REVISITING PHYSICS OF QUANTUM BITS

BY

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DISSERTATION

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Abstract

Advancements in the technology of quantum bits invoke more precise calculations for decoherence and dissipative effects. In this thesis, the physics of truncated two level systems is revisited and it is shown that in some systems, such as in triple-junction superconducting flux qubit, environmental noises that are usually recognized to only have dissipative effects and the trivial resultant dephasing, can also cause *pure dephasing*. Furthermore, it is demonstrated that, in the current domain of interest for quantum computation purposes, the effective Hamiltonian of single molecule magnets in interaction with a spin bath differs from the commonly quoted result in the literature. It is also shown that the topological effects in such problems are as small as transitional effects to higher excited states beyond the two-level picture. Finally, a simulation of the quantum noise by the classical noise and a simulation of spin bath by oscillator bath for a quantum bit as the principal system are presented.

To my twelfth Imam, Imam Mahdi (pbuh),

the peaceful savior of mankind, the pure, the guide, the loving, the living.

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Chapter 1

Introduction

There are world wide intensive efforts underway to build quantum computers. The building blocks of these computers are quantum bits. The latter are either *intrinsic* two level systems, such as nuclei of spin $\frac{1}{2}$, or in most cases quantum systems that can be regarded as two level systems within the regime of use, such as superconducting qubits and molecular nanomagnets. These intrinsic or, for the second kind, truncated two level systems interact with each other in a quantum chip and with their environment. As the result of interaction with the environment they lose quantum coherence and their quantum features. It is ideal to understand mechanisms of these types of interactions and find ways to decrease their effects.

The quantum dissipation and decoherence theory studies the procedure of truncation of multi-level systems to two level systems in presence of an environment and also studies the effect of the environment on the truncated two level systems. From theoretical aspects, this is a very mature area of research. The advent of Caldeira-Leggett oscillator bath theory took place in 1980's [1, 2], that of Prokofev-Stamp spin bath theory occured in 1990's [3], and major advancements in the theory of $1/f$ noise were in 2000's [4]. For many researchers, the theoretical studies of this field are rather complete, for practical purposes [5], and what is left is the engineering of better quantum bits, chips and eventually computers. Although this is, in my opinion, to a good extent correct, as we see in this thesis there are still some rooms to explore new physics on theoretical side of the quantum dissipation and decoherence theory.

My plan for this research in the last few years was to revisit and reexamine carefully some quantum dissipation/decoherence models and check whether there are any aspects that have been overlooked or not treated properly in the past. We found there are some.

The first one is that in double well potentials the energy splitting can depend on the bias energy between the bottom of the two wells linearly. We show this through a WKB calculation in Chapter 2. This in turn suggested to us that the tunneling matrix element should depend on the bias energy. The tunneling matrix element is well defined between two states in the Hilbert space. To calculate that we revisit truncation procedure in Chapter 3 and show how a multi-level system can be truncated properly to a two level system

in presence of an environment and how a bias term can contribute to the tunneling matrix element. We use the idea of Chapter 2 and the result of Chapter 3 and apply them to quasiparticles noise in Josephson junctions and superconducting two level systems with multiple junctions and find that there is a phase damping process that comes out of these considerations and has not been taken into account previously. In Chapter 5, as another application, we apply the idea of Chapters 2-3 to the Landau-Zener problem and show that the probability of transition is modified in presence of linear term in the tunneling matrix element. This completes Part I of the thesis.

In Part III (we discuss Part II momentarily) we apply the results of Part I-Chapter 3 to Molecular magnets in presence of a spin environment. Chapter 8 serves as a brief introduction to the field of molecular magnets. We show in Chapter 8-9 that the spin bath theory is solved in the regime that is not appropriate for molecular magnets. Thus, one has to repeat the truncation procedure for the regime relevant to molecular magnets. We use the results of Chapter 3 and do the truncation in Chapter 10 for the interesting case of half-odd-integer spins and compare our results with those of the spin bath theory and show differences. Finally, in Chapter 11 we discuss that the topological decoherence terms, which are the result of truncation procedure in the spin bath theory, are of the same importance as transition terms to higher excited states.

Part II concerns a different area of quantum dissipation and decoherence theory. It is on the effects of entanglement and its simulations. The entanglement between a system and environment is considered as the main cause of decoherence and as the distinctive feature of quantum theory as compared to classical theories. How much the effect of entanglement can be simulated classically and how much these effects depend on the type of the environment? We discuss these in Chapters 6-7. In chapter 6 we show that the effect of a quantum noise can be simulated classically without appealing to entanglement if one allows the classical noise fields to depend on the initial state of the universe. In Chapter 7 we demonstrate that an oscillator bath can simulate the effect of a spin bath on relaxation of a two level system in the strong coupling regime of the spin bath.

Part I

VARIABLE TUNNELING

Chapter 2

Tunnel splitting in asymmetric double well potentials : An improved WKB calculation

We present an improved Wentzel-Kramers-Brillouin (WKB) calculation of tunnel splitting in one dimensional asymmetric double well potentials. We show the tunnel splitting in general can have linear dependence to bias energy beside the well-known quadratic dependence. We demonstrate that the linear correction is greater than previously thought.

2.1 Introduction

The purpose of this paper is to calculate the energy level splitting (or tunnel splitting), ΔE, in a smooth, asymmetric, one-dimensional potential, such as that in Fig. 2.1, to first order in $\tilde{\epsilon}/\hbar\omega$ where $\tilde{\epsilon}$ is the bias energy between the bottom of the wells and ω is the order of magnitude of the small oscillation frequencies ω_R, ω_L in the right and the left wells (see Figs. 2.1-2.2).

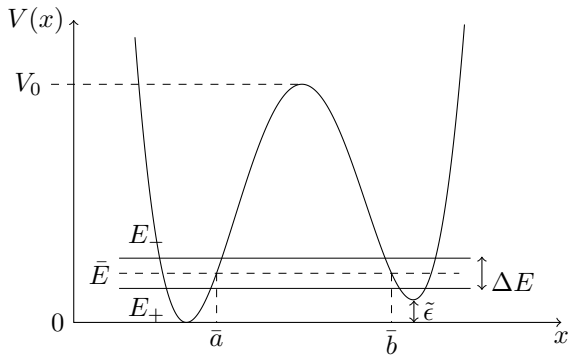

Figure 2.1: Asymmetric double well potential $V(x)$ considered in this paper. $E_\pm$ are energy levels of the ground state doublet. ΔE is the level splitting and $\tilde{\epsilon}$ is the bias in the bottoms of the wells. $\bar{E}$ is a mean energy between E_- and E_+ which is used as a mathematical tool to calculate ΔE. $\bar{a}$ and $\bar{b}$ are turning points for the fictitious energy level $\bar{E}$. The height of barrier is V_0 which is much larger than other energy quantities in the problem.

The problem of quantum mechanical tunneling in a double-well potential is ubiquitous in physics. The quantum state of the system in such problems is effectively restricted to a two-dimensional Hilbert space.

Quantum tunneling allows the state to hop between these two dimensions. Apart from the well-known microscopic example of inversion of an amonia molecule, in recent decades quantum tunneling has been observed in macroscopic phenomena such as the tunneling of magnetic flux in an rf SQUID[2, 6, 7, 8], tunneling of Bose-Einstein condensates [9, 10] and electronic spin tunneling in the nano-magnetic molecules such as Fe8 [11, 12, 13].

In some problems the height of the barrier V_0 is much larger than the energy gap $\hbar\omega$ between the ground state doublet and higher excited states. WKB approximation can be applied *under* the barrier in these problems. However, application of WKB *inside* the wells gives inaccurate results [14]. The reason is that, crudely speaking, the semi classical approximation of WKB is suitable where the classical momentum of a particle $|p(x)| = \sqrt{2m|E - V(x)|}$ is large. This is not satisfied for a particle in ground state inside a well. However, under the barrier since $V(x)$ is large the condition is satisfied and one can employ the WKB approximation. [14]

Previous works [15, 16, 17, 18] have calculated the energy splitting ΔE and tunneling amplitude Δ in an asymmetric potential to zeroth order in $\epsilon/\hbar\omega$ and ϵ/V_0. The general belief [15] is that the correction to these quantities are of order ϵ/V_0. It is also implicitly assumed in the bulk of literature that the tunneling amplitude Δ is relatively independent of the bias energy $\tilde{\epsilon}$ or ϵ.

In the present paper, however, we show that the correction to tunnel splitting is in general of order $\tilde{\epsilon}/\hbar\omega$ in the WKB limit, rather than $\tilde{\epsilon}/V_0$.

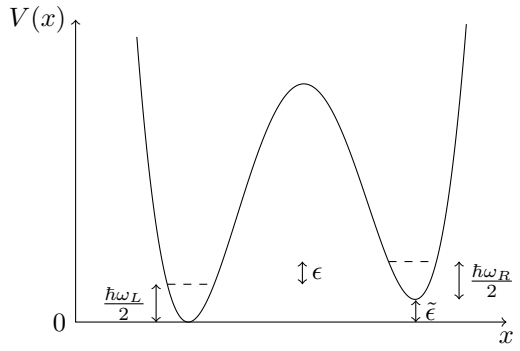

Figure 2.2: Asymmetric double well potential $V(x)$. ω_L (ω_R) is the small oscillation frequencies in the left (right) well. In the absence of tunneling, $\hbar\omega_L/2$ ($\hbar\omega_R/2 + \tilde{\epsilon}$) is the ground-state energy of the state localized in the left (right) well and ϵ is the difference between these two energies.

Calculations of this paper are more accurate than its previous counterparts. For example, we give an expression for Δ, Eq. (2.3) ,which does not depend on the value of Δ itself. The situation is rather different in Ref. [16, 17, 18]. The Gamow factor, e^{-2I}, in those references depends on the actual energy of the levels

$E_\pm$ and, hence, on the value of the ΔE, and Δ. Ref. [16] discusses that this dependence is rather weak. Here, however, we obtain Δ as a function of the Gamow factor of a fictitious energy $\bar{E}$, Eq. Eq. 2.5, independent of ΔE, Δ. Furthermore we show quantitatively that the correction to our expression is negligible.

Before embarking on detailed calculations in the following sections, let us summarize the main results of this paper for the energy splitting and tunneling amplitude of ground state doublet. We denote the energy of the near even parity state in the doublet by E_+, the lower level, and the energy of the nearly odd parity state, the upper level by E_- (Fig. 2.1). Then we shall derive that the energy splitting between these two levels $\Delta E = E_- - E_+$ is

$$\Delta E = \sqrt{\epsilon^2 + \Delta^2}\ , \tag{2.1}$$

where

$$\epsilon = \tilde{\epsilon} + \frac{\hbar(\omega_R - \omega_L)}{2}, \tag{2.2}$$

$\tilde{\epsilon}$ is the energy difference between the minima of the potential, ϵ is the energy difference between the ground states of the particle in each well in absent of tunneling (Fig. 2.1-2.2) and

$$\Delta = \frac{\hbar\sqrt{\omega_R\omega_L}}{\sqrt{e\pi}}(1 + \frac{k}{4}\frac{\epsilon}{\hbar\omega_L}\frac{\omega_R - \omega_L}{\omega_R})e^{-\bar{I}(\bar{E}(\tilde{\epsilon}))}, \tag{2.3}$$

where

$$k = \gamma - \ln 2 \simeq -0.11, \tag{2.4}$$

$$\bar{E}(\tilde{\epsilon}) = \bar{E} = \frac{\hbar(\omega_L + \omega_R)}{4} + \frac{\tilde{\epsilon}}{2}, \tag{2.5}$$

$$I(\bar{E}(\tilde{\epsilon})) = \frac{1}{\hbar}\int_{\bar{a}}^{\bar{b}} |p|dx. \tag{2.6}$$

γ is the Euler-Mascheroni constant and

$$p = \sqrt{2m(\bar{E} - V(x))}. \tag{2.7}$$

Also, $\bar{a}$ and $\bar{b}$ are the turning points for a classical particle with energy $\bar{E}$ which wishes to climb up the barrier from either well (See Fig. 2.1).

2.2 Wave functions and Energy Quantization Equations

In order to find the energy level splitting in the ground state doublet we find the wave function near the left minimum in region L, under the barrier in region B, and near the right minimum in region R as illustrated in Fig. 2.3. Then we connect these wave functions in the overlapping regions LB and BR. The connection formulas give us a constraint which determines the energy splitting.

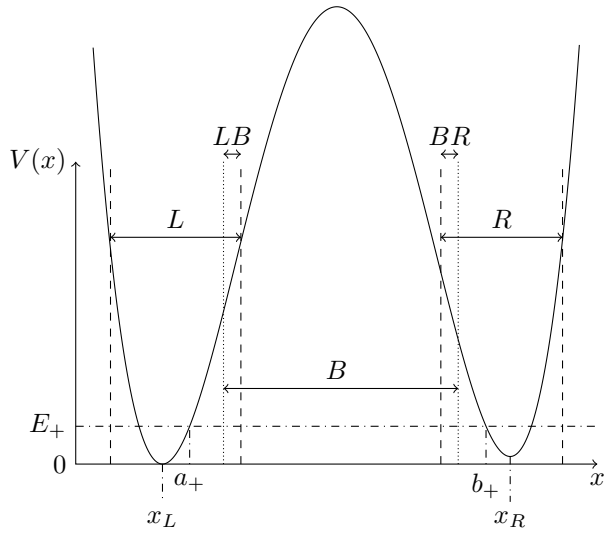

Figure 2.3: Regions L and R denote the domain of potential near the left and right minima in which the potential is sufficiently quadratic. Region B is under the barrier area away from the turning points in which the WKB approximation can be applied. LB and BR are the overlapping areas where we match the wave function of each region to that of its neighboring region. x_L and x_R denote the coordinates of the local minima.

We assume that the potential is nearly parabolic near the minima in the regions L and R. The Schrodinger equation for parabolic potentials can be solved exactly for any given energy. The solutions are parabolic cylinder functions. We find these solutions such that they do not diverge as $x \to \pm\infty$, to avoid violation of square-integrability of the wave function.

Under the barrier, in region B, we use the WKB approximation method.

2.2.1 Wave functions near the local minima of potential: Parabolic cylinder functions

As discussed earlier, near the minima of the potential x_L and x_R we can write

$$V(x) = \begin{cases} \frac{1}{2}m\omega_L^2(x-x_L)^2 + \cdots & x \in L \\ \tilde{\epsilon} + \frac{1}{2}m\omega_R^2(x-x_R)^2 + \cdots & x \in R \end{cases} \tag{2.8}$$

where

$$\omega_L^2 = \frac{V''(x_L)}{m}, \tag{2.9}$$

$$\omega_R^2 = \frac{V''(x_R)}{m}, \tag{2.10}$$

and the zero-point of potential is set such that $V(x_L) = 0$ and $V(x_R) = \tilde{\epsilon}$ (Fig. 2.3).

Neglecting the higher order terms in the potential, the Schrodinger equation in region L for the lower level in the ground state doublet with energy E_+ becomes

$$-\frac{\hbar^2}{2m}\frac{d^2}{dx^2}\psi_L(x) + \frac{1}{2}m\omega_L^2(x - x_L)^2\psi_L(x) = E_+\psi_L(x). \tag{2.11}$$

We can write the above equation in the form of differential equation of parabolic cylinder functions by defining,

$$\eta_l \equiv \frac{x - x_L}{\sqrt{\hbar/2m\omega_L}}, \tag{2.12}$$

$$\zeta_+^L \equiv \frac{E_+}{\hbar\omega_L} - \frac{1}{2}. \tag{2.13}$$

Notice that η_l is a variable and varies with x while x_L is a fixed point (We use lowercase l (r) for variables of the left (right) well and uppercase L (R) for its fixed quantities.). With the above definitions, Eq. Eq. 2.11 can be rewritten as

$$\frac{d^2}{d\eta_l^2}\psi_L(\eta_l) + \left(\zeta_+^L + \frac{1}{2} - \frac{\eta_l^2}{4}\right)\psi_L(\eta_l) = 0. \tag{2.14}$$

which is manifestly parabolic cylinder functions' differential equation [19]. This equation has two independent solutions, $D_{\zeta_+^L}(\eta_l)$ and $D_{\zeta_+^L}(-\eta_l)$. The former diverges as $\eta_l \to -\infty$ and is not allowed by square-integrability condition. Therefore, the physical solution for the wave function in region L is

$$\psi_L(\eta_l) = \alpha_L D_{\zeta_+^L}(-\eta_l) \tag{2.15}$$

where α_L is a constant to be determined by matching conditions below.

For real values of η_l the asymptotic expansion of $D_{\zeta_+^L}(-\eta_l)$ to leading order, when $|\zeta_+^L| \ll 1$, is [19]

$$D_{\zeta_+^L}(-\eta_l) =$$
$$\begin{cases} (-1)^{\zeta_+^L}\ \eta_l^{\zeta_+^L} e^{-\eta_l^2/4}, & \eta_l \ll -1 \\ \cos(\pi\zeta_+^L)|\eta_l|^{\zeta_+^L} e^{-\eta_l^2/4} + \frac{\sqrt{2\pi}}{\Gamma(-\zeta_+^L)} \frac{e^{\eta_l^2/4}}{|\eta_l|^{\zeta_+^L+1}} & \eta_l \gg 1 \end{cases} \tag{2.16}$$

which exponentially decays as $\eta_l \to -\infty$ and has a decaying and growing parts for positive large η_l. We shall see shortly that keeping both of these parts is necessary for matching the wave functions under the barrier and near the right well.

