers have analyzed the dynamics of strongly ac driven TLS by applying RWA and geometric approach and determined the boundary of validity of these approximates (Ashhab et al., 2007). Angelo and Wreszinski have studied time evolution of TLS driven by a periodic and quasi-periodic magnetic field by applying RWA that can be expressed as a function of a classical gyromagnet in these fields (Angelo and Wreszinski, 2005). Grossmann and Hanggi have shown that a periodically driven two-state system might exhibit localization at all driving frequencies (Grossmann and Hanggi, 1992). Kayanuma studied the dynamics of quantum tunneling of a particle in a double-well potential under a time-periodic external field by using the theorem of averaging approach. He has obtained analytical expressions for the occupation probability in the two asymptotic regions including rapid oscillation and large-amplitude oscillation (Kayanuma, 1994).

Liorente and Plata examined the tunneling process of a periodically driven system by using Floquet formalism and perturbative method. It is seen that this process can be controlled by adjusting external field parameters (Gomez Llorente and Plata, 1992). Xie has studied the Floquet states and quasi-energies of a two-level quantum system coupled to a periodic in time field with high frequency. It is seen that coherent destruction of tunneling (CDT) is a second-order effect with respect to the inverse of the frequency (ω) (Xie, 2009). Xie has investigated the influence of a periodic driving field on the self-trapping of two weakly coupled BECs interacting in a double-well potential. They have shown that there is exact non-linear Floquet solutions, which are valid in the limit of high frequency and strong resonance, for some values of the driving strength parameter (Xie, 2007). Verdeny and co-workers have given an argumentative review

of generalizations of Floquet theory on quasi-periodically driven quantum system (Verdeny et al., 2016).

In series papers, the influence of the environment is taken into account in order to define a more realistic system that they have investigated the dynamics of a periodically driven TLS in contact its surrounding environment. Gangopadhyay and coworkers have obtained an analytical solution of non-dissipative dynamics of a general two-level system coupled to a periodic-in-time external field. It is seen that the tunneling processes become strongly suppressed in the limit of strong driving (Gangopadhyay et al., 2010). Grifoni and co-worker have examined the effect of periodic driving field on a damped two-state system and shown that the time dependent evolution of the system has an exact solution at all temperatures and driving amplitude regimes. In addition, by using these exact solutions they have discussed the destruction of quantum tunneling (Grifoni et al., 1993). Miao and Zheng have investigated the nature of CDT for a two level system couple to across avoided crossing by using the photon counting statistics and calculated the spectra of photon emission by means of exact numerical, RWA and GVV approximations in weak relaxation limit. They have shown CDT disappears in strong dissipation (Miao and Zheng, 2016).

Kuznetsova *et. al.* have investigated the dynamics of entanglement for two spin systems subject to dipole-dipole coupling in spin locking with multi-pulse by using Magnus expansion method and the exact solution. It is seen that for an entangled state there is a critical temperature (Kuznetsova et al., 2016).

In this chapter, our aim is to obtain an analytical exact solution of two qubit system driven by a periodic driven field by using the exact results obtained from a single qubit in (Xie and Hai, 2010). We have shown that an exact solution can be found by using Bellomo method in case of two non-interacting qubits. We have also derived an analytical expression for a single qubit subject to a oscillating field by applying Magnus expansion method.

3.2 Model

In this chapter, we will present the results of an integration of the dynamics of periodically driven qubit(s). First, we will consider a qubit whose transition frequency is sinusoidally modulated with amplitude f_1 and frequency ω . We will show that time-dependence of its probability amplitude can be obtained by using Magnus expansion

method. As the second part, we will formulate a sinusoidally driven two qubit system and will discuss its dynamics in terms of the dynamics of the single qubit.

3.2.1 A Periodically Driven Qubit

We consider a two-level system whose energy levels are modulated by a time-periodic external field. The Hamiltonian of the system can be given as:

$$H(t) = \frac{f(t)}{2} \sigma_z + \frac{v}{2} \sigma_x, \quad (3.1)$$

where v is the coupling constant between $|1\rangle$ and $|0\rangle$ states and σ_i are Pauli spin matrices. The modulation is described by $f(t) = f_0 + f_1 \sin(\omega t)$, *i.e.* the level difference changes periodically between $f_0 - f_1$ and $f_0 + f_1$ with frequency ω . In the basis of σ_z eigenstates $\{|1\rangle, |0\rangle\}$ the most general time-dependent state of the two-level system is $|\Psi(\tau)\rangle = a(\tau)|1\rangle + b(\tau)|0\rangle$ with $|a(\tau)|^2 + |b(\tau)|^2 = 1$ where τ stands for dimensionless time which is defined as $\tau = \omega t$ where ω is the frequency of the driving field and f_0 , f_1 and v are defined in terms of ω . So, they are dimensionless. Hereafter, we call $f_0 = \frac{f_0}{\omega}$, $f_1 = \frac{f_1}{\omega}$, $v = \frac{v}{\omega}$, and $\varepsilon = \frac{\varepsilon}{\omega}$. We use $\hbar = 1$ units. Time evolution of such a state from Shrödinger equation can be given as:

$$\frac{d}{d\tau} |\Psi(\tau)\rangle = A(\tau) |\Psi(\tau)\rangle, \quad (3.2)$$

where

$$A(\tau) = -iH(\tau), \quad (3.3)$$

The formal solution of Eq. (3.2) is:

$$|\Psi(\tau)\rangle = T \exp \left(\int_0^\tau A(\tau_1) d\tau_1 \right) |\Psi(0)\rangle, \quad (3.4)$$

where T stands for the time-ordering operator. If $[A(\tau_1), A(\tau_2)] = 0$, *i.e.* the Hamiltonian at different times commute, the time-ordering can be dropped and the time evolution operator could be obtained as the exponential of the time-integrated Hamiltonian matrix. For the Hamiltonian in Eq. (3.1),

$$[H(\tau_1), H(\tau_2)] = \frac{i}{2} f_1 v (\sin(\omega \tau_1) - \sin(\omega \tau_2)) \sigma_y, \quad (3.5)$$

which is non-zero. So, we write the Schrödinger equation for the probability amplitudes $a(\tau)$ and $b(\tau)$ in the $\{|0\rangle, |1\rangle\}$ as:

$$\frac{d}{d\tau} \begin{pmatrix} a(\tau) \\ b(\tau) \end{pmatrix} = -\frac{i}{2} \begin{pmatrix} f_0 + f_1 \sin(\tau) & \nu \\ \nu & -(f_0 + f_1 \sin(\tau)) \end{pmatrix} \begin{pmatrix} a(\tau) \\ b(\tau) \end{pmatrix}, \quad (3.6)$$

which is in the form

$$\frac{dY(\tau)}{d\tau} = A(\tau)Y(\tau) \text{ where } Y(\tau) = \begin{pmatrix} a(\tau) \\ b(\tau) \end{pmatrix} \quad (3.7)$$

and

$$A(\tau) = -\frac{i}{2} \begin{pmatrix} f_0 + f_1 \sin(\tau) & \nu \\ \nu & -(f_0 + f_1 \sin(\tau)) \end{pmatrix}. \quad (3.8)$$

Eq. (3.6) is a set of two coupled first order differential equations with time-periodic coefficients and attempts to solve such equations have a long history. Xie and Hai have transformed the system to a second-order ordinary differential equation (ODE) and by a change of variable method has shown that the probability amplitudes could be expressed as combination of Heun confluent and double confluent functions (Xie and Hai, 2010). Another method developed to solve ODEs with periodic coefficients is the Magnus expansion method (Magnus, 1954) which will be used here to find a solution to Eq. (3.1). In Appendix B, we present a short review of Magnus expansion method. We find the approximate solution of Eq. (3.2) by using the recursive generation of the higher-order terms in Eq. (B.4). We may express the first four terms, Ω_m , up to $m = 4$, of the Magnus expansion without applying Taylor expansion of the transcendental

functions, such as $\sin(\tau)$, $\cos(\tau)$ as follows:

$$\begin{aligned}
\Omega_1 &= -\frac{1}{2}i\tau v \sigma_1 + \frac{i}{2}(f_1 \cos(\tau) - f_1 - f_0 \tau) \sigma_3, \\
\Omega_2 &= \frac{1}{4}i f_1 v [\tau + \tau \cos(\tau) - 2 \sin(\tau)] \sigma_2, \\
\Omega_3 &= i \frac{f_1 v}{48} [2 f_0 (12 - \tau^2) + 9 f_1 \tau + 2 [4 f_1 \tau + f_0 (\tau^2 - 12)] \cos(\tau) \\
&\quad - 12 (f_1 + f_0 \tau) \sin(\tau) + f_1 \tau \cos(2\tau) - 3 f_1 \sin(2\tau)] \sigma_1 \\
&\quad + i \frac{f_1 v^2}{24} [(\tau^2 - 12) (1 - \cos(\tau)) + 6 \tau \sin(\tau)] \sigma_3, \\
\Omega_4 &= i \frac{f_1 v}{576} [6 (24 f_0^2 \tau + f_0 f_1 (\tau^2 - 9) - 8 \tau (f_1^2 - 3 v^2)) \\
&\quad + 12 \tau (12 f_0^2 - 5 f_1^2 + 12 v^2) \cos(\tau) \\
&\quad - 6 f_1 [2 f_1 \tau + f_0 (\tau^2 - 9)] \cos(2\tau) \\
&\quad + 3 [19 f_1^2 + 16 f_0 f_1 \tau + 8 (f_0^2 + v^2) (\tau^2 - 12)] \sin(\tau) \\
&\quad + 30 f_1 (f_1 + f_0 \tau) \sin(2\tau) + f_1^2 \sin(3\tau)] \sigma_2.
\end{aligned} \tag{3.9}$$

Note that Ω_m for odd (even) m contains only odd (even) indexed Pauli matrices. As m increases, the argument of $\cos(\dots)$ and $\sin(\dots)$ terms goes as $\tau, 2\tau, 3\tau, \dots$ and their coefficients get complicated. To find a simpler structure, we use the Taylor expansion of the trigonometric functions and order the results as powers of τ for an initial state $|\Psi(0)\rangle = [1, 0]^T$ where T is the transpose operator. In this sense, we can obtain the solution of probability amplitudes $a(\tau)$ and $b(\tau)$ by including the first nine terms of Magnus expansion as function of the external field's the amplitude f_1 , splitting constant f_0 , coupling constant v between levels and dimensionless time τ . However, we will present only the solution of $a(\tau)$:

$$a(\tau) = 1 + a_1 \tau + a_2 \tau^2 + a_3 \tau^3 + a_4 \tau^4 + a_5 \tau^5 + a_6 \tau^6 + a_7 \tau^7 + O(\tau^8) \tag{3.10}$$

where

$$\begin{aligned}
a_1 &= -\frac{i f_0}{2}, \\
a_2 &= -\frac{1}{2^2 2!} [f_0^2 + 2i f_1 + v^2], \\
a_3 &= \frac{i f_0}{2^3 3!} [f_0^2 + 6i f_1 + v^2], \\
a_4 &= \frac{1}{2^4 4!} [f_0^4 - 12 f_1^2 + v^4 + 4i f_1 (v^2 + 2) + 2 f_0^2 (6i f_1 + v^2)], \\
a_5 &= -\frac{i f_0}{2^5 5!} [f_0^4 - 60 f_1^2 + v^4 + 20i f_1 (v^2 + 2) + 2 f_0^2 (10i f_1 + v^2)], \quad (3.11) \\
a_6 &= \frac{1}{2^6 6!} [-f_0^6 + 120i f_1^3 - v^6 - 3 f_0^4 (10i f_1 + v^2) + 4 f_1^2 (60 + 17 v^2) \\
&\quad - 2i f_1 (16 + 28 v^2 + 3 v^4) + 3 f_0^2 (60 f_1^2 - v^4 - 4i f_1 (10 + 3 v^2))], \\
a_7 &= \frac{i f_0}{2^7 7!} [f_0^6 - 840i f_1^3 + v^6 + 3 f_0^4 (14i f_1 + v^2) - 20 f_1^2 (84 + 13 v^2) \\
&\quad + 14i f_1 (16 + 20 v^2 + 3 v^4) + f_0^2 (-420 f_1^2 + 3 v^4 + 28i f_1 (10 + 3 v^2))].
\end{aligned}$$

We should emphasize that even if order (m) of Magnus expansion in Eq.(3.11) increases, denominator of the coefficient of τ^n is $2^n n!$ which increases factorially, *i.e.* expansion tends to converge. When Taylor expanded, H_{pm} and Eq. (3.10) agree in each coefficient of τ^n which indicates that Magnus expansion for this particular problem produces exact recurrence relations.

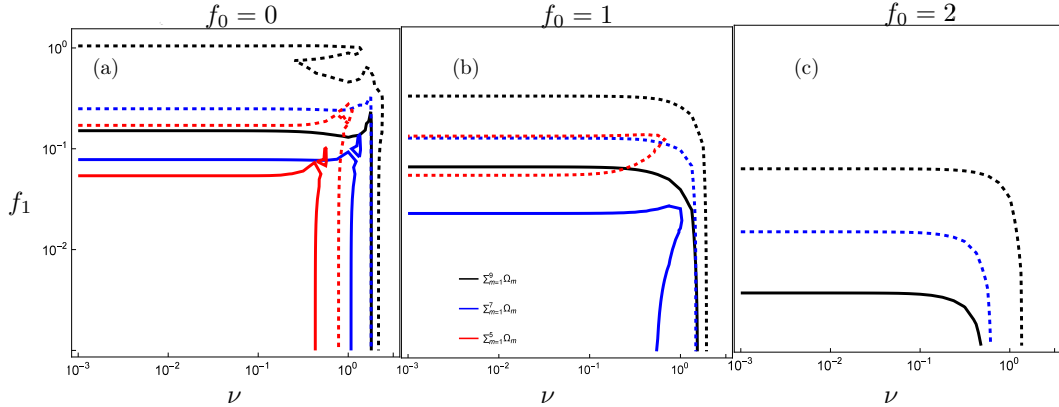


Figure 3.1. Relative error between the n^{th} order, up to order 9, Magnus expansion approximation and the exact analytical solution for probability amplitude $a(\tau)$ as a function of coupling frequency ν and the amplitude of external field f_1 . Solid lines (dashed lines) are for the relative error 0.001% (0.01%) to different contribution of Magnus expansion.

In Fig. 3.1, we display the 0.001 and 0.01 relative error contour as a function of dimensionless tunneling rate ν and driving field amplitude f_1 to find the validity range of Magnus expansion up to nine terms for three different values of qubit energy gap f_0 .

Here, the relative error is defined as the norm of difference between the exact solution and the Magnus expression up to order seven in τ for $|a(\tau)|^2$ by sampling $|a_i(\tau)|^2$ in the range $[0, 2]$ with a step length $\delta\tau = 0.01$. The plot can be divided into four equal parts as high frequency (HF, $\nu = 1/20$)- strong coupling (SC, $f_1 = 2$), low frequency (LF, $\nu = 2$)-SC, LF- weak coupling (WC, $f_1 = 1/20$), and HF-WC. Magnus expansion to low orders is an excellent approximation in HF-WC region due to the magnitude of parameters f_0 , f_1 , and ν , *i.e.*, in this region these parameter values are quite small and the contribution of higher order terms to Magnus expansion shall be small enough to ignore and the relative error is much less than 0.001. Outside of this parameter range its validity range deteriorates as one approaches both low frequency and the strong coupling parameter range. As expected, it can be seen from the figure that increasing the order of Magnus expansion pushes the validity of Magnus solution to higher values of ν and f_1 for all f_0 values considered in the study. At resonance ($\nu = 1$ and $f_1 = 1$) for no bias case, one needs at least 9 terms of the expansion to obtain 0.01 relative error in Fig. 3.1(a). It is interestingly to note that $\sum_{m=1}^5 \Omega_m$ has a rather limited region for $f_0 = 1$ that its relative error with 0.01% is located at $1/20 \leq f_1 < 1/8$. Overall, only the first five terms of this expansion is enough to compare with the analytical expression at weak field situation, high frequency limit and $f_0=0$ that provides a negligible error.

To display the features of relative errors, we plot the exact and Magnus expansion with the first 9 terms of probability amplitudes at four points of the contour graphs (HF-SC, LF-SC, LF-WC, and HF-WC) in Fig. 3.2. From the figure, it seen that agreement gets worse as function of time which might be related to the convergence time-interval of Magnus expansion which needs to be investigated further as a function of f_0 , f_1 , ν , and ω .

3.2.2 Periodically Driven Two Qubits

We consider two interacting qubits which are periodically driven independently. The Hamiltonian for this system can be written as:

$$H = \sum_{i=1,2} \left(\frac{f^i(t)}{2} \sigma_z^i + \frac{\nu^i}{2} \sigma_x^i \right) + \varepsilon \sigma \cdot \sigma, \quad (3.12)$$

where superscript i generalizes the driving field and the tunneling splitting to the i^{th} qubit from Eq. (3.1) single qubit case, $\sigma = \{\sigma_x, \sigma_y, \sigma_z\}$ and $\sigma_{x,y,z}^i$ are the Pauli spin

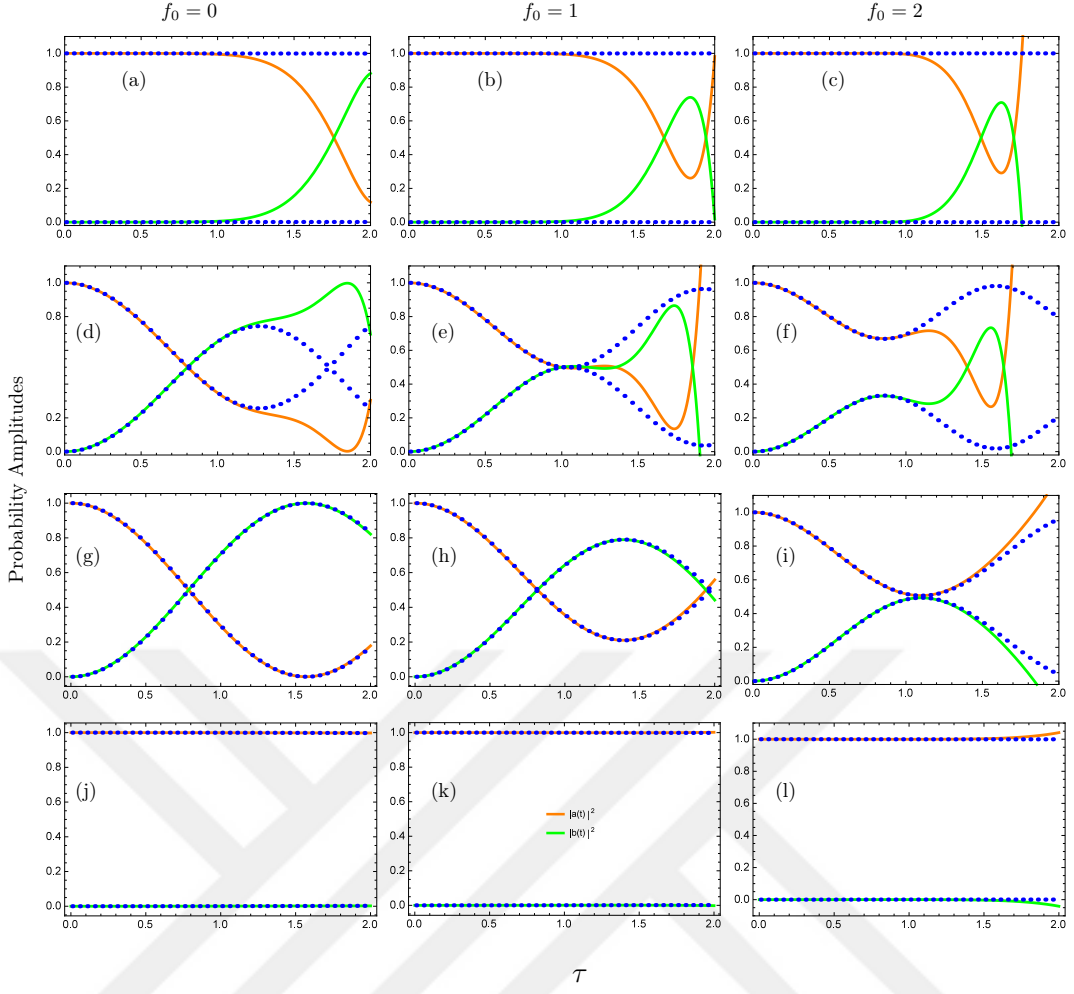


Figure 3.2. $|a_i(\tau)|^2$ versus dimensionless time τ ($= \omega t$) for a single qubit driven by a periodical external field with initial conditions $a(0) = 1$ and $b(0) = 0$ are presented for high frequency (HF, $\nu = 1/20$)- strong coupling (SC, $f_1 = 2$) (a, b, c), low frequency (LF, $\nu = 2$)-SC (d, e, f), LF- weak coupling (WC) (g, h, i), and HF-WC (j, k, l) at $f_0 = 0$, $f_0 = 1$, and $f_0 = 2$. Solid orange (green) lines indicate $|a(\tau)|^2$ ($|b(\tau)|^2$) in exact approximate solution including only Magnus expansion with 9 terms. Note that blue dots are given as exact solution of this system.

matrices for the i^{th} qubit, while v^i represents the tunneling matrix element of each qubit and ε is the strength of the spin-spin interaction between the qubits which is assumed to be along the x, y, z -direction. In the two-qubit problem, we also express all frequencies in terms ω , so the dimensionless time τ as before. The most general pure state of a two qubit system can be written as:

$$|\Psi(\tau)\rangle = a_1(\tau)|00\rangle + a_2(\tau)|01\rangle + a_3(\tau)|10\rangle + a_4(\tau)|11\rangle, \quad (3.13)$$

where $|ij\rangle$, $i, j = 0, 1$ indicates that the first qubit is in state $|i\rangle$ while second qubit's state is $|j\rangle$. The probability amplitudes $a_i(\tau)$ satisfy the normalization condition $\sum_{i=1}^4 |a_i(\tau)| =$

1. Shrödinger equation for the state $|\Psi(\tau)\rangle$ (Eq. 3.13) with Hamiltonian in Eq. (3.12) can be simplified if we transform to Bell basis with

$$|\Psi_{\pm}\rangle = (|00\rangle \pm |11\rangle)/\sqrt{2}, \quad (3.14)$$

$$|\Phi_{\pm}\rangle = (|01\rangle \pm |10\rangle)/\sqrt{2}, \quad (3.15)$$

with coefficients $k(\tau)$, $h(\tau)$, $g(\tau)$, and $w(\tau)$ as

$$|\Psi(\tau)\rangle = k(\tau)|\Psi_+\rangle + h(\tau)|\Psi_-\rangle + g(\tau)|\Phi_+\rangle + w(\tau)|\Phi_-\rangle, \quad (3.16)$$

where $k(\tau) = a_1(\tau) + a_4(\tau)$, $h(\tau) = a_1(\tau) - a_4(\tau)$, $g(\tau) = a_2(\tau) + a_3(\tau)$, and $w(\tau) = a_2(\tau) - a_3(\tau)$. Shrödinger equation

$$\frac{\partial |\Psi(\tau)\rangle}{\partial \tau} = -iH |\Psi(\tau)\rangle \quad (3.17)$$

for the states in Eqs. (3.15)-(3.14) and the Hamiltonian Eq. (3.12) can be written as a system of four coupled first-order differential equations in the Bell basis as follows:

$$\frac{dk(\tau)}{d\tau} = -i(\varepsilon k(\tau) + \nu g(\tau) + f(\tau)h(\tau)), \quad (3.18)$$

$$\frac{dh(\tau)}{d\tau} = -i(\varepsilon h(\tau) + f(\tau)k(\tau)), \quad (3.19)$$

$$\frac{dg(\tau)}{d\tau} = -i(\varepsilon g(\tau) + \nu k(\tau)), \quad (3.20)$$

$$\frac{dw(\tau)}{d\tau} = -3i\varepsilon w(\tau), \quad (3.21)$$

Note that the solution of Eq. (3.21) is quite easy which tends to evolve an exponential function of time. However, coupled first order ordinary differential equations in Eqs. (3.18)-(3.20) can be expressed as a third order differential equation in the following form:

$$\begin{aligned} \ddot{h}(\tau) &+ \left(3\varepsilon i - 2\frac{\dot{f}(\tau)}{f(\tau)}\right)\dot{h}(\tau) + \left(A - 2\varepsilon(\varepsilon + 2i)\frac{\dot{f}(\tau)}{f(\tau)} + 2\frac{\dot{f}(\tau)^2}{f(\tau)^2} - \frac{\ddot{f}(\tau)}{f(\tau)}\right)h(\tau) \\ &+ \left(\varepsilon Ai + f(\tau)\dot{f}(\tau) + 2\varepsilon^2\frac{\dot{f}(\tau)}{f(\tau)} + \varepsilon i\left(2\frac{\dot{f}(\tau)^2}{f(\tau)^2} - \frac{\ddot{f}(\tau)}{f(\tau)}\right)\right)h(\tau) = 0, \end{aligned} \quad (3.22)$$

$$\begin{aligned} \ddot{g}(\tau) &+ \left(3\varepsilon i - \frac{\dot{f}(\tau)}{f(\tau)}\right)\dot{g}(\tau) + \left(A - 2\varepsilon(\varepsilon + i)\frac{\dot{f}(\tau)}{f(\tau)}\right)g(\tau) \\ &+ \left(\varepsilon Ai - (\nu^2 - \varepsilon^2)\frac{\dot{f}(\tau)}{f(\tau)}\right)g(\tau) = 0, \end{aligned} \quad (3.23)$$

where $A(\tau) = v^2 - \varepsilon^2 + f(\tau)^2$. It should be noticed that one does not need to derive any further equation for $k(\tau)$. Because once $h(\tau)$ and $g(\tau)$ are obtained as the solution of Eq. (3.22) and Eq. (3.23), they can be plugged-into Eq. (3.18) to obtain $k(t)$.

First, we will consider the set of coupled differential equations in Eqs. (3.18)-(3.21) in the case of non-interacting qubits by setting $\varepsilon = 0$. For such a system, we will follow the procedure developed in Ref. (Bellomo et al., 2008) which expresses the time dependent density matrix elements of N independent qudits coupled to their own reservoir in terms of the density matrix elements of a single qudit. Since the present study involves only the pure states, we will formulate Bellomo's method for probability amplitudes of N qubit system in terms of those of a single qubit. Consider a qubit whose state evolves in time as:

$$|\psi(\tau)\rangle = \hat{U}(\tau, 0) |\Psi(0)\rangle \quad (3.24)$$

where $\hat{U}(\tau, 0)$ represents the time evolution operator and $|\Psi(0)\rangle$ is the initial state of the qubit. For N independent qubits, the state vector $|\Psi(\tau)\rangle$ can be written in tensor product form as

$$|\Psi(\tau)\rangle = \hat{U}_1 \otimes \hat{U}_2 \otimes \cdots \otimes \hat{U}_n |\Psi(0)\rangle, \quad (3.25)$$

where $|\Psi(\tau)\rangle$ has dimension 2^N and we have suppressed the time-dependence of time evolution operator $\hat{U}(\tau, 0)$. The identity operator in the basis of N qubit is

$I = \sum_{l_1, l_2, \dots, l_n} |l_1 l_2 \dots l_n\rangle \langle l_1 l_2 \dots l_n|$ with $l_1 = 0$ or 1 . After inserting the identity operator into Eq. (3.25), the state of N qubit system at time t can be written as:

$$|\Psi(\tau)\rangle = \hat{U}_1 \otimes \hat{U}_2 \otimes \cdots \otimes \hat{U}_n |l_1 l_2 \dots l_n\rangle \langle l_1 l_2 \dots l_n | \Psi(0)\rangle, \quad (3.26)$$

with $\hat{U}_1 \dots \hat{U}_n$ are the evolution operators of the qubits. According to Eq. (3.26), the elements of the reduced time evolution state $\langle j_1 j_2 \dots j_n | \Psi(t) \rangle = \Psi_{j_1, \dots, j_n}(t)$ have the form

$$\Psi_{j_1, j_2, \dots, j_n}(\tau) = \sum_{l_1, l_2, \dots, l_n} A_{j_1, l_1} \dots A_{j_n, l_n} \Psi_{l_1, l_2, \dots, l_n}(0), \quad (3.27)$$

where for each qubit $A_{j_n, l_n} = \langle j_n | \hat{U}_n | l_n \rangle$ represent the element of j_n^{th} row and l_n^{th} column of time evolution operator. Here, each elements of the set of j_n and l_n can be two different values, 0 or 1. Note that this procedure helps to obtain a full time dynamics of the general state of two independent qubits by using an analytical expression of a

periodically driven single qubit, initially characterized as entangled state. However, there is no need to use this method, when the initial state of the present system is prepared as a product state, since neither the Hamiltonian nor the initial state of the system contains interaction terms, so it remains as the product state.

Our aim is to reach the analytical solution of probability amplitudes of independently two qubits driven by periodically external field by using this procedure. We will start to reduce this procedure for two qubits where time-dependent general state $\Psi(\tau)$ in Eq. (3.13) can be written as

$$\begin{aligned}\langle j_1, j_2 | \Psi(\tau) \rangle &= \langle j_1, j_2 | \hat{U}_1 \hat{U}_2 | \Psi(0) \rangle, \\ &= \sum_{k,m} \langle j_1, j_2 | \hat{U}_1 \hat{U}_2 | k, m \rangle \langle k, m | \Psi(0) \rangle, \\ \Psi_{j_1, j_2}(\tau) &= \sum_{k,m} \langle j_1 | \hat{U}_1 | k \rangle \langle j_2 | \hat{U}_2 | m \rangle \langle k, m | \Psi(0) \rangle,\end{aligned}\quad (3.28)$$

where $\sum_{k,m} |k, m\rangle \langle k, m|$ is equal to unity for a single qubit. For example, we can give an expression of $a_1(\tau)$ in Eq. (3.13) with respect to elements of time evolution operator $\hat{U}_{1,2}(\tau, \tau_0)$ which Xie and Hai evaluated analytically (Xie and Hai, 2010) as follow:

$$\begin{aligned}a_1(\tau) &= \Psi_{0,0}(t) = \langle 0 | \hat{U}_1 | 0 \rangle \langle 0 | \hat{U}_2 | 0 \rangle \langle 0, 0 | \Psi(0) \rangle + \langle 0 | \hat{U}_1 | 0 \rangle \langle 0 | \hat{U}_2 | 1 \rangle \langle 0, 1 | \Psi(0) \rangle, \\ &+ \langle 0 | \hat{U}_1 | 1 \rangle \langle 0 | \hat{U}_2 | 0 \rangle \langle 1, 0 | \Psi(0) \rangle + \langle 0 | \hat{U}_1 | 1 \rangle \langle 0 | \hat{U}_2 | 1 \rangle \langle 1, 1 | \Psi(0) \rangle.\end{aligned}\quad (3.29)$$

The rest of probability amplitudes can be obtained similarly. The time evolution operator for the n^{th} qubit can be written as:

$$\hat{U}_n(\tau, \tau_0) = \begin{pmatrix} u_{11}^n & u_{12}^n \\ u_{21}^n & u_{22}^n \end{pmatrix}. \quad (3.30)$$

Since $\hat{U}$ needs to be a unitary operator, its elements need to satisfy certain symmetry properties. Namely, $u_{11} = u_{22}^*$ and $u_{21} = -u_{12}^*$. Also, the eigenvalues of $\hat{U}$ should have unit norm.

$$u_{11} = H_{pm}, \quad u_{21} = i \frac{V}{2f_1} V_{pm}, \quad (3.31)$$

where for the present problem,

$$H_{pm} = \frac{1}{2} (e^{-\varphi} h_+ + e^{\varphi} h_-) + i \frac{f_0}{2f_1} V_{pm}, \quad (3.32)$$

$$V_{pm} = e^{-\varphi} h_+ - e^{\varphi} h_-, \quad \varphi = \frac{f_1}{2} \sin(\tau). \quad (3.33)$$

Here, h_+ and h_- are double confluent Heun functions which can be given as:

$$h_{\pm} = \text{HD} \left[\pm \alpha, \beta, \gamma, \delta, -i \tan\left(\frac{\tau}{2}\right) \right], \quad (3.34)$$

where $\alpha = 2i f_1$, $\beta = -(f_0^2 + f_1^2 + \gamma^2) + 2i f_1$, $\gamma = 4i f_0 f_1$ and $\delta = f_0^2 + f_1^2 + \gamma^2 + 2i f_1$. HD is described by a standard power series around $z = 0$, i.e. $\sum_{n=0}^{\infty} d_n z^n$, where d_n stands for the six-fold recurrence relation (Xie, 2011) as follows:

$$\begin{aligned} d_n = & \frac{1}{n(n-1)} ((n-5)(n-6)d_{n-6} + \alpha(n-5)d_{n-5} + (\beta - (n-4)[3(n-6) + 7])d_{n-4} \\ & + (2\gamma + \alpha)d_{n-3} + (\delta + (n-2)[3(n-6) + 11])d_{n-2} + \alpha(n-1)d_{n-1}). \end{aligned} \quad (3.35)$$

In this sense, the analytical solution of a general state $|\Psi(\tau)\rangle$ of the two independent qubit system can be found from the exact solution obtained to a single qubit by means of these findings and the time evolution operator $U(\tau, \tau_0)$ of the single qubit which is valid for $0 \leq \tau_0 \leq \tau < \pi/2$ and $3\pi/2 < \tau_0 \leq \tau \leq 2\pi$ in the existence of the static part f_0 in (Xie and Hai, 2010). To present time evolution of the general state $|\Psi(\tau)\rangle$ of the system, we shall use Bell-like states as the initial states of the system

$$|\Phi(0)\rangle = \chi |01\rangle + \zeta |10\rangle, \quad (3.36)$$

$$|\Psi(0)\rangle = \chi |00\rangle + \zeta |11\rangle, \quad (3.37)$$

where χ is real, $\zeta = |\zeta| e^{i\theta}$ with $|\chi|^2 + |\zeta|^2 = 1$. The transition probability amplitudes of the present system can be expressed by inserting Eq. (3.31) into Eq. (3.28) for the

initial states $|\Phi(0)\rangle$ that are as follows:

$$\begin{aligned} a_1(\tau) &= \chi u_{11}^A u_{12}^B + \zeta u_{12}^A u_{11}^B, \\ &= \chi i \left(\frac{v^B}{2 f_1^B} \right) H_{pm}^A V_{pm}^B + \zeta i \left(\frac{v^A}{2 f_1^A} \right) H_{pm}^B V_{pm}^A, \end{aligned} \quad (3.38)$$

$$\begin{aligned} a_2(\tau) &= \chi u_{11}^A u_{22}^B + \zeta u_{12}^A u_{21}^B, \\ &= \chi H_{pm}^A H_{pm}^{*B} - \zeta \left(\frac{v^A}{2 f_1^A} \right) \left(\frac{v^B}{2 f_1^B} \right) V_{pm}^A V_{pm}^{*B}, \end{aligned} \quad (3.39)$$

$$\begin{aligned} a_3(\tau) &= \chi u_{21}^A u_{12}^B + \zeta u_{22}^A u_{11}^B, \\ &= -\chi \left(\frac{v^A}{2 f_1^A} \right) \left(\frac{v^B}{2 f_1^B} \right) V_{pm}^{*A} V_{pm}^B + \zeta H_{pm}^{*A} H_{pm}^B, \end{aligned} \quad (3.40)$$

$$\begin{aligned} a_4(\tau) &= \chi u_{21}^A u_{22}^B + \zeta u_{22}^A u_{21}^B, \\ &= \chi i \left(\frac{v^A}{2 f_1^A} \right) V_{pm}^{*A} H_{pm}^{*B} + \zeta i \left(\frac{v^B}{2 f_1^B} \right) H_{pm}^{*A} V_{pm}^B, \end{aligned} \quad (3.41)$$

where A and B stands for two different qubits. For the second initial state $|\Psi(0)\rangle$, the exact expressions of these probability amplitudes become:

$$\begin{aligned} a_1(\tau) &= \chi u_{11}^A u_{11}^B + \zeta u_{12}^A u_{12}^B, \\ &= \chi H_{pm}^A H_{pm}^B - \zeta \left(\frac{v^A}{2 f_1^A} \right) \left(\frac{v^B}{2 f_1^B} \right) V_{pm}^A V_{pm}^B, \end{aligned} \quad (3.42)$$

$$\begin{aligned} a_2(\tau) &= \chi u_{11}^A u_{21}^B + \zeta u_{12}^A u_{22}^B, \\ &= \chi i \left(\frac{v^B}{2 f_1^B} \right) H_{pm}^A V_{pm}^{*B} + \zeta i \left(\frac{v^A}{2 f_1^A} \right) V_{pm}^A H_{pm}^{*B}, \end{aligned} \quad (3.43)$$

$$\begin{aligned} a_3(\tau) &= \chi u_{21}^A u_{11}^B + \zeta u_{22}^A u_{12}^B, \\ &= \chi i \left(\frac{v^A}{2 f_1^A} \right) V_{pm}^{*A} H_{pm}^B + \zeta i \left(\frac{v^B}{2 f_1^B} \right) V_{pm}^B H_{pm}^{*A}, \end{aligned} \quad (3.44)$$

$$\begin{aligned} a_4(\tau) &= \chi u_{21}^A u_{21}^B + \zeta u_{22}^A u_{22}^B, \\ &= -\chi \left(\frac{v^A}{2 f_1^A} \right) \left(\frac{v^B}{2 f_1^B} \right) V_{pm}^{*A} V_{pm}^{*B} + \zeta H_{pm}^{*A} H_{pm}^{*B}. \end{aligned} \quad (3.45)$$

The analytical exact solutions of third order differential equations in Eq. (3.22)-Eq. (3.23) are represented as the expressions in Eqs. (3.38)-(3.45) for the initial states $|\Phi(0)\rangle$ and $|\Psi(0)\rangle$ in absence of interaction constant term ε . We should note that one can constitute the full time-evolution dynamics by connecting different evolution operators which are valid at different time intervals by means of the well-known Floquet operator.