The quantity $|\zeta_+^L|$ is much smaller than unity. One can observe this fact by noting that in the unbiased symmetric case this quantity is half of the tunneling amplitude Δ which is exponentially small. In the asymmetric case definition Eq. 2.13 implies that $|\zeta_+^L|$ is at most of order $\tilde{\epsilon}/\hbar\omega$ which is much smaller than unity by our convention in this paper (We demonstrate this fact rather more rigorously in Sec. 2.3 when we find the energy levels).

Similarly we can find the wave function near the right well in region R. The Schrodinger equation to second order approximation of the potential is

$$-\frac{\hbar^2}{2m}\frac{d^2}{dx^2}\psi_R(x) + \left(\frac{1}{2}m\omega_L^2(x-x_R)^2 + \tilde{\epsilon}\right)\psi_R(x) = E_+\psi_R(x). \tag{2.17}$$

By defining,

$$\eta_r \equiv \frac{x - x_R}{\sqrt{\hbar/2m\omega_R}}, \tag{2.18}$$
$$\zeta_+^R \equiv \frac{E_+ - \tilde{\epsilon}}{\hbar\omega_R} - \frac{1}{2}, \tag{2.19}$$

Eq. Eq. 2.17 can be written as

$$\frac{d^2}{d\eta_r^2}\psi_R(\eta_r) + \left(\zeta_+^R + \frac{1}{2} - \frac{\eta_r^2}{4}\right)\psi_R(\eta_r) = 0, \tag{2.20}$$

which has two solutions $D_{\zeta_+^R}(\eta_r)$ and $D_{\zeta_+^R}(-\eta_r)$. This time we reject the latter as it diverges when $\eta_r \to \infty$. So the physical solution of Eq. 2.20 is

$$\psi_R(\eta_r) = \alpha_R D_{\zeta_+^R}(\eta_r) \tag{2.21}$$

where α_R is a coefficient to be determined and the asymptotic expansion of $D_{\zeta_+^R}(\eta_r)$ for $|\zeta_+^R| \ll 1$ is

$$D_{\zeta_+^R}(\eta_r) =$$
$$\begin{cases} \cos(\pi\zeta_+^R)|\eta_r|^{\zeta_+^R} e^{-\eta_r^2/4} + \frac{\sqrt{2\pi}}{\Gamma(-\zeta_+^R)} \frac{e^{\eta_r^2/4}}{|\eta_r|^{\zeta_+^R+1}} & \eta_r \ll -1 \\ (-1)^{\zeta_+^R}\ \eta_r^{\zeta_+^R} e^{-\eta_r^2/4} & \eta_r \gg 1 \end{cases} \tag{2.22}$$

This again decays nicely for large positive η_r and has growing and decaying components for large negative η_r as expected. $|\zeta_+^R|$ is also much smaller than unity for the problem we consider for the same reasons mentioned above for $|\zeta_+^L|$.

The particular regions of interest are LB and BR (Fig. 2.3). We use these regions to match ψ_L and ψ_R to the WKB solution under the barrier. These regions are reasonably far from the turning points to satisfy validity condition of the WKB approximation, but yet close enough to the bottom of the wells to allow parabolic approximation of the potential to be employed. In these regions the wave functions that we found in this section is as follows

$$\psi_+^{par}(x) =$$
$$\begin{cases} \alpha_L \frac{\cos(\pi\zeta_+^L)|\eta_l|^{\zeta_+^L}}{e^{\eta_l^2/4}} + \alpha_L \frac{\sqrt{2\pi}}{\Gamma(-\zeta_+^L)} \frac{e^{\eta_l^2/4}}{|\eta_l|^{\zeta_+^L+1}} & x \in LB \\ \alpha_R \frac{\cos(\pi\zeta_+^R)|\eta_r|^{\zeta_+^R}}{e^{\eta_r^2/4}} + \alpha_R \frac{\sqrt{2\pi}}{\Gamma(-\zeta_+^R)} \frac{e^{\eta_r^2/4}}{|\eta_r|^{\zeta_+^R+1}} & x \in BR \end{cases} \tag{2.23}$$

where superscript *par* is for *parabolic cylinder* and the relation between x and η_l, η_r is given in Eqs. (2.12) and (2.18). In the next subsection we shall find WKB wave function under the barrier and match it with Eq. Eq. 2.23.

2.2.2 Wave functions under the barrier: WKB approximation limit

Under the barrier, far enough from the turning points in region B, we can apply the WKB approximation. With the usual ansatz of $\psi = exp(i\sigma/\hbar)$ one obtains [20]

$$\psi_+^{WKB}(x) = \frac{C}{\sqrt{|v(x)|}} e^{-\int_a^x |p|dx/\hbar} + \frac{C'}{\sqrt{|v(x)|}} e^{+\int_b^x |p|dx/\hbar} \tag{2.24}$$

where C, C' are constants,

$$p(x) = \sqrt{2m(E_+ - V(x))}\ , \tag{2.25}$$
$$v(x) = \frac{p(x)}{m}, \tag{2.26}$$

and a_+,b_+ are classical turning points for energy E_+ as shown in Fig. 2.3. The choice of lower bounds of the integrals in Eq. 2.24 is arbitrary. We chose to use a_+,b_+ to simplify future equations. This choice is different from what previous authors have used [14, 17, 18].

The particular regions of interest are again LB and BR. In these regions we can approximate the potential with parabolic functions of Eq. Eq. 2.8. Under this approximation, e.g.,

$$E_+ = V(a_+) \simeq \frac{1}{2} m\omega_L^2 (a_+ - x_L)^2 \tag{2.27}$$

and one obtains in region LB for the classical momentum

$$|p(x)| \simeq m\omega_L \sqrt{(x - x_L)^2 - (a_+ - x_L)^2} \tag{2.28}$$

The first integral in Eq. 2.24 can now be taken for $x \in LB$. We follow methods developed in Ref. [14] in taking this integral. The result is

$$\begin{aligned} \frac{1}{m\omega_L} \int_{a_+}^{x} |p| dx &\simeq \frac{1}{2}(x - x_L)^2 - \frac{1}{4}(a_+ - x_L)^2 \\ &- \frac{1}{2}(a_+ - x_L)^2 \ln\left(\frac{2(x - x_L)}{a_+ - x_L}\right). \end{aligned} \tag{2.29}$$

We used the fact that in the region LB, $(x - x_L) \gg (a_+ - x_L)$. Nevertheless we kept the second term in Eq. Eq. 2.29 as the left hand side integral appears in the *exponent* of the first term in Eq. Eq. 2.24. However, one does not need to keep the similar term in calculating $v(x)$ from Eq. Eq. 2.28, as $v(x)$ appears in the denominators in Eq. Eq. 2.24 (not in the exponents [14]),

$$v(x) \simeq \omega_L (x - x_L). \tag{2.30}$$

For the second integral in Eq. Eq. 2.24 and $x \in LB$ we note that

$$\int_{b_+}^{x} |p| dx = -\int_{a_+}^{b_+} |p| dx + \int_{a_+}^{x} |p| dx. \tag{2.31}$$

We define,

$$I_+ \equiv \int_{a_+}^{b_+} |p| dx, \tag{2.32}$$

$$g(\zeta) \equiv \sqrt{2\pi}\left(\zeta + \frac{1}{2}\right)^{\zeta+\frac{1}{2}} e^{-(\zeta+\frac{1}{2})}. \tag{2.33}$$

Now by substituting from Eqs. (2.29-2.31) into Eq. Eq. 2.24 and using Eqs. (2.12-2.13) and (2.27) we obtain for $x \in LB$,

$$\psi_+^{WKB}(x) \simeq K_L \eta_l^{\zeta_+^L} e^{-\eta_l^2/4} + K'_L \eta_l^{-(\zeta_+^L+1)} e^{-\eta_l^2/4}, \tag{2.34}$$

where

$$K_L = (\frac{\hbar\omega_L}{\pi m})^{-\frac{1}{4}} (\frac{g_+^L}{2})^{-\frac{1}{2}}\ C, \tag{2.35}$$

$$K'_L = (\frac{\hbar\omega_L}{\pi m})^{-\frac{1}{4}} (\frac{g_+^L}{\pi})^{\frac{1}{2}} e^{-I_+}\ C', \tag{2.36}$$

and $g_+^L = g(\zeta_+^L)$.

Similar procedure can be used for region BR under the barrier and near the right well. Most of the equations transform trivially if we make the substitution $E_+ \to E_+ - \tilde{\epsilon}$. For the energy and momentum one has

$$E_+ - \tilde{\epsilon} = V(b_+) - \tilde{\epsilon} \simeq \frac{1}{2} m\omega_R^2 (x_R - b_+)^2, \tag{2.37}$$

$$|p(x)| \simeq m\omega_R \sqrt{(x - x_R)^2 - (x_R - b_+)^2}. \tag{2.38}$$

These can be used to take the second integral in Eq. (2.24) as follows

$$\begin{aligned} \frac{1}{m\omega_R} \int_{b_+}^{x} |p| dx &\simeq -\frac{1}{2}(x - x_R)^2 + \frac{1}{4}(x_R - b_+)^2 \\ &+ \frac{1}{2}(x_R - b_+)^2 \ln\left(\frac{2(x_R - x)}{x_R - b_+}\right). \end{aligned} \tag{2.39}$$

Note that all the signs in the right hand side are flipped in comparison to Eq. Eq. 2.29. In taking the integrals Eq. 2.29 and Eq. 2.39 one may use Eq. (2.27) of Ref. [19]. In BR the velocity is approximately

$$v(x) \simeq \omega_R(x_R - x) \tag{2.40}$$

and the first integral in Eq. Eq. 2.24 for $x \in BR$ can be calculated by the identity

$$\int_{a_+}^{x} |p|dx = \int_{a_+}^{b_+} |p|dx + \int_{b_+}^{x} |p|dx. \tag{2.41}$$

Now by substituting from Eqs. (2.39-2.41) into Eq. Eq. 2.24 and using Eqs. (2.18-2.19) and (2.37) we obtain for $x \in BR$,

$$\psi_+^{WKB}(x) \simeq L_R|\eta_r|^{\zeta_+^R} e^{-\eta_r^2/4} + L'_R|\eta_r|^{-(\zeta_+^R+1)} e^{-\eta_r^2/4}, \tag{2.42}$$

where

$$L_R = (\frac{\hbar\omega_R}{\pi m})^{-\frac{1}{4}} (\frac{g_+^R}{\pi})^{\frac{1}{2}} e^{-I_+} C, \tag{2.43}$$

$$L'_R = (\frac{\hbar\omega_R}{\pi m})^{-\frac{1}{4}} (\frac{g_+^R}{2})^{-\frac{1}{2}}\ C', \tag{2.44}$$

and $g_+^R = g(\zeta_+^R)$.

2.2.3 Matching WKB and parabolic cylinder wave functions : Energy quantization equation for the lower energy level

Now we are ready to match the WKB wave functions Eq. 2.34,Eq. 2.42 in regions LB and BR, respectively, with the parabolic cylinder wave functions Eq. 2.23 in those regions. By matching the wave functions in LB we obtain relations between C, C' and α_L,

$$C = (\frac{\hbar\omega_L}{\pi m})^{\frac{1}{4}} (\frac{g_+^L}{2})^{\frac{1}{2}} \cos\pi\zeta_+^L\ \alpha_L, \tag{2.45}$$

$$C' = \pi(\frac{\hbar\omega_L}{\pi m})^{\frac{1}{4}} (\frac{g_+^L}{2})^{-\frac{1}{2}} e^{I_+}\Gamma^{-1}(-\zeta_+^L)\ \alpha_L. \tag{2.46}$$

Matching the wave functions in BR relate C, C' to α_R,

$$C = \pi(\frac{\hbar\omega_R}{\pi m})^{\frac{1}{4}} (\frac{g_+^R}{2})^{-\frac{1}{2}} e^{I_+}\Gamma^{-1}(-\zeta_+^R)\ \alpha_R, \tag{2.47}$$

$$C' = (\frac{\hbar\omega_R}{\pi m})^{\frac{1}{4}} (\frac{g_+^R}{2})^{\frac{1}{2}} \cos\pi\zeta_+^R\ \alpha_R. \tag{2.48}$$

In order to find the energy quantization equation we find the ratio C/C' from Eqs. (2.45-2.46) and from Eqs.(2.47-2.48) and equate them. This gives us

$$\zeta_+^L \zeta_+^R = f(\zeta_+^L) f(\zeta_+^R) e^{-2I_+} \tag{2.49}$$

where

$$f(\zeta) = (2\pi)^{-1} \cos\pi\zeta \ \Gamma(1-\zeta) \ g(\zeta). \tag{2.50}$$

and we used the identity $t\Gamma(t) = \Gamma(1+t)$. Eq.Eq. 2.49 is the fundamental equation of this section we were seeking.

2.2.4 Energy quantization equation for the upper energy level

For the upper level in the doublet with energy E_- one can similarly define

$$\zeta_-^L \equiv \frac{E_-}{\hbar\omega_L} - \frac{1}{2}, \tag{2.51}$$

$$\zeta_-^R \equiv \frac{E_- - \tilde{\epsilon}}{\hbar\omega_R} - \frac{1}{2}. \tag{2.52}$$

and do the previous procedure to obtain identically the energy equation

$$\zeta_-^L \zeta_-^R = f(\zeta_-^L) f(\zeta_-^R) \ e^{-2I_-} \tag{2.53}$$

where

$$I_- \equiv \int_{a_-}^{b_-} |p| dx. \tag{2.54}$$

2.3 Energy Splitting and Tunneling Amplitude to First order

Eqs.Eq. 2.49,Eq. 2.53, and in short

$$\zeta_\pm^L \zeta_\pm^R = f(\zeta_\pm^L) f(\zeta_\pm^R) e^{-2I_\pm}, \tag{2.55}$$

are transcendental equations. We can only solve them approximately. For small energy bias, $\tilde{\epsilon}/\hbar\omega \ll 1$, definitions of $\zeta_\pm^L$ and $\zeta_\pm^R$ imply that $\zeta_\pm^L, \zeta_\pm^R \ll 1$. For small values of $\zeta_\pm^L$ and $\zeta_\pm^R$, $f(\zeta_\pm^L) f(\zeta_\pm^R)$ is of order one hundredth. One can see this by expanding $f(\zeta)$ about zero and obtaining

$$f(\zeta) = \frac{1}{\sqrt{4e\pi}}(1 + k\ \zeta + \mathcal{O}(\zeta^2)) \tag{2.56}$$

where $k \simeq .11$ is defined in Eq. Eq. 2.4. One then notes that for $f(\zeta)f(\zeta')$ the leading order term is $1/4e\pi \simeq 0.02$. Now since $exp(-2I_\pm)$ is exponentially small in the WKB limit, the left hand side of Eq. Eq. 2.55 is also exponentially small. Therefore we can expand the right hand sides of Eqs.Eq. 2.49,Eq. 2.53 to first order in $\zeta_\pm^L$ and $\zeta_\pm^R$ to find the energy levels in the ground state doublet, $E_\pm$,

$$\zeta_\pm^L \zeta_\pm^R = \left[1 + k(\zeta_\pm^L + \zeta_\pm^R) + \mathcal{O}(\zeta_\pm^{L^2}, \zeta_\pm^{R^2})\right] \frac{e^{-2I_\pm}}{4e\pi}. \tag{2.57}$$

Now instead of engaging with $E_\pm$ which is very large compared to the tunnel splitting we define exponentially small quantities $\Delta E_\pm$ as follows and try to find them

$$\Delta E_\pm \equiv E_\pm - \bar{E} \tag{2.58}$$

where $\bar{E}$ is defined in Eq. Eq. 2.5. In terms of $\Delta E_\pm$, one can write

$$\zeta_\pm^L = (\Delta E_\pm + \epsilon/2)/\hbar\omega_L, \tag{2.59}$$

$$\zeta_\pm^R = (\Delta E_\pm - \epsilon/2)/\hbar\omega_R \tag{2.60}$$

Please observe the appearance of ϵ instead of $\tilde{\epsilon}$. One can also expand $I_\pm(E_\pm)$ around $\bar{E}$ to express both $\Delta E_\pm$ in terms of quantities defined at the mean energy $\bar{E}$,

$$I_\pm = I(E_\pm) = \bar{I} + \bar{I}'\Delta E_\pm + \mathcal{O}(\Delta E_\pm^2) \tag{2.61}$$

where $\bar{I} = I(\bar{E})$ and $\bar{I}' = \frac{\partial I}{\partial E}(\bar{E})$.[21]