Now we consider the presence of the coupling term ε between two different qubits. The time-dependent Schrödinger equation for Hamiltonian in Eq. (3.12) gives us the coupled ordinary differential equations for the general state in Eq. (3.13) that have al-

ready given in Eqs. (3.18)-(3.21) and their third order linear differential equations can be also seen from Eq. (3.22)-(3.23). However, we need to apply an other way to solve these differential equations of third order. The method of Bellomo does not work for this condition, since the current system described by the Hamiltonian in Eq. (3.12) contains the spin-spin interaction. In Fig. 3.3, we present the populations of two non-

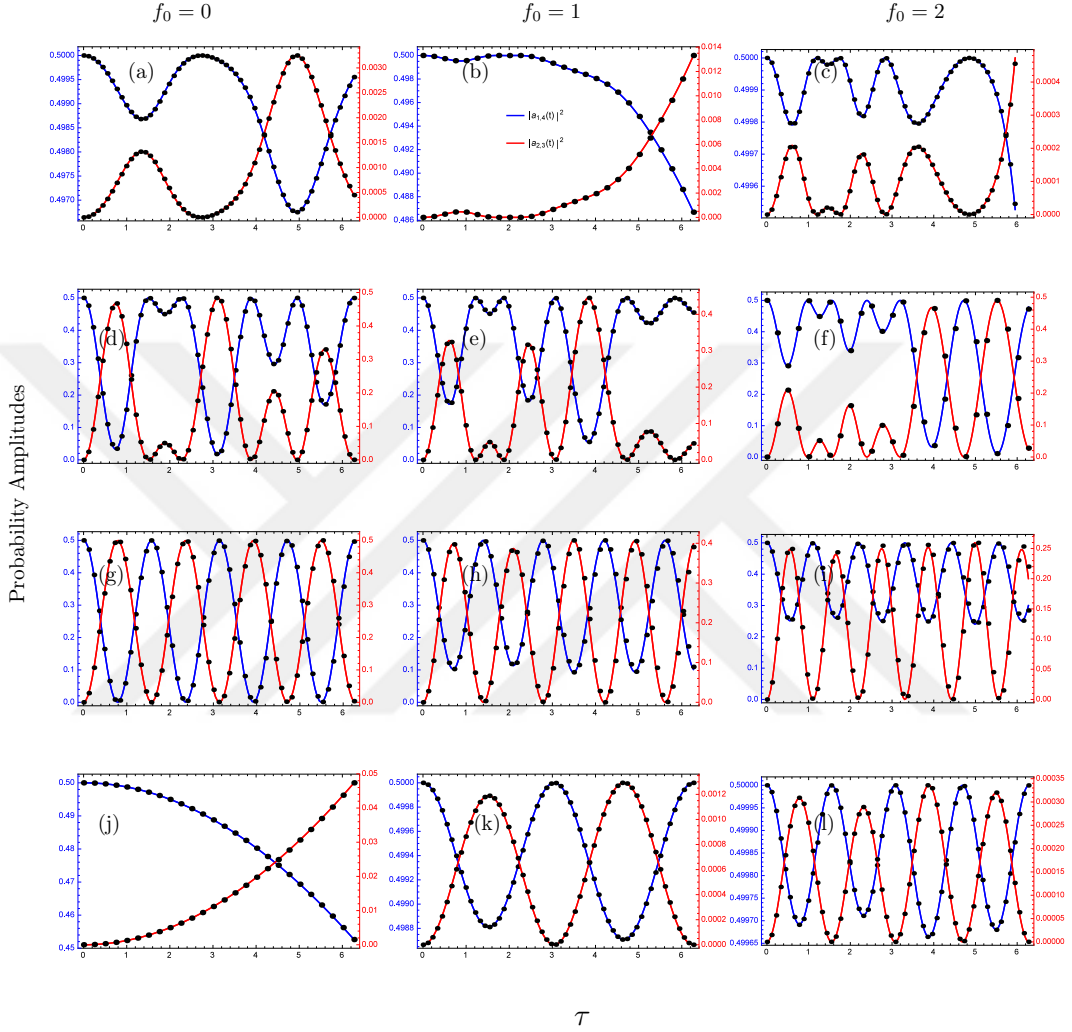


Figure 3.3. $|a_i(\tau)|^2$, $i = 1, 2, 3, 4$ versus dimensionless time $\tau(=\omega t)$ for high frequency (HF)-strong coupling (SC) $\{\frac{v}{\omega} = \frac{1}{20}, \frac{f_1}{\omega} = 2\}$ (a, b, c), low frequency (LF)-strong coupling (SC) $\{\frac{v}{\omega} = 2, \frac{f_1}{\omega} = 2\}$ (d, e, f), LF- weak coupling (WC) $\{\frac{v}{\omega} = 2, \frac{f_1}{\omega} = \frac{1}{20}\}$ (g, h, i) and HF-WC $\{\frac{v}{\omega} = \frac{1}{20}, \frac{f_1}{\omega} = \frac{1}{20}\}$ (j, k, l) in case of $a_1(0) = \frac{1}{\sqrt{2}}$, $a_2(0) = 0$, $a_3(0) = 0$, $a_4(0) = \frac{1}{\sqrt{2}}$ at different f_0 . Blue lines (left axis) indicate $|a_{1,4}(\tau)|^2$, while red lines (right axis) are for $|a_{2,3}(\tau)|^2$. Black dots are for the numerical solution.

interacting qubits for several energy gap f_0 , field amplitude f_1 , and tunneling splitting v . Similar to the single qubit case, the change in populations is seen to be negligible at high frequencies ($v \ll 1$) regardless of the magnitude of the driving field amplitude which indicates that the coherent destruction of tunneling is also present for the two

qubit case (Grossmann and Hanggi, 1992). We should note that at large f_0 s, the tunneling is also reduced, but this phenomena is not a CDT process; it just indicates that the tunneling is weak due to large energy gap of the qubit. This tendency can clearly be seen from (a,b,c) and (j,k,l) in the same figure.

3.3 Conclusions

In conclusion, we have obtained the exact approximation of dynamics of a periodically driven single qubit by using Magnus expansion method whose usage is indicated to be quite simple in comparison with the study of Xie and Hai (Xie and Hai, 2010) and investigated the magnitude of relative error between the exact solution and Magnus expansion including only first nine terms at different parameter values such as amplitude of driving field f_1 , energy gap f_0 between energy levels, and coupling constant v . As a second work is studied the dynamics of a two qubit system driven by a sinusoidal external field. By means of Bellomo's method we have obtained an exact analytical solution of probability amplitudes of the two qubits system when there is no interaction between them. An interesting result for these two studies is that both a periodically driven single qubit and non-interacting two qubits system indicate a same phenomena which is a coherent destruction of tunneling.

4. FOURTH-ORDER MASTER EQUATION

4.1 Introduction

Research activity in dissipation and decoherence of quantum systems has accelerated in the last two decades with developments in quantum information technologies, progress in detection and control of ultra-cold atom and ions, atom lasers and photonic band-gap materials as well as the research on the role of decoherence in quantum-to-classical transition (Lewenstein et al., 2012; Massignan et al., 2015; Breuer and Petruccione, 2002; Schlosshauer, 2007; Weiss, 2008). Theoretical efforts to investigate dissipation and decoherence phenomena due to interaction between a quantum system and its environment use either exact or perturbative treatments. While exact approaches can be used to treat a restricted class of problems for the whole interaction parameter range, the perturbative treatments can be applied to almost all problems, but only for a restricted range of parameters which is, generally, the weak coupling regime. A plethora of different perturbation based formalisms have been developed to treat such system-bath problems over the last six decades (Breuer and Petruccione, 2002; Schlosshauer, 2007; Weiss, 2008). The most prominent among them are Nakajima-Zwanzig projector operator method, path-integral based influence functional method (Hu et al., 1992), self-consistent hybrid schemes, Monte Carlo methods and hierarchical approaches (Tanimura, 2006; Liu et al., 2014). Most of the perturbative approaches express the equation of motion of the reduced density matrix of the system as a quantum master equation (QME) which contains second-order interaction terms.

One way to increase the validity range of the perturbative methods might be increasing the order of master equation. Actually, it is shown by Fleming and Cummings (Fleming and Cummings, 2011) that an order- $2n$ accuracy in the full-time solutions of master equations requires an order- $(2n + 2)$ master equation. A number of groups investigated the effects of the fourth and higher-order terms in detail (Budimir and Skinner, 1987; Laird et al., 1991; Reichman et al., 1997; Thingna et al., 2014; Ford

et al., 1996; Reichman and Silbey, 1996; Breuer et al., 1999a,b, 2001; Jang et al., 2002; Doll et al., 2008; Ishizaki and Tanimura, 2008; Fleming and Cummings, 2011; Singh and Brumer, 2012; Liu et al., 2014). In particular, Reichman *et. al.* have derived analytical expressions for dynamics of a harmonic oscillator in contact with various types of baths in the low temperature limit (Reichman and Silbey, 1996). Also, a physically sound fourth-order correction to Redfield equation has been worked out by Laird, Budimir and Skinner (Laird et al., 1991). Jang, Cao and Silbey have derived a fourth-order QME in both time local and nonlocal forms for a general system Hamiltonian and used it to study motion of a particle in a continuous potential field and the dynamics of a two-level system coupled to a bath. Fourth-order corrections were found to cause a potential renormalization for the first problem while they introduced additional coherence for the later (Jang et al., 2002). In a series of papers, Breuer *et. al.* have examined, in detail, the effects of including the fourth and higher-order terms in time convolutionless master equation for the damped Jaynes-Cummings model (JCM), atom laser and the damped harmonic oscillator (Breuer et al., 1999a,b, 2001). It was found that at weak and moderate coupling, the fourth-order QME was in good agreement with the exact solution for the JCM while for the intermediate coupling regime of atom lasers, one needs to include non-Markovian as well as the fourth- and sixth-order terms to the master equation to adequately describe the dynamics of the system. Singh and Brumer investigated the validity of the second-order Markovian Redfield theory for reorganization energy and decay rates of photon autocorrelation functions in dimer systems (Singh and Brumer, 2012). Liu *et.al.* (Liu et al., 2014) used hierarchical equation of motion method for the spin-boson problem and showed that fourth-order corrections are important for the intermediate coupling regime. Mavros and Voorhis considered fourth-order corrections to the memory kernel of the generalized master equation of the spin-boson problem and found that a numerically-exact solution was possible when the system-bath coupling is weak (Mavros and Voorhis, 2014). Interestingly, it was found that the non-perturbative treatment of certain system-bath problems is equivalent to the second-order perturbative master equations and including higher than second-order terms would be detrimental for such systems (Ishizaki and Tanimura, 2008; Doll et al., 2008). Dissipative dynamics of a charged particle in an electromagnetic field, whether a coherent laser field or an incoherent blackbody radiation, have been examined by several groups (Ford et al., 1988a; Marathe, 1989; Ford et al., 1996; Li et al., 1996;

Dattagupta and Singh, 1997; Bao and Bai, 2005; Bai et al., 2005; Kalandarov et al., 2007; Pachón and Brumer, 2013; Kumar, 2014) because of its relevance to damped transport in solid-state systems. Bao and Bai have considered the the motion of a single electron atom interacting with an electromagnetic field in the dipole coupling approximation and obtained a ballistic diffusion for the particle in the position space (Bao and Bai, 2005; Bai et al., 2005). Kalandarov *et. al.* have shown that dissipation of collective energy of a charged oscillator linearly coupled to a heat bath could be controlled by an external field (Kalandarov et al., 2007). Pachon and Brumer investigated the effect of blackbody radiation on the coherence of the dynamics of light-harvesting molecules in photosynthesis and showed that the blackbody radiation would not enhance the coherence of the system (Pachón and Brumer, 2013) .

Ford, Lewis, and O’Connell derived a master equation for a charged oscillator weakly coupled to an electromagnetic field by using Krylov averaging method which can be improved by including higher-order interaction terms in a systematical way (Ford et al., 1996). The model has several interesting features, such as use of velocity-coupling for the system-environment interaction which is not as widely studied as the position-coupling and keeping the diamagnetic term of the interaction Hamiltonian which leads to the nonzero third-order contributions to the master equation which are absent for most of similar studies. In the current work, our aim is two-fold; first, we obtain exact analytical expressions for all coefficients of the derived master equation which are obtained in the limit of infinite environmental cutoff frequency in Ref. (Ford et al., 1996). Our findings indicate that the exact treatment is crucial for the accurate high frequency behavior of the diffusion coefficients. Second purpose of the present study is to derive analytical expressions for the fourth-order corrections to the master equation of the charged oscillator in an electromagnetic field and delineate the convergence range of interaction parameters.

The structure of this paper is as follows. We will introduce the model of the charged harmonic oscillator in contact with a blackbody radiation field in Section 4.2 where we provide a short review of the derivation of the master equation by Krylov averaging method. Analytical expressions for the second-, the third- and the fourth-order corrections are, also, presented in Section 4.2 and discussed in Section 4.3. Section 4.4 concludes the paper with a short review of the main findings. We provide the details of the derivation of the fourth-order terms in the Appendix.

4.2 Model

We consider a charged harmonic oscillator in a blackbody radiation field (Ford et al., 1996; Bai et al., 2005; Ford et al., 1988a). The total Hamiltonian of the closed system formed by the oscillator and field can be written as:

$$H = H_S + H_R + \varepsilon H_I, \quad (4.1)$$

with

$$H_S = \frac{p^2}{2M} + \frac{1}{2}Kx^2, \quad (4.2)$$

$$H_R = \sum_j \left(\frac{p_j^2}{2m_j} + \frac{1}{2}m_j \omega_j^2 q_j^2 \right), \quad (4.3)$$

$$\varepsilon H_I = \frac{pA}{M} + \frac{A^2}{2M}, \quad (4.4)$$

where H_S , H_R , and εH_I describe the charged harmonic oscillator as the system, blackbody radiation field as the reservoir and the interaction between the velocity of the oscillator and the vector potential A of the field, respectively. K and M are the force constant and the mass of the charged oscillator, while m_j , p_j , q_j and ω_j describe the environmental oscillators that stand for the blackbody field. p and x for the oscillator and p_j , q_j of the reservoir obey the usual commutation rules as $[x, p] = i\hbar$, $[q_i, p_j] = i\hbar\delta_{ij}$ and $[x, p_j] = [p, q_j] = 0$. The vector potential of the field A can be expressed as:

$$A = \sum_j m_j \omega_j q_j. \quad (4.5)$$

The interaction between the oscillator and the field is assumed to be in velocity-coupling type and contains both $p.A$ and a diamagnetic A^2 terms. It is well known that the Hamiltonian in Eq. (4.1), can be transformed to a renormalized coordinate-coupling form by using Power-Zienau transformation (Babiker et al., 1974). The diamagnetic interaction A^2 is generally neglected (Castrigiano and Kokiantonis, 1987; Barone and Caldeira, 1991), because its inclusion makes calculation of path-integral based master equation difficult. But its inclusion is shown to be important for the correct derivation of the partition function of a charged oscillator in blackbody field (Ford et al., 1988a; Castrigiano and Kokiantonis, 1988) and we keep it here.

The influence of the environment on the system is determined by the spectral density $J(\omega)$ of the bath which is a measure of the number of bath oscillators at frequency ω and the coupling constant of the system-bath interaction. For the present case, we use the Drude corrected Ohmic spectral density:

$$J(\omega) = m\tau_e\omega \frac{\Omega^2}{\omega^2 + \Omega^2}, \quad (4.6)$$

In the interaction picture, the unitary dynamics of the closed system formed by the oscillator and the radiation field is described by the von Neumann equation for the density matrix ρ as:

$$\frac{\partial \rho(t)}{\partial t} = \frac{\varepsilon}{i\hbar} [H_I(t), \rho(t)], \quad (4.7)$$

where the interaction picture transformation is taken with respect to H_S and H_R as

$$\begin{aligned} \varepsilon H_I(t) &= e^{\frac{i(H_S+H_R)t}{\hbar}} \varepsilon H_I e^{-\frac{i(H_S+H_R)t}{\hbar}} \\ &= \frac{p(t)A(t)}{m} + \frac{A^2(t)}{2m}, \end{aligned} \quad (4.8)$$

$$\begin{aligned} A(t) &= e^{\frac{iH_R t}{\hbar}} A e^{-\frac{iH_R t}{\hbar}} \\ &= \sum_j \{m_j \omega_j q_j \cos(\omega_j t) + p_j \sin(\omega_j t)\}, \end{aligned} \quad (4.9)$$

$$\rho(t) = e^{\frac{i(H_S+H_R)t}{\hbar}} \rho e^{-\frac{i(H_S+H_R)t}{\hbar}}. \quad (4.10)$$

We assume that the bath and the oscillator are uncorrelated initially, that is

$$\rho(0) = \rho_S(0) \otimes \rho_R, \quad (4.11)$$

where $\rho_S = \text{Tr}_R\{\rho\}$ is the reduced density matrix of the oscillator and ρ_R is the density matrix of the reservoir which is assumed to be thermal at temperature T . Tr_R indicates partial trace over the bath degrees of freedom.

There have been many attempts to solve Eq. (4.7) by using various different approximations, such as projector operator (Chaturvedi and Shibata, 1979; Yonn et al., 1975), path integral influence functional formulations (Feynman and Vernon, 1963). When the system-environment interaction is weak, Krylov averaging method can also be used to obtain a hierarchy of equations for the dynamics of the oscillator (Ford et al., 1996). For the self-containment of the present paper, we give a short overview of the method and the details of the derivation of the fourth-order terms in the Appendix section.

For ease of comparison with Ref. (Ford et al., 1996), we will rearrange the commutators on the right-hand side of Eq. (4.49) as

$$\begin{aligned} \frac{\partial \rho_S}{\partial t} = & -\frac{1}{2m\hbar} \left(iA_1 [p(t)^2, \rho_S] + A_2 [p(t), [p(t), \rho_S]] + m\omega_0 A_3 [p(t), [x(t), \rho_S]] \right. \\ & \left. + A_4 ([p(t), \{p(t) - im\omega_0 x(t)\} \rho_S] + [\rho_S \{p(t) + im\omega_0 x(t)\}, p(t)]) \right) \end{aligned} \quad (4.12)$$

by defining

$$\begin{aligned} A_1 &= \frac{1}{\omega_0} \text{Im}(T_1), & A_2 &= \frac{1}{\omega_0} (\text{Re}(T_1) + \text{Im}(T_2)), \\ A_3 &= \frac{1}{\omega_0} \text{Re}(T_2), & A_4 &= \frac{1}{\omega_0} \text{Im}(T_2). \end{aligned} \quad (4.13)$$

At the second-order, we need to evaluate $T_1^{(2)}$ and $T_2^{(2)}$ defined in equations (4.50) and (4.51). Both of them can be calculated exactly by using the rational expansion of the hyperbolic cotangent

$$\coth(\beta\omega_0) = \frac{1}{\beta\omega_0} + \sum_{n=1}^{\infty} \frac{2\beta\omega_0}{\beta^2\omega_0^2 + n^2\pi^2} \quad (4.14)$$

as follows:

$$T_1^{(2)} = f_{\omega_0} \left(\gamma \coth(\beta\omega_0) - i\omega_0 \frac{\delta m}{m} \right), \quad (4.15)$$

$$T_2^{(2)} = f_{\omega_0} \gamma (I - i), \quad (4.16)$$

where the mass renormalization factor $\delta m/m$ and the decay constant γ are defined in Ref. (Ford et al., 1996) as

$$\frac{\delta m}{m} = \Omega \tau_e, \quad \gamma = \omega_0^2 \tau_e, \quad (4.17)$$

and

$$I = -\frac{1}{\beta\Omega} + \frac{2}{\pi} \left(\text{Re} \left\{ \psi_0 \left[1 - i \frac{\beta\omega_0}{\pi} \right] \right\} - \psi_0 \left[\frac{\beta\Omega}{\pi} \right] \right), \quad (4.18)$$

where $\psi_0[x]$ is the digamma function and Re stands for the real part of its argument. One should note that $T_1^{(2)}$ and $T_2^{(2)}$ of equations (4.50) and (4.51) correspond to C and S integrals of Ref. (Ford et al., 1996), respectively. They were evaluated at the $\Omega \rightarrow \infty$ limit in Ref. (Ford et al., 1996), which might cause some inconsistencies (Fleming et al., 2011). Using the mass renormalization and damping constants as defined in Ref. (Ford et al., 1996), one can see that the main modification from the exact integration is scal-

ing of these quantities by the electron structure factor $f_{\omega_0} = \Omega^2/(\omega_0^2 + \Omega^2)$. f_{ω_0} scaling factor here is important because although mass renormalization is obtained by infinite Ω limit, Ω needs to be finite for the definition of δm to be meaningful and f_{ω_0} provides a cutoff for the calculated quantities at high oscillator frequency ω_0 , also. I term in Eq. (4.16), which does not contribute to the master equation at the rotating wave approximation level, is also evaluated exactly here in terms of digamma functions and depends on the ratio of the temperature to the cutoff frequency of the blackbody field as well as the oscillator frequency.

Using the exact integrals of $T_1^{(2)}$ and $T_2^{(2)}$ given by equations (4.15) and (4.16) in Eq. (4.13), we get the second-order coefficients of master Eq. (4.12)

$$\begin{aligned} A_1^{(2)} &= -f_{\omega_0} \frac{\delta m}{m}, & A_2^{(2)} &= 2f_{\omega_0} \frac{\gamma}{\omega_0} N(\beta \omega_0), \\ A_3^{(2)} &= f_{\omega_0} \frac{\gamma}{\omega_0} I, & A_4^{(2)} &= -f_{\omega_0} \frac{\gamma}{\omega_0}, \end{aligned} \quad (4.19)$$

where $N(\beta \omega_0)$ is the thermal occupation at inverse temperature β and oscillator frequency ω_0 .

The third-order contributions which are due, partly, to the diamagnetic term A^2 in the interaction Hamiltonian will be obtained by evaluating complex T_{31} and T_{32} and real T_{33} and T_{34} integrals defined in the Appendix including equations (4.52)-(4.55). These integrals can, also, be evaluated exactly by using the rational expansion of $\coth(\beta \omega_0)$ function Eq. (4.14). The results are

$$T_{31} = -\gamma f_{\omega_0}^2 \left[\frac{\delta m}{m} \coth(\beta \omega_0) - \frac{\gamma}{\omega_0} I \right] - i \omega_0 f_{\omega_0}^2 \left[\left(\frac{\gamma}{\omega_0} \right)^2 - \left(\frac{\delta m}{m} \right)^2 \right], \quad (4.20)$$

$$T_{32} = -\gamma f_{\omega_0}^2 \left[\frac{\gamma}{\omega_0} \coth(\beta \omega_0) + \frac{\delta m}{m} I \right] + i 2\gamma \frac{\delta m}{m} f_{\omega_0}^2, \quad (4.21)$$

$$T_{33} = -\gamma f_{\omega_0} \frac{\delta m}{m} \left(\frac{1}{\beta \omega_0} + \coth(\beta \omega_0) \right), \quad (4.22)$$

$$T_{34} = \frac{2}{\pi} \gamma f_{\omega_0} \frac{\delta m}{m} \Psi_0[\beta, \Omega, \omega_0], \quad (4.23)$$

where I is defined in Eq. (4.18), $\Psi_0[\beta, \Omega, \omega_0] = \psi_0 \left[\frac{\beta \Omega}{\pi} \right] - \text{Re} \{ \psi_0 \left[1 - \frac{i \beta \omega_0}{\pi} \right] \}$ and $\psi_0(x)$ is the digamma function. The dominant term in all four integrals involve product of decay rate and the mass renormalization and their squares which are modulated by the cutoff factor f_{ω_0} . The real parts of all four are temperature dependent while the imaginary parts of T_{31} and T_{32} depend on only the mass renormalization and the inter-

action strength terms. One should note that, if the cutoff frequency of the blackbody field is chosen as the causally allowed highest value as $\Omega = \tau_e^{-1}$ then $\frac{\delta m}{m} = 1$. Noting that $\frac{\gamma}{\omega_0} \approx 10^{-7}$ for the optical frequencies, one can drop the $\Psi_0[\beta, \Omega, \omega_0]$ term of T_{31} and $\coth(\beta \omega_0)$ term of T_{32} .

The third-order contribution to the coefficients of Eq. (4.12) can be obtained by plugging the results displayed in equations (4.20)-(4.23) into the definitions in Eq. (4.13), which results

$$A_1^{(3)} = -f_{\omega_0}^2 \left[\frac{\gamma^2}{\omega_0^2} - \left(\frac{\delta m}{m} \right)^2 \right], \quad (4.24)$$

$$A_2^{(3)} = -f_{\omega_0}^2 \frac{\gamma}{\omega_0} \left[\frac{\delta m}{m} \left(1 + \frac{1}{f_{\omega_0}} \right) [1 + 2N(\omega_0)] - \frac{\gamma}{\omega_0} I \right] \\ + f_{\omega_0} \frac{\gamma}{\omega_0} \frac{\delta m}{m} \left[2f_{\omega_0} - \frac{1}{\beta \omega_0} \right], \quad (4.25)$$

$$A_3^{(3)} = -f_{\omega_0}^2 \frac{\gamma}{\omega_0} \left[\frac{\gamma}{\omega_0} [1 + 2N(\beta \omega_0)] - \frac{\delta m}{m} \left(-I + \frac{2}{\pi} \frac{1}{f_{\omega_0}} \right) \right], \quad (4.26)$$

$$A_4^{(3)} = 2f_{\omega_0}^2 \frac{\gamma}{\omega_0} \frac{\delta m}{m}. \quad (4.27)$$

The most important difference between equations (4.24)-(4.27) and the third-order coefficients in equation (7.16) of Ref. (Ford et al., 1996) is the $f_{\omega_0}^2$ scaling which provides frequency cutoff for each one of those coefficients. We, also, provide exact expressions for I_3 and I_4 integrals of Ref. (Ford et al., 1996) which were unevaluated there.

The fourth-order contributions are tedious but straightforward to obtain by performing the integrals defined in Eq. (4.63) and are given in equations (4.64)-(4.68) of the Appendix. Here, we collect the fourth-order contributions to the coefficients of the

Eq. (4.12):

$$A_1^{(4)} = -f_{\omega_0}^3 \left[\left(\frac{\gamma}{\omega_0} \right)^2 \left(1 - 3 \frac{\delta m}{m} \right) - \left(\frac{\gamma}{\Omega} \right)^2 + \left(\frac{\delta m}{m} \right)^3 \right], \quad (4.28)$$

$$\begin{aligned} A_2^{(4)} = & -f_{\omega_0}^3 \frac{\gamma}{\omega_0} \left[\left(-\frac{5}{2} \frac{\gamma}{\Omega} + \frac{\delta m}{m} \left(\frac{1}{2} + \frac{\gamma}{\Omega} - 3 \frac{\delta m}{m} \right) \right) [1 + 2N(\beta \omega_0)] \right. \\ & \left. + \frac{\gamma}{\omega_0} I + 2 \frac{\gamma}{\Omega} + \frac{\delta m}{m} \left(3 \frac{\delta m}{m} - \frac{\gamma}{\Omega} \right) \right] \\ & - f_{\omega_0}^2 \gamma \beta \left[\frac{1}{\pi^2} \frac{\gamma}{\omega_0} \text{Im}[\psi_1(\beta, \omega_0)] - \frac{1}{2} \frac{\delta m}{m} \text{csch}^2(\beta \omega_0) \right], \end{aligned} \quad (4.29)$$

$$\begin{aligned} A_3^{(4)} = & -f_{\omega_0}^3 \frac{\gamma}{\omega_0} \left[\frac{1}{2} \frac{\gamma}{\omega_0} \left(\left(\frac{\omega_0}{\Omega} \right)^2 - 1 \right) [1 + 2N(\beta \omega_0)] - I \left(3 \frac{\gamma}{\Omega} + \frac{\delta m}{m} \left(3 \frac{\delta m}{m} - \frac{\gamma}{\Omega} \right) \right) \right. \\ & \left. + \frac{1}{\beta \Omega} \left(\frac{1}{f_{\omega_0}} \frac{\delta m}{m} \left(3 \frac{\delta m}{m} + \frac{\gamma}{\Omega} \right) + \frac{\gamma}{\Omega} \left(1 + \left(\frac{\omega_0}{\Omega} \right)^2 \right) \right) \right] \\ & - f_{\omega_0}^2 \gamma \beta \frac{\gamma}{\omega_0} \left[\frac{1}{2} \text{csch}^2(\beta \omega_0) - \frac{2}{\pi^2} \left(1 + 2 \frac{\Omega}{\gamma} \left(\frac{\delta m}{m} \right)^2 \right) \psi_1 \left[\frac{\beta \Omega}{\pi} \right] \right. \\ & \left. + \frac{1}{\pi^2} \frac{\omega_0}{\gamma} \left(\frac{\delta m}{m} \right) \left(\text{Im}\{\psi_1(\beta, \omega_0)\} + \frac{1}{\pi} \frac{\beta \Omega}{f_{\omega_0}} \left(\frac{\Omega}{\omega_0} \right) \left(\frac{\delta m}{m} \right) \psi_2 \left[\frac{\beta \Omega}{\pi} \right] \right) \right] \end{aligned} \quad (4.30)$$

$$A_4^{(4)} = -f_{\omega_0}^3 \frac{\gamma}{\omega_0} \left[2 \frac{\gamma}{\Omega} + \frac{\delta m}{m} \left(3 \frac{\delta m}{m} - \frac{\gamma}{\Omega} \right) \right], \quad (4.31)$$

where $\psi_i(x)$ is the polygamma function of order i and $\text{Im}\{\psi_1(\beta, \omega_0)\} = \text{Im}\{\psi_1(1 - i\beta \omega_0/\pi)\}$. All four coefficients have an overall $f_{\omega_0}^3$ scaling and are function of various combinations of interaction strength γ/ω_0 , mass renormalization $\delta m/m$ and γ/Ω . $A_1^{(4)}$ and $A_4^{(4)}$ are independent of the temperature while $A_2^{(4)}$ and $A_3^{(4)}$ have complicated temperature dependence through thermal occupation factor as well as various polygamma functions.

4.3 Discussion

We will discuss the convergence of the expansion and the relative importance of various terms in Eq. (4.12) by a slight change of the master equation to

$$\begin{aligned} \frac{\partial \rho_S}{\partial t} = & -\frac{1}{2m\hbar} \{ i\Delta[p^2, \rho_S] + D_{xx}[p, [p, \rho_S]] \\ & + im\omega_0 \lambda[p, \{x, \rho_S\}] + m\omega_0 D_{xp}[p, [x, \rho_S]] \}, \end{aligned} \quad (4.32)$$

where $\Delta = \sum_i A_1^{(i)}$ is mass renormalization, $\lambda = \sum_i A_4^{(i)}$ is decay constant, $D_{xx} = \sum_i (A_2^{(i)} - A_4^{(i)})$ is normal diffusion and $D_{xp} = \sum_i A_3^{(i)}$ is anomalous diffusion coefficients. To dis-

cuss the relevance of various terms in Eq. (4.32), we will state the adjoint master equation that governs the dynamics of the moments of position and momentum operators of the oscillator as (Breuer and Petruccione, 2002)

$$\begin{aligned} \frac{d}{dt} \langle O \rangle_t = & \frac{i}{\hbar} \langle [H_S, O] \rangle_t + \frac{1}{2m\hbar} \{ i\Delta \langle [p^2, O] \rangle_t - D_{xx} \langle [p, [p, O]] \rangle_t \\ & + im\omega_0 \lambda \langle \{x, [p, O]\} \rangle_t - m\omega_0 D_{xp} \langle [p, [x, O]] \rangle_t \}, \end{aligned} \quad (4.33)$$

where D_{xx} term causes diffusion of the variance of the oscillator position operator, D_{xp} plays the same role for $x \cdot p$, Δ term is a change in the mass and λ is decay constant of the position. It is obvious that while diffusion constants depend on the temperature, drift terms, mass renormalization and the damping constants, are temperature independent.

By rearranging equations (4.19), (4.24) and (4.28) the renormalization constant Δ can be expressed in terms of τ_e , ω_0 and Ω in a more suggestive form as

$$\Delta = -\tilde{\Omega} f_{\tilde{\omega}_0} \{ 1 + \tilde{\Omega} f_{\tilde{\omega}_0} [(\tilde{\omega}_0^2 - 1) + f_{\tilde{\omega}_0} (\tilde{\Omega} (1 - 3\tilde{\omega}_0^2) + \tilde{\omega}_0^2 (1 - \tilde{\omega}_0^2))] \}, \quad (4.34)$$

where $\tilde{\omega}_0 = \omega_0/\Omega$ and $\tilde{\Omega} = \Omega \tau_e$ are the dimensionless oscillator and the bath cutoff frequency, respectively and $f_{\tilde{\omega}_0} = 1/(1 + \tilde{\omega}_0^2)$ is the bath cutoff function. Note that the inverse of $\tilde{\omega}_0$ can be considered as a resonance factor, its a measure of the effectiveness of the interaction between the charged oscillator and the environment, a large or small value of $\tilde{\omega}_0$ indicates that the oscillator is detuned from the peak of the environmental spectral distribution. The successive terms in the nested parenthesis of the expression for Δ come from the second-, the third- and the fourth-order corrections, respectively. These terms are in the form of product of mass renormalization and cutoff factor modulated with the ratio of the oscillator frequency to the bath cutoff. The ratio of the third- and the fourth-order to the second-order contribution can be expressed as $\tilde{\Omega}(\tilde{\omega}_0^2 - 1)/(\tilde{\omega}_0^2 + 1)$ and $\tilde{\Omega}^2(1 - 3\tilde{\omega}_0^2)/(\tilde{\omega}_0^2 + 1)^2$ which approach $\tilde{\Omega}$ and 0 at the high frequency limit $\omega_0 \gg \Omega$, respectively. In the low frequency limit $\tilde{\omega}_0 \ll 1$, these ratios tend to $-\tilde{\Omega}$ and $\tilde{\Omega}^2$.

Similar to Δ , we can rewrite λ in a simpler form as

$$\lambda = -f_{\tilde{\omega}_0} \tilde{\omega}_0 \tilde{\Omega} \{ 1 + f_{\tilde{\omega}_0} \tilde{\Omega} [-2 + f_{\tilde{\omega}_0} (\tilde{\Omega}(3 - \tilde{\omega}_0^2) + 2\tilde{\omega}_0^2)] \}. \quad (4.35)$$

The perturbative terms enter into the expression of λ as powers of $f_{\tilde{\omega}_0} \tilde{\Omega}$. A comparison

of equations (4.34) and (4.35) shows that the leading terms of Δ and λ are $f_{\tilde{\omega}_0} \tilde{\Omega}$ and $f_{\tilde{\omega}_0} \tilde{\omega}_0 \tilde{\Omega}$, respectively. So, when the oscillator is in resonance with the peak frequency of the environment, the magnitude of renormalization and the damping constants would be comparable while off-resonance λ is expected to be small compared to Δ for low frequencies. At high frequencies, $f_{\tilde{\omega}_0} \rightarrow 0$ and Δ and λ tend to zero.

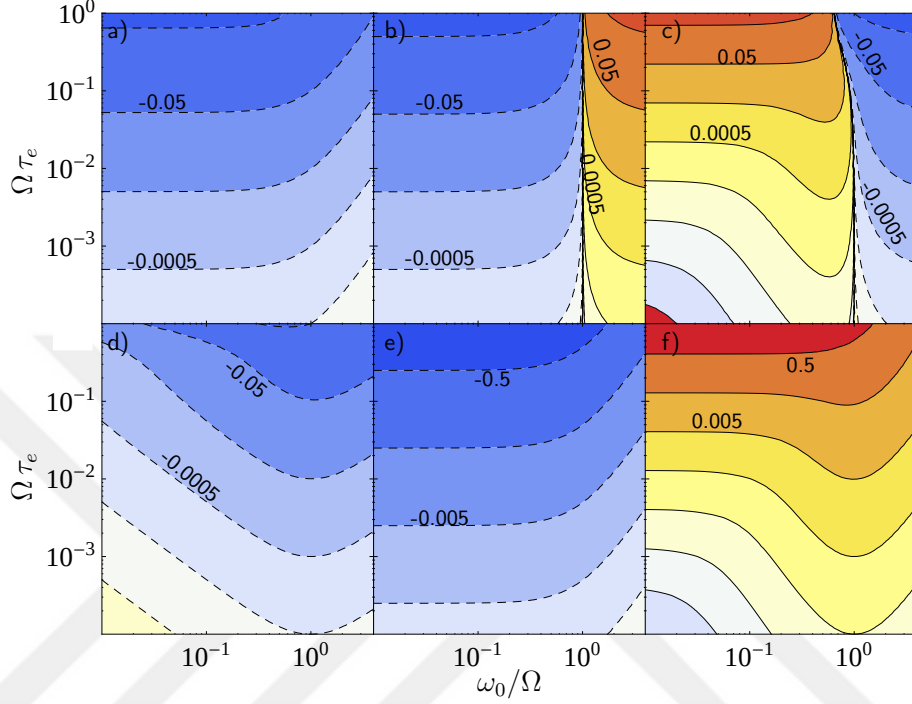


Figure 4.1. Magnitude (a) and ratio of the third- (b) and the fourth-order contributions (c) to the second-order term for Δ and γ as function of the oscillator frequency ω_0 and the cutoff frequency of the blackbody environment Ω . Both axis scales and the contour separations are logarithmic and the dashed lines indicate negative contours.

The magnitude of Δ and λ along with the ratio of the third- and the fourth-order contributions to their leading term, as function of $\Omega \tau_e$ and ω_0/Ω , are presented in Fig. 4.1(a)-(c) and Fig. 4.1(d)-(f), respectively. Both the renormalization and the decay constant are negative for the considered frequency range. $\Delta^{(3)}/\Delta$ and $\Delta^{(4)}/\Delta$ changes sign at $\omega_0 = \Omega$ while $\lambda^{(3)}/\lambda < 0$ and $\lambda^{(4)}/\lambda > 0$ for the whole range of ω_0/Ω and Ω considered here. These ratios can be considered as a proxy for the convergence of the perturbative expansion of the master equation. It is clear that $B^{(3)}$ and $B^{(4)}$ contributions partially cancel each other and only the second-order term $B^{(2)}$ might be sufficient to account for the dynamics of the oscillator for the most part of the considered parameter range. Only very near the upper limit of the cutoff frequency ($\Omega \approx \tau_e^{-1}$), the whole perturbative treatment might be divergent because the magnitude as well as the contributions approach -1 and ± 1 , respectively. It is probable that including even

higher-order terms in the master equation will not lead to a converged Δ and λ in this parameter regime. One should note that the interaction strength in the current model is $\gamma/\omega_0 = \omega_0 \tau_e$ and the non-convergence issue is independent of its value for high cutoff Ω . We display the ω_0 dependent Δ and λ at three different $\Omega \tau_e$ values in Fig. (4.2) which indicates that while $B^{(3)}$ and $B^{(4)}$ contributions are significant at $\Omega \tau_e = 0.5$, they decrease substantially when $\Omega \tau_e$ is reduced to 0.1 and become insignificant at $\Omega \tau_e = 10^{-3}$. It can be deduced from Fig. 4.1(b), (c), (e) and (f) that the relative contributions of the third- and the fourth-order terms are less than 1% for $\Omega \tau_e < 0.01$ and including only the second-order terms in master equation might be sufficient for an accurate description of the dynamics of the charged oscillator.