Now by using Eqs. (2.58-2.61), we can write Eq. (2.57) as

$$\begin{aligned} \frac{\Delta E_\pm^2}{\hbar^2\omega_R\omega_L} - \frac{(\epsilon/2)^2}{\hbar^2\omega_R\omega_L} &= \frac{e^{-2\bar{I}}}{4\pi e}\left(1 + \frac{k\epsilon(\hbar\omega_R - \hbar\omega_L)}{2\hbar^2\omega_R\omega_L}\right) \\ &- \frac{e^{-2\bar{I}}}{4\pi e}\left(u\Delta E_\pm + \mathcal{O}(\Delta E_\pm^2)\right) \end{aligned} \tag{2.62}$$

where

$$u \equiv 2\bar{I'} - \frac{k\hbar(\omega_R + \omega_L)}{\hbar^2\omega_R\omega_L} \tag{2.63}$$

Since $e^{-2\bar{I}}$ is exponentially small, we can reasonably neglect terms of order $e^{-2\bar{I}}\Delta E_{\pm}^2$ in the right hand side of Eq. Eq. 2.62 while keeping the term of order $\Delta E_{\pm}^2$ in the left hand side of the equation. Eq. Eq. 2.62 then becomes a quadratic equation with two solutions as follows

$$\Delta E_{\pm} = -b' \mp \sqrt{(\frac{\epsilon}{2})^2 + (\frac{\Delta}{2})^2 + b'^2} \tag{2.64}$$

where

$$b' = \frac{\hbar^2 \omega_L \omega_R e^{-2\bar{I}} u}{8\pi e}, \tag{2.65}$$

$$\Delta^2 = \frac{\hbar^2 \omega_R \omega_L e^{-2\bar{I}}}{e\pi} \left(1 + k\frac{\epsilon(\omega_R - \omega_L)}{2\hbar\omega_R\omega_L}\right). \tag{2.66}$$

The level splitting ΔE can now be obtained,

$$\begin{aligned} \Delta E &= E_- - E_+ = \Delta E_- - \Delta E_+ \\ &= \sqrt{\epsilon^2 + \Delta^2 + (2b')^2} \end{aligned} \tag{2.67}$$

The last term above, $(2b')^2$, is of order $e^{-4\bar{I}}$ and can be neglected in favor of the second term, Δ^2, which is of order $e^{-2\bar{I}}$. This is irrespective of the value of ϵ. Therefore, we obtain

$$\Delta E \cong \sqrt{\epsilon^2 + \Delta^2} \tag{2.68}$$

The second term above can be interpreted as the square of tunneling matrix element. To take square root from right hand side of Eq. Eq. 2.66 we note that $\epsilon/\hbar\omega_L \ll 1$ and $k, \frac{\omega_L - \omega_R}{\omega_R} < 1$, so we can keep terms to first order in $\epsilon/\hbar\omega_L \ll 1$ and obtain

$$\Delta = \frac{\hbar\sqrt{\omega_R\omega_L}}{\sqrt{e\pi}}(1 + \frac{k}{4}\frac{\epsilon}{\hbar\omega_L}\frac{\omega_R - \omega_L}{\omega_R})e^{-\bar{I}} \tag{2.69}$$

which is the same as Eq. Eq. 2.3 as promised.

2.4 Dependence of tunnel splitting on bias energy

It has been believed [15] that the dependence of Δ on ϵ is only through the quantity ϵ/V_0 which is negligible in the WKB limit. We are going to illustrate in this section that the dependence is also through the quantity $\epsilon/\hbar\omega$ which is much larger than ϵ/V_0 and may not be neglected. This fact is rather clear from Eq. Eq. 2.69

if $\omega_R \neq \omega_L$ in the unbiased double well potential when $\tilde{\epsilon} = 0$. That is to say if the potential in the absence of energy bias is not perfectly symmetric.

The above argument is quite irrespective to the way the exponential factor $e^{-\bar{I}}$ in Eq. Eq. 2.69 varies with ϵ. What we wish to illustrate below, in addition, is that $e^{-\bar{I}}$ *also* varies with $\epsilon/\hbar\omega$ or $(\epsilon/\hbar\omega)^2$ as its largest correction.

We are going to analyze below the Gamow factor $e^{-\bar{I}}$ analytically as much as possible and also illustrate numerical results for the dependence of the factor to $\epsilon/\hbar\omega$. However in order to build intuition and also give a counter example for the claim that correction to tunneling amplitude is of order ϵ/V_0 let us begin by considering a simple example of a double oscillator potential.

2.4.1 Example

Consider a biased double oscillator potential

$$V(x) = \begin{cases} \frac{1}{2}m\omega_L^2(x - x_L)^2 & x \leq 0 \\ \tilde{\epsilon} + \frac{1}{2}m\omega_R^2(x - x_R)^2 & x \geq 0 \end{cases} \tag{2.70}$$

for appropriate values of $x_L < 0$, $x_R > 0$, ω_L and ω_R. The height of the potential barrier is

$$V_0 = \frac{1}{2}m\omega_L^2 x_L^2 = \tilde{\epsilon} + \frac{1}{2}m\omega_R^2 x_R^2 \tag{2.71}$$

We can freely choose V_0, ω_L, ω_R, $\tilde{\epsilon}$ and let the above constraint determine x_L and x_R. To satisfy the WKB condition we just need to make sure that V_0 and $x_R - x_L$ are sufficiently large and ω_R and ω_L are not too large. Otherwise these quantities can be chosen freely. The potential of Eq.(2.70) has a spike at the peak, at $x = 0$, which violates the WKB condition $\frac{m\hbar}{p^3}\frac{dV}{dx} \ll 1$ [20]. However, one can smooth the potential near the spike such that the WKB condition is satisfied and the integrals of momentum stays almost intact. We continue with the potential of Eq. Eq. 2.70 for its simplicity in calculations of the integrals and that we are only interested here in the mathematical properties of $e^{-\bar{I}}$.

The integral

$$\bar{I} = \frac{1}{\hbar}\int_{\bar{a}}^{\bar{b}} |p| dx \tag{2.72}$$

can be divided into two parts

$$\bar{I} = \bar{I}_L + \bar{I}_R \tag{2.73}$$

where

$$\bar{I}_L = \frac{1}{\hbar}\int_{\bar{a}}^{x_m} |p|dx \ , \quad \bar{I}_R = \frac{1}{\hbar}\int_{x_m}^{\bar{b}} |p|dx \tag{2.74}$$

and where x_m is the coordinate of the maximum potential. This is the strategy we shall use in the next subsection too for analytic study of the general case. For the potential of Eq.(2.70), $\bar{I}_L$ can be easily calculated. The result in terms of the energy quantities is

$$\bar{I}_L = \frac{V_0}{\hbar\omega_L}\left(\sqrt{1-\lambda_L} - \lambda_L \log\frac{\sqrt{1-\lambda_L}+1}{\lambda_L}\right) \tag{2.75}$$

where $\lambda_L = \bar{E}/V_0$. Similarly for $\bar{I}_R$ one obtains,

$$\bar{I}_R = \frac{V_0-\tilde{\epsilon}}{\hbar\omega_R}\left(\sqrt{1-\lambda_R} - \lambda_R \log\frac{\sqrt{1-\lambda_R}+1}{\lambda_R}\right) \tag{2.76}$$

where $\lambda_R = (\bar{E}-\tilde{\epsilon})/(V_0-\tilde{\epsilon})$. Since $\lambda_L, \lambda_R \ll 1$ we can expand the above expression in terms of λ_L, λ_R,

$$\bar{I}_L = \frac{V_0}{\hbar\omega_L}\left(1 + \frac{\lambda_L}{2}\{\log(\frac{\lambda_L}{2}) - \frac{1}{2}\} + \mathcal{O}(\lambda_L^2)\right) \tag{2.77}$$

$$\bar{I}_R = \frac{V_0-\tilde{\epsilon}}{\hbar\omega_R}\left(1 + \frac{\lambda_R}{2}\{\log(\frac{\lambda_R}{2}) - \frac{1}{2}\} + \mathcal{O}(\lambda_R^2)\right) \tag{2.78}$$

Now we note that

$$\frac{V_0}{\hbar\omega_L}\lambda_L = \frac{\bar{E}}{\hbar\omega_L}, \tag{2.79}$$

$$\frac{V_0-\tilde{\epsilon}}{\hbar\omega_R}\lambda_R = \frac{\bar{E}-\tilde{\epsilon}}{\hbar\omega_R}. \tag{2.80}$$

This shows that the largest correction is of $\tilde{\epsilon}/\hbar\omega$. To see it more clearly we combine Eqs. (2.77-2.78) to obtain $\bar{I}$ to leading orders

$$\begin{aligned}\bar{I} &= \frac{V_0}{\hbar\omega_L} + \frac{V_0-\tilde{\epsilon}}{\hbar\omega_R} + (\frac{1}{4} + \frac{\epsilon}{4\hbar\omega_L})\{\log(\frac{\lambda_L}{2}) - \frac{1}{2}\} \\ &+ (\frac{1}{4} - \frac{\epsilon}{4\hbar\omega_R})\{\log(\frac{\lambda_R}{2}) - \frac{1}{2}\} + \mathcal{O}(\frac{\bar{E}}{V_0}, \frac{\tilde{\epsilon}}{V_0})\end{aligned} \tag{2.81}$$

Here we used the identities

$$\bar{E} = \frac{\hbar\omega_L}{2} + \frac{\epsilon}{2} \tag{2.82}$$

$$\bar{E} - \tilde{\epsilon} = \frac{\hbar\omega_R}{2} - \frac{\epsilon}{2} \tag{2.83}$$

which can be obtained from the definitions of $\bar{E}$ and ϵ in Eqs. (2.5), (2.2). To zeroth order in $\tilde{\epsilon}/V_0$, we have $\lambda_R = \lambda_L \equiv \lambda$. Therefore Eq. (2.81) becomes

$$\begin{aligned}\bar{I} = & \frac{V_0}{\hbar\omega_L} + \frac{V_0}{\hbar\omega_R} - \frac{\tilde{\epsilon}}{\hbar\omega_R} \\ & + (\frac{1}{2} + \frac{\epsilon}{4\hbar\omega_L}\frac{\Delta\omega}{\omega_R})\{\log(\frac{\lambda}{2}) - \frac{1}{2}\} + \mathcal{O}(\frac{\bar{E}}{V_0}, \frac{\tilde{\epsilon}}{V_0})\end{aligned} \tag{2.84}$$

One now observes that the leading order correction, due to the bias, comes from the third term in the right hand side of the above equation, i.e. from $-\tilde{\epsilon}/\hbar\omega_R$. In the case that $\omega_L \neq \omega_R$ the correction from the fourth term, $\frac{\epsilon}{4\hbar\omega_L}\frac{\Delta\omega}{\omega_R}\log(\frac{\lambda}{2})$, is also important. In fact this can be the dominant correction if λ is suitably small. We did not keep terms of order $\mathcal{O}(\frac{\bar{E}}{V_0})$ above. One could keep them, but that would not alter the conclusion if one neglects terms of order $\mathcal{O}(\tilde{\epsilon}/V_0)$ and $\mathcal{O}(\epsilon/V_0)$. This completes our counter example for the statement which had expressed that the corrections are of order $\tilde{\epsilon}/V_0$.

2.4.2 General Case

For a general double well potential we again divide $\bar{I}$ into $\bar{I}_L$ and $\bar{I}_R$ as in Eqs. (2.72-2.74). Then we use the results of Sec. III of Ref. [14] which deals with a similar integral (Ref. [14] solves the problem of *symmetric* potential. However some of integral calculus done there can be used here if one does division (2.72-2.74) for the action). We combine Eqs. (3.5), (3.10), and (3.11) of [14] for $\bar{I}_L = I_L(\bar{E})$ to obtain

$$\begin{aligned} I_L(\bar{E}) &= I_L(0) - \frac{m\omega_L}{2\hbar}(\bar{a} - x_L)^2 \log\frac{2(x_m - x_L)}{\bar{a} - x_L} \\ &- \frac{m\omega_L}{2\hbar}(\bar{a} - x_L)^2(A_L + \frac{1}{2}) \\ &+ \mathcal{O}((\bar{a} - x_L)^3) \end{aligned} \tag{2.85}$$

where

$$I_L(0) = \frac{1}{\hbar}\int_{x_L}^{x_m}\sqrt{2mV(x)} \tag{2.86}$$

$$A_L = \int_{x_L}^{x_m}\{\frac{m\omega_L}{\sqrt{2mV(x)}} - \frac{1}{x-x_L}\}dx \tag{2.87}$$

A_L is of order unity. For example for a symmetric quartic double well potential $A_L = \log 2$ (see e.g. Sec. V of Ref. [14]). $I_L(0)$ would be half of the action if the potential were symmetric. In deriving Eq.(2.85) one approximates the potential with a parabola near the minimum x_L all the way to the turning point $\bar{a}$. We can use this approximation to write Eq. (2.85) in terms of $\bar{E}$ by noting that $\bar{E} \simeq \frac{1}{2}m\omega_L^2(\bar{a}-x_L)^2$:

$$\begin{aligned} I_L(\bar{E}) &\simeq I_L(0) - \frac{\bar{E}}{2\hbar\omega_L}\log\frac{2(x_m-x_L)}{\sqrt{2\bar{E}/m\omega_L^2}} \\ &- \frac{\bar{E}}{2\hbar\omega_L}(A_L+\frac{1}{2})+\cdots \end{aligned} \tag{2.88}$$

In virtue of Eq. Eq. 2.82 one can observe that $I_L(\bar{E})$ in the above equation has corrections of order $\epsilon/\hbar\omega_L$. To expand $\bar{I}_R = I_R(\bar{E})$ we use the same strategy as in Sec. III of Ref. [14]. The only change that is required is to shift the zero point of potential up by the amount $\tilde{\epsilon}$. Then all the arguments trivially follow and we obtain

$$\begin{aligned} I_R(\bar{E}) &\simeq I_R(0) - \frac{\bar{E}-\tilde{\epsilon}}{2\hbar\omega_R}\log\frac{2(x_R-x_m)}{\sqrt{2(\bar{E}-\tilde{\epsilon})/m\omega_R^2}} \\ &- \frac{\bar{E}-\tilde{\epsilon}}{2\hbar\omega_R}(A_R+\frac{1}{2})+\cdots \end{aligned} \tag{2.89}$$

where

$$I_R(0) = \frac{1}{\hbar}\int_{x_m}^{x_R}\sqrt{2m(V(x)-\tilde{\epsilon})} \tag{2.90}$$

$$A_R = \int_{x_m}^{x_R}\{\frac{m\omega_R}{\sqrt{2m(V(x)-\tilde{\epsilon})}} - \frac{1}{x_R-x}\}dx \tag{2.91}$$

Using Eq. Eq. 2.83 one can see that $I_R(\bar{E})$ also has corrections of order $\epsilon/\hbar\omega_R$. So in general first order correction of order $\tilde{\epsilon}/\hbar\omega$ or $\epsilon/\hbar\omega$ appears in the tunneling amplitude both from the prefactor and the Gamow

factor,

$$\Delta(\tilde{\epsilon}) = \frac{\hbar\sqrt{\omega_R\omega_L}}{\sqrt{e\pi}}(1 + \frac{k}{4}\frac{\epsilon}{\hbar\omega_L}\frac{\omega_R - \omega_L}{\omega_R})e^{-[\bar{I}_L(\tilde{\epsilon})+\bar{I}_R(\tilde{\epsilon})]} \tag{2.92}$$

In some circumstances all the first order corrections cancel. This happens if there is unitary transformation between the Hamiltonians of the same potential with positive and negative bias of the same magnitude as we discuss elsewhere along with the applications of first order correction in tunnel splitting. In general, however, one might expect to get such first order corrections in the tunneling amplitude.

2.5 Conclusion

In conclusion, we did a WKB calculation in this paper to find the tunnel splitting in one dimensional asymmetric potentials. We found that the tunnel splitting can in general have first order dependence to the bias energy. We showed that the dependence is of order $\tilde{\epsilon}/\hbar\omega$ which is greater than $\tilde{\epsilon}/V_0$ which was previously thought.

Chapter 3

Proper truncation of a macroscopic system to a quantum bit

The result of chapter 2 suggests that a noise that causes fluctuations in bias energy of a double well potential may cause fluctuations in the tunneling matrix elements as well. The latter is properly defined when one defines a basis for the Hilbert space of the low energy Hamiltonian of the system, truncates the high energy Hamiltonian and finds the submatrix elements of it in the space of that basis. We shall study this procedure in this chapter for a general system in interaction with quantum or classical environments. The system has an arbitrary self Hamiltonian and we shall not restrict ourselves to the case of double well potentials. We use the results of this chapter in future chapters to find two level Hamiltonian of superconducting and single-molecule magnet qubits.

3.1 Quantum Environment

In many realizations of quantum bits (qubits) the system considered as the qubit has several (more than two) energy levels. However, at low enough temperatures and weak coupling with environment these systems can effectively act as a two level systems. In order to find the effective Hamiltonian of the two level system in interaction with its surrounding environment, one has to start with the total Hamiltonian of the system-environment and truncate the higher states of the system from the Hilbert space to find the effective Hamiltonian. The strategy in practice is as follows: (1) One finds the ground state $|0\rangle$ and first excited state $|1\rangle$ of the system and their energies E_0, E_1. (2) One then evaluates submatrix elements of the total Hamiltonian of system plus environment in the basis of $\{|0\rangle, |1\rangle\}$ and forms the qubit-environment Hamiltonian.