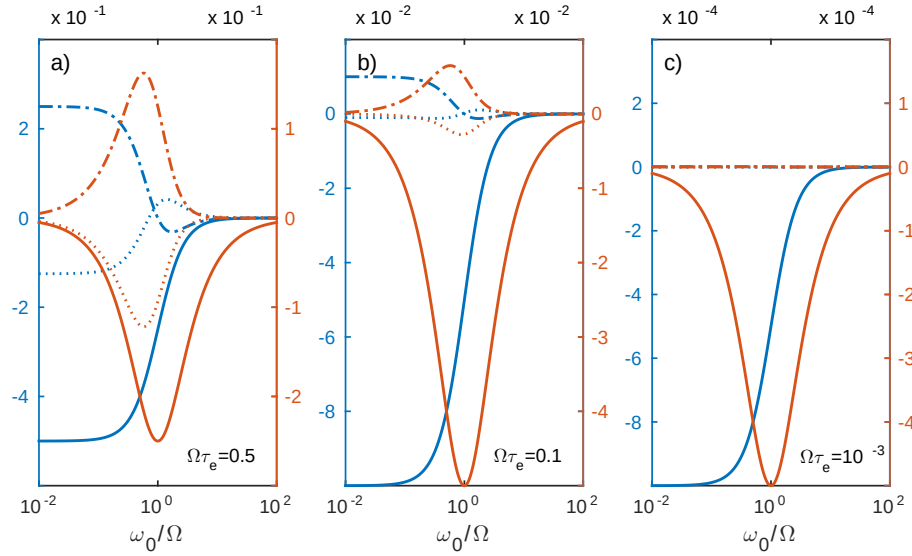


Figure 4.2. The second- (straight), the third- (dot-dashed) and the fourth- (dotted) order contributions to mass renormalization (left axis) and damping rate (right axis) as function of the dimensionless oscillator frequency at three different environmental cutoff frequencies (a) $\Omega \tau_e = 0.5$, b) $\Omega \tau_e = 0.1$ and c) $\Omega \tau_e = 0.001$.

Both the normal and the anomalous diffusion coefficients are temperature dependent. We will first discuss the high temperature limit of the higher-order contributions to D_{xx} and D_{xp} . Defining $z = \beta \Omega$, $z \ll 1$ corresponds to the high temperature regime and Taylor expanding hyperbolic cotangent and polygamma functions around zero and taking the first term of the expansion as, $\coth(\tilde{\omega}_0 z) \rightarrow 1/(\tilde{\omega}_0 z)$, $\text{csch } \tilde{\omega}_0 z \rightarrow 1/\tilde{\omega}_0 z$,

$\psi_1(1 \pm i \tilde{\omega}_0 z / \pi) \rightarrow \pi^2/6$, $\psi_1(z/\pi) \rightarrow \pi^2/z^2$ and $\psi_2(z/\pi) \rightarrow -2\pi^3/z^3$ we get

$$\begin{aligned} D_{xx}^{(2)} &= f_{\tilde{\omega}_0} \frac{\tilde{\Omega}}{z}, \\ D_{xx}^{(3)} &= -f_{\tilde{\omega}_0}^2 \frac{\tilde{\Omega}^2}{z} (3 + \tilde{\omega}_0^2), \\ D_{xx}^{(4)} &= f_{\tilde{\omega}_0}^3 \frac{\tilde{\Omega}^2}{z} [3\tilde{\Omega} + \tilde{\omega}_0^2(2 - \tilde{\Omega})], \end{aligned} \quad (4.36)$$

and

$$\begin{aligned} D_{xp}^{(2)} &= f_{\tilde{\omega}_0} \tilde{\omega}_0 \frac{\tilde{\Omega}}{z}, \\ D_{xp}^{(3)} &= -2f_{\tilde{\omega}_0}^2 \tilde{\omega}_0 \frac{\tilde{\Omega}^2}{z} (2 + \tilde{\omega}_0^2), \\ D_{xp}^{(4)} &= f_{\tilde{\omega}_0}^3 \tilde{\omega}_0 \frac{\tilde{\Omega}^2}{z} [6\tilde{\Omega} + \tilde{\omega}_0^2(3 + \tilde{\omega}_0^2)(1 + \tilde{\Omega})], \end{aligned} \quad (4.37)$$

which indicate that all contributions to D_{xx} and D_{xp} are linear in temperature at the high temperature limit. Both the second- and the fourth-order contributions to both diffusion coefficients are positive while the third-order ones are always negative.

One should note that from a comparison of equations (4.36)-(4.37) that the terms contributing to D_{xp} contain an $\tilde{\omega}_0$ factor which is a measure of interaction strength. So, anomalous diffusion coefficient is expected to be much smaller than the normal diffusion coefficient in the weak coupling limit. In this limit ($\tilde{\omega}_0 \rightarrow 0$), $f(\tilde{\omega}_0) = 1$ and the convergence of the coefficients depends solely on the cutoff frequency of the environment as can be seen from equations (4.36)-(4.37).

For the low temperature limit $z \gg 1$, we can use the approximations $\coth(z) \rightarrow 1$, $\operatorname{csch}(z) \rightarrow 0$, $\operatorname{Re}\{\psi_0(1 - i \tilde{\omega}_0 z / \pi) - \psi_0(z/\pi)\} \rightarrow \log(\tilde{\omega}_0)$, $z \operatorname{Im}[\psi_1(1 - i \tilde{\omega}_0 z / \pi)] \rightarrow \pi/\tilde{\omega}_0$, $z^2 \psi_2(z/\pi) \rightarrow -\pi^2$ and $z \psi_1(z/\pi) \rightarrow \pi$ to express D_{xx} and D_{xp} as

$$\begin{aligned} D_{xx}^{(2)} &= f_{\tilde{\omega}_0} \tilde{\omega}_0 \tilde{\Omega}, \\ D_{xx}^{(3)} &= -f_{\tilde{\omega}_0}^2 \tilde{\omega}_0 \tilde{\Omega}^2 \left[2 + \tilde{\omega}_0^2 - \frac{2}{\pi} \tilde{\omega}_0 \log(\tilde{\omega}_0) \right], \\ D_{xx}^{(4)} &= -f_{\tilde{\omega}_0}^3 \tilde{\omega}_0 \tilde{\Omega}^2 \left\{ \frac{1}{2} - 3\tilde{\Omega} + \left(\tilde{\Omega} - \frac{5}{2} \right) \tilde{\omega}_0^2 + \frac{\tilde{\omega}_0}{\pi} [1 + \tilde{\omega}_0^2 + 2 \log(\tilde{\omega}_0)] \right\}, \end{aligned} \quad (4.38)$$

and

$$\begin{aligned}
D_{xp}^{(2)} &= \frac{2}{\pi} f_{\tilde{\omega}_0} \tilde{\omega}_0 \tilde{\Omega} \log(\tilde{\omega}_0), \\
D_{xp}^{(3)} &= -f_{\tilde{\omega}_0}^2 \tilde{\omega}_0 \tilde{\Omega}^2 \left[\tilde{\omega}_0 + \frac{2}{\pi} (2 + \tilde{\omega}_0^2) \log(\tilde{\omega}_0) \right], \\
D_{xp}^{(4)} &= \frac{1}{\pi} f_{\tilde{\omega}_0}^3 \tilde{\omega}_0 \tilde{\Omega}^2 \left\{ 5\tilde{\Omega} - 1 + \tilde{\omega}_0^4 (2 + \tilde{\Omega}) + (1 + 6\tilde{\Omega}) \tilde{\omega}_0^2 + \frac{\pi}{2} \tilde{\omega}_0 (1 - \tilde{\omega}_0^2) \right. \\
&\quad \left. + 2 [3\tilde{\omega}_0^2 + \tilde{\Omega} (3 - \tilde{\omega}_0^2)] \log(\tilde{\omega}_0) \right\}.
\end{aligned} \tag{4.39}$$

The ω_0/Ω dependence of D_{xx} and D_{xp} should be the qualitatively different for the low and high environment temperatures based on the spectral distribution, $I(\omega_0) = J(\omega_0) \coth(\beta \omega_0)$, of the environment.

We have considered low ($k_B T = 0.01 \hbar \Omega$), intermediate ($k_B T = \hbar \Omega$) and high ($k_B T = 100 \hbar \Omega$) temperature limits of the environment and display the magnitude and the ratio of the third- and the fourth-order contributions to the second-order one for the normal diffusion coefficient D_{xx} in Fig. 4.3(a)-(i). From the first column of the figure, it is obvious that D_{xx} increases with increasing initial temperature as expected, because of the increase in the thermal excitations with increasing temperature. Similar to the drift terms, the third- and the fourth-order contributions to D_{xx} have opposite signs for intermediate and high T values while at low temperatures $D_{xx}^{(4)}$ is negative or positive depending on ω_0 and Ω . The interaction strength dependence of D_{xx} changes somewhat from high to low temperature region (compare Fig. 4.3(a) and Fig. 4.3(g)); while it is almost independent of $\tilde{\omega}_0 = \omega_0/\Omega \approx 1$ at high temperatures, it increases with decreasing $\tilde{\omega}_0$ in the low temperatures. Fig. 4.3(b), (e) and (h) indicate that the ratio $D_{xx}^{(3)}/D_{xx}^{(2)}$ is always negative and almost independent of the temperature. Similarly, $D_{xx}^{(4)}/D_{xx}^{(2)}$ (Fig. 4.3(c), (f) and (i)) is positive and almost identical at high and intermediate temperatures while it becomes negative at low ω_0 and Ω region at low temperatures. Since $D_{xx}^{(3)}$ and $D_{xx}^{(4)}$ have opposite signs for the most of the considered parameter range, instead of taking into account only the second- and the third-order contributions, keeping only the second-order terms might be more accurate.

To delineate the relative importance of higher-order terms for the anomalous diffusion coefficient D_{xp} , we display its value and the relative third- and the fourth-order contributions as function of ω_0 and Ω in Fig. 4.5(a)-(i) at low, intermediate and high temperatures. The magnitude of D_{xp} originating from $B^{(2)}$, $B^{(3)}$ and $B^{(4)}$ are, also, displayed in Fig. (4.4) and Fig. (4.6) for different Ω values at high and low temperatures,

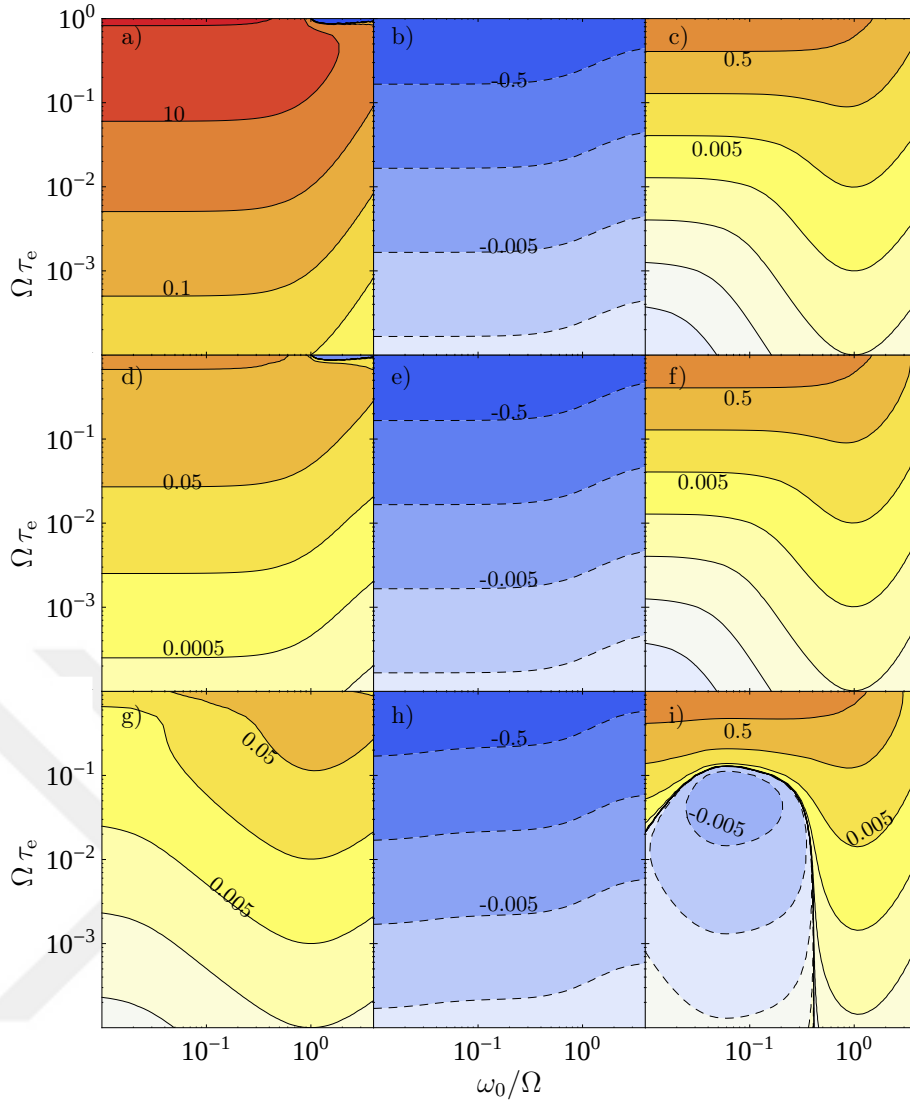


Figure 4.3. Magnitude (a, d, g) and the ratio of the third- (b, e, h) and the fourth-order contributions (c, f, i) to the second-order term for the normal diffusion coefficient D_{xx} as function of the oscillator frequency ω_0 and the cutoff frequency of the black-body environment Ω at high ($k_B T = 100 \hbar \Omega$) (a, b, c), intermediate ($k_B T = \hbar \Omega$) (d, e, f) and low ($k_B T = 0.01 \hbar \Omega$) (g, h, i) temperatures.

respectively.

Although D_{xp} at high and intermediate temperatures show similar behavior as the normal diffusion coefficient (compare the first two rows of Fig. (4.3) and Fig. (4.5)), such as being positive for all ω_0 and Ω values and $D_{xp}^{(3)}$ and $D_{xp}^{(4)}$ having opposite signs, it becomes negative for $\tilde{\omega}_0 < 1$ at low temperatures (see Fig. 4.5(g)). Sign of D_{xp} is related with the squeezing of the position or the momentum of the oscillator (Paz and Roncaglia, 2009). The changing sign indicates a change in localized dynamical variable depending on the initial temperature of the environment being high or low.

The qualitative difference in both D_{xx} and D_{xp} and the contributions to them in all

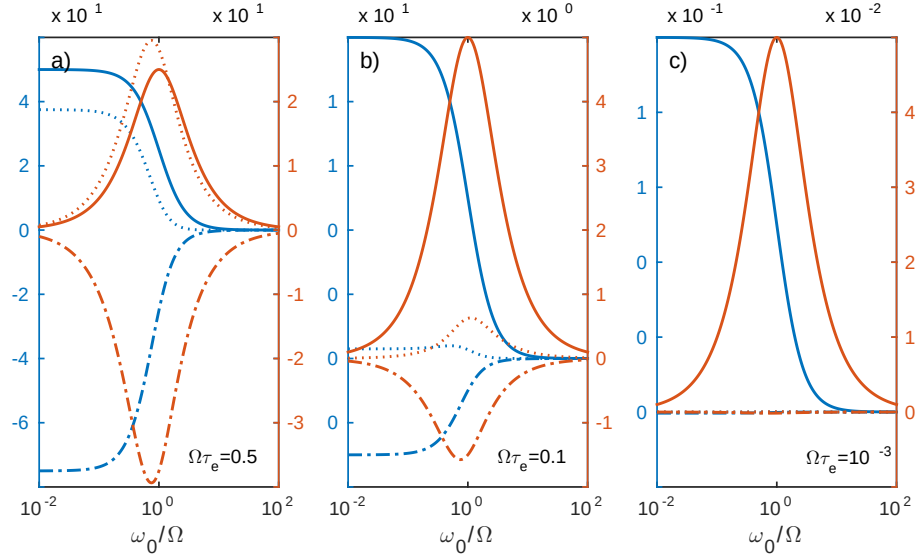


Figure 4.4. Magnitude of $B^{(2)}$ (straight), $B^{(3)}$ (dot-dashed) and $B^{(4)}$ (dotted) contributions to normal (D_{xx} : left axis) and anomalous diffusion coefficients (D_{xp} : right axis) at high temperature ($\beta \Omega = 0.01$) as function of the oscillator frequency at a) $\Omega \tau_e = 0.5$, b) $\Omega \tau_e = 0.1$ and c) $\Omega \tau_e = 10^{-3}$.

the considered orders between the high and low temperature environments is prominent when one compares Fig. (4.4) and Fig. (4.6). Independent of the temperature and the bath cutoff frequency, the shapes of the absolute values of the second-, the third- and the fourth-order diffusion terms as function of the oscillator frequency are similar: the components of D_{xp} (D_{xx}) at high (low) temperatures display a single resonance peak at $\omega_0 = \Omega$, while the low temperature behavior of D_{xp} is similar to an anti-resonance structure. Such behavior can be understood by referring to the spectral density function.

4.4 Conclusions

We have considered the exact treatment of the third- and the fourth-order corrections to the master equation of a charged harmonic oscillator interacting with a black-body radiation field by using Krylov averaging method. The oscillator-bath interaction is assumed to be in velocity-coupling form. Diamagnetic interaction term, which is generally neglected, is also taken into account. It is found that including higher than the second-order terms would not change the structure of the master equation, *i.e.* they do not introduce any new commutators.

For mass renormalization, decay constant, normal, and anomalous diffusion coefficients for the oscillator in the electromagnetic field, we obtained exact analytical expressions, to the fourth-order, which indicate that mass renormalization and decay

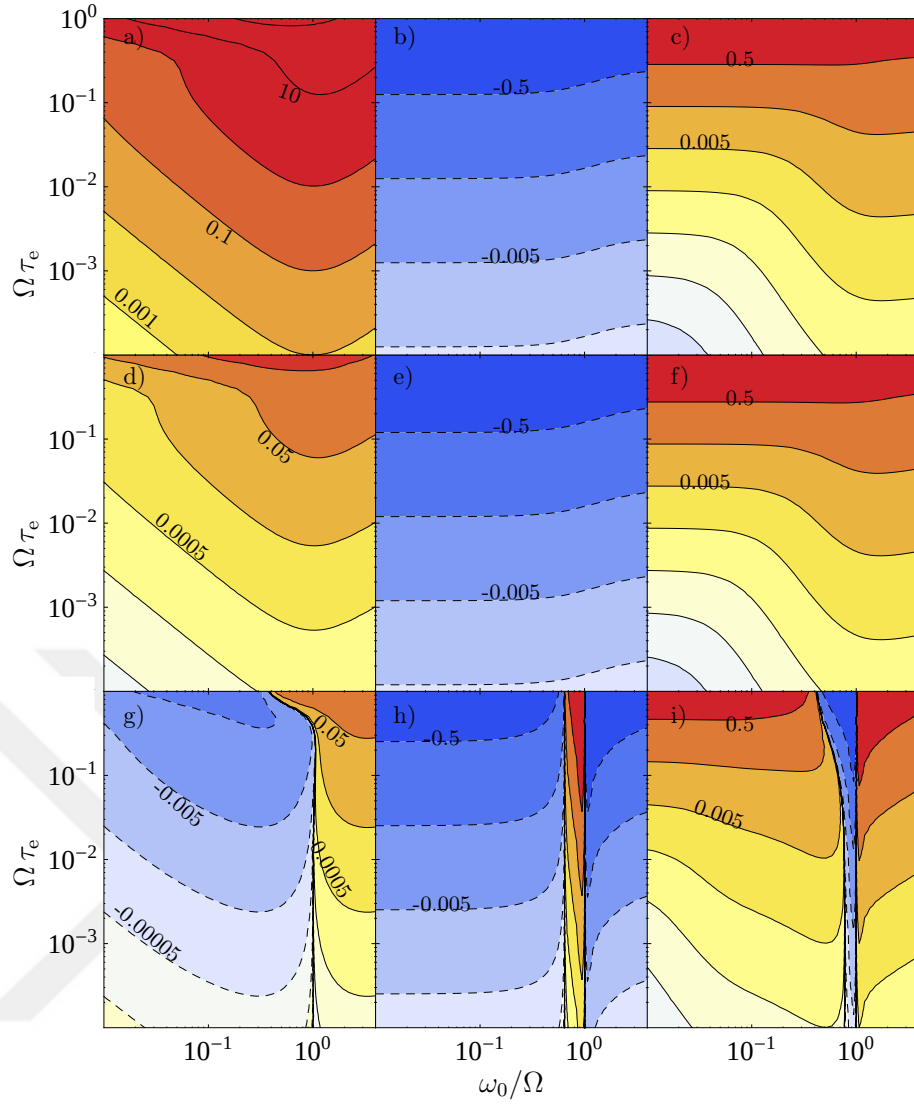


Figure 4.5. Same as figure 4.3 for D_{xp} .

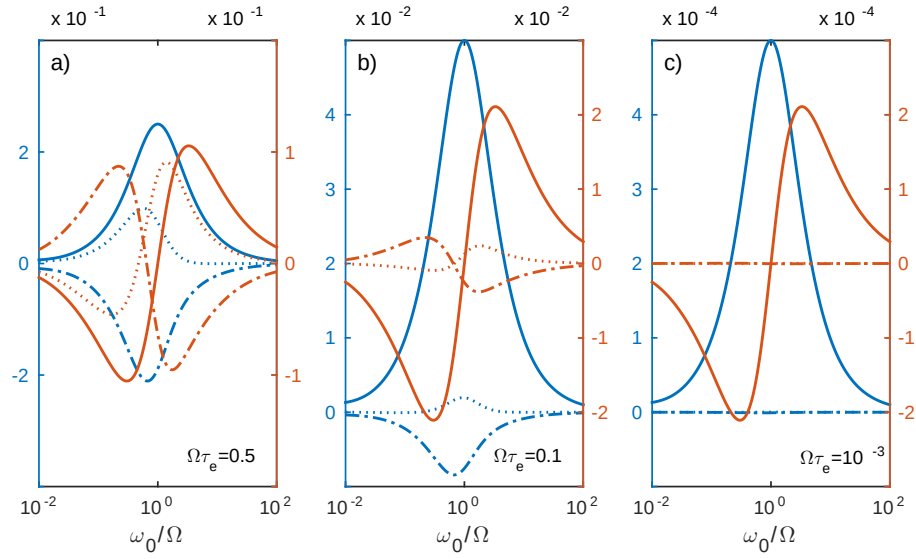


Figure 4.6. Same as figure 4.4 for low temperature ($\beta\Omega = 100$).

constant depend on the oscillator frequency and the bath cutoff frequency while the diffusion coefficients are sensitive to the bath temperature along with those two variables. Anomalous diffusion coefficient is found to go under a qualitative change between the high and the low temperature limits of the environment.

The aim of this study was to delineate the conditions under which the higher-order corrections to the master equation might make a difference in the dynamics of the charged harmonic oscillator in electromagnetic field. It is found that the single most important parameter in that context is the chosen cutoff frequency Ω compared to the inverse of electron time τ_e . We have shown that the third- and the fourth-order contributions are, mostly, opposite in sign and cancel each other. So, keeping only the second-order terms in the master equation would be accurate enough for the weak interaction regime and $\Omega \tau_e < 0.01$. For $\Omega \tau_e > 0.1$, a perturbative expansion would not be appropriate as the order of consecutive terms have comparable magnitudes.

4.5 Krylov Method of Averaging for Deriving Master Equation

Here, we summarize the Krylov averaging method used in Ref. (Ford et al., 1996), obtain exact analytical expressions for the integrals involved in the definition of $B^{(2)}(\rho_S, t)$ and $B^{(3)}(\rho_S, t)$ and give a detailed derivation of the fourth-order correction $B^{(4)}(\rho_S, t)$.

Since the system-bath interaction is assumed to be weak, one would expect that the change in the product form of the initial density matrix of the total system is minimal and $\rho(t)$ can be expressed as $\rho(t) \cong \rho_S \otimes \rho_R$ where the difference from the product terms can be approximated as a power expansion in the perturbation parameter ε and a function of the oscillator density matrix ρ_S as:

$$\rho(t) = \rho_S(t) \otimes \rho_R + \varepsilon F^{(1)}(\rho_S, t) + \varepsilon^2 F^{(2)}(\rho_S, t) + \dots, \quad (4.40)$$

where terms containing ε represent small fluctuations in the amplitude of the density matrix of the whole system around $\rho_S(t) \otimes \rho_R$. Similarly, the time rate of change of the system density matrix can be written as

$$\frac{\partial \rho_S(t)}{\partial t} = \varepsilon B^{(1)}(\rho_S, t) + \varepsilon^2 B^{(2)}(\rho_S, t) + \dots \quad (4.41)$$

For the definition $\rho_S(t) = \text{Tr}_R\{\rho(t)\}$ to hold, $\text{Tr}_R\{F^{(n)}(\rho_S, t)\} = 0$. Also, $B^{(n)}(\rho_S, t)$

should be independent of bath operators, so $\text{Tr}_R\{B^{(n)}(\rho_S, t)\rho_R\} = B^{(n)}(\rho_S, t)$.

To obtain $B^{(n)}(\rho_S, t)$ s, we insert $\rho(t)$ of Eq. (4.40) into the von Neumann equation (Eq. (4.7)) and use Eq. (4.41) for the time dependence of the reduced density matrix of the system and match the coefficients of the equal powers of ε on the left and the right sides. The process produces a hierarchy of coupled equations for $B^{(n)}(\rho_S, t)$ and $F^{(n)}(\rho_S, t)$ as:

$$\begin{aligned}\rho_R \otimes B^{(1)} + \frac{\partial F^{(1)}}{\partial t} &= \frac{1}{i\hbar} [H_I(t), \rho_S \otimes \rho_R], \\ \rho_R \otimes B^{(n)} + \frac{\partial F^{(n)}}{\partial t} &= \frac{1}{i\hbar} [H_I(t), F^{(n-1)}] - \sum_{m=1}^{n-1} F^{(n-m)}(B^{(m)}, t).\end{aligned}\quad (4.42)$$

The set of equations in (4.42) can be solved by starting from $B^{(1)}(\rho_S, t)$ and $F^{(1)}(\rho_S, t)$ and invoking the trace over the bath conditions mentioned above. We quote the results, up to order four, from Ref. (Ford et al., 1996):

$$B^{(1)}(\rho_S, t_1) = \frac{1}{i\hbar} \{1\}, \quad (4.43)$$

$$B^{(2)}(\rho_S, t_1) = \frac{1}{(i\hbar)^2} \int_{-\infty}^{t_1} dt_2 (\{12\} - \{1:2\}), \quad (4.44)$$

$$\begin{aligned}B^{(3)}(\rho_S, t_1) &= \frac{1}{(i\hbar)^3} \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \{ \{123\} - \{12:3\} - \{13:2\} - \{1:23\} \\ &\quad + \{1:2:3\} + \{1:3:2\} \},\end{aligned}\quad (4.45)$$

$$\begin{aligned}B^{(4)}(\rho_S, t_1) &= \frac{1}{(i\hbar)^4} \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \int_{-\infty}^{t_3} dt_4 \{ \{1234\} - \{12:34\} - \{13:24\} \\ &\quad - \{14:23\} - \{1:234\} - \{123:4\} - \{124:3\} - \{134:2\} \\ &\quad + \{12:3:4\} + \{12:4:3\} + \{13:2:4\} + \{13:4:2\} \\ &\quad + \{14:3:2\} + \{1:2:34\} + \{1:3:24\} + \{1:4:23\} \\ &\quad + \{1:24:3\} + \{1:34:2\} + \{14:2:3\} + \{1:23:4\} \\ &\quad - \{1:3:2:4\} - \{1:3:4:2\} - \{1:4:2:3\} \\ &\quad - \{1:2:3:4\} - \{1:2:4:3\} - \{1:4:3:2\} \},\end{aligned}\quad (4.46)$$

where the braces $\{\dots\}$ represent the trace over the reservoir degrees of freedom of the nested commutators of the closed-system Hamiltonian at times t_i with the density

matrix $\rho_S \otimes \rho_R$ while the column sign indicates a further trace operation:

$$\begin{aligned}\{1234\} &= \text{Tr}_R\{[H_I(t_1), [H_I(t_2), [H_I(t_3), [H_I(t_4), \rho_S \rho_R]]]]\} \\ \{12 : 34\} &= \text{Tr}_R\{[H_I(t_1), [H_I(t_2), \text{Tr}_R\{[H_I(t_3), [H_I(t_4), \rho_S \rho_R]]\} \rho_R]]\}.\end{aligned}$$

Thermal expectation value of the products of field operators at different times will be represented by using $\langle ij \dots \rangle$ notation as

$$\langle 123 \dots \rangle \equiv \text{Tr}_R\{\rho_R A(t_1) A(t_2) A(t_3) \dots\}.$$

Since $A(t)$ is a Gaussian operator, the thermal expectation value of the product terms that contain an odd number of field operators are zero while those that contain an even number of $A(t_i)$ can be expressed as the sum of product of pair expectations by using Isserlis' theorem:

$$\begin{aligned}\langle 1 \rangle &= \langle 123 \rangle = 0, \\ \langle 1234 \rangle &= \langle 12 \rangle \langle 34 \rangle + \langle 13 \rangle \langle 24 \rangle + \langle 14 \rangle \langle 23 \rangle.\end{aligned}\tag{4.47}$$

Explicit form of the pair expectation $\langle 12 \rangle$ for the present problem can be easily calculated by using the time dependent field operator $A(t_i)$ as defined in Eq. (4.9) (Schlosshauer, 2007):

$$\langle 12 \rangle = \frac{\hbar}{\pi} \int_0^\infty d\omega J(\omega) \left\{ \coth\left(\frac{\hbar \omega}{2k_B T}\right) \cos[\omega(t_1 - t_2)] - i \sin[\omega(t_1 - t_2)] \right\}, \tag{4.48}$$

where $J(\omega)$ denotes the spectral density of the bath and contains all the information related to the bath-oscillator interaction and is given in Eq. (4.6) for the present problem.

Before evaluating $B^{(n)}(\rho_S, t_1)$ for $n = 1, 2, 3, 4$ of equations (4.43)-(4.46), we should note that contribution of $B^{(n)}$ s to the dynamics of the oscillator density matrix has the same form of independent of n , as:

$$\begin{aligned}\frac{\partial \rho_S}{\partial t} &= -\frac{1}{2\hbar m \omega_0} ([p(t), \{T_1 p(t) + m \omega_0 T_2 x(t)\} \rho_S] \\ &\quad + [\rho_S \{T_1^* p(t) + m \omega_0 T_2^* x(t)\}, p(t)]),\end{aligned}\tag{4.49}$$

where T_1 and T_2 are time integrals of various combinations of pair correlation functions

and can be written as $T_i = \sum_n T_i^{(n)}$ for corrections up to order n . We will provide the details of calculating $T_i^{(n)}$ up to $n = 4$.

In the remainder of the Appendix, we will outline the evaluation of the contributions to the coefficients of the commutators in Eq. (4.49) up to fourth-order. First, we note that $B^{(1)}(\rho_S, t_1) = 0$ because of the Gaussian property of $A(t)$. The second-order contribution is, also, easy to calculate: From Eq. (4.44), the $\{1 : 2\}$ term is zero and $\{12\}$ term gives $T_1^{(2)} = T_{21}$ and $T_2^{(2)} = T_{22}$ as:

$$T_{21} = \left(\frac{2\omega_0}{\hbar m} \right) \int_0^\infty dt_{12} \langle 12 \rangle \cos(\omega_0 t_{12}), \quad (4.50)$$

$$T_{22} = \left(\frac{2\omega_0}{\hbar m} \right) \int_0^\infty dt_{12} \langle 12 \rangle \sin(\omega_0 t_{12}). \quad (4.51)$$

4.5.1 Third-order contributions

The contribution of $B^{(3)}(\rho_S, t_1)$ to the system master equation can be calculated by noting that only $\{123\}$ term of Eq. (4.45) is nonzero. The brace $\{123\}$ has a total of 64 terms; half of those are equal to zero because they contain odd moments. Combination of eight triple products of the diamagnetic term $A^2(t_i)$ as well as eight terms formed by different permutations of $A^2(t_1)A(t_2)A(t_3)$ cancel each other. Hence, the remaining 16 terms can be combined as four different integrals as $T_1^{(3)} = T_{31} + T_{33}$ and $T_2^{(3)} = T_{32} + T_{34}$, where

$$T_{31} = \left(\frac{4\omega_0}{\hbar^2 m^2} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \text{Im}\{\langle 12 \rangle\} \langle 23 \rangle \cos(\omega_0 t_{13}), \quad (4.52)$$

$$T_{32} = \left(\frac{4\omega_0}{\hbar^2 m^2} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \text{Im}\{\langle 12 \rangle\} \langle 23 \rangle \sin(\omega_0 t_{13}), \quad (4.53)$$

$$T_{33} = \left(\frac{4\omega_0}{\hbar^2 m^2} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \text{Im}\{\langle 13 \rangle\} \langle 23 \rangle \cos(\omega_0 t_{12}), \quad (4.54)$$

$$T_{34} = \left(\frac{4\omega_0}{\hbar^2 m^2} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \text{Im}\{\langle 13 \rangle\} \langle 23 \rangle \sin(\omega_0 t_{12}). \quad (4.55)$$

One should note that when the diamagnetic term is neglected, the third-order contribution would be zero, so the nonzero third-order contributions here are due to interactions that contain $A^2(t_2)$ and $A^2(t_3)$. These integrals are discussed in the main body of the text.