To see this more concrete, we first decompose the total system-environment Hamiltonian into three parts,

$$\hat{H} = \hat{H}_S + \hat{H}_{int} + \hat{H}_{env} \tag{3.1}$$

where $\hat{H}_S$, $\hat{H}_{int}$, and $\hat{H}_{env}$ are the system, interaction and the environment Hamiltonians respectively. Then

we notice that we can write the interaction Hamiltonian as

$$\hat{H}_{int} = \sum_{i=1}^{N} \hat{W}_i \, \hat{\mathcal{E}}_i \tag{3.2}$$

where $\hat{W}_i$ are operators on the system Hilbert space and $\hat{\mathcal{E}}_i$ are operators on the environment Hilbert space. It follows from Eq. 3.1 that $\hat{H}_{int}$ has to be Hermitian. However, for $N \geq 2$, each $\hat{W}_i\hat{\mathcal{E}}_i$ individually does not have to be Hermitian, although they have to add up to a Hermitian operator. In the practical examples that we shall consider, N is finite and small, $N = 1, \cdots, 6$. For a Molecular spin $\hat{W}_i$ are the spin operators, $\hat{S}_x$, $\hat{S}_y$, $\hat{S}_z$, and for a Josephson junctions $\hat{W}_i$ are $\cos\hat{\phi}$ and $\sin\hat{\phi}$ where $\hat{\phi}$ is the phase difference operator of the junction.

Let us apply the truncation strategy and find submatrix of $\hat{H}$, denoted by $\hat{H}_{\rm trunc}$ (for truncated Hamiltonian), in the basis of ground state and first excited states of $\hat{H}_S$, $\{|0\rangle, |1\rangle\}$,

$$\hat{H}_{trunc} = \begin{pmatrix} E_0 & 0 \\ 0 & E_1 \end{pmatrix} + \sum_{i=1}^{N} \begin{pmatrix} W_i^{00} & W_i^{01} \\ W_i^{10} & W_i^{11} \end{pmatrix} \hat{\mathcal{E}}_i + \hat{H}_{env} \tag{3.3}$$

where

$$W_i^{nm} = \langle n|\hat{W}_i|m\rangle. \tag{3.4}$$

In the following we fix the basis of the two level system to $\{|0\rangle, |1\rangle\}$ and define Pauli operators $\tau_0, \tau_x, \tau_y, \tau_z$ in this *fixed basis* as usual and write the Hamiltonians in terms of these operators,

$$\hat{\tau}_0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \quad \hat{\tau}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \hat{\tau}_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \hat{\tau}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{3.5}$$

$$\hat{\tau}^+ = 2\begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \quad \hat{\tau}^- = 2\begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}, \tag{3.6}$$

and write $\hat{H}_{trunc}$ in terms of these operators,

$$\begin{aligned} \hat{H}_{trunc} &= \frac{E_0 - E_1}{2}\hat{\tau}_z + \frac{E_0 + E_1}{2}\hat{\tau}_0 + \hat{\tau}_z \sum_{i=1}^{N} \frac{W_i^{00} - W_i^{11}}{2} \, \hat{\mathcal{E}}_i + \hat{\tau}_0 \sum_{i=1}^{N} \frac{W_i^{00} + W_i^{11}}{2} \, \hat{\mathcal{E}}_i \\ &+ \frac{\hat{\tau}^+}{2} \sum_{i=1}^{N} W_i^{01} \, \hat{\mathcal{E}}_i + \frac{\hat{\tau}^-}{2} \sum_{i=1}^{N} W_i^{10} \, \hat{\mathcal{E}}_i + \hat{H}_{env} \end{aligned} \tag{3.7}$$

A unit operator in Hamiltonian does not play any role and it is equivalent to shifting the zero point of potential which has no physical effect. So one can drop $(E_0 + E_1)\hat{\tau}_0/2$ in the above equation. The same is not true with $\hat{\tau}_0 \sum_{i=1}^{N} \frac{W_i^{00}+W_i^{11}}{2} \, \hat{\mathcal{E}}_i$ because this one is a non-unit operator on the Hilbert space of the

environment. This can be absorbed into the self Hamiltonian of the environment. For clarity we choose not to do that at this stage. We shall see later that when the environment is an oscillator bath, absorption of this term causes a shift in the equilibrium points of the oscillator. However, when the environment is a spin bath, this term acts as a Zeeman term which can be significantly larger than the self Hamiltonian of the spin bath.

3.1.1 System with non-degenerate ground state doublet

Let us first consider the case in which the lowest two states of the system, when isolated from the environment, are non-degenerate. We define the energy splitting between the lowest two levels of the system by

$$\Delta E = E_1 - E_0 \tag{3.8}$$

and after reordering and dropping the obvious $\hat{\tau}_0$ we rewrite Eq. 3.7 as

$$\begin{aligned} \hat{H}_{trunc} &= -\frac{\Delta E}{2}\hat{\tau}_z + \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \hat{\mathcal{E}}_i + \frac{\hat{\tau}^+}{2}\sum_{i=1}^{N} W_i^{01}\ \hat{\mathcal{E}}_i + \frac{\hat{\tau}^-}{2}\sum_{i=1}^{N} W_i^{10}\ \hat{\mathcal{E}}_i \\ &+ \sum_{i=1}^{N}\frac{W_i^{00} + W_i^{11}}{2}\ \hat{\mathcal{E}}_i + \hat{H}_{env} \quad \text{(Qubit-Env., General } \hat{W}_i\text{, non-degenerate case)} \end{aligned} \tag{3.9}$$

This is the truncated Hamiltonian of the system-environment. We shall call it the qubit-environment Hamiltonian. One notices that the Hamiltonian Eq. 3.9 has in general population relaxation or T_1 (as in NMR language) terms as well as pure dephasing or τ_ϕ terms. One of the emphases of this thesis is to show that in some effective Hamiltonians of qubits an existing τ_ϕ term is ignored.

We took $\hat{W}_i$'s general in Eq. 3.9. *If* in a problem, $\hat{W}_i$'s are Hermitian operators, then one can simplify Eq. 3.9 to

$$\begin{aligned} \hat{H}_{trunc} &= -\frac{\Delta E}{2}\hat{\tau}_z + \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \hat{\mathcal{E}}_i + \hat{\tau}_x\sum_{i=1}^{N} W_i^{01}\ \hat{\mathcal{E}}_i \\ &+ \sum_{i=1}^{N}\frac{W_i^{00} + W_i^{11}}{2}\ \hat{\mathcal{E}}_i + \hat{H}_{env} \quad \text{(Qubit-Env., Hermitian } \hat{W}_i\text{, non-degenerate case)} \end{aligned} \tag{3.10}$$

which is the qubit-environment Hamiltonian when the system is non-degenerate and $\hat{W}_i$ are Hermitian.

3.1.2 System with Degenerate ground state doublet

When the ground state doublet of the isolated system is degenerate, the energy splitting vanishes $\Delta E = 0$ in Eq. 3.9 and Eq. 3.10 and the qubit-environment Hamiltonian becomes, for general $\hat{W}_i$

$$\begin{aligned} \hat{H}_{trunc} &= \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \hat{\mathcal{E}}_i + \frac{\hat{\tau}^+}{2}\sum_{i=1}^{N} W_i^{01}\ \hat{\mathcal{E}}_i + \frac{\hat{\tau}^-}{2}\sum_{i=1}^{N} W_i^{10}\ \hat{\mathcal{E}}_i \\ &+ \sum_{i=1}^{N}\frac{W_i^{00} + W_i^{11}}{2}\ \hat{\mathcal{E}}_i + \hat{H}_{env} \quad \text{(Qubit-Env., General } \hat{W}_i\text{, degenerate case)} \end{aligned} \tag{3.11}$$

and for Hermitian $\hat{W}_i$,

$$\begin{aligned} \hat{H}_{trunc} &= \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \hat{\mathcal{E}}_i + \hat{\tau}_x\sum_{i=1}^{N} W_i^{01}\ \hat{\mathcal{E}}_i \\ &+ \sum_{i=1}^{N}\frac{W_i^{00} + W_i^{11}}{2}\ \hat{\mathcal{E}}_i + \hat{H}_{env} \quad \text{(Qubit-Env., Hermitian } \hat{W}_i\text{, degenerate case)} \end{aligned} \tag{3.12}$$

One should note that there is some freedom in choosing states $|0\rangle$ and $|1\rangle$ in the degenerate case because any linear combination of these two is also a ground state. The choice of $|0\rangle$ and $|1\rangle$ will affect the submatrix elements W_i^{nm}.

3.2 Classical Environment

Under certain conditions, the effect of the environment can be considered classically. For example when the environment has only low frequency modes such that for each mode $hf \ll k_B T$, the temperature is high enough that one can adapt classical approximation and treat the environment classically. The total Hamiltonian of system-environment hence simplifies to

$$H(t) = H_S + \sum_{i=1}^{N}\hat{W}_i\ \epsilon_i(t) \tag{3.13}$$

where $\epsilon_i(t)$ is the noise of the environment coupled to $\hat{W}_i$ and is a c-number.

We can apply the same strategy of truncation as before to these cases.

3.2.1 Non-degenerate ground state doublet

The argument follows up to Eq. 3.9 similar to that of preceding sections if we replace $\hat{\mathcal{E}}_i$ by $\epsilon_i(t)$,

$$\begin{aligned}\hat{H}_{trunc}(t) &= -\frac{\Delta E}{2}\hat{\tau}_z + \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \epsilon_i(t) + \frac{\hat{\tau}^+}{2}\sum_{i=1}^{N} W_i^{01}\ \epsilon_i(t) + \frac{\hat{\tau}^-}{2}\sum_{i=1}^{N} W_i^{10}\ \epsilon_i(t) \\ &+ \sum_{i=1}^{N}\frac{W_i^{00} + W_i^{11}}{2}\ \epsilon_i(t) \end{aligned} \tag{3.14}$$

The last term above is a time dependent c-number. Effect of such terms is a global phase factor in the total wavefunction which has no physical significance. So it can be safely dropped:

$$\begin{aligned}\hat{H}_{trunc}(t) &= -\frac{\Delta E}{2}\hat{\tau}_z + \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \epsilon_i(t) + \frac{\hat{\tau}^+}{2}\sum_{i=1}^{N} W_i^{01}\ \epsilon_i(t) + \frac{\hat{\tau}^-}{2}\sum_{i=1}^{N} W_i^{10}\ \epsilon_i(t) \\ &\text{(Qubit-Classical Env., General } \hat{W}_i\text{, non-degenerate case)}\end{aligned} \tag{3.15}$$

For Hermitian $\hat{W}_i$'s the Hamiltonian simplifies to

$$\begin{aligned}\hat{H}_{trunc} &= -\frac{\Delta E}{2}\hat{\tau}_z + \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \epsilon_i(t) + \hat{\tau}_x\sum_{i=1}^{N} W_i^{01}\ \epsilon_i(t) \\ &\text{(Qubit-Classical Env., Hermitian } \hat{W}_i\text{, non-degenerate case)}\end{aligned} \tag{3.16}$$

3.2.2 Degenerate ground state doublet

For the degenerate case $\Delta E = 0$ so we obtain for general $\hat{W}_i$'s

$$\begin{aligned}\hat{H}_{trunc}(t) &= \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \epsilon_i(t) + \frac{\hat{\tau}^+}{2}\sum_{i=1}^{N} W_i^{01}\ \epsilon_i(t) + \frac{\hat{\tau}^-}{2}\sum_{i=1}^{N} W_i^{10}\ \epsilon_i(t) \\ &\text{(Qubit-Classical Env., General } \hat{W}_i\text{, degenerate case)}\end{aligned} \tag{3.17}$$

and for Hermitian $\hat{W}_i$'s

$$\begin{aligned}\hat{H}_{trunc} &= \frac{\hat{\tau}_z}{2}\sum_{i=1}^{N}(W_i^{00} - W_i^{11})\ \epsilon_i(t) + \hat{\tau}_x\sum_{i=1}^{N} W_i^{01}\ \epsilon_i(t). \\ &\text{(Qubit-Classical Env., Hermitian } \hat{W}_i\text{, degenerate case)}\end{aligned} \tag{3.18}$$

Chapter 4

Pure dephasing noise of quasiparticles in Josephson junctions

As an application of the discussions of chapters 2-3, we truncate the interaction Hamiltonian of single and triple Josephson junction flux qubits in this chapter and show that quasiparticles cause pure dephasing (τ_ϕ) noise beside the well recognized population relaxation (T_1) noise. In the literature the quasiparticle noise is recognized as a population relaxation (T_1) noise, when the qubit operates at its optimum point, with usually an ohmic power spectral density function [2, 22]. We show here that the noise produce pure dephasing as well. For the interaction Hamiltonian, we use the results of the study of the microscopic theory done by Eckern , Schön, and Ambegaokar [23]. This result has been cited by Leggett et. al. [2, p. 9], Prokofev and Stamp [3, p. 678], and Weiss [22, p. 21], however the spin-boson model, which has only a T_1 process, has dominated the literature of quasiparticle noise.

A possible reason for this may be a discussion in [2, p. 14] which casts a general form of noise in flux qubits into the spin boson model. It seems, however, that the discussion of [2, p. 14] is not appropriate for at least modern flux qubits. There are two points that one can make in this regard: (1) Ref. [2] studies the flux qubit problem in the WKB approximation limit. But many applications of Josephson junctions do not operate in this limit [24]. (2) The discussion of [2, p. 14] incorporates a pure dephasing term into the renormalized tunnel splitting of the flux qubit. This, however, may not be all accurate. Because a renormalized Hamiltonian of the flux qubit in absence of any other noise will undergo coherent tunneling with no dephasing. But, the the original unnormalized Hamiltonian of the flux qubit with a pure dephasing noise term will undergo phase damping. The resultant evolutions are different, suggesting that the incorporation of pure dephasing into renormalization of tunnel splitting may not be quite accurate, at least in non-WKB limit.

Instead of incorporating pure dephasing terms into renormalization of tunnel splitting, we shall derive the truncated Hamiltonian with all of its population relaxation and pure dephasing noise terms for single and triple junction flux qubits in this chapter and discuss the significance of pure dephasing terms in each case. We shall see that the effect of such terms is more important in the triple junction flux qubits and in

the non-WKB limit.

4.1 Single Josephson junction flux qubit

Consider a flux qubit with single junction in interaction with environment. The system Hamiltonian is

$$H_{sys} = \frac{\hat{Q}^2}{2C} + \frac{(\hat{\Phi} - \Phi_{ext})^2}{2L} + E_J(1 - \cos\hat{\phi}) \tag{4.1}$$

where C, $\hat{\Phi}$, Φ_{ext} , L, E_J and $\hat{\phi}$ are respectively the capacitance of the junction, total flux in the loop, the external flux in the loop, the self geometrical inductance of the ring, the Josephson junction energy and the phase difference of two superconductors on two side of the junction. $\hat{Q}$ is the half of charge difference of the two superconductors and can be written in terms of half of the number difference of Cooper pairs on two superconductors $\hat{N}$ as

$$\hat{Q} = 2e\hat{N} \tag{4.2}$$

It can be shown[25] that

$$\hat{N} = i\frac{\partial}{\partial\hat{\phi}} \quad [\hat{\phi}, \hat{N}] = -i \tag{4.3}$$

The flux quantization relationship requires

$$\frac{\Phi}{\Phi_0} = \frac{\phi}{2\pi} \tag{4.4}$$

where $\Phi_0 = h/2e$ is the flux quantum. For $\Phi_{ext} = \Phi_0/2$ the potential in Eq. (4.1) becomes a symmetric double well potential if E_J is suitably large. One can define $\phi_{ext} = 2\pi\Phi_{ext}/\Phi_0$ to write the potential in terms of the phase of the junction

$$V_{sys} = E_L\frac{(\phi - \phi_{ext})^2}{2} + E_J(1 - \cos\phi) \tag{4.5}$$

where $E_L = \Phi_0^2/4\pi^2 L$. This potential is symmetric for $\phi_{ext} = \pi$.

We consider the case when the single junction qubit operates at its optimum point here ($\phi_{ext} = \pi$) and study quasiparticles effects. Eckern , Schön, and Ambegaokar [23] showed that the interaction of a Josephson junction with qusiparticles can be casted in the following form

$$H_{int} = \cos(\frac{\phi}{2})\sum_i C_{i,1}R_{i,1} + \sin(\frac{\phi}{2})\sum_j C_{j,2}R_{j,2} \tag{4.6}$$

where ϕ is the phase of the Josephson junction, $\{R_{i,1}\}$ and $\{R_{i,2}\}$ are coordinates of two independent ensembles of harmonic oscillators, $\{C_{i,1}\}$, $\{C_{i,2}\}$ are real-valued coefficients. The self Hamiltonian of such environment is

$$\begin{aligned} H_{env} &= \sum_i \frac{P_{i,1}^2}{2M_{i,1}} + \frac{1}{2}M_{i,1}\omega_{i,1}^2 R_{i,1}^2 \quad (4.7) \\ &+ \sum_j \frac{P_{j,2}^2}{2M_{j,2}} + \frac{1}{2}M_{j,2}\omega_{j,1}^2 R_{j,2}^2 \quad (4.8) \end{aligned}$$

The spectral densities of these two bath are relatively equal and ohmic in many cases when the variations of phase are slow on a time scale given by the inverse of the superconducting gap [23, p. 6426-6427].

To truncate the interaction Hamiltonian of Eq. 4.6 we notice that each term in this Hamiltonian is individually Hermitian. Thus, we can use Eq. 3.10 for the truncation procedure. The result is

$$\begin{aligned} \hat{H}_{trunc} &= -\frac{\Delta E}{2}\hat{\tau}_z + \frac{\hat{\tau}_z}{2}(c^{00}-c^{11})\sum_i C_{i,1}R_{i,1} + \hat{\tau}_x\ c^{01}\sum_i C_{i,1}R_{i,1} \\ &+ \frac{\hat{\tau}_z}{2}(s^{00}-s^{11})\sum_i C_{i,2}R_{i,2} + \hat{\tau}_x\ s^{01}\sum_i C_{i,2}R_{i,2} \\ &+ \frac{c^{00}+c^{11}}{2}\ \sum_i C_{i,1}R_{i,1} + \frac{s^{00}+s^{11}}{2}\ \sum_i C_{i,2}R_{i,2} + \hat{H}_{env} \quad (4.9) \end{aligned}$$

where

$$c^{nm} = \langle n|\cos\frac{\hat{\phi}}{2}|m\rangle \quad (4.10)$$

$$s^{nm} = \langle n|\sin\frac{\hat{\phi}}{2}|m\rangle \quad (4.11)$$

The term $\frac{c^{00}+c^{11}}{2}\ \sum_i C_{i,1}R_{i,1} + \frac{s^{00}+s^{11}}{2}\ \sum_i C_{i,2}R_{i,2}$ can be absorbed into H_{env} by appropriate shift of the center of oscillation of each oscillator in the baths, $R_{j,n} \rightarrow R'_{j,n}$. This, of course, produces an overall constant, c-number, term which can always be dropped since it does not affect the dynamics. Therefore, one reaches to

$$\begin{aligned} \hat{H}_{trunc} &= -\frac{\Delta E}{2}\hat{\tau}_z + \frac{\hat{\tau}_z}{2}(c^{00}-c^{11})\sum_i C_{i,1}R'_{i,1} + \hat{\tau}_x\ c^{01}\sum_i C_{i,1}R'_{i,1} \\ &+ \frac{\hat{\tau}_z}{2}(s^{00}-s^{11})\sum_i C_{i,2}R'_{i,2} + \hat{\tau}_x\ s^{01}\sum_i C_{i,2}R'_{i,2} + \hat{H'}_{env} \quad (4.12) \end{aligned}$$

Some of the coefficients are zero in the above equation. One can see this easily by using shifted phase

quantity $\bar{\phi} = \phi - \pi$. In terms of this quantity,

$$\sin\phi/2 = \cos\bar{\phi}/2 \tag{4.13}$$

$$\cos\phi/2 = -\sin\bar{\phi}/2 \tag{4.14}$$

The ground state $\langle\bar{\phi}|0\rangle$ is an even function of $\bar{\phi}$ and the first excited state $\langle\bar{\phi}|1\rangle$ is an odd function of $\bar{\phi}$. Since, $\sin\bar{\phi}/2$ is odd and $\cos\bar{\phi}/2$ is even, we conclude that

$$s^{01} = c^{00} = c^{11} = 0. \tag{4.15}$$

Using Eq. 4.15 the truncated Hamiltonian 4.12 is simplified to

$$\hat{H}_{trunc} = -\frac{\Delta E}{2}\hat{\tau}_z + \frac{\hat{\tau}_z}{2}(s^{00} - s^{11})\sum_i C_{i,2}R'_{i,2} + \hat{\tau}_x\ c^{01}\sum_i C_{i,1}R'_{i,1} + \hat{H'}_{env}. \tag{4.16}$$

The second term above is a pure dephasing term and the third term is a population relaxation term. $s^{00} - s^{11}$ is not zero, although it may be much smaller than c^{01}. In the non-WKB limit it is more accurate to include this term in noise models.

In the weak noise limit, we can add the effect of each noise term in Eq. 4.16 to obtain total relaxation rate, $1/T_1$, and total dephasing rate, $1/T_2$. If we denote the power spectral density function of the first oscillator bath by $S_1(\omega)$ and that of the second bath by $S_2(\omega)$ then [26, 4, 22]

$$\frac{1}{T_1} = \pi(c^{01})^2\ S_1(\Delta E) \tag{4.17}$$

$$\frac{1}{\tau_\phi} = \pi(\Delta s)^2\ S_2(0) \tag{4.18}$$

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{\tau_\phi} \tag{4.19}$$

where $\Delta s = (s^{00} - s^{11})/2$. Compared to previous analyses of flux qubits with single Josephson junctions that operates at its optimum point, the new term that we found in this section is the pure daphasing $1/\tau_\phi$ term above. This term is negligible in the WKB limit. However in non-WKB limit, where most of current qubtis operate, one may need to include it in their noise models to obtain more accurate models to interpret experimental data.

4.2 Triple Josephson junction flux Qubit

In most applications of Josephson junctions in quantum information processing there are more than one junction involved. In this section we study dissipation and decoherence in triple junction flux qubits. Consider a three-junction qubit with junctions 1 and 2 that have Josephson energies E_J and capacitance C and junction 3 that has a Josephson energy and capacitance α times larger (Fig. 4.1). The node islands 1 and 2 in Fig.

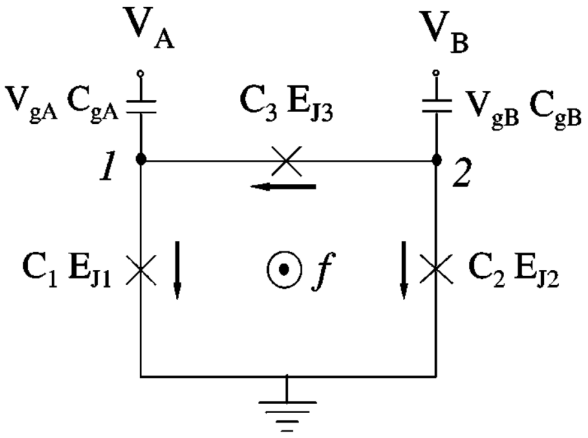

Figure 4.1: Triple Junction flux qubit. Courtesy of Ref. [27].

4.1 are connected to gate voltages V_A and V_B through capacitors with capacitances $C_{gA} = C_{gB} = \gamma C$. The superconducting loop is threaded by an external magnetic flux $f\Phi_0$, where $f \in [0, 1]$ and $\Phi_0 = h/2e$ is the superconducting flux quantum. The inductance of the loop is assumed to be negligible compared to $L_J = \Phi_0/2\pi I_0$, where I_0 is critical current of the junction 1. The potential energy of this system is [27]

$$U = E_J[2 + \alpha - \cos\phi_1 - \cos\phi_2 - \alpha\cos(2\pi f + \phi_1 - \phi_2)] \tag{4.20}$$

where ϕ_1 and ϕ_2 are phase differences in junctions 1 and 2, respectively. The total Hamiltonian of this system can be quantized and written as follows [27]

$$H_{sys} = \frac{P_p^2}{2M_p} + \frac{P_m^2}{2M_m} + E_J\{2 + \alpha - 2\cos\phi_p\cos\phi_m - \alpha\cos(2\pi f + 2\phi_m)\} \tag{4.21}$$

where $\phi_p = (\phi_1 + \phi_2)/2$, $\phi_m = (\phi_1 - \phi_2)/2$, $P_p = -i\hbar\partial/\partial\phi_p$, $P_m = -i\hbar\partial/\partial\phi_m$, $M_p = (\Phi_0/2\pi)^2 2C(1 + \gamma)$ and $M_m = (\Phi_0/2\pi)^2 2C(1 + 2\alpha + \gamma)$. The optimum point of operation of this qubit is at $f = 1/2$. At that point the potential makes a double well landscape in two dimensions when $\alpha > 1/2$. The minima in energy occur at $(\phi_p, \phi_m) = (0, \pm\arccos\frac{1}{2\alpha})$. The ground state $\Psi_0(\phi_p, \phi_m)$ is an even function in ϕ_m, $\Psi_0(\phi_p, \phi_m) =$

$\Psi_0(\phi_p, -\phi_m)$ and the first excited state $\Psi_1(\phi_p, \phi_m)$ is an odd function in ϕ_m, $\Psi_1(\phi_p, \phi_m) = -\Psi_1(\phi_p, -\phi_m)$.

As before, each Josephson junction interacts with two independent oscillator baths. The interaction Hamiltonian for all three junctions is

$$\begin{aligned} H_{int} &= \cos\frac{\phi_1}{2}\sum_i C_{i,1}R_{i,1} + \sin\frac{\phi_1}{2}\sum_j C_{j,2}R_{j,2} \\ &+ \cos\frac{\phi_2}{2}\sum_i C_{i,3}R_{i,3} + \sin\frac{\phi_2}{2}\sum_j C_{j,4}R_{j,4} \\ &+ \cos\frac{\phi_3}{2}\sum_i C_{i,5}R_{i,5} + \sin\frac{\phi_3}{2}\sum_j C_{j,6}R_{j,6} \end{aligned} \tag{4.22}$$

We can use the flux quantization relation $\phi_3 = -2\pi f - \phi_1 + \phi_2$ [27] as in Eq. 4.20 to eliminate the phase of the third junction ϕ_3. Also, we can utilize the relation between ϕ_1, ϕ_2 and ϕ_p, ϕ_m to write H_{int} as follows

$$\begin{aligned} H_{int} &= \cos\frac{\phi_p+\phi_m}{2}\sum_i C_{i,1}R_{i,1} + \sin\frac{\phi_p+\phi_m}{2}\sum_j C_{j,2}R_{j,2} \\ &+ \cos\frac{\phi_p-\phi_m}{2}\sum_i C_{i,3}R_{i,3} + \sin\frac{\phi_p-\phi_m}{2}\sum_j C_{j,4}R_{j,4} \\ &- \sin\phi_m\sum_i C_{i,5}R_{i,5} - \cos\phi_m\sum_j C_{j,6}R_{j,6} \end{aligned} \tag{4.23}$$

Here we set $f = 1/2$, since we are interested in new effects that have not been recognized in the literature for the optimum point, which has the minimum decoherence.

We can now truncate H_{int} and find its matrix elements in the basis of the ground state and first excited state of the system. The result is

$$\begin{aligned} H_{int,trunc} &= \frac{\hat{\tau}_z}{2}(c_+^{00} - c_+^{11})\sum_i C_{i,1}R_{i,1} + \hat{\tau}_x c_+^{01}\sum_i C_{i,1}R_{i,1} \\ &+ \frac{\hat{\tau}_z}{2}(s_+^{00} - s_+^{11})\sum_i C_{i,2}R_{i,2} + \hat{\tau}_x s_+^{01}\sum_i C_{i,2}R_{i,2} \\ &+ \frac{\hat{\tau}_z}{2}(c_-^{00} - c_-^{11})\sum_i C_{i,3}R_{i,3} + \hat{\tau}_x c_-^{01}\sum_i C_{i,3}R_{i,3} \\ &+ \frac{\hat{\tau}_z}{2}(s_-^{00} - s_-^{11})\sum_i C_{i,4}R_{i,4} - \hat{\tau}_x s_-^{01}\sum_i C_{i,4}R_{i,2} \\ &- \frac{\hat{\tau}_z}{2}(s_m^{00} - s_m^{11})\sum_i C_{i,5}R_{i,5} - \hat{\tau}_x s_m^{01}\sum_i C_{i,5}R_{i,5} \\ &- \frac{\hat{\tau}_z}{2}(c_m^{00} - c_m^{11})\sum_i C_{i,6}R_{i,6} + \hat{\tau}_x c_m^{01}\sum_i C_{i,6}R_{i,6} \end{aligned} \tag{4.24}$$

plus terms that can be absorbed into the self Hamiltonian of the oscillator baths. In Eq. 4.24

$$
\begin{aligned}
c_{\pm}^{mn} &= \langle m| \cos \frac{\phi_p \pm \phi_m}{2} |n\rangle && (4.25)\\
s_{\pm}^{mn} &= \langle m| \sin \frac{\phi_p \pm \phi_m}{2} |n\rangle && (4.26)\\
c_{m}^{mn} &= \langle m| \cos \phi_m |n\rangle && (4.27)\\
s_{m}^{mn} &= \langle m| \sin \phi_m |n\rangle. && (4.28)
\end{aligned}
$$

Since the ground state is even and the first excited state is odd with respect to ϕ_m, we have

$$c_m^{01} = s_m^{00} = s_m^{11} = 0. \tag{4.29}$$

We cannot extend this argument to $c_{\pm}^{mn}$ and $s_{\pm}^{mn}$ because $\cos \frac{\phi_p \pm \phi_m}{2}$, $\sin \frac{\phi_p \pm \phi_m}{2}$ are neither even nor odd with respect to ϕ_m.

For brevity, let us define

$$
\begin{aligned}
\Delta c_l &= (c_l^{00} - c_l^{11})/2 && (4.30)\\
\Delta s_l &= (s_l^{00} - s_l^{11})/2 && (4.31)\\
& && (4.32)
\end{aligned}
$$

where $l = \pm, m$. The total truncated Hamiltonian can now be written as

$$
\begin{aligned}
H_{trunc} &= -\frac{\Delta E}{2}\hat{\tau}_z \\
&+ \hat{\tau}_z\ \Delta c_+ \sum_i C_{i,1}R_{i,1} + \hat{\tau}_x\ c_+^{01} \sum_i C_{i,1}R_{i,1} \\
&+ \hat{\tau}_z\ \Delta s_+ \sum_i C_{i,2}R_{i,2} + \hat{\tau}_x\ s_+^{01} \sum_i C_{i,2}R_{i,2} \\
&+ \hat{\tau}_z\ \Delta c_- \sum_i C_{i,3}R_{i,3} + \hat{\tau}_x\ c_-^{01} \sum_i C_{i,3}R_{i,3} \\
&+ \hat{\tau}_z\ \Delta s_- \sum_i C_{i,4}R_{i,4} + \hat{\tau}_x\ s_-^{01} \sum_i C_{i,4}R_{i,4} \\
&- \hat{\tau}_z\ \Delta c_m \sum_i C_{i,6}R_{i,6} - \hat{\tau}_x\ s_m^{01} \sum_i C_{i,5}R_{i,5} \\
&+ H_{env} && (4.33)
\end{aligned}
$$

where H_{env} composed of six oscillator baths self Hamiltonians.

We expect the extra pure dephasing terms that is found in Eq. 4.33 to be more significant than the one

in the single qubit junction because the triple junction wave functions in two dimensions have less symmetry than the ones of single junction in one dimension. Therefore, the quantities Δc_l, Δs_l should be somewhat larger than Δs in the single junction.

As in the case of the single junction, in the weak coupling limit we can add the effects of each oscillator bath above to find the relaxation and dephasing rate as follows

$$\frac{1}{T_1} = \pi\{(c_+^{01})^2 S_1(\Delta E) + (s_+^{01})^2 S_2(\Delta E) + (c_-^{01})^2 S_3(\Delta E) + (s_+^{01})^2 S_4(\Delta E) + (s_m^{01})^2 S_5(\Delta E)\} \quad (4.34)$$

$$\frac{1}{\tau_\phi} = \pi\{\Delta c_+^2 S_1(0) + \Delta s_+^2 S_2(0) + \Delta c_-^2 S_3(0) + \Delta s_-^2 S_4(0) + \Delta c_m^2 S_6(0)\} \quad (4.35)$$

In comparison with the literature all the terms in Eq. 4.35 are new and have not been recognized to my knowledge. These terms have non-negligible effects in the non-WKB limit.

This completes our discussion of extra pure dephasing terms in superconduting qubits.

4.3 Conclusion

In conclusion, in this chapter we used the idea of chapter 2 and the method of chapter 3 to find the two level system Hamiltonian of single and triple junction flux qubits in interaction with quasiparticles. We utilized the high energy Hamiltonian of quasiparticles interaction found by Eckern , Schön, and Ambegaokar [23] and truncated it to find the submatrix elements of the interaction Hamiltonian in the basis of ground state and first excited state of the flux qubit. We found that in addition to population relaxation noise, the interaction causes pure dephasing noise for two level systems. This pure dephasing noise has not been recognized, or well treated in my opinion, in the literature. We discussed that the pure dephasing noise is somewhat more significant for triple junction qubits than is for single junction ones and is more important in the non-WKB limit. The application of this study is in experiments that temperature is comparable with the superconducting energy gap.