4.5.2 Fourth-order contributions

Evaluation of $B^{(4)}(\rho_S, t_1)$, given in Eq. (4.46), is more involved and tedious compared to that of the lower order terms. Luckily, most of the braces in Eq. (4.46) contain a single element separated with columns and those are zero because of the Gaussian character of the bath. So, the nonzero fourth-order terms can be written as:

$$B^{(4)}(\rho_S, t_1) = \frac{1}{(i\hbar)^4} \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \int_{-\infty}^{t_3} dt_4 \{ \{1234\} - \{12 : 34\} - \{13 : 24\} - \{14 : 23\} \}, \quad (4.56)$$

where the brace $\{1234\}$ has a total of 256 terms; half of those are zero because they involve the odd powers of the vector potential $A(t_i)$. Furthermore, 16 (48) terms which are in the form of product of four diamagnetic $A(t_i)^2$ (vector potential $A(t_1)^2 \dots$) terms at times t_1, t_2, t_3, t_4 add up to zero. The remaining terms can be written as four and six time averages as:

$$\begin{aligned} \{1234\} = & \frac{1}{m^4} [\langle 4213 \rangle [p(t_1), p(t_3) \rho_S p(t_4) p(t_2)] + \langle 1234 \rangle [p(t_1), p(t_2) p(t_3) p(t_4) \rho_S] \\ & - \langle 2134 \rangle [p(t_1), p(t_3) p(t_4) \rho_S p(t_2)] - \langle 4321 \rangle [p(t_1), \rho_S p(t_4) p(t_3) p(t_2)] \\ & - \langle 3124 \rangle [p(t_1), p(t_2) p(t_4) \rho_S p(t_3)] - \langle 4123 \rangle [p(t_1), p(t_2) p(t_3) \rho_S p(t_4)] \\ & + \langle 4312 \rangle [p(t_1), p(t_2) \rho_S p(t_4) p(t_3)] + \langle 3214 \rangle [p(t_1), p(t_4) \rho_S p(t_3) p(t_2)]] \\ & + \frac{1}{4m^4} [(\langle 122334 \rangle - \langle 221334 \rangle + \langle 332214 \rangle - \langle 331224 \rangle) [p(t_1), p(t_4) \rho_S] \\ & + (\langle 422133 \rangle - \langle 433221 \rangle + \langle 433122 \rangle - \langle 412233 \rangle) [p(t_1), \rho_S p(t_4)] \\ & + (\langle 122344 \rangle - \langle 221344 \rangle + \langle 442213 \rangle - \langle 441223 \rangle) [p(t_1), p(t_3) \rho_S] \\ & + (\langle 322144 \rangle - \langle 443221 \rangle + \langle 443122 \rangle - \langle 312244 \rangle) [p(t_1), \rho_S p(t_3)] \\ & + (\langle 123344 \rangle - \langle 331244 \rangle + \langle 443312 \rangle - \langle 441233 \rangle) [p(t_1), p(t_2) \rho_S] \\ & + (\langle 332144 \rangle - \langle 213344 \rangle + \langle 442133 \rangle - \langle 443321 \rangle) [p(t_1), \rho_S p(t_2)]]], \quad (4.57) \end{aligned}$$

where four-time terms are due to $p(t) \cdot A(t)$ interaction, while six-time terms originate from $p(t_i) A(t_i) A^2(t_j) \dots$ type couplings that involve both diamagnetic and velocity interactions. The remaining braces in Eq. (4.46) involve only velocity-coupling terms and

can be expressed as:

$$\begin{aligned}
\{12 : 34\} = & \frac{1}{m^4} [\langle 12 \rangle \langle 34 \rangle [p(t_1), p(t_2) [p(t_3), p(t_4) \rho_S]] \\
& + \langle 12 \rangle \langle 43 \rangle [p(t_1), p(t_2) [\rho_S p(t_4), p(t_3)]] \\
& - \langle 21 \rangle \langle 34 \rangle [p(t_1), [p(t_3), p(t_4) \rho_S] p(t_2)] \\
& - \langle 21 \rangle \langle 43 \rangle [p(t_1), [\rho_S p(t_4), p(t_3)] p(t_2)]], \quad (4.58)
\end{aligned}$$

$$\begin{aligned}
\{13 : 24\} = & \frac{1}{m^4} [\langle 13 \rangle \langle 24 \rangle [p(t_1), p(t_3) [p(t_2), p(t_4) \rho_S]] \\
& + \langle 13 \rangle \langle 42 \rangle [p(t_1), p(t_3) [\rho_S p(t_4), p(t_2)]] \\
& - \langle 31 \rangle \langle 24 \rangle [p(t_1), [p(t_2), p(t_4) \rho_S] p(t_3)] \\
& - \langle 31 \rangle \langle 42 \rangle [p(t_1), [\rho_S p(t_4), p(t_2)] p(t_3)]], \quad (4.59)
\end{aligned}$$

$$\begin{aligned}
\{14 : 23\} = & \frac{1}{m^4} [\langle 14 \rangle \langle 23 \rangle [p(t_1), p(t_4) [p(t_2), p(t_3) \rho_S]] \\
& + \langle 14 \rangle \langle 32 \rangle [p(t_1), p(t_4) [\rho_S p(t_3), p(t_2)]] \\
& - \langle 41 \rangle \langle 23 \rangle [p(t_1), [p(t_2), p(t_3) \rho_S] p(t_4)] \\
& - \langle 41 \rangle \langle 32 \rangle [p(t_1), [\rho_S p(t_3), p(t_2)] p(t_4)]]. \quad (4.60)
\end{aligned}$$

Since Eq. (4.57) contains both four and six index terms, one needs to transform them into pair product form by using Isserlis' theorem. The formula for the four index term was given in Eq. (4.47) while the six index term can be written as:

$$\begin{aligned}
\langle 123456 \rangle = & \langle 12 \rangle \langle 34 \rangle \langle 56 \rangle + \langle 12 \rangle \langle 35 \rangle \langle 46 \rangle + \langle 12 \rangle \langle 36 \rangle \langle 45 \rangle \\
& + \langle 13 \rangle \langle 24 \rangle \langle 56 \rangle + \langle 13 \rangle \langle 25 \rangle \langle 46 \rangle + \langle 13 \rangle \langle 26 \rangle \langle 45 \rangle \\
& + \langle 14 \rangle \langle 23 \rangle \langle 56 \rangle + \langle 14 \rangle \langle 25 \rangle \langle 36 \rangle + \langle 14 \rangle \langle 26 \rangle \langle 35 \rangle \\
& + \langle 15 \rangle \langle 23 \rangle \langle 46 \rangle + \langle 15 \rangle \langle 24 \rangle \langle 36 \rangle + \langle 15 \rangle \langle 26 \rangle \langle 34 \rangle \\
& + \langle 16 \rangle \langle 23 \rangle \langle 45 \rangle + \langle 16 \rangle \langle 24 \rangle \langle 35 \rangle + \langle 16 \rangle \langle 25 \rangle \langle 34 \rangle. \quad (4.61)
\end{aligned}$$

In equations (4.57)-(4.60), we can use the time dependent momentum operator (Ford et al., 1996)

$$p(t_2) = p(t_1) \cos[\omega_0(t_1 - t_2)] + m \omega_0 x(t_1) \sin[\omega_0(t_1 - t_2)] \quad (4.62)$$

to simplify the commutators involving the system momentum at different times and the system density matrix and obtain the contribution of $B^{(4)}(\rho_S, t_1)$ to the master equation $T_1^{(4)} = T_{41} + T_{61} + T_{63}$ and $T_2^{(4)} = T_{42} + T_{62} + T_{64}$ where

$$\begin{aligned}
T_{41} &= i \left(\frac{8\omega_0^2}{\hbar^2 m^2} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \int_0^\infty dt_{34} \{ \text{Im}[\langle 14 \rangle] \langle 23 \rangle \cos(\omega_0 t_{13}) \sin(\omega_0 t_{24}) \\
&\quad + \text{Im}[\langle 13 \rangle] \langle 24 \rangle \cos(\omega_0 t_{14}) \sin(\omega_0 t_{23}) + \text{Im}[\langle 14 \rangle \langle 23 \rangle] \cos(\omega_0 t_{12}) \sin(\omega_0 t_{34}) \} , \\
T_{42} &= i \left(\frac{8\omega_0^2}{\hbar^2 m^2} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \int_0^\infty dt_{34} \{ \text{Im}[\langle 14 \rangle] \langle 23 \rangle \sin(\omega_0 t_{13}) \sin(\omega_0 t_{24}) \\
&\quad + \text{Im}[\langle 13 \rangle] \langle 24 \rangle \sin(\omega_0 t_{14}) \sin(\omega_0 t_{23}) + \text{Im}[\langle 14 \rangle \langle 23 \rangle] \sin(\omega_0 t_{12}) \sin(\omega_0 t_{34}) \} , \\
T_{61} &= \left(\frac{32\omega_0}{\hbar^3 m^3} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \int_0^\infty dt_{34} \{ \text{Im}[\langle 12 \rangle] \text{Im}[\langle 23 \rangle] \langle 34 \rangle \cos(\omega_0 t_{14}) \} , \\
T_{62} &= \left(\frac{32\omega_0}{\hbar^3 m^3} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \int_0^\infty dt_{34} \{ \text{Im}[\langle 12 \rangle] \text{Im}[\langle 23 \rangle] \langle 34 \rangle \sin(\omega_0 t_{14}) \} , \\
T_{63} &= \left(\frac{32\omega_0}{\hbar^3 m^3} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \int_0^\infty dt_{34} \{ \text{Im}[\langle 12 \rangle] \text{Im}[\langle 24 \rangle \langle 34 \rangle] \cos(\omega_0 t_{13}) \\
&\quad + (\text{Im}[\langle 23 \rangle] \text{Im}[\langle 14 \rangle \langle 34 \rangle] + \text{Im}[\langle 13 \rangle] \text{Im}[\langle 24 \rangle \langle 34 \rangle]) \cos(\omega_0 t_{12}) \} , \\
T_{64} &= \left(\frac{32\omega_0^2}{\hbar^3 m^2} \right) \int_0^\infty dt_{12} \int_0^\infty dt_{23} \int_0^\infty dt_{34} \{ \text{Im}[\langle 12 \rangle] \text{Im}[\langle 24 \rangle \langle 34 \rangle] \sin(\omega_0 t_{13}) \\
&\quad + (\text{Im}[\langle 23 \rangle] \text{Im}[\langle 14 \rangle \langle 34 \rangle] + \text{Im}[\langle 13 \rangle] \text{Im}[\langle 24 \rangle \langle 34 \rangle]) \sin(\omega_0 t_{12}) \} . \tag{4.63}
\end{aligned}$$

Here T_{6i} , where $i = 1, 2, 3$, and 4, contain the diamagnetic interaction terms while T_{41} and T_{42} are due to $p.A$ type interaction. Although tedious, all of T_{4i} and T_{6i} integrals can be carried out exactly by using the rational expansion of the hyperbolic cotangent function, as was the case for the second- and the third-order contributions. The results

are:

$$\begin{aligned}
T_{41} = & -\gamma f_{\omega_0}^3 \left[\frac{1}{2} \left(\frac{\delta m}{m} - 5 \frac{\gamma}{\Omega} \right) \coth(\beta \omega_0) + \frac{\gamma}{\omega_0} I \right] \\
& - f_{\omega_0}^2 \gamma \beta \left[\frac{1}{\pi^2} \gamma \text{Im}[\psi_1(\beta, \omega_0)] - \frac{1}{2} \omega_0 \frac{\delta m}{m} \text{csch}^2(\beta \omega_0) \right] \\
& - i \gamma^2 f_{\omega_0}^3 \left(\frac{1}{\omega_0} - \frac{\omega_0}{\Omega^2} \right), \tag{4.64}
\end{aligned}$$

$$\begin{aligned}
T_{42} = & -\gamma^2 f_{\omega_0}^3 \left[\frac{1}{2} \left(\frac{\omega_0}{\Omega^2} - \frac{1}{\omega_0} \right) \coth(\beta \omega_0) + \frac{1}{\beta \Omega^2} \left(1 + \left(\frac{\omega_0}{\Omega} \right)^2 \right) - \frac{3}{\Omega} I \right] \\
& - f_{\omega_0}^2 \gamma \beta \left[\gamma \left(\frac{1}{2} \text{csch}^2(\beta \omega_0) - \frac{2}{\pi^2} \psi_1 \left[\frac{\beta \Omega}{\pi} \right] \right) + \frac{1}{\pi^2} \omega_0 \frac{\delta m}{m} \text{Im}[\psi_1(\beta, \omega_0)] \right] \\
& - i 2 f_{\omega_0}^3 \frac{\gamma^2}{\Omega}, \tag{4.65}
\end{aligned}$$

$$\begin{aligned}
T_{61} = & -f_{\omega_0}^3 \gamma \frac{\delta m}{m} \left[\left(\frac{\gamma}{\Omega} - \frac{\delta m}{m} \right) \coth(\beta \omega_0) + 2 \frac{\gamma}{\omega_0} I \right] \\
& - i f_{\omega_0}^3 \gamma \frac{\delta m}{m} \left[\frac{\omega_0}{\gamma} \left(\frac{\delta m}{m} \right)^2 - 3 \frac{\gamma}{\omega_0} \right], \tag{4.66}
\end{aligned}$$

$$\begin{aligned}
T_{62} = & -f_{\omega_0}^3 \gamma \frac{\delta m}{m} \left[\left(\frac{\gamma}{\Omega} - \frac{\delta m}{m} \right) I - 2 \frac{\gamma}{\omega_0} \coth(\beta \omega_0) \right] \\
& - i f_{\omega_0}^3 \gamma \frac{\delta m}{m} \left[3 \frac{\delta m}{m} - \frac{\gamma}{\Omega} \right], \tag{4.67}
\end{aligned}$$

$$T_{63} = -2 f_{\omega_0}^3 \gamma \frac{\delta m}{m} \left[-\frac{\gamma}{\omega_0} I - \frac{\delta m}{m} \coth(\beta \omega_0) \right], \tag{4.68}$$

$$\begin{aligned}
T_{64} = & -2 f_{\omega_0}^3 \gamma \frac{\delta m}{m} \left[\frac{\gamma}{\omega_0} \coth(\beta \omega_0) - \frac{\delta m}{m} I \right] \\
& - f_{\omega_0} \gamma \frac{\delta m}{m} \left[\frac{f_{\omega_0}}{\beta \Omega} \left(3 \frac{\delta m}{m} + \frac{\gamma}{\Omega} \right) + \frac{\delta m}{m} \beta \Omega \left(\frac{\beta \Omega}{\pi^3} \psi_2 - \frac{4 f_{\omega_0}}{\pi^2} \psi_1 \right) \right]. \tag{4.69}
\end{aligned}$$

5. SPIN-BOSON MODEL

5.1 Introduction

The spin-boson model is one of the most prominent models used to study dissipative and decoherence effects in quantum mechanics (Weiss, 2008; Leggett et al., 1987). It describes a two-state system (TSS) coupled to an infinite array of non-interacting harmonic oscillators whose effect on the system is characterized by spectral density $J(\omega)$. The spin-boson model with a power law spectral density, which is the general setting of the model, contains only the cutoff frequency of the bath as an internal energy scale and leads to scale-free rates while the so-called structured environments provide a more non-trivial internal dynamics which might be relevant for controlling coherence and relaxation times by engineering a properly structured environment (Wilhelm et al., 2004). Garg, Onuchic and Ambegaokar (GOA) have shown that the spin-boson model with Ohmic spectral density can be mapped to the problem of a TSS interacting with a harmonic oscillator which is damped by an Ohmic environment whose effective spectral function can be approximated as Lorentzian when the cutoff frequency of the Ohmic bath is much larger than the characteristic frequency of the harmonic oscillator (Garg et al., 1985). The GOA model has been used in many studies to describe several phenomena such as electron transfer reactions in various condensed phase environments. The same Hamiltonian and $J(\omega)$, also, describe experimentally relevant quantum systems, such as flux-qubit read out by a dc-SQUID (van der Wal et al., 2000; Tian et al., 2002; Chiorescu et al., 2003), atom-based cavity quantum electrodynamics (Raimond et al., 2001), circuit quantum electrodynamics with superconducting systems (Schuster et al., 2007), semiconducting quantum dots in nano-cavities (Badolato et al., 2005) and in nano-mechanical resonators (Blencowe, 2004; O’Connell et al., 2010). The same model is employed in chemical physics context to study charge transfer (Tanaka and Tanimura, 2009; Dijkstra and Tanimura, 2015), energy transfer dynamics in photosynthesis (Escher and Ankerhold, 2011; Chin et al., 2013; Iles-Smith et al., 2016) and

linear and nonlinear spectroscopies (Dijkstra and Tanimura, 2015).

Theoretical approaches used to investigate the dynamics of the coupled TSS-damped harmonic oscillator(HO) system can be, broadly, divided into two groups; on the one hand, the infinite dimensional TSS-HO Hamiltonian is approximated by a finite dimensional system which is reduced to Jaynes-Cummings or ac-Stark Hamiltonian depending on whether one is in the resonant or dispersive regime (Blais et al., 2004). On the other hand, the problem could be considered as a spin-boson model with a peaked spectral density. A large number of computational techniques have been developed to investigate the dynamics of spin-boson model, such as hierarchical equations of motion, renormalization group techniques and path integral based formalism (Tanimura, 1990; Kleff et al., 2004; Tanaka and Tanimura, 2009; Prior et al., 2010; Chin et al., 2013; Dijkstra et al., 2015). The effects of structured spectral density on the decoherence properties of a qubit have been studied with a perturbative approach in Refs. (Goorden et al., 2004, 2005). Thorwart *et. al.* have used ab initio QUAPI technique to show that perturbative treatment breaks down when qubit-HO coupling is strong ($g \gg \Gamma$) and when the qubit and the oscillator frequencies are comparable (Thorwart et al., 2004). Gan, Huang and Zheng have studied the dynamics of the spin-boson model by using a unitary transformation method (Gan et al., 2010) while Ref. (Kleff et al., 2004) has investigated the dephasing times for the TSS-HO system by using flow-equations renormalization method and shown that harmonic oscillator frequency can be used to control qubit dynamics.

As the interaction between the TSS and its environment, whether a bath of non-interacting harmonic oscillator or the damped harmonic oscillator, is considered to be linear, the spectral density function $J(\omega)$ and the bath temperature completely characterizes the TSS-bath coupling. All relevant computational techniques make use of bath correlation function $G(t)$ which is the thermal average of the force auto-correlation of the bath degrees of freedoms or its two-times integrated form to account for the effect of the bath. So, obtaining manageable analytical expressions for $G(t)$ might be beneficial for theoretical as well as computational techniques used to treat the spin-boson problem. There has been a number of reports on the analytical expressions for correlation functions of peaked spectral density (Garg et al., 1985; Tanimura, 2006; Nesi et al., 2007; Vierheilig et al., 2011). Nesi, Grifoni and Paladino have studied the dynamics of the system for the large Q-factor oscillator at arbitrary detuning and finite

temperatures and obtained analytical expressions for the TSS population (Nesi et al., 2007) for the weak-coupling regime. Building on the findings of Ref (Nesi et al., 2007), Vierheilig, Bercieux and Grifoni have considered the dynamics of a qubit coupled to a nonlinear oscillator which is coupled to an Ohmic bath and showed that the system can be mapped to the TSS-damped harmonic oscillator model with an effective peaked spectral density (Vierheilig et al., 2011).

In the present study, our first aim is to obtain an exact analytical expression for the correlation function $G(t)$ of TSS-HO system which is characterized by a peaked spectral density. The TSS-HO system has several characteristic times set by the energy splitting and tunneling amplitude of the TSS, frequency and the damping of the harmonic oscillator, the temperature of the HO's environment and the coupling constant between the HO and the TSS. The complicated interplay among these rate constants make it difficult to develop a universal computational technique which is applicable for all parameter values. Using the derived $G(t)$ expression, we investigate the population dynamics of the TSS and map the validity range of various approximations as function of TSS tunneling splitting, coupling strength between the TSS and the harmonic oscillator, damping of the oscillator and the temperature of the bath. It is found that for a range of parameters, one can use Markov approximation and obtain an exact expression for the population difference.

The outline of the paper is follows: In Section 5.2, we describe the problem and derive the expression for $G(t)$ and present several approximations to it. The validity range of the approximations is discussed in Section 5.3 and a brief summary of the study is presented in the conclusions.

5.2 Model

Let the total Hamiltonian of the closed system be

$$H_t = \frac{\varepsilon_0}{2} \sigma_z + \frac{V}{2} \sigma_x + \sum_k \omega_k b_k^\dagger b_k + \sigma_z \sum_k g_k (b_k^\dagger + b_k), \quad (5.1)$$

where ε_0 is the splitting of the energy levels of the system, V is the tunneling matrix element and σ_i are the Pauli spin matrices. The bath is modeled as a collection of harmonic oscillators with creation (annihilation) operators $b_k^\dagger$ (b_k) and the mode frequency ω_k . The dynamics of the two-state system are characterized by the population

difference $P(t)$ which is defined as the expectation value of σ_z as:

$$P(t) = \text{Tr}_{\text{TLS}} [\sigma_z \text{Tr}_B (e^{-iHt} \rho(0) e^{iHt})]. \quad (5.2)$$

Here, $\rho(0)$ is the initial density matrix of the total system which is assumed to be in factorized form $\rho(0) = \rho_{\text{TLS}}(0) \otimes \exp(-\beta H_B)/Z$ and the two partial trace operations refer to trace over the bath (B) and system (TLS) degrees of freedom. $P(t)$ for the spin-boson problem obeys the generalized master equation (Weiss, 2008):

$$\frac{dP(t)}{dt} = - \int_0^t dt' (K^s(t, t') P(t') + K^a(t, t')), \quad (5.3)$$

where $K^a(t, t')$ and $K^s(t, t')$ are the asymmetric and symmetric parts of the kernel, respectively. They are derived from the two-time integrated bath correlation function $G(t)$

$$G(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \langle F(t_2) F(0) \rangle_T + iE_r t, \quad (5.4)$$

and can be represented as a series in tunneling amplitude V . In Eq. (5.4), E_r is the reorganization energy of the bath and $\langle F(t) F(0) \rangle_T$ is the thermal average of the force auto-correlation function of the bath modes which is defined as:

$$\langle F(t) F(0) \rangle_T = \frac{1}{2\pi} \int_0^\infty J(\omega) \frac{\cosh(\omega/2k_B T - i\omega t)}{\sinh(\omega/2k_B T)} d\omega. \quad (5.5)$$

The effect of the environment on the system is characterized by bath spectral function $J(\omega)$, which we assume to be of the form

$$J(\omega) = 8\kappa^2 \frac{\gamma \omega_0 \omega}{(\omega^2 - \omega_0^2)^2 + 4\gamma^2 \omega^2}, \quad (5.6)$$

for the present study. $J(\omega)$ in Eq. 5.6 is a structured spectral function and can be used to describe various types of environments, such as the traditional spin-boson model with an Ohmic environment (Garg et al., 1985), a TSS coupled to a nonlinear, damped-oscillator (Vierheilig et al., 2011), or an environment that contains both a background vibrations and one prominent vibrational mode. Depending on the model it describes, its parameters (ω_0 , κ , and γ) would have slightly different meanings. Here, we have a two-state system in contact with a harmonic oscillator which damped by an Ohmic thermal bath in mind. So, ω_0 is the frequency of the central harmonic oscillator, κ is

the TSS-HO coupling constant, γ is the broadening of the oscillator levels due to its interaction with the Ohmic environment. One should note that $J(\omega)$ of Eq. 5.6 reduces to so called Debye form in the over-damped limit $\omega_0 \ll \gamma/2$.

The reorganization energy E_r is defined as

$$E_r = \frac{1}{2\pi} \int_0^\infty \frac{J(\omega)}{\omega} d\omega, \quad (5.7)$$

and is equal to κ^2/ω_0 for the spectral density given in Eq. 5.6. The bath correlation function $G(t)$ can be simplified for the strong coupling regime $\kappa \gg \omega_0$, where one can invoke the short-time approximation by noting that the kernel function $G(t)$ is non-zero only for a very short time and obtain (Garg et al., 1985):

$$G_{\text{st}}(t) = \langle F(0)^2 \rangle t^2 + iE_r t, \quad (5.8)$$

where $\langle F(0)^2 \rangle$ can be evaluated from Eq. 5.4 by using Eq. 5.6 as

$$\langle F(0)^2 \rangle = 2\frac{E_r}{\beta} + \frac{1}{\pi} \frac{E_r \omega_0^2}{\Delta} \text{Im} \left[\psi \left(1 + i\tilde{\beta}(\Delta - i\gamma) \right) \right], \quad (5.9)$$

where $\Delta = \sqrt{\omega_0^2 - \gamma^2}$, $\tilde{\beta} = \beta/(2\pi) = 1/(2\pi k_B T)$ is the inverse temperature scaled by $1/(2\pi)$ and $\psi(z)$ is the complex di-gamma function. To evaluate $G(t)$ for the general case, we write the force auto-correlation function as:

$$\langle F(t)F(0) \rangle_T = \frac{1}{2\pi} \int_0^\infty J(\omega) (\coth(\beta\omega/2) \cos(\omega t) - i \sin(\omega t)) d\omega. \quad (5.10)$$

One should note that the auto-correlation function is the difference of the Fourier cosine and sine transforms of $J(\omega) \coth(\beta\omega/2)$ and $J(\omega)$. Fourier sine transform of the spectral function is straightforward, while the cosine transform part can be accomplished by using the series form of the hyperbolic cotangent function:

$$\coth(\beta\omega/2) = \frac{2}{\beta\omega} + \frac{4}{\beta} \sum_{m=1}^{\infty} \frac{\omega}{\omega^2 + 4m^2\pi^2/\beta^2}. \quad (5.11)$$

Using Eqs. (5.6) and (5.11) in Eq. (5.10) and carrying out the double integral in Eq. (5.4),

it is possible to obtain an exact expression for $G(t)$ as:

$$G_{\text{all}}(t) = -(a+b) + a \exp(-it z^*) + b \exp(-it z) - ct + \sum_{n=1}^3 K_n, \quad (5.12)$$

where $z = \Delta + i\gamma$, $\Delta = \sqrt{\omega_0^2 - \gamma^2}$ and

$$a = \frac{E_r}{\Delta} \frac{e^{-2i\theta}}{e^{\beta z} - 1}, \quad b = \frac{E_r}{\Delta} \frac{e^{2i\theta}}{1 - e^{-\beta z^*}}, \quad c = 4E_r \frac{\gamma}{\beta \omega_0^2},$$

where $\theta = \arctan(\gamma/\Delta)$. The last three terms of Eq. 5.12, which are important for the low temperature regime, are:

$$K_1 = -\frac{E_r}{\pi\Delta} \text{Im} \left[e^{-2i\theta} \left(H(-i\tilde{\beta}z) + H(i\tilde{\beta}z) \right) \right], \quad (5.13)$$

$$K_2 = 4E_r \frac{\gamma}{\pi\omega_0^2} \log \left(1 - e^{-\frac{t}{\tilde{\beta}}} \right), \quad (5.14)$$

$$K_3 = -\frac{E_r}{\pi\Delta} \text{Im} \left[e^{-2i\theta} \left(B(t, -i\tilde{\beta}z) + B(t, i\tilde{\beta}z) \right) \right], \quad (5.15)$$

where $\tilde{\beta} = \beta/(2\pi)$, $H(z)$ is the complex Harmonic number, $B(t, z) = e^{tz} \mathcal{B} \left(e^{-\frac{2\pi}{\tilde{\beta}}t}, 1+z \right)$ and $\mathcal{B}$ denotes incomplete Beta function. One should note that all three K_n s are real.

The correlation function at $T = 0$ can not be obtained from the $T = 0$ limit of Eq. 5.12 because of the form chosen for the $\coth(x)$ in Eq. 5.11. Instead, we use the identity $\coth(\infty) = 1$ in Eq. 5.10 and carry out the Fourier sine and cosine transforms to get:

$$\begin{aligned} G_{T=0}(t) = & \frac{2\gamma E_r}{\pi^2 \Omega^2} (\gamma_E + \log(\Omega t)) + \frac{E_r}{\pi \Delta \Omega^2} \left(\left(\frac{1}{2} - \frac{\theta}{\pi} \right) (\Delta^2 - \gamma^2) + i\gamma\Delta \right) \\ & - \frac{E_r}{2\pi\Delta} \left(e^{2i\theta} \cos(-z^*t) - i e^{-2i\theta} \sin(zt) \right) \\ & - \frac{E_r}{2\pi^2\Delta} \text{Im} \left[e^{2i\theta} \left(e^{-iz^*t} \text{Ei}(iz^*t) + e^{iz^*t} \text{Ei}(-iz^*t) \right) \right], \end{aligned} \quad (5.16)$$

where γ_E is the Euler gamma constant and $\text{Ei}(z)$ is the complex exponential integral. The $\gamma = \Omega$ special case of $G(t)$ at zero temperature can not be derived in a similarly as:

$$\begin{aligned} G_{\text{sp}}(t) = & -\frac{E_r}{\pi\Omega} \left((2 - \Omega t) e^{\Omega t} \text{Ei}(-\Omega t) + (\Omega t + 2) e^{-\Omega t} (\text{Ei}(\Omega t) + i\pi) \right. \\ & \left. - 4 \left(\log(\Omega t) + \frac{i\pi}{2} + \gamma_E \right) \right). \end{aligned} \quad (5.17)$$

5.3 Discussion

At the zero bias-case ($\epsilon_0 = 0$ in Eq. 5.1), the dynamics of $P(t)$ simplifies to

$$\frac{dP(t)}{dt} = - \int_0^t K(t-t')P(t') dt'. \quad (5.18)$$

Although the relation between the bath correlation function $G(t)$ and the kernels $K(t, t')$ in Eq. 5.18 is, generally, complicated, it can be simplified in certain parameter regimes. For instance, non-interacting blip approximation (NIBA) allows one to truncate the exact kernels to first order in the square of tunneling splittings (V^2) and obtain

$$K(t) = V^2 e^{-G_s(t)} \cos(G_a(t)) = \text{Re} [V^2 \exp(-G(t))] , \quad (5.19)$$

NIBA is exact at zero damping, otherwise it works best for zero bias and/or large damping and/or high temperature (Leggett et al., 1987). NIBA form of the population dynamics can, also, be derived for the polaron transformed spin-boson problem (Orth et al., 2013; Wilhelm et al., 2004). NIBA is, generally, justified whenever the bath relaxation time is fast compared to the time-scales of the TSS, such that bath correlation function averages out in a short time which leads to weak damping of the system in longer times. Although NIBA can not be justified for all the range of V , ω_0 , κ , γ and β values considered below, we will use its kernel form as proxy to map the validity range of approximate $G(t)$ s.

The standard way to solve the integro-differential Eq. 5.18 is to exploit its convolution form and use Laplace transform method to express it as:

$$P(s) = \frac{1}{s + K(s)}, \quad (5.20)$$

where the $K(s)$ is the Laplace transform of the kernel function. For the remainder of the paper, we will try to determine the validity range of various approximations to $G_{\text{all}}(t)$ as function of ω_0 , γ , κ , and inverse temperature β .

The first approximation we consider is to neglect the K_n terms in $G_{\text{all}}(t)$ of Eq. 5.12. Using only the first three terms of $G_{\text{all}}(t)$ and expressing the exponential of exponential

term that contains z^* in series form, one gets the dynamical equation:

$$\frac{dP(t)}{dt} = -\text{Re} \left[e^{-(a+b)} \sum_{n=0}^{\infty} \frac{a^n}{n!} \int_0^t dt' \exp \left(b e^{-(t-t')z} - (nz^* + c)(t-t') \right) P(t') \right]. \quad (5.21)$$

It is interesting to note that Eq. 5.21 indicates that the rate of change of the population difference show similarities to sampling from a type of Gumbel extreme value distribution. Such a distribution indicates dissipation of information on the log-linear scale while the observation variable is exponential (Frank, 2014). At low temperatures ($k_B T < V$), a is small and the sum can be approximated as the first term ($n = 0$). One needs to include more and more terms of the sum as the temperature increases. The Laplace transform of Eq. 5.21 can be evaluated exactly in the form of infinite series sum as:

$$P_{F3}(s) = \frac{1}{s + K_{F3}(s)}, \quad (5.22)$$

where

$$K_{F3}(s) = \frac{1}{z} e^{-(a+b)} \sum_{n=0}^{\infty} \frac{a^n}{n!} (-b)^{\frac{1}{z}(nz^* + c + s)} \Gamma \left[\frac{1}{z}(nz^* + c + s), -b \right], \quad (5.23)$$

where $\Gamma[z, b]$ is the lower incomplete Gamma function. As shown below, for a large range of parameters, a is negligible and including only the $n = 0$ term in definition of $K_{F3}(s)$ is adequate. Such solutions will be denoted by subscript F3b below. Similarly, in the short-time approximation, Eq. 5.18 can be solved by using Laplace transform method as

$$P_{st}(s) = \frac{2\sqrt{a}}{2\sqrt{a}s + w \left(\frac{E_r + is}{2\sqrt{a}} \right)}, \quad (5.24)$$

where $w(z)$ is the plasma dispersion function. Although $P(s)$ of Eqs. 5.23 and 5.24 have no inverse Laplace transform which can be expressed in terms of known elementary or special functions, its singularities and branch properties can be used to deduce the characteristics of the dynamics of the TLS for the given parameter values.

Although the dynamical Eq. 5.18 has no known exact analytical solution for the peak spectral density (Eq. 5.6) for arbitrary parameters, such as the frequency ω_0 and the dispersion γ of the central oscillator, the system-oscillator coupling κ and the bath inverse temperature β , it is possible to obtain approximate analytical solutions for certain parameter regimes. For instance, one can derive an exact analytical expression

for $P(t)$ in the strong coupling regime $\kappa \gg \omega_0$, under "Markovian" regime by writing Eq. 5.18 as

$$\frac{dP(t)}{dt} = -K(t)P(t), \quad (5.25)$$

where

$$K(t) = \int_0^t K(t')dt' = \frac{1}{2}\sqrt{\frac{\pi}{a}}e^{-\xi^2}\text{Im}[\text{erfi}(z)], \quad (5.26)$$

where $z = \xi - i\sqrt{a}t$ and $\text{erfi}(z)$ is the complex error function. Eq. 5.25 can be integrated with $K(t)$ defined as Eq. 5.26 to obtain:

$$P(t) = P(0) \exp \left\{ \frac{V^2}{2a} \left[1 - 2\xi \mathcal{D}(\xi) - e^{-\xi^2} (\cos(E_r t) - \sqrt{\pi} \text{Re}[z \text{erfi}(z)]) \right] \right\}, \quad (5.27)$$

where $\mathcal{D}(\xi)$ is the Dawson integral.

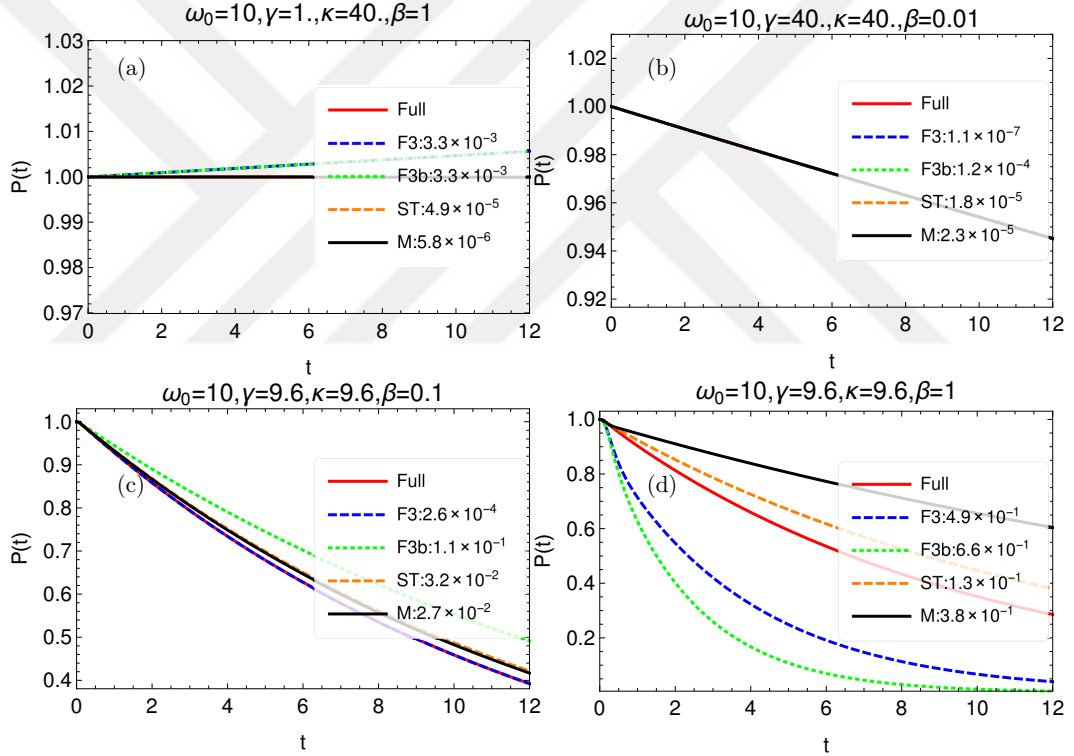


Figure 5.1. Population dynamics for their relative error values of F3, F3b, short time (ST), and Markov approximations with respect to exact expression (Full) in non-adiabatic regime ($\omega_0 = 10 \gg V$) at strong coupling (SC), overdamped case $\gamma = 10 \omega_0$ (a,b) and at intermediate damping (ID) and coupling case $\gamma \approx \omega_0$ (c,d).

In Fig. 5.1, there is either no tunneling (a) or very weak incoherent decay (b) that is almost independent of the damping and the temperature at strong coupling. F3 approximation has the smallest error at high temperature regardless of damping and coupling constants, while decreasing temperature of the bath increases the error value, except at low temperature, intermediate damping, up to order of 10^{-3} that is still considerable

small error for this approximation. In Fig.5.1 (c,d) when the interaction strength κ is considerable with the oscillator frequency ω_0 , the relative error of various approximations we have considered gets large at low temperatures and one needs to take into account the Harmonic number and Beta function parts Eq. (5.12) for the correlation function of the bath. In Fig.5.2, at intermediate (a,c) and low (b,d) temperatures one

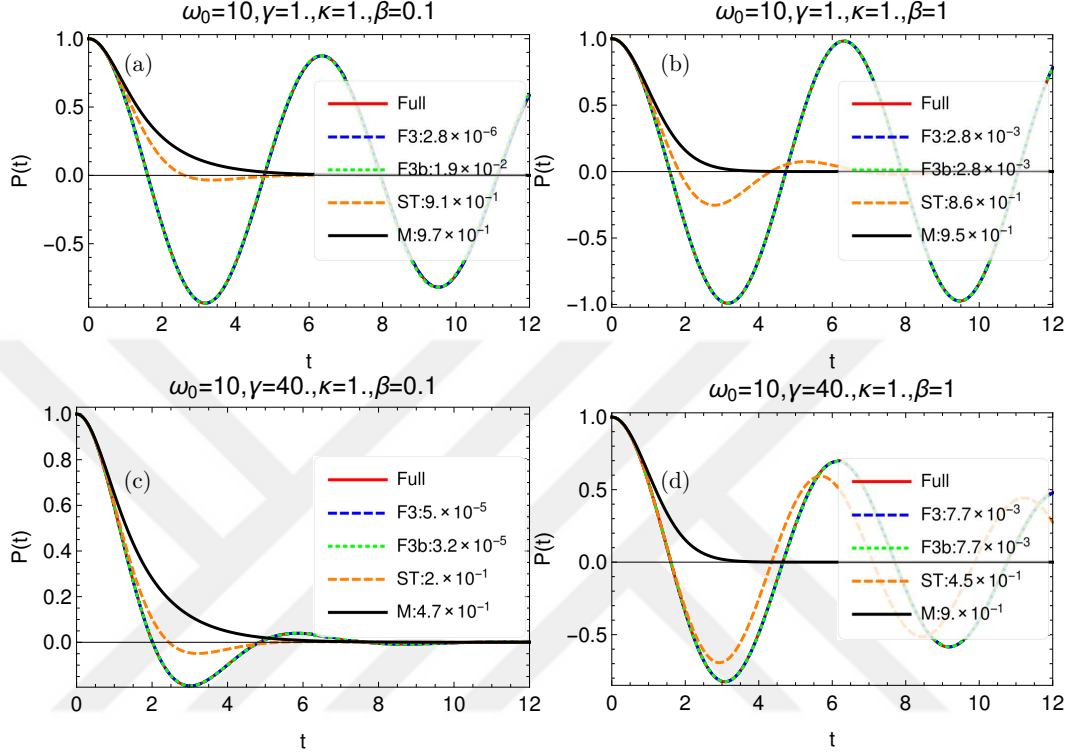


Figure 5.2. Same as figure 5.1 in case of weak coupling (WC) at underdamped $\gamma = 0.1 \omega_0$ (a,b) and at ID case $\gamma = 4 \omega_0$ (c,d).

can observe the effect of the damping of the oscillator γ on the dynamics. We observe that $P_z(t)$ dynamics changes to almost pure incoherent decay when the oscillator is overdamped, while it has a coherent decay character when γ is underdamped. From the same figure, one can conclude that the F3b approximation is a valid description of the spectral density for weak coupling, intermediate and low temperature and under- as well as overdamped cases for $\omega_0 = 10 \ll V$, *i.e.*, non-adiabatic regime.

In Fig. 5.3, $\omega_0 = 0.1$ *i.e.*, adiabatic regime, the parameter dependence of $P_z(t)$ is similar to the non-adiabatic case with same modifications. For the strong coupling case, *i.e.*, $\kappa = 1$, we have either incoherent decay, as was the case for $\omega_0 = 10$, but at lower temperatures, there is a small coherent decay at the start which settles into a non-tunneling mode as can be observed from the same figure (a, b). In addition, Markovian as well as short-time approximations are found to well describe the popula-

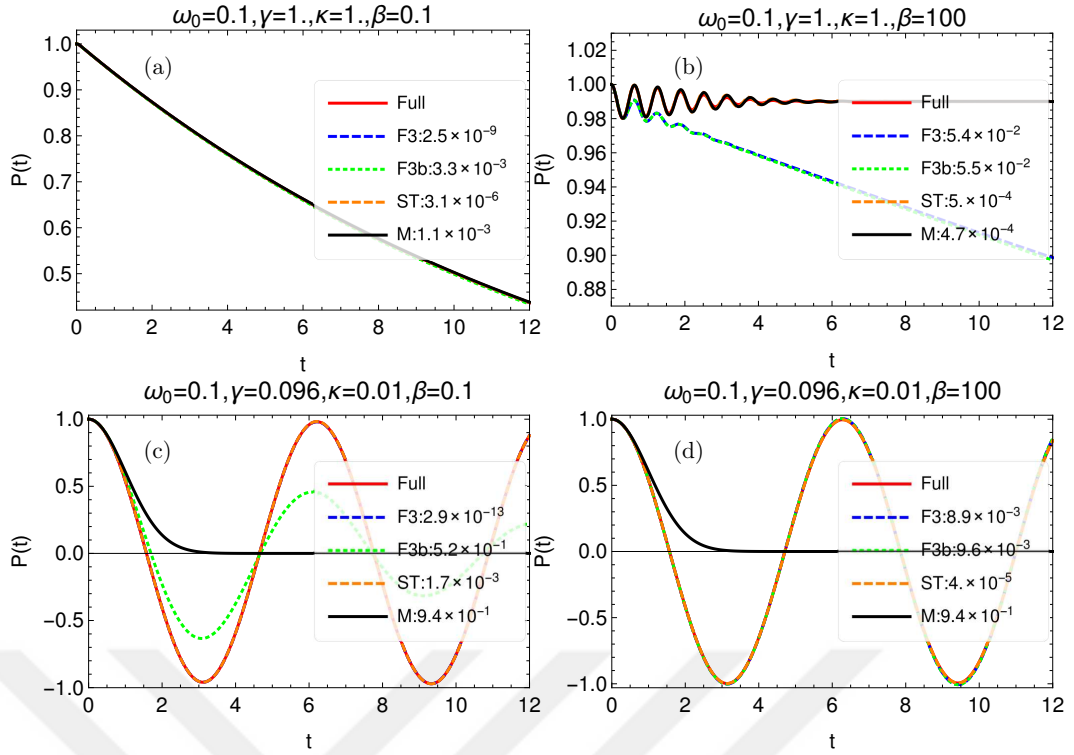


Figure 5.3. Same as figure 5.1 in adiabatic regime ($\omega_0 = 0.1 \ll V$) at high (a,c)(low (b,d)) temperatures with $\kappa = 1$ and $\gamma = 1$ ($\kappa = 0.01$ and $\gamma = 0.096$) respectively.

tion dynamics for this parameter region as well. In Fig. 5.4 at intermediate as well as

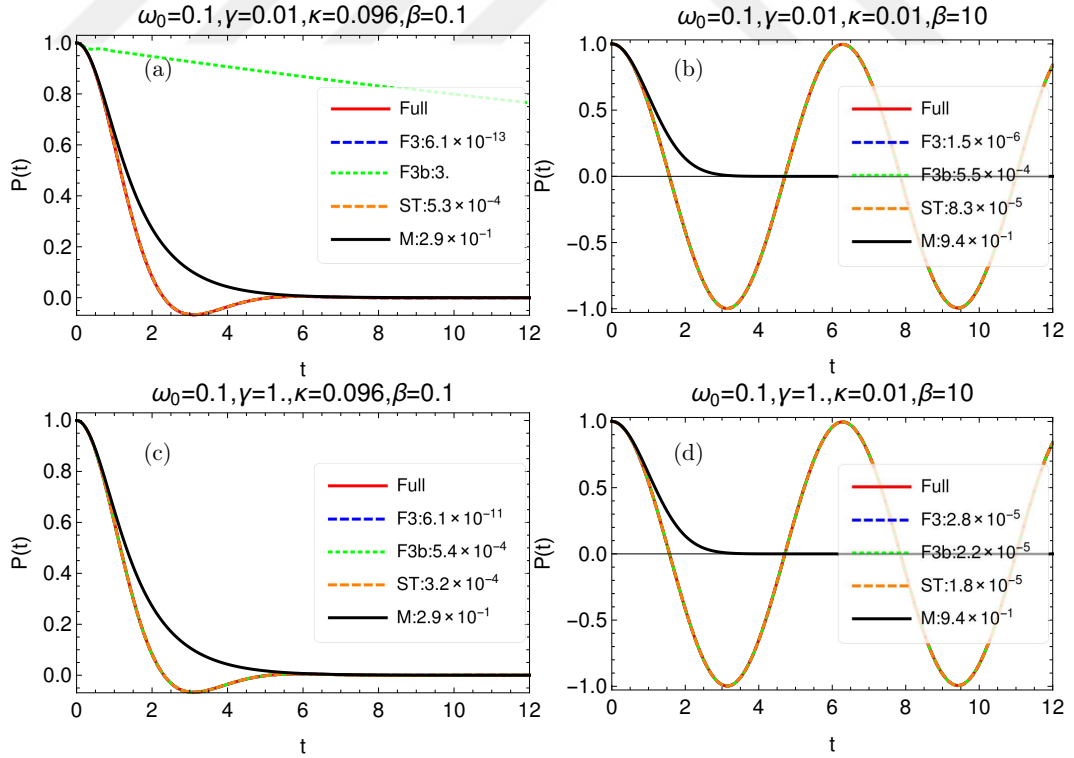


Figure 5.4. Same as figure 5.1 in adiabatic regime ($\omega_0 = 0.1 \ll V$) at high (a,c)(intermediate (b,d)) temperatures respectively.

weak coupling regimes, $P_z(t)$ mostly display coherent decay with a decay constant that

increases with the coupling constant and the temperature of the bath, as can be seen from (a,b) and (c,d). Also, the $P_z(t)$ is found to be insensitive to the magnitude of the damping constant γ (a,c) and (b,d). Moreover, short-time approach works well for the intermediate coupling case, while F3b approximation provides a reasonable description of the bath correlation function.

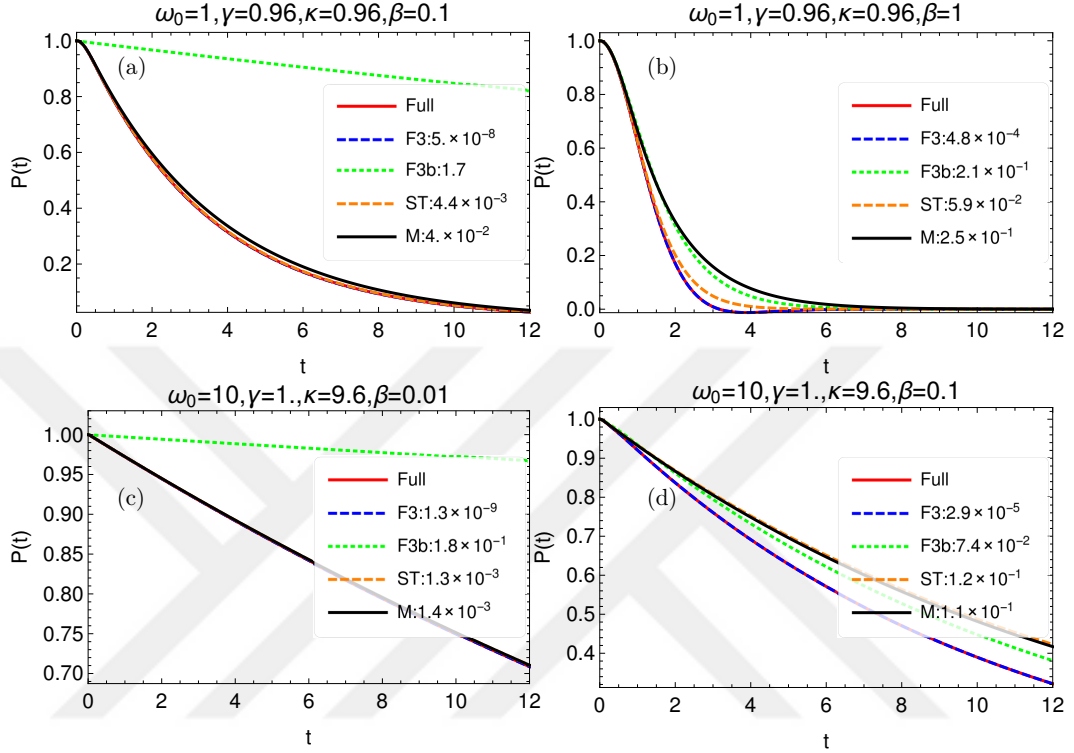


Figure 5.5. Same as figure 5.3 for both $\omega_0 = 1$ (a,b) and $\omega_0 = 10$ (c,d).

In Fig. 5.5 one can observe that increasing frequency of the harmonic oscillator leads to work perfectly Markovian approximation in case of intermediate ($\kappa = 0.96$)(a,b) as well as strong coupling ($\kappa = 9.6$)(c,d) at high and intermediate temperature. Also, we can say that decay constant is greater by increasing ω_0 that tunneling substantiates more rapidly.

As all the expressions we have found for $G(t)$ are rather involved for symbolic comparisons, we will use a heuristic approach to delineate the validity of approximations for various parameter regimes as follows: We solve Eq. 5.18 for $P(t)$ with $G_{\text{full}}(t)$ and its approximate forms $G_{\text{F3b}}(t)$, $G_{\text{F3}}(t)$ and $G_{\text{st}}(t)$ for a time interval $[0, t_f]$ where t_f depends on the problem parameters ω_0 , γ , κ , and β . t_f is chosen in such a way that $P_{\text{full}}(t)$ is almost constant at the last 1/10th of the interval. The Markovian approximation solution $P_M(t)$ is evaluated from Eq. 5.25 directly. The relative error in the norm $\varepsilon = |P_{\text{app}}(t) - P_{\text{full}}(t)| / |P_{\text{full}}(t)|$ which is calculated at 1000 points of the interval $[0, t_f]$

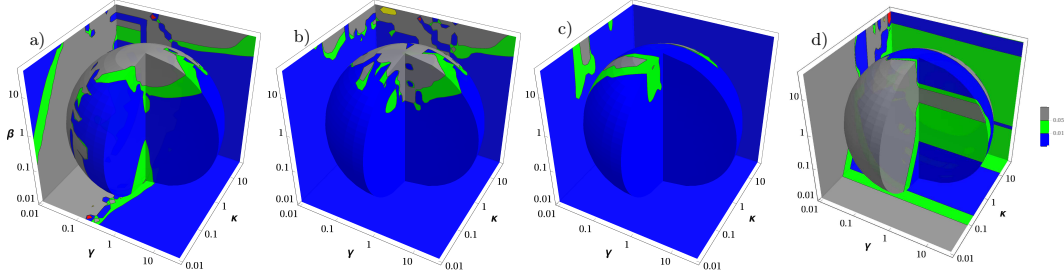


Figure 5.6. Relative error contours of F3b (a), F3 (b), short-time (c) and Markov (d) approximations for the correlation function $G(t)$ as function of γ , κ and β for adiabatic ($\omega_0 = 0.1V$) regime.

is used as the criteria to judge the validity of the approximation. Visual comparison suggests that for $\varepsilon < 0.01$, there is no discernible difference between the two solutions.

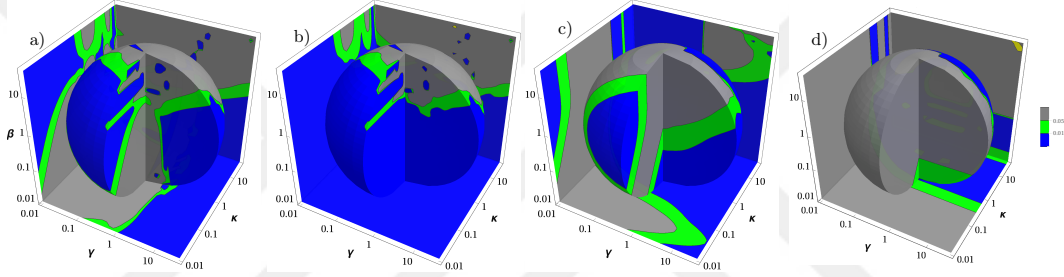


Figure 5.7. Same as Fig. 5.6 for the harmonic oscillator frequency equal to the tunneling rate of the system ($\omega_0 = 1V$)

We choose the time scales of the system (inverse tunneling rate) and the central oscillator (ω_0^{-1}) as the base and display the relative errors for the F3b, F3, ST and M approximations in Figs.5.6- 5.8(a-d) for the adiabatic ($\omega_0 \ll V$), intermediate ($\omega_0 = V$) and the non-adiabatic ($\omega_0 \gg V$) regimes, respectively. These figures display the constant relative error surfaces ($\varepsilon < 0.01$ (in blue), $0.01 < \varepsilon < 0.05$ (green) and $\varepsilon > 0.05$ (gray)) as function of oscillator damping γ , system-oscillator interaction strength κ and the inverse temperature of the bath β . We should emphasize that the blue regions in the figures means that the relative error ε is less than the arbitrarily chosen value of 0.01 for the given γ , κ and β and in the blue region magnitudes of ε for the F3b, F3, and ST approximations are not equal everywhere. One can observe from all these three figures that there are appreciable ranges of κ , γ and β s that F3b, F3, and ST approximations produce reasonably accurate dynamics as evidenced by the appearance of the blue regions while the Markovian approximation has validity only for the strong coupling regime.

In the adiabatic regime ($\omega_0 = 0.1V$), the harmonic oscillator is slow compared to the dynamics of the two-state system and acts as a static disorder and the short-time approximation is found to be the approximation that has a reasonable accuracy for the widest range of κ , γ and β values, as can be deduced from Fig. 5.6-c. Figure 5.7 suggests that as the oscillator response time and the characteristic time of the system become comparable, F3 approximation is justified for a large range of parameters, which means that the population dynamics is driven by a combination of Gumbel type extreme value statistics. A similar situation with a single extreme value distribution dominated dynamics is observed for the non-adiabatic regime where the oscillator response time is much faster compared to that of the system.

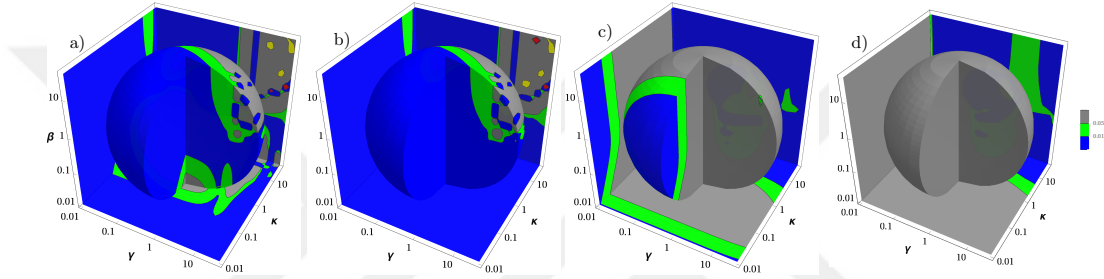


Figure 5.8. Same as Fig. 5.6 for non-adiabatic regime ($\omega_0 = 10V$).

Figure 5.9a-c is a summary of Figs.1-3 and displays the regions where relative error in $P(t)$ for F3b, F3, ST, and M approximations is less than 0.01 as function of inverse temperature β , coupling κ and damping γ for the adiabatic ($\omega_0 = 0.1V$), intermediate ($\omega_0 = V$) and non-adiabatic ($\omega_0 = 10V$) regimes, respectively. Color-coding in the figure is such that the blue regions indicate the validity region of the Markovian solution with short time approximation, green regions show where the solution for the short-time approximation kernel has $\varepsilon < 0.01$, the gray and yellow colored parameter regions indicate the validity region of F3b and F3 approximations while the orange regions indicate that one needs to include all the terms in Eq. 5.12 in the bath correlation function. One should note that in some parameter regions, more than one approximation might satisfy the inequality $\varepsilon < 0.01$. Since we are not searching for the best approximation at the given parameters, but the validity range of various approximations, coloring is done by checking the inequality criteria in the short-time Markovian, short-time, F3b and F3 approximations order. Changing this order leads to somewhat different coloring.

The first observation from Fig. 5.9 is that the short-time approximation whether in Markovian or non-Markovian form provides an adequate description of the population

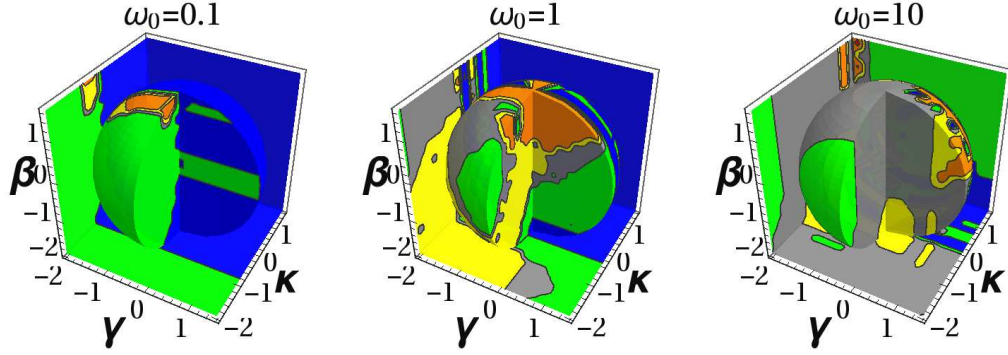


Figure 5.9. Validity regions of various approximations for the $G(t)$ as function of γ , κ and β for adiabatic ($\omega_0 = 0.1V$), intermediate ($\omega_0 = V$) and non-adiabatic ($\omega_0 = 10V$) regimes. Note that all three axis are scaled logarithmically and color coding is as blue:Markov, green:short-time, gray:F3b and yellow:F3 approximations, while the orange color indicates that one needs to include all terms in $G(t)$.

dynamics for the most values of γ , κ , and β parameters in the adiabatic case. As the adiabaticity is reduced, i.e. the bath response time gets faster, short-time approximation cease to describe the dynamics faithfully and gamma-type distribution becomes the valid approximation. There exist two prominent regimes in the figure; for the strong coupling between the TSS and the oscillator ($\kappa \gg \omega_0$), the short-time approximation is valid for a wide temperature range at both under- and over-damped limits of γ . For the weak coupling regime, the first three terms of the kernel provides an adequate description of the dynamics. As the reorganization energy is increased, there is a phase transition to state where there is no tunneling. The full description of the kernel function is found to be necessary at low temperatures for both adiabatic and non-adiabatic regimes as indicated by the orange regions in Fig. 5.9.

5.4 Conclusions

We have studied the bath correlation function $G(t)$ for the spin-boson model with a thermal bath which is characterized by a peaked spectral density. An analytical expression which is exact for all the system parameters, such as the temperature of the bath, central frequency of the bath oscillator, system-bath coupling constant and the oscillator damping constant, is obtained. We have analyzed the population dynamics of a two-level system under various approximations to $G(t)$ in adiabatic as well as non-adiabatic conditions and found that the short-time approximation provides an accurate description of the dynamics for a large range of model parameters. Also, an

exact expression for $P(t)$ is obtained under Markovian conditions which is found to be a valid description of the population dynamics for the strong TSS-harmonic oscillator interaction.



6. EFFECT OF DICHOTOMOUS NOISE ON DISSIPATIVE TUNNELING

6.1 Introduction

Charge transfer plays an important role in a number of biological and chemical reactions as well as physical systems Garg et al. (1985); May and Kühn (2003); Albinsson and Martensson (2008); Benderskii et al. (1993); Morillo and Cukier (1993). Such reactions take place mostly in condensed phases but also in molecular environments and environmental fluctuations and dissipation is necessary for the reaction to succeed (Yoshimori et al., 1989; Bagchi and Fleming, 1990).

The spin-boson model is the simplest model to examine the behavior of a two level-system (TLS) in contact with a surrounding bath. It is, also, widely used to model charge transfer for various systems. In this model, the environment is usually considered as a thermal bath of a collection of harmonic oscillators as proposed by Caldeira and Leggett (Caldeira and Leggett, 1983). In this case, the oscillators are in thermal equilibrium and interact linearly with the system. The effect of the environment on the TLS is fully encoded in the spectral density $J(\omega)$, which contains information about both the number of oscillators in the bath at a given frequency and also the interaction strength between harmonic oscillators and the TLS.

The dynamics of TLS coupled to a bath is one of the most studied subjects of quantum physics in the last fifty years and numerous distinct approximate techniques have been developed to handle such system-bath coupling problems. The most important among those methods, one can count the path-integral (Hu et al., 1992; Escher and Ankerhold, 2011; Evans and Coalson, 1995), Nakajima Zwanzig projector operator (Zwanzig, 1960; Nakajima, 1958), non-interacting blip approximation (Goychuk et al., 1997, 1995a), Monte Carlo and hierarchical methods (Iles-Smith et al., 2014; Tanaka and Tanimura, 2009; Tanimura, 2012; Liu et al., 2014). However, a variety of systems including photosynthetic complexes, tunneling in amorphous metals, electron

transfer reaction, are described by the dissipative two-level system driven by a dichotomous noise. To obtain a consistent mathematical expression for such systems, the description of the environment as a collection of harmonic oscillators only might not be adequate. Such systems can be modeled by using a discrete stochastic process. The simplest example of such a process is the dichotomous Markovian process (telegraph noise) (Bourret et al., 1973; Horsthemke and Lefever, 2002; Goychuk et al., 1994, 1995a, 1997; Xie et al., 2013; Vierheilig et al., 2009; Goychuk et al., 1995b). Such a noise is characterized by two properties, frequency of the noise which determines auto-correlation time of the noise and its amplitude. Goychuk *et. al.* have derived a generalization of the Pauli master equation for a quantum system which is strongly coupled to a stochastic external field in a thermal bath by using cumulant expansion method (Goychuk et al., 1994). Goychuk, Petrov and May have derived a set of integro-differential equations for the population difference between the donor and the acceptor sites averaged over the stochastic terms for the dichotomously driven spin-boson model including a fluctuating tunneling coupling, under the effect of a strong periodic field by using the non-interacting blip approximation and calculated an exact expression for the transition rate of the problem with respect to these results. They have shown that the oscillating field can lead electron transition in the quasi-adiabatic as well as the non-adiabatic regimes regardless of the strength of the field (Goychuk et al., 1997).

Goychuk *et. al.* have investigated the effect of dichotomous driving on the long-range charge transfer for the donor-acceptor system by deriving the kinetic equations of motion for the time evolution of the level difference, correlator between the populations and the coherence elements. They have found that the transfer rate of the system can be expressed analytically as a function of correlation time of fluctuations of the bath in the Markovian approximation (Goychuk et al., 1995a). Xie *et. al.* have calculated electron transfer rate constants in both the adiabatic and the non-adiabatic regime for the spin-boson model consisting of distinct parameters by using Ehrenfest and surface hopping methods and compared them with numerical exact results beyond the non-adiabatic limit. They have obtained that calculated rates with the short time dynamics might be more precise compared with ones with long time dynamics (Xie et al., 2013). Vierheilig *et. al.* have studied the dynamics of TLS interacting with a nonlinear oscillator in a surrounding environment defined by an Ohmic spectral density in case of weak decaying, weak non-linearity and intermediate coupling between the oscillator

and a TLS by applying Van Vleck perturbation theory in the second order and treating the non-linearity in the first order coupling. Analytical expressions for the population difference of the qubit are derived in case of weak qubit-bath interaction. They have shown that the long-time dynamics is modulated by a single decay rate, when TLS is in resonance with the nonlinear oscillator (Vierheilig et al., 2009).

In this chapter, we analyzed the problem of a TLS which is stochastically driven and is placed in a thermal bath. The driving term has the dichotomous form. We derive average expressions for both the population difference and coherence in the form of integro-differential equations by using an approach similar to the non-interacting-blip approximation (NIBA). For this model there is still no an exact analytical solution. In the present study, we were able to find an exact analytical solution for certain parameter regimes when the environment is described by a structured spectral density.

6.2 Model

We consider a dichotomously driven two level system in a thermal bath. The total Hamiltonian of the closed system formed by the TLS and its environment can be written as:

$$H = H_S(t) + H_B + H_{int}, \quad (6.1)$$

with

$$H_S(t) = \frac{\hbar}{2}\varepsilon(t)\hat{\sigma}_z + \frac{\hbar}{2}V\hat{\sigma}_x, \quad (6.2)$$

$$H_B = \sum_{\lambda} \hbar \omega_{\lambda} b_{\lambda}^{\dagger} b_{\lambda}, \quad (6.3)$$

$$H_{int} = \sum_{\lambda} g_{\lambda} (b_{\lambda}^{\dagger} + b_{\lambda}) \hat{\sigma}_z, \quad (6.4)$$

where $H_S(t)$, H_B , and H_{int} describe the two level system driven by the telegraph noise, the thermal bath that is composed of independent harmonic oscillators with natural frequencies ω_{λ} and the interaction between the system and thermal environment, respectively. Here σ_i with $i = x, y, z$ are the Pauli spin matrices, $\hbar V$ is the tunneling splitting between two states of the TLS, $\hbar \varepsilon(t) = \hbar \varepsilon_0 + \hbar \Omega \alpha(t)$, where $\hbar \varepsilon_0$ is an energy gap of the TLS, and $\hbar \Omega$ represents the amplitude of fluctuations of the stochastic external field, while $\alpha(t)$ denotes the dichotomous Markov process (DMP) which has possible values ± 1 and its average is $\langle \alpha(t) \rangle = 0$. Besides, autocorrelation function behaves

as an exponential decay, i.e., $\langle \alpha(t) \alpha(t') \rangle = \exp(-\nu(t-t'))$ with the autocorrelation time corresponding to the inverse of ν . $b_\lambda^\dagger$ and b_λ are the annihilation and creation operators of the environmental oscillator, while g_λ in Eq. (6.4) denotes the coupling constant between the qubit and the harmonic oscillator at the bath with frequency ω_λ .

The state of the closed system (TLS + environment) is described by the density matrix $\rho(t)$ whose dynamics is governed by the von-Neumann equation

$$\frac{d\rho(t)}{dt} = \frac{1}{i\hbar} [H(t), \rho(t)]. \quad (6.5)$$

$\rho(t)$ is an infinite dimensional matrix and it contains information on the state of the TLS as well as the state of all the environmental oscillators. Since we are interested only in the dynamics of the TLS, we need to separate the TLS degrees of freedom from the density matrix $\rho(t)$. The standard way to obtain the density matrix of a subsystem from a total system is to take partial trace over the irrelevant degrees of freedom as:

$$\rho_S(t) = \text{Tr}_B[\rho(t)], \quad (6.6)$$

where Tr_B represents the partial trace over the bath degrees of freedom and $\rho_S(t)$ is the density matrix of the TLS. Here the partial trace over the bath dimensions is akin to an averaging process which reflects the effect of the environment on the state of the TLS. We assume that the bath and the two-level system are uncorrelated initially, that is $\rho(0) = \rho_S(0) \otimes \rho_B$, where ρ_B is the density matrix of the environment which is assumed to be in thermal equilibrium at temperature T which can be written as:

$$\rho_B = e^{-\beta H} / \text{Tr}_B[e^{-\beta H}], \quad (6.7)$$

where the inverse temperature $\beta = 1/k_B T$ with k_B as the Boltzmann constant and T is the temperature of the environment. The von Neumann equation is impossible to solve exactly. Various master equations have been developed to find an approximate solution to Eq. (6.5) by expanding it in terms of a small "interaction" parameter in a perturbative way. Since, we will deal with a strong interaction system, one possible way to obtain a master equation is to transform the Hamiltonian into a frame that has "weak" interaction. Polaron transform provides one such frame. Below, we will present a brief explanation of the polaron transform and transform the Hamiltonian Eq. (6.1)

into this frame.

6.2.1 Polaron Transformation

In case of strong coupling between the system and the bath, the polaron transformation can be used to either decrease the efficiency of interaction term or destroy the system-bath coupling constant by transforming the total Hamiltonian into the polaron frame.

This transformation is created by the unitary operator (Leggett et al., 1987)

$$\hat{U} = \exp \left(\sum_{\lambda} \frac{g_{\lambda}}{\omega_{\lambda}} (b_{\lambda}^{\dagger} - b_{\lambda}) \hat{\sigma}_z \right), \quad (6.8)$$

which leads to a shift in the position of bath oscillator based on the state of the TLS.

Applied to the Hamiltonian in Eq. (6.1) and leaving aside the thermal average of interaction constant in the transformed one which leads to

$$\begin{aligned} H'_{tot} = & \frac{\hbar}{2} \left(\varepsilon(t) - \sum_{\lambda} \frac{g_{\lambda}^2}{\omega_{\lambda}} I \right) \hat{\sigma}_z + \sum_{\lambda} \hbar \omega_{\lambda} b_{\lambda}^{\dagger} b_{\lambda} + \frac{\hbar}{2} V_r \hat{\sigma}_x \\ & + \frac{\hbar}{2} V \left[\frac{\hat{\sigma}_x}{2} (B_- + B_+ - 2B) + i \frac{\hat{\sigma}_y}{2} (B_- - B_+) \right], \end{aligned} \quad (6.9)$$

where $\sum_{\lambda} \frac{g_{\lambda}^2}{\omega_{\lambda}} I$ is a constant term that causes no effect on the dichotomous dynamics. The operators B_- and B_+ are the polaron operators which can be written as $B_{\pm} = e^{\mp \hat{S}}$ with $\hat{S} = \sum_{\lambda} \frac{2g_{\lambda}}{\omega_{\lambda}} (b_{\lambda}^{\dagger} - b_{\lambda})$ and their thermal averages are equal to

$$B = \langle e^{\mp \hat{S}(t)} \rangle_B = \langle e^{\mp \hat{S}(0)} \rangle_B = e^{-\Phi}, \quad (6.10)$$

with

$$\Phi = \frac{1}{2\pi} \int_0^{\infty} d\omega \frac{J(\omega)}{\omega^2} \coth \left(\frac{\beta \omega}{2} \right) \text{ with } \beta = \frac{1}{k_B T}. \quad (6.11)$$

The term V_r represents the reduced tunneling frequency

$$V_r = B V = e^{-\Phi} V. \quad (6.12)$$

Taking into account an Ohmic spectral density $J(\omega)$ leads to the infrared divergence of the corresponding integral, Eq. (6.11) that yields to $\Phi \rightarrow \infty$ and $V_r \rightarrow 0$, so the total

Hamiltonian, Eq. (6.9), in the polaron frame becomes

$$H'_{tot} = \frac{\hbar}{2} \varepsilon(t) \hat{\sigma}_z + \sum_{\lambda} \hbar \omega_{\lambda} b_{\lambda}^{\dagger} b_{\lambda} + \frac{\hbar}{2} V (\hat{\sigma}_+ B_- + \hat{\sigma}_- B_+), \quad (6.13)$$

where $\hat{\sigma}_{\pm} = (\hat{\sigma}_x \pm i\hat{\sigma}_y)/2$. In the polaron frame, H_B and σ_z -dependent part of H_S are the same as the original frame, while the interaction term has a new form $\frac{\hbar}{2} V (\hat{\sigma}_+ B_- + \hat{\sigma}_- B_+)$ which is reduced from its original form. B_+ and B_- are the thermal exponential of the correlation functions of the bath and they tend to be zero at long times, so the interaction term becomes smaller.

The coherent dynamics of the TLS whose Hamiltonian is the first term of Eq. (6.13) can be described by the time evolution operator $U(t, t')$ which satisfies

$$i \frac{\partial U}{\partial t} = H_S(t) U \quad \text{with} \quad H_S = \frac{\hbar}{2} \varepsilon(t) \sigma_z, \quad (6.14)$$

which can be solved easily because H_S is diagonal as:

$$\begin{aligned} U_s(t, t') &= T \left[\exp \left(-\frac{i}{\hbar} \int_{t'}^t d\tau H_S(\tau) \right) \right] = T \left[\exp \left(-\frac{i}{2} \int_{t'}^t d\tau \varepsilon(\tau) \right) \hat{\sigma}_z \right], \\ &= \cos \left[\frac{\varepsilon_0(t-t') + f(t, t')}{2} \right] I + i \sin \left[\frac{\varepsilon_0(t-t') + f(t, t')}{2} \right] \hat{\sigma}_z, \end{aligned} \quad (6.15)$$

where T is time-ordering, I is 2×2 unit matrix, and $f(t, t') = -i\Omega \int_{t'}^t d\tau \alpha(\tau)$.

To derive the master equation for the closed system, we transform H'_{tot} of Eq. (6.13) to interaction picture with respect to $H_0 = H_S + H_B = \frac{\hbar}{2} \varepsilon(t) \sigma_z + \sum_{\lambda} \hbar \omega_{\lambda} b_{\lambda}^{\dagger} b_{\lambda}$ as:

$$e^{\frac{iH_S t}{\hbar}} \sigma_{\pm} e^{-\frac{iH_S t}{\hbar}} = \sigma_{\pm} e^{\pm i\varepsilon(t)t}, \quad (6.16)$$

$$e^{\frac{iH_B t}{\hbar}} B_{\pm} e^{-\frac{iH_B t}{\hbar}} = e^{\mp 2 \sum_{\lambda} \frac{g_{\lambda}}{\omega_{\lambda}} (b_{\lambda}^{\dagger} e^{i\omega_{\lambda} t} + b_{\lambda} e^{-i\omega_{\lambda} t})}. \quad (6.17)$$

The master equation in Shrödinger picture can be written as (Nazir and McCutcheon, 2016):

$$\begin{aligned} \dot{\rho}_{S,ij}(t) &= -\frac{i}{\hbar} \sum_{kl} \int_0^t dt' [H_{S,ik}(t') \delta_{lj} - \delta_{ik} H_{S,lj}(t')] \rho_{S,kl}(t') \\ &\quad - \sum_{kl} \int_0^t dt' \Re_{ijkl}(t') \rho_{S,kl}(t'), \end{aligned} \quad (6.18)$$

where the Redfield relaxation tensor $\mathfrak{R}_{ij,kl}(t)$ is given by (Blum, 1996):

$$\mathfrak{R}_{ijkl}(t) = \delta_{lj} \sum_r \Gamma_{irrk}^+(t) + \delta_{ik} \sum_r \Gamma_{lrrj}^-(t) - \Gamma_{ljik}^+(t) - \Gamma_{ljik}^-(t). \quad (6.19)$$

Here, the rates $\Gamma_{ijkl}^\pm(t)$ can be calculated by using expressions in the below:

$$\Gamma_{lj,ik}^+(t) = \frac{1}{\hbar^2} \sum_{\alpha,\beta} \langle B_\alpha(t) B_\beta(0) \rangle_B A_{\alpha,lj} \sum_{m,n} U_{S,im}(t,0) A_{\beta,mn} U_{S,kn}^*(t,0), \quad (6.20)$$

$$\Gamma_{lj,ik}^-(t) = \frac{1}{\hbar^2} \sum_{\alpha,\beta} \langle B_\beta(0) B_\alpha(t) \rangle_B \sum_{m,n} U_{S,lm}(t,0) A_{\beta,mn} U_{S,jn}^*(t,0) A_{\alpha,ik}, \quad (6.21)$$

where A and B are the system and bath operators that enter into interaction term of the Hamiltonian in Eq. (6.13), respectively.

6.2.2 The Non-Markovian Master Equation

For the definition of the dynamics of two-level system, it is convenient to use the Bloch vector $P(t) = \{P_x(t), P_y(t), P_z(t)\} \in R^3$ which is described by

$$P(t) = \text{Tr}[\sigma \rho(t)] = \begin{pmatrix} \rho_{12}(t) + \rho_{21}(t) \\ i(\rho_{21}(t) - \rho_{12}(t)) \\ \rho_{11}(t) - \rho_{22}(t) \end{pmatrix}. \quad (6.22)$$

Here $\sigma = \{\sigma_x, \sigma_y, \sigma_z\}$ is the vector representation of Pauli matrices.

To obtain the equations satisfied by the elements of $P(t)$, we use Eq. (6.18) along with Eqs. (6.19)-(6.22) and obtain the dynamics of the population difference and coherence of the two-level system coupled to the thermal bath and driven by the stochastic external field (Jirari and Pötz, 2006) as follows:

$$\dot{P}_x(t) = \varepsilon(t) P_y(t), \quad (6.23)$$

$$\dot{P}_y(t) = -\varepsilon(t) P_x(t) - \int_0^t dt' \zeta(t, t') P_x(t') - \int_0^t dt' \gamma(t, t') P_y(t'), \quad (6.24)$$

$$\dot{P}_z(t) = -\int_0^t dt' \gamma(t, t') P_z(t') - \int_0^t dt' \chi(t, t'), \quad (6.25)$$

where

$$\zeta(t, t') = V^2 \zeta_0(t - t') \sin [\varepsilon_0(t - t') + f(t, t')], \quad (6.26)$$

$$\gamma(t, t') = V^2 \gamma_0(t - t') \cos [\varepsilon_0(t - t') + f(t, t')], \quad (6.27)$$

$$\chi(t, t') = V^2 \chi_0(t - t') \sin [\varepsilon_0(t - t') + f(t, t')], \quad (6.28)$$

with

$$\zeta_0(t - t') = \gamma_0(t - t') = e^{-G_s(t - t')} \cos [G_a(t - t')], \quad (6.29)$$

$$\chi_0(t - t') = e^{-G_s(t - t')} \sin [G_a(t - t')]. \quad (6.30)$$

Note that the dynamics of the population difference and the coherence are uncoupled in this formulation. Here the function $f(t, t')$ is given in Eq. (6.15), while the quantities $G_a(t)$ and $G_s(t)$ result from the autocorrelation of the polaron operators $B_{\pm}(t)$ at different times which are given by

$$\langle B_{\pm}(t) B_{\pm}(0) \rangle = e^{-G_s(t) + i G_a(t)}, \quad (6.31)$$

$$\langle B_{\pm}(t) B_{\mp}(0) \rangle = e^{-G_s(t) - i G_a(t)}, \quad (6.32)$$

and

$$G_a(t) = \frac{1}{2\pi} \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} \sin(\omega t), \quad (6.33)$$

$$G_s(t) = \frac{1}{2\pi} \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} \coth\left(\frac{\beta \omega}{2}\right) (1 - \cos(\omega t)). \quad (6.34)$$

Since every realization of the noise term would be somewhat different, such equation should be expressed as the average over the different realizations of the noise process. Below, we outline the noise averaging of the dynamical equations (6.23)-(6.25) over the dichotomous noise.