Chapter 5

Landau-Zener effect in presence of time dependent off-diagonal terms in Hamiltonian

In this chapter I present an analytical solution of the Landau-Zener type problem when an extra linear term is added to the off-diagonal element of the Hamiltonian. I show such term qualitatively leave the Landau-Zener effect unaffected while quantitatively modifies the probability of transition. I also construct an example and present numerical solution of a sweep problem in presence of an oscillatory term in the off diagonal matrix element of the Hamiltonian. I demonstrate that a frequency of oscillation as low as twice the minimum gap can dramatically affect the result of the evolution and make an otherwise adiabatic evolution, diabatic. This effect as we see is more influential than increasing the speed of the sweep for a similar factor in the Landau-Zener problem.

5.1 Introduction

Landau-Zener theory [28, 29] provides a powerful method for measuring the minimum gap between two energy levels of a system. It was this method which enabled researchers to measure the extremely small tunnel splitting in single molecule magnets with relatively large spin S, such as Fe8 ($S = 10$), Mn4 ($S = 8$), Mn4 ($S = 9/2$), at the first place [11, 30]. Landau-Zener (LZ) effect has recently gained renewed attention in the context of adiabatic quantum computation [31, 32].

In this chapter, we study two problems: (1) The effect of an extra linear off-diagonal term, proportional to the bias energy, in the Hamiltonian of Landau-Zener problem. (2) The effect of an oscillatory off-diagonal term in the Hamiltonian of LZ problem.

We show that in the first problem the LZ effect is qualitatively unaffected, however, it is quantitatively modified. In the second problem, we illustrate that for some choice of the frequency of the oscillatory term, the effect is even qualitatively influenced.

5.2 Effect of Linearly time dependent off-diagonal term in the Landau-Zener Hamiltonian

In problems that the linear effect occurs, as discussed in the previous chapters, the Hamiltonian for the truncated two level system is

$$H = -\frac{\hbar\Delta}{2}(1 + c\frac{\tilde{\epsilon}}{\Delta})\ \sigma_x - \frac{\hbar\tilde{\epsilon}}{2}\ \sigma_z \tag{5.1}$$

where c is a constant and typically $c \ll 1$.

Here we study the correction that the extra term above $-c\ \hbar\tilde{\epsilon}\ \sigma_x/2$ makes in Landau-Zener effect. We vary the bias by a constant rate as

$$\tilde{\epsilon}(t) = vt \tag{5.2}$$

The Hamiltonian becomes, in the σ_z-basis,

$$H(t) = -\frac{\hbar}{2}\begin{pmatrix} vt & \Delta + c\ vt \\ \Delta + c\ vt & -vt \end{pmatrix}. \tag{5.3}$$

Suppose the system is in the ground state of $H(-T/2)$ at $t = -T/2$, for T being arbitrarily large. What is the probability of finding the system in the excited state of $H(T/2)$ at $t = T/2$?

When $c = 0$ the problem becomes the standard Landau-Zener problem and the answer is

$$P = \exp\left[-\frac{\pi\Delta^2}{2v}\right] \tag{5.4}$$

For $v \to \infty$ the probability is one. This is because the excited state of $H(T/2)$ is the same as the ground state of $H(-T/2)$ and in a fast sweep the state does not change. For $v \to 0$ the probability is zero. This is a result of the adiabatic theorem which states that the system stays in the instantaneous ground state of the Hamiltonian if the Hamiltonian varies slowly by time [33, 34, 35]. For a moderate sweep rate the probability of transition is according to Eq. 5.4. Kayanuma shows that the transition time is of the order of $\Delta/2v$ around $t = 0$ [36].

We show below that when $c \neq 0$, Eq. Eq. 5.4 is modified and the probability of transition to the excited state of the final Hamiltonian becomes

$$P = \exp\left[-\frac{\pi\Delta^2}{2v(1+c^2)^{3/2}}\right]. \tag{5.5}$$

Eq. Eq. 5.5 well satisfies the limiting cases, as before. As $v \to 0$, $P \to 1$ in accordance with the adiabatic

theorem. Also, as $v \to \infty$, $P \to 0$ in accordance with the fast sweep argument discussed above.

The (unnormalized) ground state of $H(-T/2)$ and excited state of $H(T/2)$ for arbitrary large T, in σ_z-basis, is

$$|g_{-\infty}\rangle = |e_{+\infty}\rangle = \begin{pmatrix} \frac{1-(1+c^2)^{1/2}}{c} \\ 1 \end{pmatrix} \tag{5.6}$$

We note that in the limit of $c \to 0$ this state becomes $|\downarrow\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$, which is the spin down state as expected in the standard LZ problem. The unnormalized excited state of $H(-T/2)$ and ground state of $H(T/2)$ is

$$|e_{-\infty}\rangle = |g_{+\infty}\rangle = \begin{pmatrix} 1 \\ \frac{c}{1+(1+c^2)^{1/2}} \end{pmatrix} \tag{5.7}$$

which becomes the spin up state $|\uparrow\rangle$ in $c \to 0$ limit.

In solving the problem, we follow the method of Landau and Lifshitz given in Secs. 52, 53 and 90 of Ref. [29] for the standard LZ problem. We denote the instantaneous eigenenergies of Hamiltonian $H(t)$, by $U_g(t)$ and $U_e(t)$. The instantaneous energy splitting between the two levels is

$$\Delta U(t) = U_e(t) - U_g(t) = \hbar\sqrt{(vt)^2 + (\Delta + cvt)^2} \tag{5.8}$$

According to Ref. [29, p. 349-350], quite generally, the probability P of transition to the excited state of $H(T/2)$ is

$$P = \exp\left[-\frac{2}{\hbar}\,\mathrm{Im}\int_{t_1}^{t_0} \Delta U(t)dt\right] \tag{5.9}$$

where t_1 may be taken to be any point on the real axis, t_0 is the complex number in the upper half plane for which

$$\Delta U(t_0) = 0 \tag{5.10}$$

and integration is on the path that starts at t_1, goes vertically on the real axis to $\mathrm{Re}(t_0)$ and from there horizontally parallel to imaginary axis to t_0 (See e.g. Fig. 19 of Sec. 52 of Ref. [29])

To find t_0 we first complete the square in $\Delta U(t)$ as follows,

$$\Delta U(t) = \hbar\sqrt{v^2(1+c^2)\left(t + \frac{c\Delta}{v(1+c^2)}\right)^2 + \frac{\Delta^2}{1+c^2}} \tag{5.11}$$

Then we choose t_1 to be

$$t_1 = -\frac{c\Delta}{v(1+c^2)}. \tag{5.12}$$

and define

$$\tilde{v} \equiv v\,(1+c^2)^{1/2} \tag{5.13}$$

$$\tilde{\Delta} \equiv \Delta\,(1+c^2)^{-1/2} \tag{5.14}$$

to rewrite $\Delta U(t)$ as

$$\Delta U(t) = \hbar\sqrt{\tilde{v}^2(t-t_1)^2 + \tilde{\Delta}^2}. \tag{5.15}$$

Now t_0 can be easily obtained as follows,

$$t_0 = t_1 + i\frac{\tilde{\Delta}}{\tilde{v}} \equiv t_1 + i\tau_0. \tag{5.16}$$

To take the required integral

$$\int_{t_1}^{t_1+i\tau_0} \sqrt{\tilde{v}^2(t-t_1)^2 + \tilde{\Delta}^2}\; dt \tag{5.17}$$

we first substitute $t - t_1 = s$,

$$\int_0^{i\tau_0} \sqrt{\tilde{v}^2 s^2 + \tilde{\Delta}^2}\; ds, \tag{5.18}$$

then, since the integration on s is entirely on the imaginary axis, we substitute $s = i\tau$ to get

$$i\int_0^{\tau_0} \sqrt{-\tilde{v}^2\tau^2 + \tilde{\Delta}^2}\; d\tau = i\frac{\pi\tilde{\Delta}^2}{4\tilde{v}}. \tag{5.19}$$

Substituting Eq. 5.19 into Eq. 5.9 we find the probability of transition,

$$P = \exp\left[-\frac{\pi\tilde{\Delta}^2}{2\tilde{v}}\right] = \exp\left[-\frac{\pi\Delta^2}{2v(1+c^2)^{3/2}}\right]. \tag{5.20}$$

In comparison to the transition probability Eq. 5.4 in the standard LZ problem, the above expression has an extra factor of $(1+c^2)^{-3/2}$ in the exponent. Thus, if one conducts an experiment to measure Δ, and one is unaware of the presence of the extra term $-c\;\hbar\tilde{\epsilon}\;\sigma_x/2$ in Hamiltonian Eq. 5.1, one will underestimate Δ by a factor of $(1+c^2)^{3/4}$.

5.3 Effect of oscillatory off-diagonal term in the Landau-Zener Hamiltonian

In this section we construct an example that illustrates that the adiabatic quantum computation is strongly influenced in presence of oscillatory terms in the Hamiltonian. It is usually considered in adiabatic quantum computing that the important factor which determines the adiabaticity of the evolution is the size of the minimum gap between lowest energy levels (at least when considering the closed system) [32]. We, however, show that in presence of oscillatory terms this is not true.

We first construct a Landau-Zener type example in the slow sweep rate regime which behaves adiabatically. Then we turn on an oscillatory term in the off-diagonal element of the Hamiltonian with oscillation frequency as slow as twice the minimum gap. We show this small change makes the evolution diabatic.

Strictly speaking, we do not strikingly violate the adiabatic theorem here. However, we show that what determines the speed of sweep for an adiabatic evolution in a typical situation that deals with an avoided crossing, is not only the size of the gap but also the existence of oscillatory terms in the Hamiltonian.

Consider the following time dependent Hamiltonian

$$H(t) = -\frac{\hbar\Delta}{2}\left(1 + a[1 - \cos(2\Delta t)]\right)\ \sigma_x - \frac{\hbar v t}{2}\ \sigma_z \tag{5.21}$$

For $a = 0$ this is the standard Landau-Zener Hamiltonian. The ground state of $H(-T/2)$ for large T is $|\downarrow\rangle$. Using dimensionless parameter $s = t/T$, we denote the probability of the spin to be found in state $|\downarrow\rangle$ at time t by $P_{\downarrow\downarrow}(s)$. This becomes at $t = T/2$ equal to the probability of transition to the excited state of the final Hamiltonian defined in the previous section

$$P = P_{\downarrow\downarrow}(1/2) \tag{5.22}$$

because the excited state of $H(T/2)$ is the same as the ground state of $H(-T/2)$, as mentioned before. In this section we solve the Schrodinger equation for Hamiltonian Eq. 7.3 by use of numerical techniques. Fig. 5.1 illustrates $P_{\downarrow\downarrow}(s)$ for $a = 0$ and a slow sweep rate $v = \Delta^2/4$. The probability of staying in the spin down state at late times $P(s = 1/2)$ matches very well with analytical expression of Eq. Eq. 5.4, $P = e^{-2\pi} \simeq 0.0018$.

Now we turn on the oscillatory term in Eq. Eq. 7.3 and set $a = 1$. The off-diagonal term oscillates between $-\hbar\Delta/2$ and $-3\hbar\Delta/2$. It never becomes zero, so the gap between the two energy levels is always open. It is modulated between two values of $\hbar\Delta$ and $3\hbar\Delta$ throughout the process, periodically. This modulation, however, has a strong effect on the evolution of the system. It excites the system out of its instantaneous

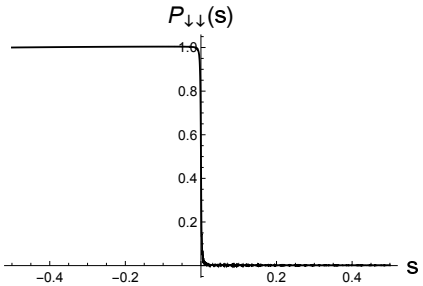

Figure 5.1: Probability of being in spin down state $P_{\downarrow\downarrow}(s)$ in the standard Landau-Zener problem as a function of dimensionless time variable s, for $v = \Delta^2/4$. Parameters of the simulation are set at $T = 2000$, $a = 0$, and $v = \Delta^2/4 = 0.25$.

ground state. Fig. 5.2 illustrates the result of such evolution.

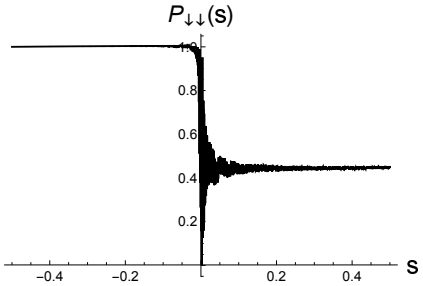

Figure 5.2: Probability of being in spin down state $P_{\downarrow\downarrow}(s)$ in the Landau-Zener problem with extra off-diagonal oscillatory term, Eq. Eq. 7.3, as a function of $s = t/T$, for $v = \Delta^2/4$. Parameters of the simulation are set at $T = 2000$, $a = 1$, and $v = \Delta^2/4 = 0.25$.

The system remains in the instantaneous ground state until it reaches the avoided crossing. At that place it gets partially excited and transitions partly to the instantaneous excited state. As the levels diverge the part that transitioned to the excited state remains there until the evolution ends. At the end, the probability of transition becomes over 45% and the probability of remaining in the instantaneous ground state is less than 55%. Thus, although the sweep rate is slow, the system does not remain in the instantaneous ground state.

In adiabatic quantum computing, the main factor that is usually considered as a measure of slowness of the process is the minimum gap between the ground level and excited levels [32]. If the sweep rate v is small compared to this gap squared Δ^2 then the process is recognized as adiabatic (for a closed system). That is the evolution takes the system from a trivial initial ground state to the desired final ground state [31]. However, we observed in this section that Δ^2 alone is not determinant. The presence of oscillatory terms in the Hamiltonian can strongly affect the result and lead to diabatic evolution even if the sweep rate is slow.

We shall discuss below the validity of adiabatic theorem for our example. However, the point we wish to make here is not whether the adiabatic theorem is technically violated or not. But, the observation that hidden oscillatory terms in a Hamiltonian can ruin adiabatic computation. One, who conducts adiabatic quantum processing, may not be, a priori, aware of existence of such oscillatory terms. This will lead them to make unsuitable choices for the sweep rate that in turn leads to incorrect results for adiabatic quantum computation!

Let us now examine the validity of the adiabatic theorem in the above problem. The adiabatic theorem states [33, 34, 35] that if the system starts in an instantaneous energy eigenstate $|E_n(-T/2)\rangle$, it will remain in that energy level $|E_n(t)\rangle$ throughout the evolution as long as

$$I = \max_{-T/2 \le t \le T/2} \left| \frac{\langle E_m(t)|dH/dt|E_n(t)\rangle}{E_{nm}^2(t)/\hbar} \right| \ll 1 \quad \text{for all } m \neq n \tag{5.23}$$

where $E_{nm}(t) = E_n(t) - E_m(t)$. As we see in Fig. 5.2 the major non-adiabatic events occur near $s = 0 - 0.03$. Thus, if the adiabatic theorem Eq. 5.23 is violated it is most likely violated in this region. Time derivative of Hamiltonian Eq. 7.3 is

$$\frac{dH(t)}{dt} = -\hbar\Delta^2 a \sin(2\Delta t)\ \sigma_x - \frac{\hbar v}{2}\ \sigma_z \tag{5.24}$$

At $t = 0$, the ground state and excited state are the eigenstates of σ_x, $|\pm\rangle$, for both $a = 0, 1$. At that time the gap between the two levels is Δ for both values of a. The condition Eq. 5.23 becomes

$$I = \frac{v/2}{\Delta^2} \ll 1 \tag{5.25}$$

We notice on passing that the Landau-Zener probability can be written as

$$P = \exp(-\pi/4I) \tag{5.26}$$

For $v = \Delta^2/4$, I is

$$\frac{1}{8} = 0.125 \ll 1 \tag{5.27}$$

which we consider as being satisfactory enough. Here

$$P = e^{-\pi/4I} = e^{-2\pi} \sim 0.0018. \tag{5.28}$$

For $a = 0$, $I = 1/8$ in Eq. 5.23. For $a = 1$, I comes from a time near the time that $vt = \Delta$,

$$I = \frac{\hbar\Delta^2/\sqrt{2}}{2\hbar\Delta^2} = \frac{1}{2\sqrt{2}} \simeq 0.35. \tag{5.29}$$

If this was a Landau Zener problem with $I = 0.35$ the probability P would be

$$P = e^{-\pi/4I} \sim 0.10 \tag{5.30}$$

However, as one sees from Fig. 5.2, $P_{\downarrow\downarrow}$ for late times reaches to about 0.45. This means that the oscillation more violate the adiabaticity than a fast sweep in a Landau Zener type experiment.

The other point, as mentioned above, is that what determines adiabaticity is not the minimum gap, but also existence of oscillatory terms and one a priori may not be aware of such existence because one does not know the instantaneous ground state and first excited state of the system at all times. The goal of adiabatic quantum computation is to find the final ground state of the system at late times. In both cases above, $a = 0, 1$, the minimum gap was Δ. The sweep rate was $\Delta^2/4$. But one case showed an adiabatic evolution ($a = 0$) while the other ($a = 1$) exhibited a diabatic one. One important feature of our example is that the frequency of oscillation is small (twice the minimum gap). Thus, even a slow oscillation can ruin adiabaticity. For a large array of qubits this can be a problem since the gap becomes small.

We conclude that oscillatory terms in adiabatic quantum computation are more detrimental than an equivalent fast sweeps and the computation may not give the expected result unless the Hamiltonians are designed such that they preclude generation of oscillatory terms.

Part II

SIMULATION OF QUANTUM NOISE

Chapter 6

Classical simulation of arbitrary quantum noise

I present an explicit classical simulation of arbitrary quantum noise for quantum models in which one qubit interacts with a quantum bath. The classical model simulates the interaction of the bath and the qubit by random unitary evolutions. I show that any arbitrary quantum dynamics, including quantum dissipation, recurrence, and dephasing, can be simulated classically when one allows the unitary operators in the classical model to depend on the initial state of the system and bath. For initial mixed states of the system and non-product states of the system and bath, I demonstrate that random unitary expansion is still possible, in terms of a set of pure states.

6.1 Introduction

Entanglement is '*the* trait of quantum mechanics' that 'enforces its entire departure from classical lines of thought', according to Erwin Schrodinger [37]. It is the growth of entanglement with an environment, in open quantum systems, that is commonly stated as the root cause of decoherence [38, 39, 40, 41]. The loss of coherence in open quantum systems, or decoherence, itself is considered to be the reason of the appearance of classical traits in quantum systems and to be connected with the quantum-to-classical transition [42, 43].

However, recently it has been demonstrated that certain types of decoherence can be simulated classically by random unitary dynamics without appealing to entanglement with an environment and the idea of transformation of information, which comes with it [44, 45, 46, 47, 48, 49]. Specifically, the pure dephasing decoherence, for principle systems with two dimensional Hilbert space, and depolarizaing noise, for all dimensionalities, has been simulated classically [47].

Still, the degree of entanglement is been thought as a decisive feature which can classify environments. In microscopic system-bath models, the strength of coupling between the system and each degrees of freedom of the environment, which leads to the degree of entanglement with and within the environment, is the decisive factor. In oscillator bath models each degrees of freedom of the environment is only weakly perturbed [1, 2].

In spin bath models, however, each degrees of freedom can be strongly perturbed, too [3, 50] . The spin bath models which have weak couplings with the principle system can be mapped onto oscillator baths [1, 51, 3, **?**, 52] . It has been stated that the spin baths with strong couplings cannot be mapped onto oscillator baths and they make totally different effects on principle systems [3, 53] .

However, it has recently been recognized that by an appropriate choice of spectral density function, an oscillator bath can simulate the effect of the spin bath in the strong coupling limit as well [54].

What is the role of entanglement in quantum decoherence? And what is the distinction between quantum and classical noises? This paper attempts to investigate these questions further.

For a single qubit, it is well known that every doubly stochastic (or unital) channel can be represented as a random unitary channel [44, 55]. A doubly stochastic channel is a completely positive map on the Hilbert space of the principle system that maps the completely mixed state onto itself, whereas a random unitary channel is a convex combination of unitary transformations:

$$\mathcal{E}(\rho) = \sum_i p_i U_i \rho U_i^\dagger \qquad \left(p_i > 0, \sum_i p_i = 1 \right). \tag{6.1}$$

We extend the idea of random unitary channels to random unitary expansions by letting the unitary operators U_i depend on the initial state of the system-plus-environment. We show that for a single qubit with initial pure states $\rho(t_i)$ not only doubly stochastic operations but all quantum evolutions have random unitary expansions:

$$\rho(t) = \sum_\alpha p_\alpha U_\alpha \rho(t_i) U_\alpha^\dagger \tag{6.2}$$

where U_α is a function of time and the initial state of the system-plus-environment. A Lebesgue integral over an infinitely uncountable set of index α is intended by the sum above.

For mixed initial state $\rho(t_i)$ of a single qubit we show that a random unitary expansion is possible in the following sense:

$$\rho(t) = \sum_\alpha p_\alpha U_\alpha \rho_\alpha(t_i) U_\alpha^\dagger \tag{6.3}$$

where

$$\rho(t_i) = \sum_\alpha p_\alpha \rho_\alpha(t_i) \tag{6.4}$$

and $\rho_\alpha(t_i)$ are some pure states.

We construct an explicit time-continues classical model that simulates the effect of entanglement and

derives the above results. For the sake of simplicity we first introduce the classical simulation of quantum models in which the system starts in pure states and the universe (system plus environment) starts in product states. We devote Sec. 6.2-6.4 to this case and describe the quantum and classical models and prove their equivalence. In Sec. 6.5 we relax the initial state assumption and let the system start in a mixed state and the universe in a non-product state. We show that a random unitary expansion is still possible in this case. Finally, in Sec. 6.6 we give three examples for the case of initial pure states. The first example is the simulation of quantum recurrence in which the entropy decreases in the intermediate stage of evolution from almost maximum value of $\ln 2$ to zero! The second example simulates pure dephasing decoherence. The last example is a simulation of amplitude damping, which could not be achieved in the previous classical models, , except for short times and at high temperatures [48].

6.2 Quantum Model for Initial pure states

In this section through Sec. 6.4 we consider all quantum models with the following four properties: (1) The central system S is a single qubit. (2) The qubit interacts with an arbitrary quantum bath B. (3) The initial state is a product state

$$\rho_U(t_i) = \rho^Q(t_i) \otimes \rho_B(t_i) \tag{6.5}$$

where t_i is the initial time, ρ_U is the density matrix of the universe (system plus bath), and $\rho^Q(t_i)$, $\rho_B(t_i)$ are the initial density matrices of the system and the bath (We use superscript Q for the density matrix of the system, instead of subscript S, to emphasize that this density matrix is associated with the quantum model). (4) The system is initially in a pure state,

$$\rho^Q(t_i) = |\Psi_i\rangle\langle\Psi_i|. \tag{6.6}$$

The total Hamiltonian of the above quantum models can be decomposed into three parts as usual:

$$H_U = H_S + H_{int} + H_B \tag{6.7}$$

where H_U, H_S, H_B and H_{int} are the Hamiltonian of the universe, the qubit, the bath, and the interaction Hamiltonian respectively. The density matrix of the universe evolves by the evolution unitary operator

$$U(t, t_i) = e^{-iH_U(t-t_i)}. \tag{6.8}$$

where we have set $\hbar = 1$. At each t the density matrix of the universe is

$$\rho_U(t) = U(t,t_i)\rho_U(t_i)U(t,t_i)^\dagger. \tag{6.9}$$

The quantity of interest here is the *reduced* density matrix of the system, which can be obtained by taking the trace of $\rho_U(t)$ over a basis of the bath

$$\rho^Q(t) = \mathrm{Tr}_B\left[\rho_U(t)\right] \tag{6.10}$$

Choosing some basis for the Hilbert space of the qubit, one can write $\rho^Q(t)$ in its matrix form

$$\rho^Q(t) = \begin{pmatrix} \rho^Q_{00}(t) & \rho^Q_{01}(t) \\ \rho^Q_{10}(t) & \rho^Q_{11}(t) \end{pmatrix} \tag{6.11}$$

Since the evolution is quite arbitrary there are only a few general statements that one can make about $\rho^Q(t)$. Three of them are particularly useful in our discussion:

$$\rho^Q_{00}(t), \rho^Q_{11}(t) \geq 0 \tag{6.12}$$

$$\rho^Q_{00}(t) + \rho^Q_{11}(t) = 1 \tag{6.13}$$

$$\left|\rho^Q_{10}(t)\right| \leq \sqrt{\rho^Q_{00}(t)\rho^Q_{11}(t)}. \tag{6.14}$$

The first two are well known. The third one can be derived from the positivity condition of the reduced density matrix, which implies $\det(\rho^Q) \geq 0$. The positivity of the reduced density matrix itself can be derived easily from the positivity of the universe density matrix. In App. A we give a proof for Eq. 6.14, which is finer than the positivity condition and may be used for generalization of the argument in higher dimensions. We shall use (6.12)-(6.14) in constructing the classical model in the next section.

6.3 Classical Model for Initial pure states

The classical model consists of a stochastic magnetic field, which acts on the qubit with Hamiltonian,

$$H_{Cl}(t) = \frac{1}{2}\vec{B}(t)\cdot\vec{\sigma} \tag{6.15}$$

where $\vec{B}(t) = (B_x(t), B_y(t), B_z(t))$ and $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ are Pauli matrices. Each $B_j = (B_j(t) : t_i \leq t)$, for $j = x, y, z$, is a random process in the standard sense [56]. That is, each B_j is a family of random variables $B_j(t)$ defined on a probability space $(\Omega, \mathcal{F}, P)$ where Ω is the sample space, $\mathcal{F}$ is a set of subsets of Ω, and P is a probability measure on $\mathcal{F}$. By definition, for each t fixed, the random variable $B_j(t)$ is a function from the sample space Ω to the real line: $\omega \mapsto B_j(t,\omega)$. Here ω are elements of Ω. For each ω fixed, $\vec{B}(t,\omega)$ is a

function of t, called the sample path (or noise history) corresponding to ω, so ω's label the sample paths.

On each sample path ω, the qubit evolves from an initial state $\rho^{Cl}(t_i)$ to state $\rho^{Cl}_\omega(t)$ at each $t \geq t_i$. The initial state does not depend on ω, however at any later time the state depends on ω. The evolution operator for sample path ω is

$$U^{Cl}_\omega = \mathcal{T} \exp[-i \int_{t_i}^{t} H^{Cl}_\omega(t) dt] \tag{6.16}$$

where $\mathcal{T}$ denotes time ordering and

$$H^{Cl}_\omega(t) = \frac{1}{2} \vec{B}(t, \omega) \cdot \vec{\sigma}. \tag{6.17}$$

The evolved state at time t can be written as

$$\rho^{Cl}_\omega(t) = U^{Cl}_\omega \rho^{Cl}(t_i) U^{Cl\dagger}_\omega. \tag{6.18}$$

In a classical noise model, the standard approach [47] is to consider the density matrix of the qubit at time t as the expectation value of $\rho^{Cl}_\omega(t)$ over all sample paths,

$$\rho^{Cl}(t) = \left\langle \rho^{Cl}_\omega(t) \right\rangle_\omega, \tag{6.19}$$

where $\langle \rangle_\omega$ denotes the expectation function.

Our goal is to construct the random magnetic field process $\vec{B}(t)$ such that $\rho^{Cl}(t) = \rho^{Q}(t)$ when the two models start from the same initial state $\rho^{Cl}(t_i) = \rho^{Q}(t_i)$.

To this end, we begin with introducing, on a probability space $(\Omega, \mathcal{F}, P)$, a random process $\Phi = (\Phi(t) : t_i \leq t)$ with the following properties:

1. For all $\omega \in \Omega$, $\Phi(t_i, \omega) = 0$

2. For each ω fixed, $\Phi(t, \omega)$ is differentiable with respect to t.

3. For each t fixed, the probability density function of the random variable $\Phi(t)$ is a Gaussian with mean zero and variance $\sigma^2(t)$,

$$p_\Phi(\phi, t) = \frac{1}{\sqrt{2\pi\sigma^2(t)}} \exp\left[-\frac{\phi^2}{2\sigma^2(t)}\right]. \tag{6.20}$$

4. The variance $\sigma^2(t)$ is

$$\sigma^2(t) = -2 \lim_{s \to t} \ln \frac{\left|\rho_{10}^Q(s)\right|}{\sqrt{\rho_{11}^Q(s)\rho_{00}^Q(s)}} \tag{6.21}$$

Inequality (6.14) guarantees that the right hand side of Eq. 6.21 is nonnegative. Hence, it can be considered as the variance of a Gaussian distribution. In the case that $\sigma^2(t) = 0$ the Gaussian distribution becomes a delta function. For the case $\sigma^2(t) = \infty$ the distribution Eq. 6.20 is interpreted as a uniform distribution over the entire real line. App. B gives a construction of $\Phi(t, \omega)$ which satisfies the above properties.

The meaning of probability density function $p_\Phi(\phi, t)$ for each t fixed is

$$p_\Phi(\phi, t) = \lim_{\epsilon \to 0} \frac{1}{\epsilon} P\{\omega : \phi \leq \Phi(t, \omega) \leq \phi + \epsilon\} \tag{6.22}$$

Knowing the probability density function, we can calculate the expectation of any function of $\Phi(t, \omega)$ for each t fixed. As we shall see, it is particularly useful to calculate $\langle \exp[\pm i\Phi(t, \omega)] \rangle_\omega$:

$$\begin{aligned} \left\langle e^{\pm i\Phi(t,\omega)} \right\rangle_\omega &= \int_{-\infty}^{\infty} e^{\pm i\phi} P\{\omega : \phi \leq \Phi(t, \omega) \leq \phi + d\phi\} \\ &= \int_{-\infty}^{\infty} e^{\pm i\phi}\ p(\phi, t)\ d\phi = e^{-\sigma^2(t)/2} \\ &= \frac{\left|\rho_{10}^Q(t)\right|}{\sqrt{\rho_{11}^Q(t)\rho_{00}^Q(t)}} \end{aligned} \tag{6.23}$$

where the first integral above is a Lebesgue integral that is written in terms of an ordinary Reimann integral on the second line by use of the probability density function.

We define for each ω fixed, functions $a(t, \omega)$, $b(t, \omega)$:

$$a(t, \omega) = \sqrt{\rho_{00}^Q(t)} \tag{6.24}$$

$$b(t, \omega) = \sqrt{\rho_{11}^Q(t)}\ e^{i\ \mathrm{Arg}[\rho_{10}^Q(t)]} e^{i\Phi(t,\omega)} \tag{6.25}$$

Here Arg is the argument function over complex numbers (e.g. $z = |z|e^{i\mathrm{Arg}[z]}$). Note that $a(t, \omega)$ is a deterministic function of t and is independent of sample path ω, however, $b(t, \omega)$ is a random function and depends on the sample path. Nevertheless, on each sample path one always has the identity

$$|a(t, \omega)|^2 + |b(t, \omega)|^2 = \rho_{00}^Q(t) + \rho_{11}^Q(t) = 1 \tag{6.26}$$

where we used Eq. Eq. 6.13 in the last equality.

We are now ready to give the explicit form of $\vec{B}(t,\omega)$:

$$B_z(t,\omega) = i[\dot{a}(t,\omega)\ a(t,\omega)^* + \dot{b}(t,\omega)^*\ b(t,\omega)] \tag{6.27}$$
$$B_+(t,\omega) = -i[\dot{a}(t,\omega)^* b(t,\omega) - \dot{b}(t,\omega) a(t,\omega)^*] \tag{6.28}$$
$$B_x(t,\omega) = \text{Re}\ B_+(t,\omega) \tag{6.29}$$
$$B_y(t,\omega) = \text{Im}\ B_+(t,\omega) \tag{6.30}$$

where dots denote derivatives with respect to t. $B_x(t,\omega)$ and $B_y(t,\omega)$ are by definition real-valued. It is easy to show that $B_z(t,\omega)$ is also real-valued:

$$\begin{aligned} B_z(t,\omega) - B_z(t,\omega)^* &= i[\dot{a}a^* + \dot{a}^*\ a + \dot{b}^*\ b + \dot{b}\ b^*] \\ &= i\frac{\partial}{\partial t}[aa^* + bb^*] = 0 \end{aligned} \tag{6.31}$$

Here we omitted (t,ω) dependences of $a(t,\omega)$, $b(t,\omega)$ for brevity and used Eq. 6.26 in the last step. Eq. Eq. 6.31 implies that $B_z(t,\omega)$ is real-valued. Thus (6.27)-(6.30) describe a well-defined stochastic magnetic field. Substituting them in Eq. Eq. 6.17 one obtains the Hamiltonian $H_\omega^{Cl}(t)$ of the classical model on each sample path.

We note that the classical Hamiltonian $H_\omega^{Cl}(t)$ depends on the total Hamiltonian of the quantum model H_U, the time elapsed from the beginning of the evolution and the initial state of the universe $\rho_U(t_i)$. This is because $a(t,\omega)$ and $b(t,\omega)$, which constitute $B(t,\omega)$, depend on $\rho^Q(t)$ which in turn depends on $\rho_U(t)$. The latter depends on H_U, t and $\rho_U(t_i)$ (See Eqs. Eq. 6.7-Eq. 6.11). Thus $H_\omega^{Cl}(t)$ is not only a function of the Hamiltonian of the quantum model, but also depends of the initial state of the universe.