6.2.3 Averaging over the Stochastic Process

Integro-differential kinetic equations for both the populations difference and the coherence of the states of the dichotomously driven two-level system, Eqs. (6.23)-(6.25), contain stochastic term $f(t, t')$. Cumulant expansion method (Kubo, 1963; Fox, 1978)

is one of the most widely used averaging method for the Gaussian type noise processes but for the dichotomous type noise, it is impractical. Two theorems due to Bourret and Frisch (Bourret et al., 1973) and Shapiro and Loginov (Shapiro and Loginov, 1978) make it possible to noise average the dynamical equations of $P_x(t)$, $P_y(t)$, and $P_z(t)$ over the dichotomous noise. Here, we outline those theorems and perform the averaging. This section follows the method of Goychuk *et al.* (Goychuk et al., 1995b) and generalizes their method to the equations for $P_x(t)$ and $P_y(t)$. Let us redefine the kernels $\zeta(t, t')$, $\gamma(t, t')$, and $\chi(t, t')$ in Eqs. (6.26)-(6.28) in the form

$$\gamma(t, t') = \gamma_0(t - t') \text{Re} \left[e^{-i\varepsilon_0(t-t')} S(t, t') \right], \quad (6.35)$$

$$\zeta(t, t') = -\zeta_0(t - t') \text{Im} \left[e^{-i\varepsilon_0(t-t')} S(t, t') \right], \quad (6.36)$$

$$\chi(t, t') = -\chi_0(t - t') \text{Im} \left[e^{-i\varepsilon_0(t-t')} S(t, t') \right], \quad (6.37)$$

where $S(t, t') = \exp \left[-i\Omega \int_{t'}^t d\tau \alpha(\tau) \right]$ is the time evolution operator of the Kubo oscillator and satisfies the stochastic evolution equation:

$$\frac{d}{dt} S(t, t') = -i\Omega \alpha(t) S(t, t'), \quad S(t', t') = 1. \quad (6.38)$$

To average the set of equations in Eqs. (6.23)-(6.25), one needs to consider the formal expression (Goychuk et al., 1995b):

$$\langle S(t, t' + \tau) \alpha(t' + \tau) \alpha(t') P_i(t') \rangle \quad (\tau > 0) \text{ and } i = x, y, z. \quad (6.39)$$

Here both operators $S(t, t' + \tau)$ and $P_i(t')$ are functionals of DMP $\alpha(t)$ implying only times posterior to $t' + \tau$ and prior to t' , respectively. Therefore, one can use this expression with the theorem of Bourret and Frisch (Bourret et al., 1973), which provide the essential tool to calculate the autocorrelation functions. According to the theorem we have:

$$\begin{aligned} \langle S(t, t' + \tau) \alpha(t' + \tau) \alpha(t') P_i(t') \rangle &= \langle S(t, t' + \tau) \rangle \langle \alpha(t' + \tau) \alpha(t') \rangle \langle P_i(t') \rangle \\ &+ \langle S(t, t' + \tau) \alpha(t' + \tau) \rangle \langle \alpha(t') P_i(t') \rangle \end{aligned} \quad (6.40)$$

In the limit of $\tau \rightarrow 0$, and by using the special property of DMP, $\alpha^2(t) = 1$, Eq.(6.40) can be written as:

$$\langle S(t, t') P_i(t') \rangle = S_0(t - t') \langle P_i(t') \rangle + S_1(t - t') \langle \alpha(t') P_i(t') \rangle, \quad (6.41)$$

with $S_0(t - t') = \langle S(t, t') \rangle$ and $S_1(t - t') = \langle \alpha(t') S(t, t') \rangle$. By using the result in Eq. (6.41), the next expression can be written

$$\langle \alpha(t) S(t, t') P_i(t') \rangle = S_1(t - t') \langle P_i(t') \rangle + S_2(t - t') \langle \alpha(t') P_i(t') \rangle, \quad (6.42)$$

where $S_2(t - t') = \langle \alpha(t) \alpha(t') S(t, t') \rangle$. We should note that one can obtain $S_1(t - t')$ and $S_2(t - t')$ in terms of $S_0(t - t')$ by taking into account Eq. (6.38) which leads to

$$S_1(t - t') = \frac{i}{\Omega} \frac{d}{dt} S_0(t - t'), \quad (6.43)$$

$$S_2(t - t') = -\frac{1}{\Omega^2} \frac{d^2}{dt^2} S_0(t - t'). \quad (6.44)$$

Before finding the average of Pauli spin operators, one needs to evaluate $S_{0,1,2}(\tau)$ with $\tau = t - t'$, which are the averaged evolution operator of the noise by using the theorem of Shapiro and Loginov (Shapiro and Loginov, 1978):

$$\frac{d}{dt} \langle \alpha(t) F(t) \rangle = -\nu \langle \alpha(t) F(t) \rangle + \langle \alpha(t) \frac{d}{dt} F(t) \rangle, \quad (6.45)$$

where $F(t)$ is any functional of DMP $\alpha(t)$. Using Eqs. (6.38) and (6.43)-(6.45), one gets a set of coupled differential equation for $S_0(\tau)$ and $S_1(\tau)$

$$\frac{d}{dt} S_0(\tau) = -i\Omega S_1(\tau), \quad (6.46)$$

$$\frac{d}{dt} S_1(\tau) = -\nu S_1(\tau) - i\Omega S_0(\tau). \quad (6.47)$$

These first order coupled differential equation with initial conditions $S_0(0) = 1$ and $S_1(0) = 0$ can be solved easily to obtain

$$S_0(\tau) = \exp\left(-\frac{v\tau}{2}\right) \left[\cosh\left(\frac{1}{2}\sqrt{v^2 - 4\Omega^2}\tau\right) + \frac{v}{\sqrt{v^2 - 4\Omega^2}} \sinh\left(\frac{1}{2}\sqrt{v^2 - 4\Omega^2}\tau\right) \right], \quad (6.48)$$

$$S_1(\tau) = -\frac{i2\Omega}{\sqrt{v^2 - 4\Omega^2}} \exp\left(-\frac{v\tau}{2}\right) \sinh\left(\frac{1}{2}\sqrt{v^2 - 4\Omega^2}\tau\right). \quad (6.49)$$

We should note that an other way to find $S_1(\tau)$ is to take derivative of $S_0(\tau)$ in Eq. (6.44) with respect to τ . $S_2(\tau)$ is found by using Eq. (6.48) in Eq.(6.44):

$$S_2(\tau) = \exp\left(-\frac{v\tau}{2}\right) \left[\cosh\left(\frac{1}{2}\sqrt{v^2 - 4\Omega^2}\tau\right) - \frac{v}{\sqrt{v^2 - 4\Omega^2}} \sinh\left(\frac{1}{2}\sqrt{v^2 - 4\Omega^2}\tau\right) \right]. \quad (6.50)$$

Now, we will use the results Eqs. (6.48)-(6.50) along with Shapiro-Loginov theorem to calculate the noise averages of the coupled set of differential equations (6.23)-(6.25) that determine the dynamics of the population difference $P_z(t)$ and the coherences $P_x(t)$ and $P_y(t)$. Let start with Eq. (6.25). Taking the noise average of the both sides of the equation, one gets:

$$\langle \dot{P}_z(t) \rangle = - \int_0^t dt' \langle \gamma(t, t') P_z(t') \rangle - \int_0^t dt' \langle \chi(t, t') \rangle, \quad (6.51)$$

where $\langle \gamma(t, t') P_z(t') \rangle = \langle \gamma_0(t - t') \text{Re} \left[e^{-i\varepsilon_0(t-t')} S(t, t') \right] P_z(t') \rangle$ and this average can be calculated by taking into account of Bourret and Frisch theorem (Bourret et al., 1973) and Eq (6.41) as:

$$\begin{aligned} \langle \gamma(t, t') P_z(t') \rangle &= \text{Re} \left[\gamma_0(t - t') e^{-i\varepsilon_0(t-t')} \langle S(t, t') P_z(t') \rangle \right], \\ &= \gamma_0(t - t') \left(S_0(t - t') \cos [\varepsilon_0(t - t')] \langle P_z(t') \rangle \right. \\ &\quad \left. + i S_1(t - t') \sin [\varepsilon_0(t - t')] \langle \alpha(t') P_z(t') \rangle \right). \end{aligned} \quad (6.52)$$

Since $\chi(t, t')$ contains only the noise term and no TLS operator, its average is easier to find:

$$\langle \chi(t, t') \rangle = \chi_0(t - t') S_0(t - t') \sin [\varepsilon_0(t - t')]. \quad (6.53)$$

Plugging Eq. (6.52) and Eq. (6.53) into Eq. (6.51) leads to integro-differential equation for the noise-averaged population difference

$$\begin{aligned}\langle \dot{P}_z(t) \rangle = & - \int_0^t dt' S_0(t-t') \gamma_0(t-t') \cos [\varepsilon_0(t-t')] \langle P_z(t') \rangle \\ & - i \int_0^t dt' S_1(t-t') \gamma_0(t-t') \sin [\varepsilon_0(t-t')] \langle \alpha(t') P_z(t') \rangle \\ & - \int_0^t dt' S_0(t-t') \chi_0(t-t') \sin [\varepsilon_0(t-t')] .\end{aligned}\quad (6.54)$$

Since Eq. (6.54) indicates that $\langle \dot{P}_z(t) \rangle$ now depends on both the integral of $\langle P_z(t) \rangle$ over a kernel that contains noise properties and the average of the noise signal times $P_z(t)$ over the same noise, *i.e.*, $\langle \alpha(t') P_z(t') \rangle$ term, we need to derive an expression for the time-rate of change of $\langle \alpha(t') P_z(t') \rangle$ also. Applying Shapiro-Loginov theorem to $\langle \alpha(t) P_z(t) \rangle$, we get

$$\frac{d}{dt} \langle \alpha(t) P_z(t) \rangle = -\nu \langle \alpha(t) P_z(t) \rangle + \langle \alpha(t) \frac{d}{dt} P_z(t) \rangle. \quad (6.55)$$

Inserting Eq. (6.54) into Eq. (6.55) leads to obtain following expression:

$$\begin{aligned}\frac{d}{dt} \langle \alpha(t) P_z(t) \rangle = & -\nu \langle \alpha(t) P_z(t) \rangle - i \int_0^t dt' S_1(t-t') \chi_0(t-t') \cos [\varepsilon_0(t-t')] \\ & - i \int_0^t dt' S_1(t-t') \gamma_0(t-t') \sin [\varepsilon_0(t-t')] \langle P_z(t') \rangle \\ & - \int_0^t dt' S_2(t-t') \gamma_0(t-t') \cos [\varepsilon_0(t-t')] \langle \alpha(t') P_z(t') \rangle.\end{aligned}\quad (6.56)$$

Note that the results in Eq. (6.54) and Eq. (6.56) are consistent with the ones in Ref. (Goychuk et al., 1995b) that can be interpreted as a generalization of NIBA to the problem of spin-boson model driven by dichotomous noise.

The dynamics of the $P_x(t)$ and $P_y(t)$ terms can be, similarly, averaged over the

dichotomous noise as follows:

$$\langle \dot{P}_x(t) \rangle = \varepsilon_0 \langle P_y(t) \rangle + \Omega \langle \alpha(t) P_y(t) \rangle, \quad (6.57)$$

$$\begin{aligned} \langle \dot{P}_y(t) \rangle = & -\varepsilon_0 \langle P_x(t) \rangle - \Omega \langle \alpha(t) P_x(t) \rangle \\ & - \int_0^t dt' S_0(t-t') \gamma_0(t-t') \cos[\varepsilon_0(t-t')] \langle P_y(t') \rangle \\ & - i \int_0^t dt' S_1(t-t') \gamma_0(t-t') \sin[\varepsilon_0(t-t')] \langle \alpha(t') P_y(t') \rangle \\ & - i \int_0^t dt' S_1(t-t') \zeta_0(t-t') \cos[\varepsilon_0(t-t')] \langle \alpha(t') P_x(t') \rangle \\ & - \int_0^t dt' S_0(t-t') \zeta_0(t-t') \sin[\varepsilon_0(t-t')] \langle P_x(t') \rangle, \end{aligned} \quad (6.58)$$

$$\frac{d}{dt} \langle \alpha(t) P_x(t) \rangle = -\nu \langle \alpha(t) P_x(t) \rangle + \varepsilon_0 \langle \alpha(t) P_y(t) \rangle + \Omega \langle P_y(t) \rangle, \quad (6.59)$$

$$\begin{aligned} \frac{d}{dt} \langle \alpha(t) P_y(t) \rangle = & -\nu \langle \alpha(t) P_y(t) \rangle - \varepsilon_0 \langle \alpha(t) P_x(t) \rangle - \Omega \langle P_x(t) \rangle \\ & - \int_0^t dt' S_2(t-t') \gamma_0(t-t') \cos[\varepsilon_0(t-t')] \langle \alpha(t') P_y(t') \rangle \\ & - i \int_0^t dt' S_1(t-t') \gamma_0(t-t') \sin[\varepsilon_0(t-t')] \langle P_y(t') \rangle \\ & - \int_0^t dt' S_2(t-t') \zeta_0(t-t') \sin[\varepsilon_0(t-t')] \langle \alpha(t') P_x(t') \rangle \\ & - i \int_0^t dt' S_1(t-t') \zeta_0(t-t') \cos[\varepsilon_0(t-t')] \langle P_x(t') \rangle. \end{aligned} \quad (6.60)$$

One should note that while the averages in Eq. (6.54) and Eq. (6.56) are also derived in Ref. (Goychuk et al., 1995b), Eqs. (6.57)-(6.60) are derived for the first time in the present study. A complete characterization of the state of the TLS as function of parameters that describe its dynamics, properties of its thermal environmental and the noise process that effects its transition frequencies requires exact solution of the set of coupled integro-differential equations in (6.54)-(6.56) for the population differences and (6.57)-(6.60) for the coherences. As the integrals in these equations are in convolution form, Laplace transform techniques can be utilized to find a solution to the integro-differential equations. Below, we derive formal Laplace transformations for the set of those integro-differential equations.

6.2.4 Laplace Transformation of Population Difference And Coherence Process

Let define the Laplace transforms of the $\langle P_i(t) \rangle$ and the $\langle \alpha_i(t) \rangle = \langle \alpha(t) P_i(t) \rangle$:

$$\bar{P}_i(s) = \int_0^\infty dt e^{-st} \langle P_i(t) \rangle, \quad (6.61)$$

$$\bar{\alpha}_i(s) = \int_0^\infty dt e^{-st} \langle \alpha(t) P_i(t) \rangle, \quad (6.62)$$

with $i = x, y, z$, we get:

$$\bar{P}_x(s) = \frac{1}{s} (\langle P_x(0) \rangle + \varepsilon_0 \bar{P}_y(s) + \Omega \bar{\alpha}_y(s)), \quad (6.63)$$

$$\begin{aligned} \bar{P}_y(s) = & \left(\langle P_y(0) \rangle + \left(\frac{i}{4} \bar{H}_1(s) + \frac{i\nu}{4m} \bar{H}_2(s) - \varepsilon_0 \right) \bar{P}_x(s) + \left(\frac{\Omega}{2m} \bar{H}_3(s) - \Omega \right) \bar{\alpha}_x(s) \right. \\ & \left. + \frac{i\Omega}{2m} \bar{F}_3(s) \bar{\alpha}_y(s) \right) / \left(s + \frac{1}{4} \bar{F}_4(s) - \frac{\nu}{4m} \bar{F}_3(s) \right), \end{aligned} \quad (6.64)$$

$$\begin{aligned} \bar{P}_z(s) = & \left(\frac{i}{4} \bar{G}_1(s) + \frac{i\nu}{4m} \bar{G}_2(s) + \frac{i\Omega}{2m} \bar{F}_2(s) \bar{\alpha}_z(s) + \langle P_z(0) \rangle \right) / \left(s + \frac{1}{4} \bar{F}_4(s) - \frac{\nu}{4m} \bar{F}_3(s) \right), \\ \bar{\alpha}_x(s) = & \frac{1}{s + \nu} (\langle \alpha(0) P_x(0) \rangle + \varepsilon_0 \bar{\alpha}_y(s) + \Omega \bar{P}_y(s)), \end{aligned} \quad (6.65)$$

$$\begin{aligned} \bar{\alpha}_y(s) = & \left(\langle \alpha(0) P_y(0) \rangle + \left(\frac{i}{4} \bar{H}_1(s) - \frac{i\nu}{4m} \bar{H}_2(s) - \varepsilon_0 \right) \bar{\alpha}_x(s) + \left(\frac{\Omega}{2m} \bar{H}_3(s) - \Omega \right) \bar{P}_x(s) \right. \\ & \left. + \frac{i\Omega}{2a} \bar{F}_2(s) \bar{P}_y(s) \right) / \left(s + \nu + \frac{\nu}{4m} \bar{F}_3(s) + \frac{1}{4} \bar{F}_4(s) \right), \end{aligned} \quad (6.66)$$

$$\begin{aligned} \bar{\alpha}_z(s) = & \left(\langle \alpha(0) P_z(0) \rangle + \frac{\Omega}{2m} \bar{G}_3(s) + \frac{i\Omega}{2m} \bar{F}_2(s) \bar{P}_z(s) \right) / \left(s + \nu + \frac{1}{4} \bar{F}_4(s) \right. \\ & \left. + \frac{\nu}{4m} \bar{F}_3(s) \right), \end{aligned} \quad (6.67)$$

where $m = \sqrt{\nu^2 - 4\Omega^2}$,

$$\bar{A}_1(s) = \bar{a}_0(s - \beta_1) - \bar{a}_0(s - \beta_1^*) + \bar{a}_0(s + \beta_2^*) - \bar{a}_0(s + \beta_2), \quad (6.68)$$

$$\bar{A}_2(s) = \bar{a}_0(s - \beta_1) - \bar{a}_0(s - \beta_1^*) - \bar{a}_0(s + \beta_2^*) + \bar{a}_0(s + \beta_2), \quad (6.69)$$

$$\bar{A}_3(s) = -\bar{a}_0(s - \beta_1) - \bar{a}_0(s - \beta_1^*) + \bar{a}_0(s + \beta_2^*) + \bar{a}_0(s + \beta_2). \quad (6.70)$$

Form of $\bar{H}_i(s)$, $\bar{G}_i(s)$, and $\bar{F}_i(s)$ for $i = 1, 2, 3$ have the same form with $\bar{A}_i(s)$ and can be obtained by using $\bar{h}_0(s)$, $\bar{g}_0(s)$, and $\bar{f}_0(s)$ instead of $\bar{a}_0(s)$ in Eqs. (6.68)-(6.70). $\bar{F}(s)$

has a fourth component which equal to the sum of all four $\bar{f}_0(s)$ factors:

$$\bar{F}_4(s) = \bar{f}_0(s - \beta_1) + \bar{f}_0(s - \beta_1^*) + \bar{f}_0(s + \beta_2^*) + \bar{f}_0(s + \beta_2). \quad (6.71)$$

Here $\bar{h}_0(s)$, $\bar{g}_0(s)$, and $\bar{f}_0(s)$ represent the Laplace transformation of $\zeta_0(t)$, $\chi_0(t)$, $\gamma_0(t)$ respectively. β_1 and β_2 can be expressed as:

$$\beta_1 = \frac{m}{2} - \frac{\nu}{2} + i\varepsilon_0, \quad (6.72)$$

$$\beta_2 = \frac{m}{2} + \frac{\nu}{2} + i\varepsilon_0. \quad (6.73)$$

Note that the Laplace transform of averaged population difference in Eq. (6.65) is the general form of Eq. (29) in Ref. (Goychuk et al., 1995b) in case of zero level splitting $\varepsilon_0 = 0$. In addition, the formal solutions of this problem correspond to Eqs. (6.63)-(6.67) that can be characterized by coherent and incoherent regime depending on the bath, system, their interaction parameters, and the stochastic field parameters. In case of incoherent regime, the average relaxation time τ can be written as

$$\tau = \int_0^\infty dt \langle P_z(t) \rangle = \bar{P}_z(0). \quad (6.74)$$

Inverse of $\bar{P}_z(0)$ gives us the effective relaxation rate Γ which is

$$\Gamma = \left(\frac{1}{4}\bar{F}_4(0) - \frac{\nu}{4a}\bar{F}_3(0) \right) / \left(\frac{i}{4}\bar{G}_1(0) + \frac{i\nu}{4a}\bar{G}_2(0) + \frac{i\Omega}{2a}\bar{F}_2(0)\bar{\alpha}_z(0) + \langle P_z(0) \rangle \right). \quad (6.75)$$

We should note that Eq. (6.75) indicates the effect of the external stochastic field on the population difference of the dichotomous driven two-level system.

The most important factor that defines the interaction between the TLS and the environment is the spectral density of the bath which also determines the complexity of the kernel of the integro-differential equations. In the following subsection, we will provide a detailed analysis of the various forms of spectral functions that are used to describe bath descriptions and derive analytical expressions for the kernel functions $e^{-G_s(t)} \cos[G_a(t)]$ and $e^{-G_s(t)} \sin[G_a(t)]$ for those spectral functions.

6.2.5 Bath Correlation Functions

The effect of the environment on the reduced system can be described by using a spectral density $J(\omega)$ containing the information about the strength of interaction between the system and the bath as well as frequency distribution of the environmental oscillators. The formal definition of $J(\omega)$ is (Weiss, 2008; Breuer and Petruccione, 2002)

$$J(\omega) = 2\pi \sum_{\lambda} g_{\lambda}^2 \delta(\omega - \omega_{\lambda}), \quad (6.76)$$

where g_{λ} represents the coupling strength between the system and the bath oscillator with frequency ω_{λ} and the Dirac delta function counts the environmental vibration modes with frequency ω_{λ} .

Although the two-level system is an abstraction and the dynamics of any two physically different realizations of TLS would be similar for the same parameter magnitudes, the form of $J(\omega)$ depends strongly on the underlying physical system of the TLS and the character of the interaction between the TLS and its environment. In physics literature, $J(\omega)$ is chosen either in power form with a cutoff or a structured form. In this subsection, we will provide an in-depth study of bath correlation functions $G_s(t)$ and $G_a(t)$ that enter into dynamical equations for power, semi-empirical, and structured spectral densities.

6.2.5.1 Power Spectral Density

The power spectral densities have the general form

$$J(\omega) = \alpha \frac{\omega^s}{\omega_c^{s+1}} f(\omega, \omega_c), \quad (6.77)$$

where α is the dimensionless interaction strength, s is the power exponent, $f(\omega, \omega_c)$ is the cutoff function that ensures $J(\omega)$ smoothly goes to zero at high frequencies and ω_c is the cutoff frequency of the bath oscillators. The most widely used cutoff functions are exponential ($f(\omega, \omega_c) = e^{-\omega/\omega_c}$), Gaussian ($f(\omega, \omega_c, \omega_0) = e^{-(\omega-\omega_0)^2/2\omega_c^2}$), abrupt cut ($\theta(\omega_c - \omega)$) is the Heaviside step function and the Lorentzian cutoff ($f(\omega, \omega_c) = \omega_c^2/(\omega^2 + \omega_c^2)$). Such spectral densities are called as sub-Ohmic ($s < 1$), Ohmic ($s = 1$), and super-Ohmic ($s > 1$). Physical system such as $1/f$ noise or lossy inductor-capacitor-resistor systems are claimed to be better modelled as sub-Ohmic dissipa-

tion (Tong and Vojta, 2006). The effect of environment in Cooper-pair boxes, quantum dots coupled to Bose-Einstein condensate are examples of Ohmic spectral density, while quantum dots coupled with transverse acoustic and optical phonons of the environment are good example to systems that can be described by super-Ohmic spectral functions with $s = 3$ and $s = 5$, respectively. The most widely studied spin-boson model is the one with the Ohmic spectral density.

We will, first, consider an Ohmic bath and calculate the bath correlation functions $G_a(t)$ and $G_s(t)$ from Eq. (6.33) and Eq. (6.34) by using $J(\omega)$ of Eq. (6.77) with $s = 1$. Expressing $\coth(x)$ as $1 + 2 \sum_{n=1}^{\infty} e^{-2nx}$, bath integrals can be carried out exactly to obtain:

$$G_a(t) = \frac{\alpha}{2\pi} \arctan[\omega_c t], \quad (6.78)$$

$$G_s(t) = \frac{\alpha}{2\pi} \log \left[\frac{\sqrt{1+t^2\omega_c^2} \Gamma \left[1 + \frac{1}{2\beta\omega_c} \right]^2}{\Gamma \left[1 + \frac{1}{2\beta\omega_c} + i\frac{t}{2\beta} \right] \Gamma \left[1 + \frac{1}{2\beta\omega_c} - i\frac{t}{2\beta} \right]} \right], \quad (6.79)$$

where a function Γ is Euler's gamma function and β is the inverse temperature parameter. Note that these results are consistent with the ones reported by Ref. (Jirari and Pötzt, 2006). By using these results $\gamma_0(t)$ of Eq. (6.77) can be obtained as:

$$\begin{aligned} \gamma_0(t) &= e^{-G_s(t)} \cos[G_a(t)], \\ &= \frac{1}{2} \left(\frac{\Gamma \left[1 + \frac{1}{2\beta\omega_c} + i\frac{t}{2\beta} \right] \Gamma \left[1 + \frac{1}{2\beta\omega_c} - i\frac{t}{2\beta} \right]}{\sqrt{1-it\omega_c} \Gamma \left[1 + \frac{1}{2\beta} \right]} \right)^{\frac{\alpha}{2\pi}} \\ &\quad + \frac{1}{2} \left(\frac{\Gamma \left[1 + \frac{1}{2\beta\omega_c} + i\frac{t}{2\beta} \right] \Gamma \left[1 + \frac{1}{2\beta\omega_c} - i\frac{t}{2\beta} \right]}{\sqrt{1+it\omega_c} \Gamma \left[1 + \frac{1}{2\beta} \right]} \right)^{\frac{\alpha}{2\pi}}. \end{aligned} \quad (6.80)$$

The expression for $\gamma_0(t)$ can be reduced to simpler forms for low ($\beta \rightarrow \infty$), intermediate ($\beta \rightarrow \omega_c^{-1}$), high ($\beta \rightarrow 0$) temperature limits which are displayed in the first column of Table 6.1.

The Laplace transform of $\gamma_0(t)$ enters into the integro-differential equations of $P_i(t)$, Eqs. (6.23)-(6.25), and can be analytically evaluated at low and intermediate tempera-

tures as:

$$\bar{\gamma}_0(s) = \frac{1}{\omega_c} \left(\frac{\pi}{2} - \text{SinIntegral} \left[\frac{s}{\omega_c} \right] \right) \cos \left[\frac{s}{\omega_c} \right] + \frac{1}{\omega_c} \sin \left[\frac{s}{\omega_c} \right] \text{CosIntegral} \left[\frac{s}{\omega_c} \right], \quad (6.81)$$

$$\bar{\gamma}_0(s) = \frac{1}{\pi \omega_c} \left(\psi_0 \left[\frac{3}{4} + \frac{s}{2\pi \omega_c} \right] - \psi_0 \left[\frac{1}{4} + \frac{s}{2\pi \omega_c} \right] \right), \quad (6.82)$$

where the symbol $\text{SinIntegral}[x]$ ($\text{CosIntegral}[x]$) represents the sine integral (the cosine integral) and $\psi_0[x]$ corresponds to the digamma function. Although the inverse Laplace transform of the full expression for $P_i(t)$ s are difficult to find for this form of $\gamma_0(s)$, physical properties such as transport coefficients are just the zero frequency value of the Laplace transform of $P_i(t)$ and equations (6.81) and (6.82) are useful for such calculations. Moreover, γ_E is the Euler's constant which is equal to 0.57721 and CoshIntegral (SinhIntegral) gives the hyperbolic cosine (sine) integral in table 6.1.

Table 6.1. The form of both $G_a(t)$ and $G_s(t)$ obtained in terms of different spectral densities as well as distinct temperatures

$J_C(\omega)(\text{Low T.})$	$J_{DL}(\omega)(\text{Low T.})$
$G_a(t) = \frac{\alpha}{2\pi} \arctan[t \omega_c]$	$G_a(t) = \frac{\pi}{4} (1 - e^{-t \omega_c})$
$G_s(t) = \frac{\alpha}{4\pi} \log[1 + t^2 \omega_c^2]$	$G_s(t) = \frac{\alpha}{2\pi} \gamma_E + \frac{\eta}{2\pi} \log[t \omega_c]$ $-\frac{\alpha}{2\pi} \cosh[t \omega_c] \text{CoshIntegral}[t \omega_c]$ $+\frac{\alpha}{2\pi} \sinh[t \omega_c] \text{SinhIntegral}[t \omega_c]$
$J_C(\omega)(\text{High T.})$	$J_{DL}(\omega)(\text{High T.})$
$G_a(t) = \frac{\alpha}{2\pi} \arctan[t \omega_c]$	$G_a(t) = \frac{\alpha}{4} (1 - e^{-t \omega_c})$
$G_s(t) = \frac{\alpha}{4\pi\beta \omega_c} \log[1 + t^2 \omega_c^2]$ $+\frac{\alpha t}{2\pi\beta} \arctan[t \omega_c]$ $+\frac{\alpha\beta}{6\pi} \frac{t^2 \omega_c^3}{1+t^2 \omega_c^2}$	$G_s(t) = \frac{\alpha}{4\beta \omega_c} (t \omega_c - 1 + e^{-t \omega_c})$ $+\frac{\alpha\beta \omega_c}{12} e^{-t \omega_c} (e^{t \omega_c} - 1)$

As the second class of special functions, we have considered a system that is used to study charge and energy transport in biological systems where the environmental is modelled as a background of vibrations with a wide frequency distribution which is modelled as log-normal distribution and a prominent vibrational mode which is modelled as a structured spectral function with Lorentzian shape.

6.2.5.2 Semi-Empirical Spectral Density

Based on studies in coherent energy transfer in biological system such as photosynthesis, a special form of bath spectral density which is a combination of distinct forms has been considered in Ref. (Ritschel and Alexander, 2014). In their approach, the spectral function $J(\omega)$ is fitted to experimentally obtained data with a form

$$J(\omega) = \omega^{n-1} \sum_k p_k (J_k(\omega) - J_k(-\omega)), \quad (6.83)$$

where n is an odd number, p_k are positive weights and $J_k(\omega) = \prod_{k_j} \frac{1}{(\omega - \omega_{k_j})(\omega - \omega_{k_j}^*)}$ with $\omega_{k_j} = \Omega_{k_j} + i\gamma_{k_j}$ here Ω_{k_j} and γ_{k_j} are positive real numbers. The main motivation in choosing the form of $J(\omega)$ in Eq. (6.83) is to have simple poles in its definition, so that $G_a(\tau)$ and $G_s(\tau)$ integrals would be easy to carry out.

We are interested in finding a representation of a physically relevant spectral density as Eq. (6.83). Towards that end, we will consider a TLS in an environment that can be considered as combination of two distinct spectral density. One can imagine an acceptor-donor system in a molecular environment where one or more vibrational modes of the surrounding molecules are strongly coupled to the the acceptor-donor system while the remaining environmental vibrations could be considered as the background noise with some structure. The prominent vibrational mode can be modelled as a harmonic oscillator itself coupled to an Ohmic environment which broadens its energy levels and cause damping. Such a damped harmonic mode can be described by the spectral function (Ritschel and Alexander, 2014):

$$J_{vib}(\omega) = \frac{X \omega^2 J_{ohm}(\omega)}{(\omega - \tilde{g}(\omega))^2 + J_{ohm}^2(\omega)}. \quad (6.84)$$

where $\tilde{g}(\omega) = \Omega - \alpha\Lambda/\pi + J_{ohm}(\omega)\text{Ei}(\omega/\Lambda)/\pi$, $\text{Ei}(z) = -\int_{-z}^{\infty} dt e^{-t}/t$, and $J_{ohm}(\omega) = \alpha \omega e^{-\omega/\Lambda}$. the Huang-Rhys factor X is the strength of the coupling between the two-level system and the oscillator, Ω is the characteristic frequency of the oscillator, while Λ is the cutoff frequency of the bath. The numerical value of those parameters are $\alpha = 0.3$, $\omega_c = 100 \text{ cm}^{-1}$, $\Omega = 180 \text{ cm}^{-1}$, and $X = 0.03$.

Vibrational modes of the background can be modelled with an Ohmic spectral den-

sity with log-normal cutoff function of the form:

$$J_{ln}(\omega) = \frac{\pi S \omega}{\sqrt{2\pi}\sigma} e^{-\log[\omega/\omega_c]^2/2\sigma^2}, \quad (6.85)$$

where ω_c is the cutoff frequency and σ is the standard deviation. In a number of photosynthetic complexes (Kell et al., 2013) this type spectral density is used to describe experimental $J(\omega)$ that converges at large frequencies. The numerical values for the parameters of $J_{ln}(\omega)$ are $S = 0.3$, $\sigma = 0.7$, and $\omega_c = 38 \text{ cm}^{-1}$, which are deduced from the experimental results in (Kell et al., 2013). To find the parameters of the empirical spectral density $J(\omega)$ of Eq. (6.83), we perform nonlinear least square fitting of the data obtained from $J_{vib}(\omega) + J_{ln}(\omega)$ with parameters given in the paragraphs above to the form Eq. (6.83) with $n = 5$. The resulting weights and frequency and damping constants are displayed in Table 6.2. We also plot the fitted spectral density along with the data as well as the fit errors in Fig. 6.1. Also, a comparison of the errors in the Ref. (Ritschel and Alexander, 2014) and the present study is shown in the same graphs which indicates our fit is slightly better. These figures also indicate that representing a complicated spectral density with a function with simple poles is quite satisfactory.

Table 6.2. The magnitude of the parameters Ω_{k_j} , γ_{k_j} , and p_k to fit the vibrational, log-normal and the combined spectral density.

	Vibration modes	Log-normal modes	Combined
n	5	5	5
p_1	5109.12	168.73×10^3	192.82×10^3
p_2			1208.1
ω_{1_1}	$183.03 + i9.18$	$23.58 + i35.49$	$6.82 + i10.84$
ω_{1_2}	$151.20 + i167.25$	$66.22 + i106.93$	$58.71 + i98.60$
ω_{1_3}	$41.23 + i22.29$	$9.65 + i9.91$	$17.57 + i34.33$
ω_{2_1}			$236.06 + i130.87$
ω_{2_2}			$183.04 + i9.18$
ω_{2_3}			$170.70 + i37.23$

Now, we use the definition of $G_a(\tau)$ and $G_s(\tau)$, Eq. (6.33) and Eq. (6.34), with $J_{emp}(\omega)$ of Eq. (6.83) to calculate the bath spectral functions as:

$$G_a(t) = \sum_{k=1}^2 \sum_{j=1}^3 \left(c_{k_j}^+ e^{i\omega_{k_j} t} - c_{k_j}^- e^{-i\omega_{k_j}^* t} \right) \quad (6.86)$$

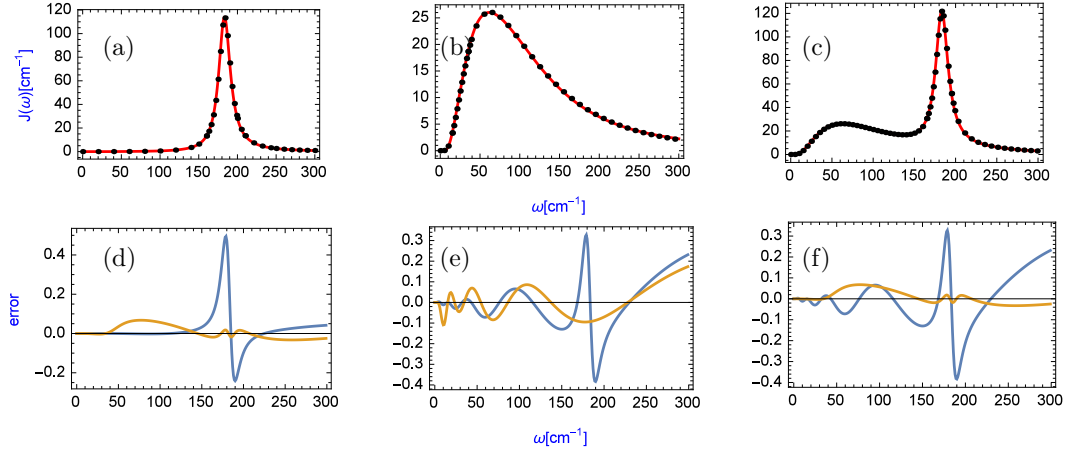


Figure 6.1. The behavior of vibrational (left column), log-normal (middle column) and the sum of those spectral densities (right column) as a function of frequency ω under the optimal parameter values. Black dots show the data that is obtained from the analytical expressions Eq. (6.84) and Eq. (6.85), while the red curve refers to the fit function in Eq. (6.83) with $\Omega_{k_j}, \gamma_{k_j}, p_k$ given in Table 6.2. Second row represents the fit errors and in Ref. (Ritschel and Alexander, 2014)'s parameters (blue) and the ones we obtained (yellow).

where

$$c_{k_j}^+ = -\frac{p_k \omega_{k_j}^2}{4 \gamma_{k_j}} \prod_{m=1, j \neq m}^3 \frac{1}{(\omega_{k_j} - \omega_{k_m})(\omega_{k_j} - \omega_{k_m}^*)} \quad (6.87)$$

$$c_{k_j}^- = c_{k_j}^{+*}. \quad (6.88)$$

We should note that $G_s(t)$ contains two different class of poles; ones due to the poles of $J_k(\omega)$ as well as the contributions coming from the poles of $\coth[\beta \omega/2]$ factor in the integral. These simple pole contributions can be summed to obtain:

$$G_s(t) = \sum_{k=1}^2 \sum_{j=1}^3 \left(d_{k_j}^+ (e^{i\omega_{k_j} t} - 1) - d_{k_j}^- (e^{-i\omega_{k_j}^* t} - 1) \right), \\ + i \sum_{k=1}^2 \frac{\pi^2 \beta^3 p_k}{2} (df - df^*), \quad (6.89)$$

where

$$d_{k_j}^+ = -\frac{p_k \omega_{k_j}^2}{4 \gamma_{k_j}} \coth[\beta \omega_{k_j}] \prod_{m=1, j \neq m}^3 \frac{1}{(\omega_{k_j} - \omega_{k_m})(\omega_{k_j} - \omega_{k_m}^*)} \quad (6.90)$$

$$df = \prod_{j=1}^3 \frac{1}{(l\pi - i\beta \omega_{k_j})(l\pi - i\beta \omega_{k_j}^*)}. \quad (6.91)$$

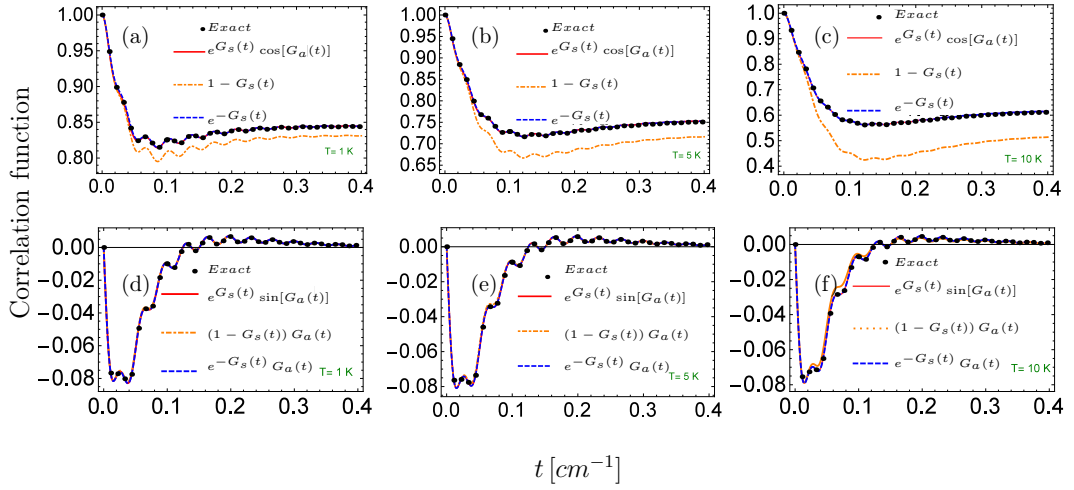


Figure 6.2. The time-dependent environmental correlation functions (CF) $\gamma_0(t)$ and $\chi_0(t)$ at different temperatures (time is in units of cm) for the combined spectral densities of Eq.(6.85) and Eq.(6.84). Red curve is the expression obtained from residue theorem, while blue and orange ones refer the Taylor approximation of these CF based on the result of Eq. (6.86)-Eq. (6.89). Black dotted is the exact expression of CF.

In Fig. 6.2 we display a comparison of the analytical form of the kernel functions $\gamma_0(t)$ and $\chi_0(t)$ with their numerical form as well as approximating those with $e^{-G_s(t)}$, $1 - G_s(t)$ instead of using the full form. Also, the second row plots in Fig. 6.2 indicate that $\chi_0(t) = e^{-G_s(t)} \sin(G_a(t))$ can be approximated as $e^{-G_s(t)} G_a(t)$ as well as $(1 - G_s(t)) G_a(t)$ at low temperatures. The dots in those plots show the exact $\gamma_0(t)$ and $\chi_0(t)$ calculated by numeric integration of $G_a(t)$ and $G_s(t)$, while thick lines represent the values obtained from Eq. (6.86) and Eq. (6.89). Overall, the agreement between those two is excellent both for $\gamma_0(t)$ and $\chi_0(t)$. It is also possible to deduce that at low temperatures $e^{-G_s(t)} \cos(G_a(t))$ can be approximated as $e^{-G_s(t)}$ and to some degree as $1 - G_s(t)$.

6.2.5.3 Structured Spectral Density

As the third example of spectral densities, we will consider so called structured spectral density which was used as the prominent vibrational mode of the previous section. We will provide details of the derivation of exact bath correlation functions for such spectral density which is new for the literature.

We are trying to get an analytical expression for the two-time integrated force auto-correlation function for a thermal environment whose spectral density is defined as

$$J(\omega) = 8 \frac{\kappa^2}{\hbar^2} \frac{\gamma \omega_0 \omega}{(\omega^2 - \omega_0^2)^2 + 4\gamma^2 \omega^2}, \quad (6.92)$$

where κ is the magnitude of the coupling between the two-level system and the oscillator, ω_0 is the characteristic frequency of the oscillator, while γ represents the broadening of the levels of the oscillator due to its surrounding Ohmic bath.

The two-time integrated correlation function is $G(t)$:

$$G(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \langle F(t_2)F(0) \rangle_T + i \frac{E_r t}{\hbar}, \quad (6.93)$$

where

$$\langle F(t_2)F(0) \rangle_T = \frac{1}{2\pi} \int_0^\infty J(\omega) \frac{\cosh(\hbar\omega/2k_B T - i\omega t)}{\sinh(\hbar\omega/2k_B T)} d\omega, \quad (6.94)$$

and

$$E_r = \frac{\hbar}{2\pi} \int_0^\infty \frac{J(\omega)}{\omega} d\omega. \quad (6.95)$$

The force auto-correlation can be written as:

$$\langle F(t_2)F(0) \rangle_T = \frac{1}{2\pi} \int_0^\infty J(\omega) (\coth(\hbar\omega/2k_B T) \cos(\omega t) - i \sin(\omega t)) d\omega. \quad (6.96)$$

which can be divided into two parts containing $\cos(\omega t)$ and $\sin(\omega t)$ terms as

$$C(t_2) = \frac{1}{2\pi} \int_0^\infty J(\omega) \coth(\beta\omega/2) \cos(\omega t) d\omega, \quad (6.97)$$

$$S(t_2) = \frac{1}{2\pi} \int_0^\infty J(\omega) \sin(\omega t) d\omega, \quad (6.98)$$

which are Fourier cosine and sine transforms of $J(\omega) \coth(\beta\omega/2)$ and $J(\omega)$, respectively. Difficulty of the problem is due to the cosine transform of the term that contains hyperbolic cotangent term. So, we write the $\coth(x)$ term as series expansion

$$\coth(\beta\omega/2) = \frac{2}{\beta\omega} + \frac{4}{\beta} \sum_{m=1}^\infty \frac{\omega}{\omega^2 + 4m^2\pi^2/\beta^2}. \quad (6.99)$$

So, the symmetric part of Eq. 6.93, $G_s(t)$ can be written as:

$$G_s(t) = \frac{1}{2\pi} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^\infty d\omega J(\omega) \left\{ \frac{2}{\beta\omega} + \frac{4}{\beta} \sum_{m=1}^\infty \frac{\omega}{\omega^2 + 4m^2\pi^2/\beta^2} \right\} \cos(\omega t).$$

Fourier cosine transform and the sum can be carried out exactly. The result of the triple

integral for the $2/\beta\omega$ term is:

$$G_{s,1}(t) = 2 \frac{\kappa^2 [\Delta^2 - 3\gamma^2]}{\beta\omega_0^5} - \frac{\kappa^2}{\beta\Delta\omega_0^5} e^{-\gamma t} \left[(\Delta - i\gamma)^3 e^{i\Delta t} + (\Delta + i\gamma)^3 e^{-i\Delta t} \right] + 4 \frac{\gamma\kappa^2}{\beta\omega_0^3} t,$$

where $\Delta = \sqrt{\omega_0^2 - \gamma^2}$. The triple integral of the sum term has four different types of sums of the form:

$$T_1 = \sum_{n=1}^{\infty} \frac{1}{n^4 + a^2 n^2 + b^4}, \quad (6.100)$$

$$T_2 = \sum_{n=1}^{\infty} \frac{n^2}{n^4 + a^2 n^2 + b^4}, \quad (6.101)$$

$$T_3 = \sum_{n=1}^{\infty} \frac{1}{n(n^4 + a^2 n^2 + b^4)}, \quad (6.102)$$

$$T_4 = \sum_{n=1}^{\infty} \frac{\exp(-2\pi n t / \beta)}{n(n^4 + a^2 n^2 + b^4)}, \quad (6.103)$$

where $a^2 = \beta^2 (\Delta^2 - \gamma^2) / (2\pi)$ and $b = \beta\omega_0 / (2\pi)$.

The coefficient of n^2 term is:

$$c_2(t) = -\frac{\beta\kappa^2}{2\pi^2\Delta\omega_0} e^{-\gamma t} \left\{ (\Delta + i\gamma) e^{-i\Delta t} + (\Delta - i\gamma) e^{i\Delta t} - 2\Delta \right\}. \quad (6.104)$$

The sum in Eq. 6.101 is:

$$T_2 = \frac{\pi^2}{2\beta\gamma\Delta} \frac{\Delta \sin(\beta\gamma) - \gamma \sinh(\beta\Delta)}{\cos(\beta\gamma) - \cosh(\beta\Delta)}. \quad (6.105)$$

The contribution of this term is:

$$G_{s,2}(t) = -\frac{1}{4\Delta^2\gamma\omega_0} \frac{\kappa^2 \Delta \sin(\beta\gamma) - \gamma \sinh(\beta\Delta)}{\cos(\beta\gamma) - \cosh(\beta\Delta)} \left[e^{-\gamma t} \left\{ (\Delta + i\gamma) e^{-i\Delta t} + (\Delta - i\gamma) e^{i\Delta t} \right\} - 2\Delta \right]. \quad (6.106)$$

The coefficient of n^0 term is:

$$c_1(t) = \frac{\kappa^2\beta^3}{8\pi^4\Delta\omega_0} \left[2\Delta(\Delta^2 - 3\gamma^2) - e^{-\gamma t} \left\{ (\Delta + i\gamma)^3 e^{-i\Delta t} + (\Delta - i\gamma)^3 e^{i\Delta t} \right\} \right]. \quad (6.107)$$

The sum in Eq. 6.100 is:

$$T_1 = -\frac{2\pi^4}{\beta^4\gamma\Delta\omega_0^4} \left(4\gamma\Delta + \beta\omega_0^2 \frac{\Delta \sin(\beta\gamma) + \gamma \sinh(\beta\Delta)}{\cos(\beta\gamma) - \cosh(\beta\Delta)} \right). \quad (6.108)$$

So, the contribution to $G_s(t)$ is:

$$G_{s,3}(t) = \frac{1}{4\beta\gamma\Delta^2\omega_0^4} \frac{\kappa^2}{\omega_0} \left(4\gamma\Delta + \beta\omega_0^2 \frac{\Delta \sin(\beta\gamma) + \gamma \sinh(\beta\Delta)}{\cos(\beta\gamma) - \cosh(\beta\Delta)} \right) \times \\ \left[e^{-\gamma t} \left\{ (\Delta + i\gamma)^3 e^{-i\Delta t} + (\Delta - i\gamma)^3 e^{i\Delta t} \right\} - 2\Delta(\Delta^2 - 3\gamma^2) \right]. \quad (6.109)$$

The coefficient of n^{-1} term is:

$$c_{3-4}(t) = \frac{\beta^4}{4\pi^5} \gamma \kappa^2 \omega_0. \quad (6.110)$$

Similarly, T_3 and T_4 can be expressed as:

$$T_3 = \frac{2i\pi^4}{\beta^4\gamma\Delta\omega_0^4} \left\{ (\gamma - i\Delta)^2 \left[H\left(-\frac{\beta}{2\pi}(\gamma + i\Delta)\right) + H\left(\frac{\beta}{2\pi}(\gamma + i\Delta)\right) \right] \right. \\ \left. - (\gamma + i\Delta)^2 \left[H\left(-\frac{\beta}{2\pi}(\gamma - i\Delta)\right) + H\left(\frac{\beta}{2\pi}(\gamma - i\Delta)\right) \right] \right\},$$

and

$$T_4 = \frac{2i\pi^4}{\beta^4\gamma\Delta\omega_0^4} \left\{ (\gamma + i\Delta)^2 \left[e^{-(\gamma - i\Delta)t} \mathcal{B}\left(e^{-\frac{2\pi t}{\beta}}, 1 - \frac{\beta}{2\pi}(\gamma - i\Delta), 0\right) \right. \right. \\ \left. \left. + e^{(\gamma - i\Delta)t} \mathcal{B}\left(e^{-\frac{2\pi t}{\beta}}, 1 + \frac{\beta}{2\pi}(\gamma - i\Delta), 0\right) \right] \right. \\ \left. - (\gamma - i\Delta)^2 \left[e^{-(\gamma + i\Delta)t} \mathcal{B}\left(e^{-\frac{2\pi t}{\beta}}, 1 - \frac{\beta}{2\pi}(\gamma + i\Delta), 0\right) \right. \right. \\ \left. \left. + e^{(\gamma + i\Delta)t} \mathcal{B}\left(e^{-\frac{2\pi t}{\beta}}, 1 + \frac{\beta}{2\pi}(\gamma + i\Delta), 0\right) \right] \right\} \\ - \frac{16\pi^4}{\beta^4\omega_0^4} \log\left(1 - e^{-\frac{2\pi t}{\beta}}\right).$$

The anti-symmetric part of the correlation function, which is the Fourier sine is easy to calculate as:

$$S(t) = -i \frac{\kappa^2}{2\omega_0^3\Delta} e^{-\gamma t} \left((\gamma - i\Delta)^2 e^{-i\Delta t} - (\gamma + i\Delta)^2 e^{i\Delta t} \right) + 2 \frac{\kappa^2\gamma}{\omega_0^3} + \frac{\kappa^2}{\omega_0} t. \quad (6.111)$$

The sum of the first three terms of the symmetric part of the correlation functions

is:

$$G_{S3}(t) = -\frac{\kappa^2}{2\Delta\omega_0^3}e^{-\gamma t} \left\{ (\gamma - i\Delta)^2 \frac{\sinh(\beta\Delta) + i\sin(\beta\gamma)}{\cos(\beta\gamma) - \cosh(\beta\Delta)} e^{-i\Delta t} + \right. \\ \left. (\gamma + i\Delta)^2 \frac{\sinh(\beta\Delta) - i\sin(\beta\gamma)}{\cos(\beta\gamma) - \cosh(\beta\Delta)} e^{i\Delta t} \right\} \\ + \frac{4\kappa^2\gamma}{\beta\omega_0^3}t + \frac{\kappa^2}{\omega_0^3} \frac{2\gamma\Delta\sin(\beta\gamma) + (\gamma^2 - \Delta^2)\sinh(\beta\Delta)}{\cos(\beta\gamma) - \cosh(\beta\Delta)}.$$

We need kernel functions of the form $e^{-G_s(t)} \cos(G_a(t))$ and $e^{-G_s(t)} \sin(G_a(t))$ which can be written as sum $(e^{K_1(t)} + e^{K_2(t)})/2$ and difference $(e^{K_1(t)} - e^{K_2(t)})/2i$, respectively. $K_1(t)$ and $K_2(t)$ are defined as :

$$K_1(t) = \frac{E_r}{\Delta} \left[\frac{e^{-i\theta}}{1 - e^{-\beta z}} (e^{itz} - e^{-\beta z}) - \frac{e^{i\theta}}{1 - e^{\beta z^*}} (e^{-itz^*} - e^{\beta z^*}) \right] + 4 \frac{\gamma E_r}{\beta \omega_0^2} (i\beta - t),$$

and

$$K_2(t) = \frac{E_r}{\Delta} \left[\frac{e^{i\theta}}{1 - e^{-\beta z^*}} (e^{-itz^*} - 1) - \frac{e^{-i\theta}}{1 - e^{\beta z}} (e^{itz} - 1) \right] - 4 \frac{\gamma E_r}{\beta \omega_0^2} t, \quad (6.112)$$

where $z = \Delta + i\gamma$, $\theta = 2 \arctan(\gamma/\Delta)$ and $E_r = \kappa^2/\omega_0$ is the reorganization energy.

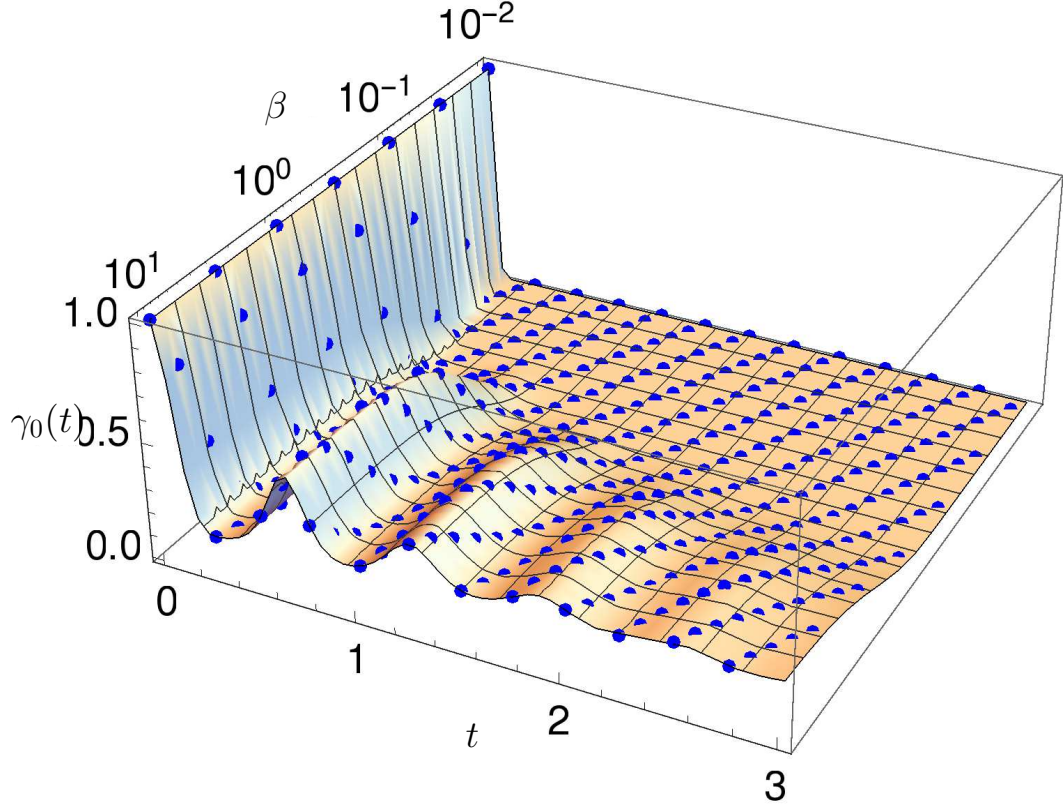


Figure 6.3. Comparison between analytical and numerical expression for $\gamma_0(t) = e^{-G_s(t)} \cos[G_a(t)]$

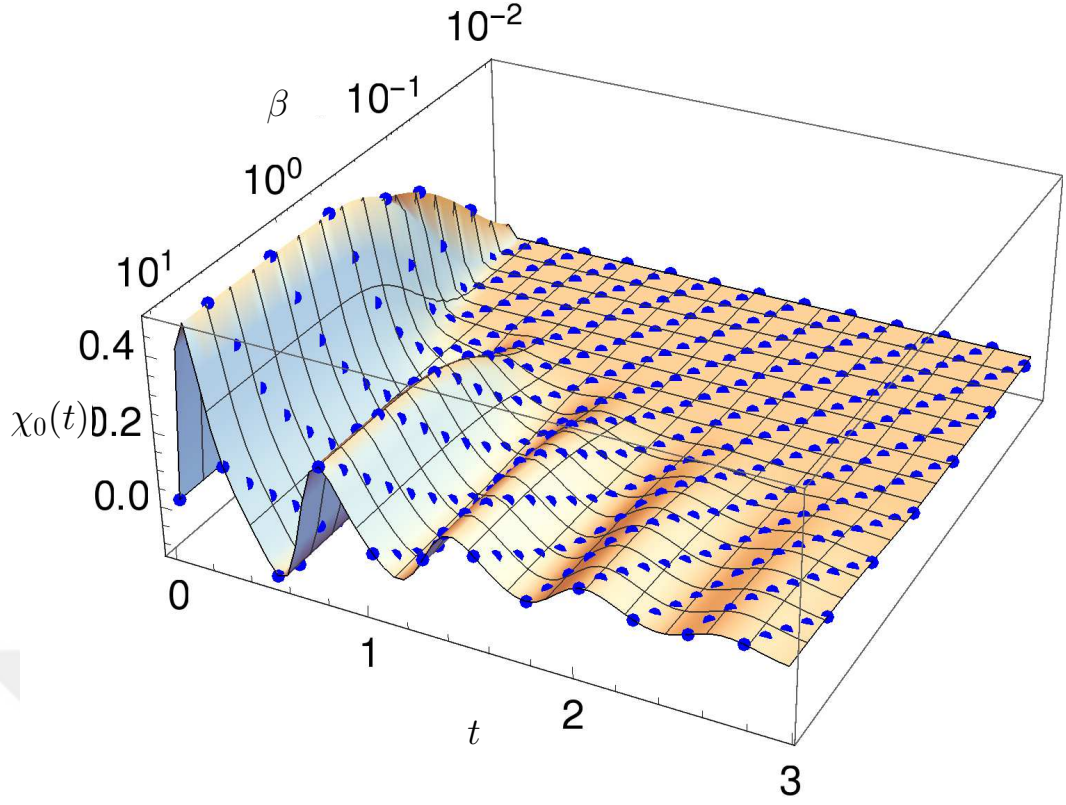


Figure 6.4. Comparison between analytical and numerical expression for $\chi_0(t) = e^{-G_s(t)} \sin[G_a(t)]$

In Figs. 6.3 and 6.4, we display the analytical and numerical kernel functions $\chi_0(t) = e^{-G_s(t)} \cos[G_a(t)]$ and $\chi_0(t) = e^{-G_s(t)} \sin[G_a(t)]$ for comparison purposes. The surface is the analytical expression while the dots are the results of the numerical calculations. As can be seen from the figures the agreement between the two is perfect. So, analytical results is exact.

6.3 Results

Our aim in this chapter was to investigate the effect of dichotomous noise on the population and coherence of a two-level system in a thermal bath. In the Results section, we will present and discuss our findings for a bath that can be described by a structured spectral density as outlined in sub-section 6.2.5.

The dichotomous noise we consider is characterized by two energy scales; its amplitude Ω and its inverse auto-correlation time ν which are used to define characteristic parameters of the noise, namely Kubo number $K = \Omega/\nu$ and the noise intensity $\eta = \Omega^2/\nu$. Noise with low Kubo number ($K \ll 1$) are called weakly colored while $K \gg 1$ indicates strongly colored noise. One compares the intensity η to the reorgani-

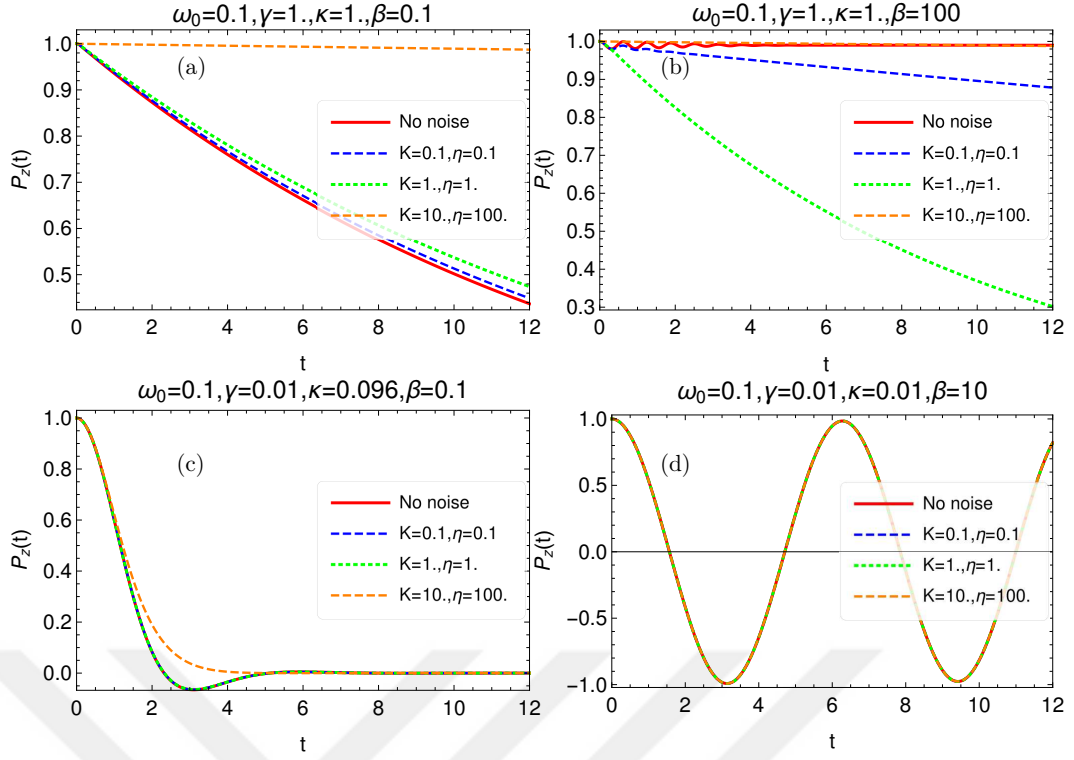


Figure 6.5. Population dynamics under different noise strength ($\eta = 100, \eta = 1, \eta = 0.1$) at $\omega_0 = 0.1$

zation energy (E_r) of the system to decide whether the noise intensity is low or high; if $\eta \gg E_r$, one talks of strong noise, while $\eta \ll E_r$ is weak noise.

For the problem we consider, the number of parameters that effect the dynamics is quite large; on the one hand, we have two time scales determined by the inverse of the TLS transition energy ε_0 and the tunneling splitting V . Environmental system, *i.e.*, the damped harmonic oscillator, has also two time scales proportional to the inverse of its central frequency ω_0 and damping γ . TLS-environment coupling constant κ contributes another time scale to the dynamics. When one adds two time scales associated with the noise (ν^{-1} and Ω^{-1}) to the time scales of the problem, the total number of the time parameters that influence the TLS dynamics becomes seven which makes disentangling the effect of each parameter quite difficult. The standard way to deal with such multi-parameter cases is to fix one of the time scales and express all the other times in terms of the fixed parameter which reduces the number of parameters by one. Here, we fix tunneling matrix element to $V = 1$. So, all parameters ($\omega_0, \gamma, \beta, \kappa, \varepsilon_0, \nu, \Omega$) are expressed as multiples of V . We also consider only the symmetric TLS, *i.e.*, TLS with no bias so $\varepsilon_0 = 0$. The choice of $\varepsilon_0 = 0$ is motivated by the fact that for the acceptor-donor system we have in mind, generally, $\varepsilon_0 = 0$ is a reasonable choice. Also, NIBA

like dynamics we are considering perform well under no-bias condition.

Having reduced the number of time constants of the system to five, we analyze the dynamics of the population difference and the coherence by solving the coupled integro-differential equations of the average $P_{x,y,z}(t)$ and their noise-multiplied correlators (Eqs. 6.57, 6.58, 6.54), respectively. After this point, we will drop the average sign form $\langle P_i(t) \rangle$ but $P_i(t)$ still refers to its noise-averaged form. In this section, we provide numerical solutions that are valid for all parameter ranges. The technique used to solve those equations is somewhat involved and we provide Mathematica functions used to find those solutions in Appendix A.

One of the most important factors that determines how the environment fluctuations would effect the TLS dynamics is the relation between the relaxation time of the bath which is proportional to ω_0^{-1} and characteristic time of the TLS (V^{-1} here). When the relaxation time of the bath is much faster than the time it takes for the system to change its state, the total system is said to be in non-adiabatic regime and the bath dissipates the disturbance from the TLS in a short time. In the opposite limit of slow bath-fast TLS one has the adiabatic dynamics which indicates that TLS moving in the static disorder of the environmental oscillators. We have considered there adiabaticity regimes in the present work, namely $\omega_0 = 10V$ (non-adiabatic regime), $\omega_0 = 0.1V$ (adiabatic regime).

For the noise parameters, we have also, considered three distinct cases: weakly colored low intensity noise ($K = 0.1, \eta = 0.1E_r$), intermediate ($K = 1, \eta = E_r$) and colored, strong intensity noise ($K = 10, \eta = 100E_r$).

The noisy population dynamics of the TLS are displayed in Fig. 6.5(a-d) for the adiabatic regime for the different combinations of the oscillator damping γ , TLS-oscillator coupling coefficient κ , and the inverse environmental temperature β . One can observe four distinct effect of the noise from these figures. Figure 6.5(a) shows $P_z(t)$ in adiabatic regime with strong damping for oscillator ($\gamma > \omega_0$), strong TLS-oscillator coupling ($\kappa = 1$) and relatively high temperature. Because of the high-temperature and the strong coupling one would expect the TLS without noise to display an incoherent damping type dynamics which is observed (full line). Under these conditions, increasing noise color and strength is seen to decrease transport coefficient and at the strong intensity limit, the tunneling is found to completely destroyed by the noise.

Figure 6.5(b) indicates another aspect of the effect of the noise. Here, the temperature is low and the non-noise dynamics are non-tunneling because of the strong

coupling, *i.e.* TLS very strongly coupled to the environment can not tunnel from donor side to the acceptor side. It can be seen that, low intensity noise can assist the TLS tunneling while dynamics under high intensity noise is again no tunneling which is same as the no-noise case.

Figure 6.5(c) and (d) are for the underdamped ($\gamma < \omega_0$) and weak coupling ($\kappa \ll \omega_0$) case and indicate that the effects of the noise in those cases are rather limited even for the strong intensity case. Figure 6.6(a-d) presents dynamics of $P_z(t)$ for the non-

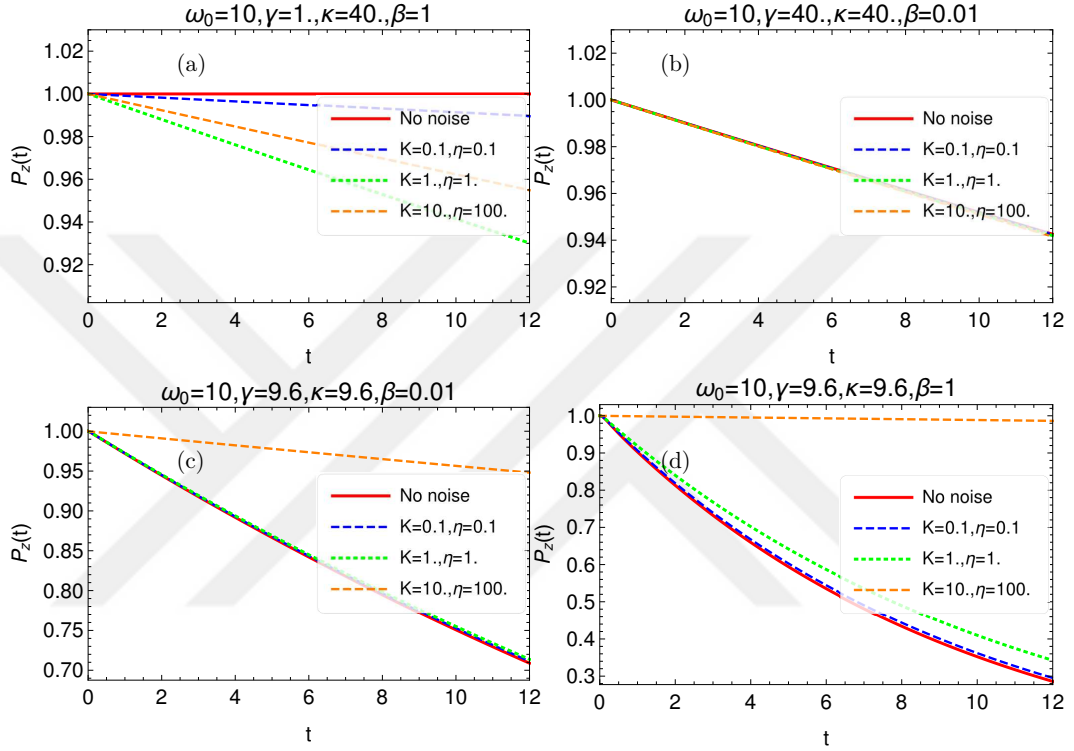


Figure 6.6. Population dynamics under different noise strength ($\eta = 100$, $\eta = 1$, $\eta = 0.1$) at $\omega_0 = 10$

adiabatic regime. For the under-damped, strong coupling case, the noise is found to change the state from the non-tunneling to incoherent damping dynamics which increases with the noise strength, *i.e.* noise helps the system tunnel from the donor to the acceptor site (Fig.6.6a) at low temperature. At high temperature ($\beta = 0.01$) one can observe two distinct cases depending on γ and κ ; at overdamped-strong coupling limit, even the highest noise strength ($\eta = 100E_r$) has no effect on the dynamics (Fig.6.6b) while for the intermediate damping and the coupling, high intensity noise tends to destroy the tunneling (Fig.6.6c). Figure 6.6(d) indicates that as the temperature is lowered, destruction of the tunneling by strong noise is complete (compare Fig. 6.6c and 6.6d). To distinguish the individual roles of γ , β , and κ in the effect of the noise, we display some further situations of $P_z(t)$ dynamics for the non-adiabatic regime in Figs.6.7(a-d).

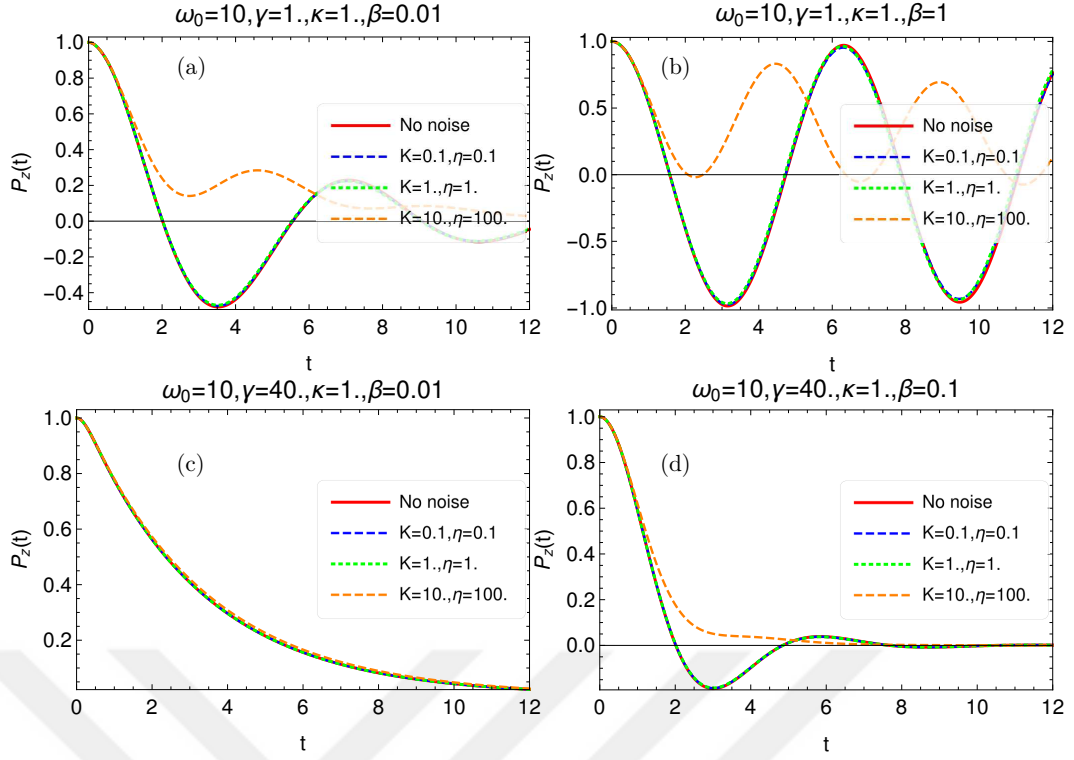


Figure 6.7. Population dynamics under different noise strength ($\eta = 100$, $\eta = 1$, $\eta = 0.1$) at $\omega_0 = 10$

The first row of Fig. 6.7 which displays $P_z(t)$ at underdamped-weak coupling regime for high and low temperatures suggest an interesting affect of noise; at low temperatures dynamics are found to be oscillatory, similar to the non-noise case, but while the TLS states oscillates between the acceptor and donor sites completely, noisy case the minimum of $P_z(t)$ is zero which indicates that the site populations are at most equal, *i.e.* the tunneling is incomplete. The last row of Fig. 6.7 shows that the effect of noise in overdamped-weak coupling case is rather weak.

To delineate the effects of dichotomous noise on the coherence of the TLS, we solve Eqs. (6.57)-(6.60) for initial state $P_x(0) = 1$, $\alpha(0)P_x(0) = \alpha(0)P_y(0) = P_y(0) = 0$ and discuss the results below. One should note that, the dynamical equations are in the Shrödinger picture but in polaron frame. It can be shown that $P_z(t)$ in polaron frame is the same as the σ_z -frame, but transforming the coherences from the polaron frame to the measurement base is difficult (Nazir and McCutcheon, 2016) and we present the polaron frame coherences here.

Since we are considering the TLS with zero bias $\varepsilon_0 = 0$, $P_x(t)$ does not change with time when there is no noise as can be seen from the Figs. (6.8-6.10). It is obvious from Figs. (6.8-6.10) that the destructive role of the noise is much more prominent on

the coherence compared to that on the population; except for the adiabatic case with underdamped oscillator and weak-coupling case, noise destroys the coherence.

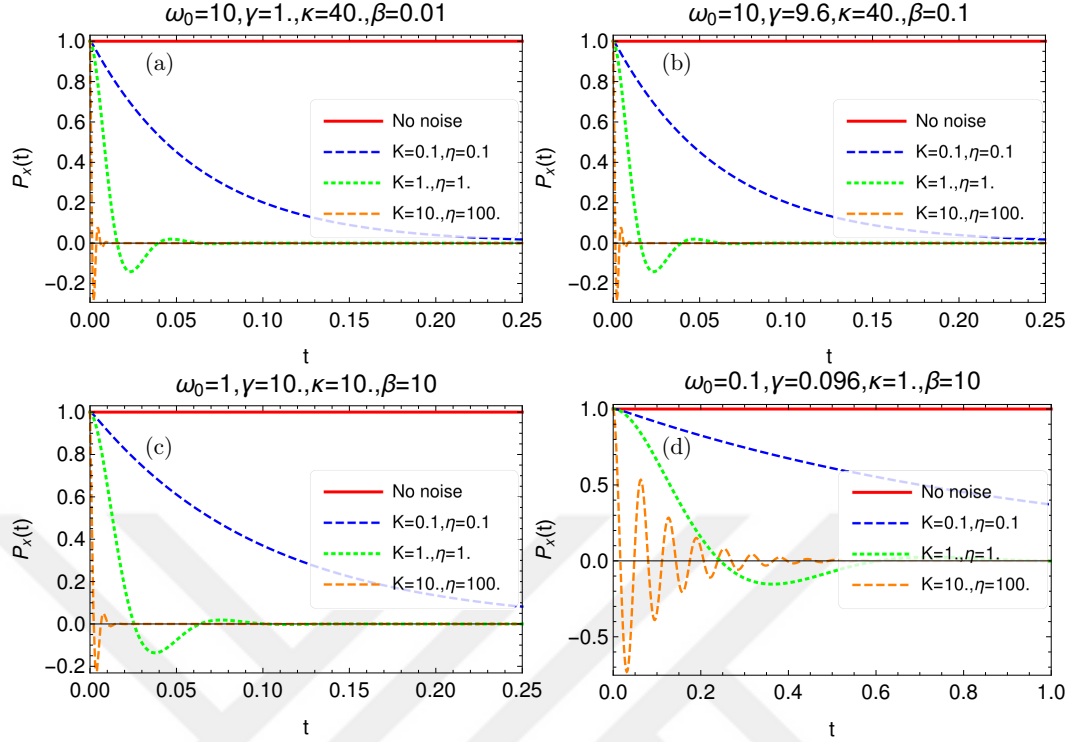


Figure 6.8. Dynamics of coherence process in case of strong coupling ($\kappa > \omega_0$)

In the strong TLS-oscillator coupling case, low intensity noise leads to an exponential decay of the quantumness while increasing the noise intensity leads to a coherent decay type dynamics for high intensity noise (Figs. 6.8 a-d). For the intermediate coupling case, one can see that for the adiabatic regime, low and intermediate strength noise's effect on $P_x(t)$ is small (Fig. 6.9a) while for the non-adiabatic region, the quantumness of the TLS state is washed out quite rapidly by even the low intensity noise (Figs. 6.9 b-d). One can observe that oscillatory decay of $P_x(t)$ for the high intensity noise when the characteristic time of oscillator and the TLS are in resonance ($\omega_0 = V$) that can be seen from Figs. 6.9(c)-(d).

We present the dynamics of $P_x(t)$ in Figs. 6.10(a)-(d) for the weak TLS-oscillator coupling case. For the adiabatic case, independent of the bath temperature and the oscillator damping it is found that the noise has no effect on the quantumness of the TLS (Figs. 6.10a-b).

As the bath response time increases compared to the TLS tunneling rate, one observes the destructive role of the noise on the coherence of the TLS (Figs. 6.10c-d)

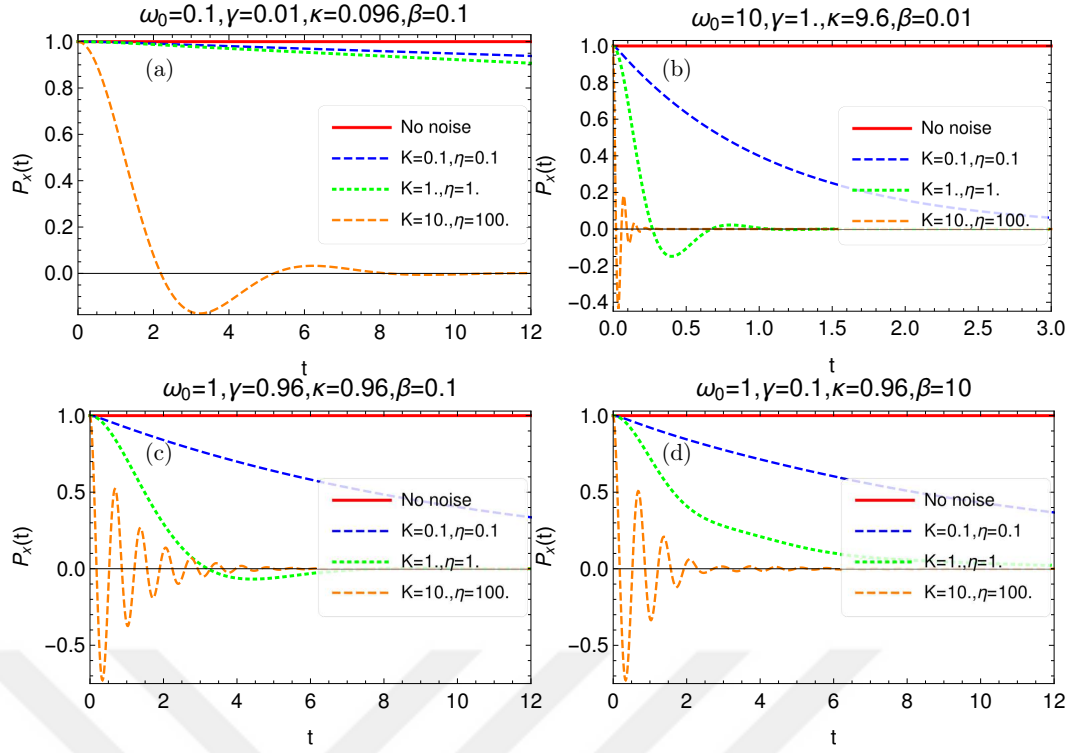


Figure 6.9. Dynamics of coherence process in case of intermediate coupling ($\kappa \approx \omega_0$)

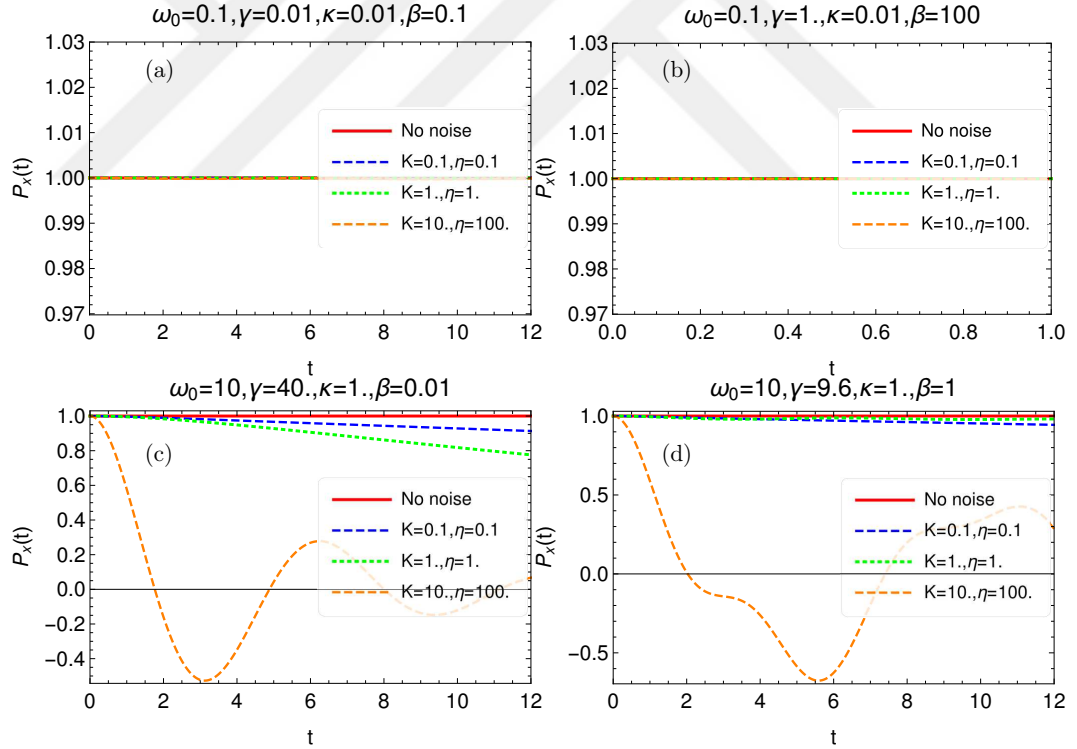


Figure 6.10. Dynamics of coherence process in case of weak coupling ($\kappa < \omega_0$)

6.4 Conclusions

In this part of the PhD research project, we have considered the effect of dichotomous noise on the dynamics of a two-level system which is in contact with a thermal

bath of harmonic oscillators. The noise drives the transition energy of the TLS. We have approached the problem by using polaron transform method and obtained a non-Markovian master equation for the density matrix of the TLS. The noise averaging of the dynamical equations for the population difference and the coherence of the TLS were performed and their Laplace transform forms were obtained. We have analyzed a number of different type of thermal baths with power, semi-empirical and structured spectral densities. The effect of the noise for the structured spectral density in the adiabatic, resonance and non-adiabatic regimes were characterized for a large range of TLS-harmonic oscillator coupling strength, oscillator damping constant and the temperature of the bath. The general findings of the study can be summarized as follows: in absence of the dichotomous noise there are four different dynamics on the population difference of the TLS such as no-tunneling, incoherent, coherent, and oscillating dynamics. For the non-adiabatic case, the dynamics without noise prevents to the tunneling in the case of strong coupling with intermediate and low temperatures at all damping constants which can be interpreted that TLS very strongly coupled to the environment can not tunnel from donor side to the acceptor side, while in the same conditions the charge transfer begins in a state of the two-level system with increasing noise intensity and decreases after a certain threshold of the noise, *i.e.* the noise depending on its strength plays an constructive role on the population difference of the TLS. On the other hand, under the high-temperature and the strong coupling one obtains that increasing color and strength of the noise decrease transport coefficient and the tunneling completely destroyed by the noise at the strong intensity limit.

We have, also, examined how the noise effects the dynamics of the coherence of the TLS and we have seen that in case of intermediate coupling, low and intermediate strength noise's influence on the coherence of the TLS is weak for the adiabatic regime while for the non-adiabatic region, the quantumness of the TLS state is destroyed quite rapidly by even the low intensity noise. Moreover, one can see that the noise has no effect on the quantumness of the TLS in adiabatic region and regardless of the bath temperature and the oscillator damping.

7. CONCLUSIONS AND OUTLOOK

In chapter 3, we have examined the dynamics of a two qubit system driven by a periodically external field and obtained an exact analytical solution of probability amplitudes of the two qubits system by means of Bellomo's method in case of zero coupling constant. However, if there is an interaction between these qubits, the third order ordinary differential equations for each amplitudes are found that needs to be solved. As a second work for the problem is obtained the exact approximation of dynamics of a periodically driven single qubit by using Magnus expansion method whose usage is shown to be fairly simple in comparison with the study of Xie and Hai Xie and Hai (2010). In addition, we have evaluated the strength of relative error between them as a function of amplitude of driving field f_1 , energy gap f_0 , and interaction term v .

In chapter 4, we have investigated the exact expression of the third- and the fourth-order contributions to the master equation of a charged harmonic oscillator coupled to a black-body radiation field by using Krylov averaging method. The interaction between the oscillator and bath is considered to be in velocity-coupling form and includes diamagnetic interaction term, which is mostly ignored. We have obtained that regarding higher than the second-order terms would not lead any new commutators in structure of the master equation. We have found that normal and anomalous diffusion coefficients for the harmonic oscillator in the thermal bath depend on temperature of the bath, while mass renormalization and decay constant change with the oscillator and the environment cutoff frequency. In addition, one can say that the third- and the fourth-order contributions cancel each other in case of weak coupling regime that including only the second-order contribution to the master equation would be appropriate enough. However, a perturbative approximation would not be accurate as the order of consecutive terms have comparable magnitudes for $\Omega \tau_e > 0.1$.

In chapter 5, we have studied the bath correlation function $G(t)$ for the spin-boson model in a thermal bath with a peaked spectral density. For the temperature of the bath, central frequency of the bath oscillator, system-bath coupling constant and the oscilla-

for damping constant the $G(t)$ is obtained as an exact analytical expression. Population dynamics $P(t)$ of a two-level system we have examined under four different approximations, such as F3b, F3, short time, and Markovian, to $G(t)$ in adiabatic as well as non-adiabatic cases. According to this dynamics, the short-time approximation can be used for a wide range of system parameters. Besides, we have obtained that there is an exact treatment for $P(t)$ under Markovian conditions that is valid for the strong TSS-harmonic oscillator interaction.

In chapter 6, we have investigated the effect of a dichotomous noise on the dynamics of population difference and coherence for a two-level system in a thermal bath as different system parameters by using the polaron transformation and derived average treatments of integro-differential equations for both population and coherence process by means of Laplace transform method. We have observed that for the dynamics without noise in the case of low temperature strong coupling there is no charge transfer from one state to other state of the TLS, while increasing noise intensity increases transport coefficient which is an interesting result for this problem. On the other hand, we have found kernel functions entering these integro-differential equations by applying the residue theorem.

As first outlook, in third problem using variational polaron transformation in steady of polaron transform leads to more accurate way to examine dynamics of master equation for any problem at not only strong coupling but also weak and intermediate coupling regimes. Secondly, we analyzed the effect of fourth-order contribution to the master equation for an oscillator in the electromagnetic field and shown that the third and fourth order corrections have to be taken into account when successive terms' order is comparable together, thus a second order approximation does not work for a such system.

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APPENDICES

9. APPENDICES

9.1 Appendix A

Here we provide the Mathematica code for the solution of the integro-differential equations that arise in the dichotomously driven two level system in contact with a thermal bath.

```
spectral[w_, w0_, g_, k_] := 8 k^2 g w0 w / ((w^2 - w0^2)^2 + 4 g^2 w^2);

numericGs[t_?NumericQ, w0_, g_, k_, beta_] :=
  1/(2 Pi) NIntegrate[
    spectral[w, w0, g, k] Coth[beta w/2]*(1 - Cos[w t])/w^2,
    {w, 0, Infinity}, MaxRecursion -> 25];

numericGa[t_?NumericQ, w0_, g_, k_] :=
  1/(2 Pi) NIntegrate[
    spectral[w, w0, g, k] Sin[w t]/w^2,
    {w, 0, Infinity}, MaxRecursion -> 25];

numericKernel[t_?NumericQ, w0_, g_, k_, beta_, wga_:2] :=
  If[Abs[t] < 10^(-10), 1,
    If[wga == 1,
      Exp[-numericGs[t, w0, g, k, beta]] Cos[numericGa[t, w0, g, k]],
      Exp[-numericGs[t, w0, g, k, beta]] Cos[ga[t, w0, g, k]]
    ]
  ];

anaKernelCos13[t_, w0_, g_, k_, beta_, ith_:1] :=
  Module[{de, Er, a, b, c, tt},
    de = Sqrt[w0^2 - g^2]; Er = k^2/w0;
    {a, b} = k^2/(de w0^3) {- (de - I g)^2/(1 - Exp[beta (de + I g)]),
      (de + I g)^2/(1 - Exp[-beta (de - I g)])};
    c = 4 Er g/(beta w0^2);
    a = If[ith == 1, 0, a];
    tt = -(a + b) + a Exp[I t (de + I g)] + b Exp[-I t (de - I g)] - c t;
    (Exp[tt] + Exp[Conjugate[tt]])/2
  ];

ga[t_, w0_, g_, k_] :=
  Module[{de, ged, gad},
    de = Sqrt[w0^2 - g^2];
    ged = g - I de; gad = g + I de;
    -I k^2/(2 w0^3 de)
    Exp[-t g] (ged^2 Exp[-I de t] - gad^2 Exp[I de t])
    + 2 k^2 g/w0^3
  ];

anaKernelCos[t_, w0_, g_, k_, beta_, ith_:1] :=
  Module[{ged, gad, de, Er, term, harmonicTerm, betaTerm, tot,
    a, b, c, ipiob, gaTerm},
    de = Sqrt[w0^2 - g^2]; Er = k^2/w0;
    {a, b} = k^2/(de w0^3) {- (de - I g)^2/(1 - Exp[beta (de + I g)]),
      (de + I g)^2/(1 - Exp[-beta (de - I g)])};
    c = 4 Er g/(beta w0^2);
    term[1] = -(a + b) + a Exp[I t (de + I g)]
```

```

+b Exp[-I t (de-I g)]-c t;
ipioB=2 Pi/beta;
term[2]=-4 Er g/(Pi w0^2)Log[1-Exp[-ipioB t]];
ged=g-I de; gad=g+I de;
harmonicTerm:=- (gad)^2 (HarmonicNumber[-ged/ipioB]
+HarmonicNumber[gad/ipioB])+
(ged)^2 (HarmonicNumber[-gad/ipioB]
+HarmonicNumber[gad/ipioB]);
term[3]:=-I k^2/(2 Pi de w0^3)(harmonicTerm);
betaTerm:=gad^2(Exp[-t ged]
Beta[Exp[-ipioB t],1-ged/ipioB,0]+
Exp[t ged] Beta[Exp[-ipioB t],1+ged/ipioB,0])-
ged^2(Exp[-t gad]Beta[Exp[-ipioB t],1-gad/ipioB,0]+
Exp[t gad]Beta[Exp[-ipioB t],1+gad/ipioB,0]);
term[4]:=I k^2/(2 Pi de w0^3) betaTerm;
gaTerm:=ga[t,w0,g,k];
tot=Which[ith<=4,Sum[term[i],{i,1,ith}],ith==5,
Sum[term[i],{i,2,4}]];
If[Abs[t]<10^(-12),1,Which[ith<=4,
(Exp[tot]+Exp[Conjugate[tot]])/2,ith==5,
Exp[tot]Cos[gaTerm]]];
];

kernelFunction[t_,w0_,g_,k_,beta_,wh_]:=
Module[{d2,int1,int2},
Which[wh<=5,anaKernelCos[t,w0,g,k,beta,wh],wh==6,
d2=Sqrt[w0^2-g^2]; int1=k^2/(beta w0);
int2=k^2/(Pi d2) w0
Im[PolyGamma[0,1+beta/(2 Pi) (g+I d2)]];
Exp[-(int1+int2) t^2] Cos[k^2 t/w0],
wh==7,
Exp[-k^2 t^2/(beta w0)] Cos[k^2 t/w0],
wh==8,
anaKernelCos13[t,w0,g,k,beta,1]]
];

solvePopulation[w0_,g_,k_,b_,V_,dt_,tend_,wh_]:=
Module[{M,ec,sig,K,Kk,integralsig,eq,kernel},
M=Round[tend/dt];
ec[t_]:=If[Abs[t]<10^(-10),1,
Re[kernelFunction[t,w0,g,k,b,wh]]];
sig[0]=1; kernel[1][n_]:=4 V^2 ec[n dt];
Kk[1]=Table[kernel[1][n],{n,0,M}];
K[m_][n_]:=Kk[m][[n+1]];
integralsig[m_]:=Module[{dataSig},dataSig=Table[
sig[i] K[1][m-i]+sig[i+1] K[1][m-i-1],{i,0,m-1}];
Sum[dataSig[[i]],{i,1,Length[dataSig]}]
];
eq[n_]:={sig[n]==sig[n-1]-dt^2/2 integralsig[n]};
Do[sig[i]=sig[i]/. Solve[eq[i],sig[i]][[1]],{i,1,M}];
Table[{n dt,Re[sig[n]]},{n,0,M}];
];

anaKernelCos13Noise[t_,w0_,g_,k_,beta_,ith_:1]:=
Module[{de,Er,a,b,c,tt}, de=Sqrt[w0^2-g^2]; Er=k^2/w0;
{a,b}=k^2/(de w0^3) {- (de-I g)^2/(1-Exp[beta (de+I g)]),
(de+I g)^2/(1-Exp[-beta (de-I g)])};
c=4 Er g/(beta w0^2); a=If[ith==1,0,a];
tt=-(a+b)+a Exp[I t (de+I g)]+b Exp[-I t (de-I g)]-c t;
Exp[tt]

```

```

];

kernelFunctionNoise[t_, w0_, g_, k_, beta_, wh_] :=
Module[{d2, int1, int2},
Which[wh <= 5, anaKernelNoise[t, w0, g, k, beta, wh],
wh == 6,
d2 = Sqrt[w0^2 - g^2]; int1 = k^2 / (beta w0);
int2 = k^2 / (Pi d2) w0
Im[PolyGamma[0, 1 + beta / (2 Pi) (g + I d2)]];
Exp[-(int1 + int2) t^2 + I k^2 t / w0],
wh == 7,
Exp[-k^2 t^2 / (beta w0) + I k^2 t / w0],
wh == 8,
anaKernelCos13Noise[t, w0, g, k, beta]]];

anaKernelNoise[t_, w0_, g_, k_, beta_, ith_: 1] :=
Module[{ged, gad, de, Er, term, harmonicTerm,
betaTerm, tot, ipiob, a, b, c},
de = Sqrt[w0^2 - g^2]; Er = k^2 / w0;
{a, b} = k^2 / (de w0^3) {-(de - I g)^2 / (1 - Exp[beta (de + I g)]),
(de + I g)^2 / (1 - Exp[-beta (de - I g)])};
c = 4 Er g / (beta w0^2);
term[1] = -(a + b) + a Exp[I t (de + I g)]
+ b Exp[-I t (de - I g)] - c t;
ipiob = 2 Pi / beta;
term[2] = -4 Er g / (Pi w0^2) Log[1 - Exp[-ipiob t]];
ged = g - I de; gad = g + I de;
harmonicTerm = -(gad)^2 (HarmonicNumber[-ged / ipiob]
+ HarmonicNumber[ged / ipiob]) +
(ged)^2 (HarmonicNumber[-gad / ipiob]
+ HarmonicNumber[gad / ipiob]);
term[3] = -I k^2 / (2 Pi de w0^3) (harmonicTerm);
betaTerm = gad^2 (Exp[-t ged] *
Beta[Exp[-ipiob t], 1 - ged / ipiob, 0] + Exp[t ged] *
Beta[Exp[-ipiob t], 1 + ged / ipiob, 0]) -
ged^2 (Exp[-t gad] *
Beta[Exp[-ipiob t], 1 - gad / ipiob, 0] +
Exp[t gad] Beta[Exp[-ipiob t], 1 + gad / ipiob, 0]);
term[4] = I k^2 / (2 Pi de w0^3) betaTerm;
tot = Which[ith <= 4, Sum[term[i], {i, 1, ith}], ith == 5,
Sum[term[i], {i, 2, 4}]];
If[Abs[t] < 10^(-12), 1, Exp[tot]]
];

solvePopulationNoise[eps_, w0_, g_, k_, b_, nu_, delta_,
wh_: 4, dt_: 0.01, tend_: 3, V_: 1/2] :=
Module[{M, ec, sig, K, Kk, integSig, integSigA, eqn,
nuDel, S, es, kernel, aSig},
M = Round[tend / dt];
ec[t_] := If[Abs[t] < 10^(-10), 1,
Re[kernelFunctionNoise[t, w0, g, k, b, wh]]];
es[t_] := If[Abs[t] < 10^(-10), 0,
Im[kernelFunctionNoise[t, w0, g, k, b, wh]]];
nuDel = Sqrt[nu^2 - 4 delta^2];
S[0][t_] := If[Abs[nuDel] < 10^(-12),
Exp[-nu t / 2] (1 + nu t / 2),
Exp[-nu t / 2] (Cosh[t nuDel / 2]
+ nu Sinh[t nuDel / 2] / nuDel)];
S[1][t_] := If[Abs[nuDel] < 10^(-12),
-2 I delta t Exp[-nu t / 2] / 2,

```

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-2 I delta Exp[-nu t/2] Sinh[t nuDel/2]/nuDel];
S[2][t_]:=If[Abs[nuDel]<10^(-12),
  Exp[-nu t/2] (1-nu t/2),
  Exp[-nu t/2](Cosh[t nuDel/2]
  -nu Sinh[t nuDel/2]/nuDel)];
kernel[1][n_]:=4 V^2 ec[n dt]S[0][n dt] Cos[eps n dt];
kernel[2][n_]:=V^2 I S[1][n dt] ec[n dt] Sin[eps n dt];
kernel[3][n_]:=V^2 S[0][n dt] es[n dt] Sin[eps n dt];
kernel[4][n_]:=V^2 S[2][n dt] ec[n dt] Cos[eps n dt];
kernel[5][n_]:= V^2 I S[1][n dt] es[n dt] Cos[eps n dt];
sig[0]=1; aSig[0]=0;
Do[Kk[i]=Table[kernel[i][n],{n,0,M}],{i,1,5}];
K[m_][n_]:=Kk[m][[n+1]];
integSig[m_]:= Module[{datSig,datASig,datC},
  datSig=Table[sig[i] K[1][m-i]+sig[i+1]K[1][m-i-1],
    {i,0,m-1}];
  datASig=Table[aSig[i] K[2][m-i]+aSig[i+1] K[2][m-i-1],
    {i,0,m-1}];
  datC=Table[K[3][m-i]+K[3][m-i-1],{i,0,m-1}];
  Sum[datSig[[i]]+datASig[[i]]+datC[[i]],
    {i,1,Length[datSig]}];];
integSigA[m_]:= Module[{datSig,datASig,datC},
  datSig=Table[sig[i] K[2][m-i]+sig[i+1] K[2][m-i-1],
    {i,0,m-1}];
  datASig=Table[aSig[i] K[4][m-i]+aSig[i+1] K[4][m-i-1],
    {i,0,m-1}];
  datC=Table[K[5][m-i]+K[5][m-i-1],{i,0,m-1}];
  Sum[datSig[[i]]+datASig[[i]]+datC[[i]],{i,1,
    Length[datSig]}];];
eqn[n_]:= {sig[n]==sig[n-1]-dt^2/2 integSig[n],
  aSig[n]==aSig[n-1]-dt nu aSig[n]-dt^2/2 integSigA[n]};
Do[{sig[i],aSig[i]}= {sig[i],aSig[i]}/.
  Solve[eqn[i]{sig[i],aSig[i]}][[1]],{i,1,M}];
{Table[{n dt,sig[n]},{n,0,M}],
  Table[{n dt,aSig[n]},{n,0,M}]}];

kernelFunctionNoise[t_,w0_,g_,k_,beta_,wh_]:=
Module[{d2,int1,int2},
  Which[wh<=5,anaKernelNoise[t,w0,g,k,beta,wh],
    wh==6,
      d2=Sqrt[w0^2-g^2]; int1=k^2/(beta w0);
      int2=k^2/(Pi d2) w0
      Im[PolyGamma[0,1+beta/(2 Pi) (g+I d2)]];
      Exp[-(int1+int2) t^2+I k^2 t/w0],
    wh==7,
      Exp[-k^2 t^2/(beta w0)+I k^2 t/w0],
    wh==8,
      anaKernelCos13Noise[t,w0,g,k,beta]]];

findDt[w0_,g_,k_,beta_]:=
Module[{dt,tend,p,ip,diff,ic,nd,nk},
  ic=1; dt[ic]=0.1;tend[ic]=100.0;
  p[ic]=
  solvePopulation[w0,g,k,beta,1/2,dt[ic],tend[ic],4];
  ip[ic]=Interpolation[p[ic]];
  While[True,ic=ic+1; dt[ic]=dt[ic-1]/2;
    tend[ic]=tend[ic-1]/2;
  p[ic]=
  solvePopulation[w0,g,k,beta,1/2,dt[ic],tend[ic],4];

```

```

        ip[ic]=Interpolation[p[ic]];
nd=Norm[Table[ip[ic][t]-ip[ic-1][t],
{t,0,tend[ic],tend[ic]/1000}]];
nk=Norm[Table[ip[ic][t],
{t,0,tend[ic],tend[ic]/1000}]];
diff=nd/nk; If[diff<0.01||ic>30,Break[]];
{p[ic-1],2 dt[ic]}
];

populationAnalyze[w0_,g_,k_,b_]:=
Module[{convergentQ,ix,tend,dt,zero,p,pref,prefNorm},
convergentQ[w0x_,gx_,kx_,bx_,tendx_,whx_]:=
Module[{tmp},
tmp=Table[Chop[kernelFunction[t,w0x,gx,kx,bx,whx]],
{t,0,tendx,tendx/100}];
Max[Abs[tmp]]<1.1];
ix={1,6,7,8};{pref,dt}=findDt[w0,g,k,b];
prefNorm=Norm[pref[[All,2]]];tend=1000.0 dt;
calc=Table[convergentQ[w0,g,k,b,tend,ix[[i]]],
{i,1,Length[ix]}];
zero=Table[{t,0},{t,0,tend,dt}];
p=Table[If[calc[[i]],Re[Chop[solvePopulation[w0,g,k,
b,1/2,dt,tend,ix[[i]]]]],zero],
{i,1,Length[calc]}];
p=Table[If[VectorQ[Flatten[p[[i]]],NumericQ],
p[[i]],zero,zero],{i,1,Length[p]}];
diffs=Table[If[calc[[i]],Norm[pref[[All,2]]
-p[[i,All,2]]/prefNorm,10],{i,1,Length[calc]}];
<|"w0"->w0,"g"->g,"k"->k,"b"->b,
"dt"->dt,"df1"->diffs[[1]],
"df6"->diffs[[2]],"df7"->diffs[[3]],
"df8"->diffs[[4]],"solRef"->pref,
"sol1"->p[[1]],"sol6"->p[[2]],"sol7"->p[[3]],
"sol8"->p[[4]]|>
];

findPopulationHBTerms[w0_,g_,k_,beta_,dt_,tend_]:=
Module[{sole,convergentQ,zero},
convQ[w0x_,gx_,kx_,bx_,tendx_]:=
Module[{tmp},
tmp=Table[Chop[kernelFunction[t,w0x,gx,kx,bx,5]],
{t,0,tendx,tendx/100}];
Max[Abs[tmp]]<1.1
];
If[convQ[w0,g,k,beta,tend],
sole=solvePopulation[w0,g,k,beta,1/2,dt,1000 dt,5];
<|"w0"->w0,"g"->g,"k"->k,"b"->beta,"conv"->True,
"solHB"->sole|>,
zero=Table[{t,0},{t,0,tend,dt}];
<|"w0"->w0,"g"->g,"k"->k,"b"->beta,"conv"->False,
"solHB"->zero|>
];

markov[t_,w0_,g_,k_,b_]:=
Module[{del,a,c,sum},del=Sqrt[w0^2-g^2];
a=k^2/(b w0);
a=a+k^2/(Pi del)
Im[PolyGamma[0,1+b/(2 Pi)(g+I del)]];

```

```

c=k^2/w0;
sum=1/(2 a)-Exp[-c^2/(4 a)
+(c-2 I a t)^2/(4 a)]/(4 a);
sum=sum-Exp[-c^2/(4 a)+(c+2 I a t)^2/(4 a)]/(4 a);
sum=sum-(c Exp[-(c^2/(4 a))] Sqrt[Pi]*
Erfi[c/(2 Sqrt[a])])/(4 a^(3/2));
sum=sum+(c Exp[-(c^2/(4 a))] Sqrt[Pi]*
Erfi[(c-2 I a t)/(2 Sqrt[a])])/(8 a^(3/2));
sum=sum-(I Exp[-(c^2/(4 a))] Sqrt[Pi] t*
Erfi[(c-2 I a t)/(2 Sqrt[a])])/(4 Sqrt[a]);
sum=sum+(c Exp[-(c^2/(4 a))] Sqrt[Pi]*
Erfi[(c+2 I a t)/(2 Sqrt[a])])/(8 a^(3/2));
sum=sum+(I Exp[-(c^2/(4 a))] Sqrt[Pi] t*
Erfi[(c+2 I a t)/(2 Sqrt[a])])/(4 Sqrt[a]);
Exp[sum]];

solveMarkovian[w0_,g_,k_,b_,V_,dt_,tend_]:=Table[
{t,Re[markov[t,w0,g,k,b]]},{t,0,tend,dt}];

solvePopulationMarkovian[w0_,g_,k_,b_,V_,dt_,tend_]:=
Module[{ec,kernelMarkov,sol,sigz,coz},
Print["dt=",dt," tend=",tend];
ec[t_]:=If[Abs[t]<10^(-10),1,
Re[kernelFunction[t,w0,g,k,b,6]]];
kernelMarkov[t_?NumericQ]:=4 V^2 NIntegrate[ec[tx],
{tx,0,t},MaxRecursion->25];
sol=NDSolve[{sigz'[t]==-kernelMarkov[t] sigz[t],
sigz[0]==1},sigz[t],{t,0,tend}];
coz=sigz[t]/. sol[[1]];
Table[{tx,coz /. t->tx},{tx,0,tend,dt}]
];

KAll[t_,w0_,g_,k_,beta_,ith_:1]:=
Module[{ged,gad,de,Er,term,harmonicTerm,betaTerm,tot,
ipio,b,a,b,c,gaTerm},
If[Abs[t]<10^(-12),1,de=Sqrt[w0^2-g^2]; Er=k^2/w0;
{a,b}=k^2/(de w0^3){-(de-I g)^2/(1-Exp[beta(de+I g)]),
(de+I g)^2/(1-Exp[-beta(de-I g)])};
c=4 Er g/(beta w0^2);
term[1]=-(a+b)+a Exp[I t(de+I g)]+b Exp[-I t(de-I g)]
-c t;
term[1]=-a+a Exp[I t(de+I g)];
term[2]=-b+b Exp[-I t(de-I g)];
term[3]=-c t; ipio=b/2 Pi/beta;
term[4]=-4 Er g/(Pi w0^2) Log[1-Exp[-ipio b t]];
ged=g-I de; gad=g+I de;
harmonicTerm:=- (gad)^2 (HarmonicNumber[-ged/ipio b]
+HarmonicNumber[ged/ipio b])
+(ged)^2 (HarmonicNumber[-gad/ipio b]
+HarmonicNumber[gad/ipio b]);
term[5]:=-I k^2/(2 Pi de w0^3)(harmonicTerm);
betaTerm:=gad^2(Exp[-t ged] Beta[Exp[-ipio b t],
1-ged/ipio b,0]+ Exp[t ged]
Beta[Exp[-ipio b t],1+ged/ipio b,0])-
ged^2(Exp[-t gad]
Beta[Exp[-ipio b t],1-gad/ipio b,0]+
Exp[t gad] Beta[Exp[-ipio b t],
1+gad/ipio b,0]);
term[6]:=I k^2/(2 Pi de w0^3) betaTerm;
tot=Sum[term[i],{i,1,ith}];

```

$\text{Re}[\text{Exp}[\text{tot}]]];$



9.2 Appendix B

The Magnus expansion is an approximate method that developed to solve a coupled ordinary linear homogeneous equation systems including operator or matrix forms (Magnus, 1954):

$$\frac{d}{dt}Y(t) = A(t)Y(t) \quad \text{with} \quad Y(0) = I \quad (\text{B.1})$$

where $A(t)$ and $Y(t)$ are $n \times n$ matrices. The solution of Eq.(B.1) is:

$$Y(t) = \exp(\Omega(t))Y(0) \quad (\text{B.2})$$

where $\Omega(t)$ can be written as an infinite sum:

$$\Omega(t) = \sum_{m=1}^{\infty} \Omega_m(t) \quad (\text{B.3})$$

with $\Omega_m(t)$ corresponding to the m^{th} order term in the Magnus expansion and can be expressed as (Blanes et al., 2009):

$$\Omega_m = \sum_{j=1}^{m-1} \frac{B_j}{j!} \int_0^t S_m^{(j)}(t_1) dt_1, \quad m \geq 2 \quad (\text{B.4})$$

$$\Omega_1 = \int_0^t A(t_1) dt_1, \quad S_m^{(1)} = [\Omega_{m-1}, A(t)] \quad (\text{B.5})$$

$$S_m^{(j)} = \sum_{n=1}^{m-j} [\Omega_n, S_{m-n}^{(j-1)}] \quad \text{with} \quad 2 \leq j \leq m-1. \quad (\text{B.6})$$

Here B_j refers the Bernoulli numbers, i.e., $B_0 = 1$, $B_1 = -\frac{1}{2}$, $B_2 = \frac{1}{6}$, $B_3 = 0 \dots$ in a recursive form. Higher order $\Omega_m(t)$ have a complicated form, so we display only the first three terms as:

$$\Omega_1(t) = \int_0^t A(t_1) dt_1 \quad (\text{B.7})$$

$$\Omega_2(t) = \frac{1}{2} \int_0^t dt_1 \int_0^{t_1} dt_2 [A(t_1), A(t_2)] \quad (\text{B.8})$$

$$\Omega_3(t) = \frac{1}{6} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 ([A(t_1), [A(t_2), A(t_3)]] + [A(t_3), [A(t_2), A(t_1)]]) \quad (\text{B.9})$$

where the brackets stands for the commutator of X and Y , i.e., $[X, Y] = XY - YX$. The interpretation of these equations in Eq. (B.7)-(B.9) may be given as follow: If the coefficient matrix at different times commute, i.e., $[A(t_1), A(t_2)] = 0$, Eq.(B.7) could be used to construct the exact solution as $\exp[\Omega_1(t)]$. For the systems that have $[A(t_1), A(t_2)] \neq 0$, Magnus has derived the correction terms that would make the general exponential solution $e^{\Omega(t)}$ and those corrections are given in Eq. (B.4)-(B.6). One of the advantages of the Magnus expansion method over the other approximate methods, such as time dependent perturbation theory used for solving a system including varying parameters in time, is that the unitary dynamics of evolution operator $\exp[\Omega(t)]$ is preserved at every truncation of the series in Eq. (B.3).

The convergence criteria for the Magnus series have been investigated in a number of papers (Pechukas and Light, 1966; Moan and Niesen, 2008; Blanes et al., 1998). The optimal convergence condition for a bound operator $A(t)$ in the time interval $t \in [0, T)$ reported to be

$$\int_0^T \|A(s)\| ds < \pi \quad (\text{B.10})$$

where $\|\cdot\|$ denotes a matrix norm and can be calculated as the square-root of the largest eigenvalue of $A^\dagger A$. The validity of convergence boundary can be improved by transforming the original Hamiltonian to interaction picture (Casas, 2007; Blanes et al., 2009; Lopez et al., 2013).

10. CURRICULUM VITAE

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List of Publications :

Ferdi Altintas, Arzu Kurt, and Resul Eryiğit, (2012), "Classical Memoryless Noise-Induced Maximally Discordant Mixed Separable Steady States", Phys. Lett. A, 377: 53-59.

Arzu Kurt and Resul Eryiğit, (2015), "Fourth-Order Master Equation for a Charged Harmonic Oscillator Interacting with the Electromagnetic Field", Phys. Lett. A, 379: 3037-3044.

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Arzu Kurt and Resul Eryiğit, "Effect of Dichotomous Noise on Dissipative Tunneling" (In preparation).