6.4 Equivalence of the Quantum and Classical Models for initial pure states

We assume that the qubit in the classical model starts from the same initial pure state as in the quantum model,

$$\rho^{Cl}(t_i) = \rho^Q(t_i) = |\Psi_i\rangle\langle\Psi_i|. \tag{6.32}$$

Since the initial state is a pure state, on each sample path ω the qubit evolves according to the time-dependent Schrodinger equation

$$i|\dot{\Psi}(t,\omega)\rangle = H_\omega^{Cl}(t)|\Psi(t,\omega)\rangle \tag{6.33}$$

The solution of this Schrodinger equation on each sample path is

$$|\Psi(t,\omega)\rangle = \begin{pmatrix} a(t,\omega) \\ b(t,\omega) \end{pmatrix} \tag{6.34}$$

as we demonstrate below: Firstly, since $\Phi(t_i,\omega) = 0$ for all sample paths, from definitions Eq. 6.24-Eq. 6.25 one can see that

$$|\Psi(t_i,\omega)\rangle = |\Psi_i\rangle \tag{6.35}$$

for each ω, modulus an overall phase factor. Secondly, $|\Psi(t,\omega)\rangle$ of Eq. Eq. 6.34 satisfies the Schrodinger equation Eq. 6.33:

$$\begin{aligned} H_\omega^{Cl}|\Psi(t,\omega)\rangle &= \begin{pmatrix} B_z & B_+^* \\ B_+ & -B_z \end{pmatrix} \begin{pmatrix} a \\ b \end{pmatrix} \\ &= \begin{pmatrix} i\dot{a}[a^*a + b^*b] \\ i\dot{b}[a^*a + b^*b] \end{pmatrix} \\ &= \begin{pmatrix} i\dot{a} \\ i\dot{b} \end{pmatrix} = i|\dot{\Psi}(t,\omega)\rangle \end{aligned} \tag{6.36}$$

where we used Eq. 6.26. Thus, $|\Psi(t,\omega)\rangle$ is the solution of Eq. 6.33.

The density matrix of the qubit on each sample path is then

$$\rho_\omega^{Cl}(t) = |\Psi(t,\omega)\rangle\langle\Psi(t,\omega)| = \begin{pmatrix} |a|^2 & ab^* \\ ba^* & |b|^2 \end{pmatrix} \tag{6.37}$$

The density matrix of the classical model is the average of the density matrices of all sample paths

$$\rho^{Cl}(t) = \left\langle \rho^{Cl}(t) \right\rangle_\omega = \begin{pmatrix} \left\langle |a|^2 \right\rangle_\omega & \langle ab^* \rangle_\omega \\ \langle ba^* \rangle_\omega & \left\langle |b|^2 \right\rangle_\omega \end{pmatrix}. \tag{6.38}$$

Since $|a(t,\omega)|^2 = \rho_{00}^Q(t)$ and $|b(t,\omega)|^2 = \rho_{11}^Q(t)$ are deterministic functions,

$$\left\langle |a(t,\omega)|^2 \right\rangle_\omega = \rho_{00}^Q(t) \tag{6.39}$$

$$\left\langle |b(t,\omega)|^2 \right\rangle_\omega = \rho_{11}^Q(t). \tag{6.40}$$

For the off-diagonal term $\langle ba^* \rangle_\omega$ we have

$$\langle b(t,\omega)a(t,\omega)^* \rangle_\omega = \sqrt{\rho_{00}(t)\rho_{11}(t)} e^{i \ \mathrm{Arg}[\rho_{10}(t)]} \left\langle e^{i\Phi(t,\omega)} \right\rangle_\omega \tag{6.41}$$

By use of Eq. Eq. 6.23, the above expression simplifies to

$$\langle ba^* \rangle_\omega = \left| \rho^Q_{10}(t) \right| e^{i \ \mathrm{Arg}[\rho^Q_{10}(t)]} = \rho^Q_{10}(t) \tag{6.42}$$

Similarly, one obtains

$$\langle ab^* \rangle_\omega = \left| \rho^Q_{10}(t) \right| e^{-i \ \mathrm{Arg}[\rho^Q_{10}(t)]} = \rho^Q_{01}(t). \tag{6.43}$$

Finally, by substituting Eq. 6.39, Eq. 6.40, Eq. 6.42, and Eq. 6.43 into Eq. 6.38 we obtain

$$\rho^{Cl}(t) = \begin{pmatrix} \rho^Q_{00}(t) & \rho^Q_{01}(t) \\ \rho^Q_{10}(t) & \rho^Q_{11}(t) \end{pmatrix} = \rho^Q(t) \tag{6.44}$$

just as desired. Hence, the classical model simulates the quantum model exactly.

Summarizing, we built an stochastic magnetic field and hence a classical Hamiltonian for each history of noise. Then we showed that the density matrix of the qubit in this classical model at each moment of time is equal to the reduced density matrix of the quantum model at that time.

One can write Eq. Eq. 6.44 in a more familiar form

$$\rho^Q(t) = \int_{-\infty}^{\infty} d\phi \ p(\phi,t) \ U^{Cl}_\omega |\Psi_i\rangle\langle\Psi_i| U^{Cl^\dagger}_\omega \tag{6.45}$$

where $p(\phi,t)d\phi = P\{\omega : \phi \leq \Phi(t,\omega) \leq \phi + d\phi\}$ and $U^{Cl}_\omega = U^{Cl}_\omega(t; H_U; \rho_U(t_i))$. This form demonstrates that every quantum evolution of an open two dimensional system has a random unitary expansion (See Eq. Eq. 6.2).

6.5 Quantum and Classical Models for Initial Mixed States

The simulation of mixed states is similar to the one for pure states, apart from a few modifications that we mention below. For the quantum model, we relax conditions (3) and (4) of Sec. 6.2 for initial states of the universe and the principle system (a single qubit), and let them to start in any arbitrary states. That is the universe can start in a non-product state $\rho_U(t_i)$ and the system can start in a mixed state $\rho^Q(t_i) = \mathrm{Tr}_B[\rho_U(t_i)]$. The universe evolves according to the unitary evolution of Eq. Eq. 6.9. The relations Eq. 6.12-Eq. 6.14 still hold since they do not depend on the initial states. Therefore we can use them in building the classical model as before.

The classical model follows the classical model of Sec. 6.3 verbatim, except we relax the first property of the random phase $\Phi(t,\omega)$: It does not start from zero for all sample paths, rather it obeys the Gaussian distribution of Eq. Eq. 6.20 for all times including the initial time t_i. Thus, $\Phi(t)$ is a differentiable random process with Guassian distribution whose mean and variance are zero and $\sigma^2(t)$ of Eq. Eq. 6.21, respectively. Since the initial state is a mixed state, $\sigma(t_i) \neq 0$. We construct $\Phi(t,\omega)$ as in App. B, Eq. Eq. B.1. Because $\sigma(t_i) \neq 0$, not all $\Phi(t_i,\omega)$ are equal to zero, as expected. The random magnetic field and the classical Hamiltonian follows Eqs. Eq. 6.27-Eq. 6.30 and Eq. 6.17, as before.

To prove the equivalence of the classical model and the quantum model in the case of initial mixed state we begin with constructing wave functions $|\Psi(t,\omega)\rangle$ as in Eq. Eq. 6.34. By use of Eqs. Eq. 6.37-Eq. 6.44 one can see that the density matrix of the quantum system at anytime, including the initial time t_i, can be expanded in terms of these pure states:

$$\rho^Q(t) = \int_{-\infty}^{\infty} d\phi \; p(\phi,t) \; |\Psi(t,\omega)\rangle\langle\Psi(t,\omega)| \tag{6.46}$$

One notes that wave functions $|\Psi(t,\omega)\rangle$ now do not start from the same value at $t = t_i$ because for two different sample paths ω, ω', the initial value of the random phase can be different $\Phi(t_i,\omega) \neq \Phi(t_i,\omega')$. Nevertheless, $|\Psi(t,\omega)\rangle$ satisfy Schrodinger equation Eq. 6.33 on each sample path, as shown in Eq. Eq. 6.36. Therefore, one can obtain $|\Psi(t,\omega)\rangle$ by evolving $|\Psi(t_i,\omega)\rangle$ through U_ω^{Cl}:

$$|\Psi(t,\omega)\rangle = U_\omega^{Cl}|\Psi(t_i,\omega)\rangle \tag{6.47}$$

where U_ω^{Cl} is defined in Eq. Eq. 6.16. Substituting Eq. 6.47 into Eq. Eq. 6.46 we obtain

$$\rho^Q(t) = \int_{-\infty}^{\infty} d\phi \; p(\phi,t) \; U_\omega^{Cl}|\Psi(t_i,\omega)\rangle\langle\Psi(t_i,\omega)|U_\omega^{Cl^\dagger}. \tag{6.48}$$

At $t = t_i$, U_ω^{Cl}, which is a function of time, is the identity operator and Eq. Eq. 6.48 reduces to

$$\rho^Q(t_i) = \int_{-\infty}^{\infty} d\phi \; p(\phi,t) \; |\Psi(t_i,\omega)\rangle\langle\Psi(t_i,\omega)|. \tag{6.49}$$

Eqs. Eq. 6.48-Eq. 6.49 are the random unitary expansions we intended to find for arbitrary evolution of a single qubit with initial mixed state (See Eqs. Eq. 6.3-Eq. 6.4). In the sense of these equations the classical model and the quantum model are equivalent for initial mixed states as well.

6.6 Examples

We consider three examples for classical simulation of quantum models. In all the examples we assume that the universe starts in the product state $\rho_U(0) = |\Psi_i\rangle\langle\Psi_i| \otimes \rho_B(0)$ where $|\Psi_i\rangle = \alpha|0\rangle + \beta|1\rangle$ and $\rho_B(0)$ will be specified for each example. We determine $\sigma^2(t)$, $a(t,\omega)$ and $b(t,\omega)$ in each example. The stochastic magnetic field $B(t,\omega)$ and the classical Hamiltonian $H_{Cl}(t,\omega)$ can then be constructed by Eqs. Eq. 6.27-Eq. 6.30, Eq. 6.17 and the discussion of Sec. 6.3.

6.6.1 Quantum recurrence

Consider a spin-boson Hamiltonian at zero temperature

$$H_U = \frac{1}{2}\omega_0\sigma_z + \sigma_z \sum_{n=1}^{N}(g_n a_n^\dagger + g_n^* a_n) + \sum_{n=1}^{N}\omega_n a_n^\dagger a_n \tag{6.50}$$

where N is finite and the frequencies of the bath are commensurable (i.e. for each ω_n, ω_m there are integer numbers p_n , p_m such that $\omega_n/\omega_m = p_n/p_m$). The bath is initially in its ground state. The evolution of the reduced density matrix is then [52]

$$\rho^Q(t) = \begin{pmatrix} |\alpha|^2 & \alpha\beta^* \; e^{-i\omega_0 t - \Gamma(t)} \\ \alpha^*\beta \; e^{i\omega_0 t - \Gamma(t)} & |\beta|^2 \end{pmatrix} \tag{6.51}$$

where

$$\Gamma(t) = \sum_{n=1}^{N} 4\frac{|g_n|^2}{\omega_n^2}(1 - \cos\omega_n t) \tag{6.52}$$

Since ω_n's are commensurable, $\Gamma(t)$ is a periodic function. It starts at $\Gamma(0) = 0$ and returns to zero with some period P. Between two nodes of function $\Gamma(t)$, however, the value of the function can be large if N is large or coupling constants g_n are significant. For example, for $|g_n| = \omega_n = 2\pi n/P$ and $N = 30$ the average value of the function between two nodes is $\Gamma(t) \approx 120$. This gives rise to decoherence factor $\exp[-\Gamma(t)] \approx 10^{-53}$ in the off diagonal elements of Eq. 6.51. This implies that for $\alpha = \beta = 1/\sqrt{2}$ the system that started in a pure state with entropy $S[\rho^Q(0)] = Tr[\rho^Q(0)\ln\rho^Q(0)] = 0$ evolves to nearly the completely maxed state

$$\rho^Q(t) \approx \begin{pmatrix} \frac{1}{2} & 0 \\ 0 & \frac{1}{2} \end{pmatrix} \qquad 0 \ll t \ll P \tag{6.53}$$

with maximum possible entropy $S[\rho^Q(t)] \approx \ln 2$ for most of the times between, for example, $t = 0$ and $t = T$, and then the entropy of the system *decreases* and the system returns to the original pure state at $t = T$ with $S[\rho^Q(P)] = 0$.

In terms of Bloch vector the above process describes a contraction of the Bloch sphere to *almost* a point and then an expansion of it to its full size.

Although the entropy decreases in the intermediate stage in the above process, it can still be simulated classically. The variance $\sigma(t)^2$ defined in Eq. 6.21 is in this case

$$\sigma^2(t) = 2\ \Gamma(t) \tag{6.54}$$

and the functions $a(t,\omega)$, $b(t,\omega)$ of Eqs. (6.24)-(6.25) are

$$a(t,\omega) = |\alpha|, \qquad b(t,\omega) = |\beta|\ e^{i\mathrm{Arg}[\alpha^*\beta]}\ e^{i\omega_0 t} e^{i\Phi(t,\omega)} \tag{6.55}$$

6.6.2 Pure dephasing

One can also use Hamiltonian Eq. 6.50 as an example of phase damping in the limit $N \to \infty$. Suppose the bath is in thermal equilibrium at some nonzero temperature T and the spectral density function of the bath is ohmic: $J(\omega) = \sum_n |g_n|^2 \delta(\omega - \omega_n) = 4^{-1} J_0 \omega e^{-\omega/\Lambda}$, where J_0 is a dimensionless constant and Λ is a cut-off frequency. The evolution of the reduced density matrix can be described by Eq. Eq. 6.51 when one substitutes for $\Gamma(t)$ the following expression [52]

$$\begin{aligned}\Gamma(t) &= \int_0^\infty d\omega \frac{4J(\omega)}{\omega^2}(1-\cos\omega t)\coth(\omega/2k_BT) \\ &= \frac{J_0}{2}\ln(1+\Lambda^2 t^2) + J_0 \ln\left[\frac{\sinh(\pi k_B T t)}{\pi k_B T t}\right]\end{aligned} \tag{6.56}$$

Here $\Gamma(t)$ is an increasing function of time, which gives rise to the increase of the entropy of the system over time. There is no revival of coherence in this limit. The classical model is similar to the one in the previous subsection and is described by Eqs. Eq. 6.54-Eq. 6.55 where $\Gamma(t)$ is given by Eq. Eq. 6.56.

6.6.3 Amplitude damping

Finally, consider an amplitude damping channel [55]

$$\rho(t) = \begin{pmatrix} 1-(1-\gamma(t))(1-|\alpha|^2) & \alpha\beta^*\sqrt{1-\gamma(t)} \\ \alpha^*\beta\sqrt{1-\gamma(t)} & |\beta|^2(1-\gamma(t)) \end{pmatrix} \tag{6.57}$$

where the environment starts in the ground state and $\gamma(t)$ is the probability of decay of the qubit from the excited state to its ground state. For real physical processes, $\gamma(t)$ can be replaced by $(1-e^{-t/T_1})$, where T_1 is the longitudinal relaxation time constant.

Amplitude damping channel is not a unital channel and a general classical simulation of it has not been achieved in the literature, to our knowledge. The classical model of Sec. 6.3 gives such simulation by letting the unitary operators of the classical model to depend on the initial state of the universe. The ingredients of the model are as follows

$$a(t,\omega) = \left[1-(1-\gamma(t))(1-|\alpha|^2)\right]^{1/2} \tag{6.58}$$

$$b(t,\omega) = |\beta|\sqrt{1-\gamma(t)}\; e^{i\mathrm{Arg}[\alpha^*\beta]}\; e^{i\Phi(t,\omega)} \tag{6.59}$$

$$\sigma^2(t) = \ln\left(|\alpha|^{-2}-(1-\gamma(t))(|\alpha|^{-2}-1)\right). \tag{6.60}$$

6.7 Conclusion

In conclusion, we have constructed, for arbitrary quantum noises, a classical simulation of single-qubit models. We showed how entanglement between a qubit and an external bath can be modeled classically without using the bath. This was made possible by allowing the unitary operators in the classical model to depend on the initial state of the system and the bath.

We demonstrated that the reduced density matrices of quantum models that start from initial pure states have random unitary expansions. For the quantum models that start from mixed states (and even non-product state of the system and bath) we showed that the density matrices can be expressed as random unitary expansion of some pure states.

The classical model was based on utilizing a differentiable random phase that has Gaussian distribution with time-varying variance. We gave the explicit expression for the stochastic magnetic field of the classical Hamiltonian. The field depends on the Hamiltonian of the quantum model, the time elapsed from the beginning of the evolution, and the initial state of the system and bath in the quantum model.

Simulation of quantum dissipation such as amplitude damping had not been achieved in preceding classical models, except for short times and high temperatures. Here, we offered exact results for a general simulation of such process (amplitude damping), for arbitrary long times, as well as of quantum recurrence and pure dephasing.

Entanglement with an external environment plays an important role in quantum dissipation and decoherence of open quantum systems, beyond doubt. However, the result of this paper and its preceding counterparts show that the distinction between quantum and classical noises may not be apparent in systems with low dimensionality.

Chapter 7

Oscillator bath simulation of spin bath

I demonstrate that the conventional Caldeira-Leggett oscillator bath model can simulate the effect of Prokof'ev-Stamp spin bath model in the strong coupling limit of the spin bath. It has been previously recognized that the spin bath model, in the strong coupling limit, has effects on quantum systems that are strikingly different from what an oscillator bath can produce. I show, however, that by choosing appropriate parameters for an oscillator bath, the bath can simulate the effect of the spin bath on systems.

7.1 Introduction

Quantum theory was originally developed in the context of *isolated* microscopic systems whose interactions with their environments were negligible (e.g. the atoms in a beam). The theory was tested successfully in this domain in the early twentieth century and some of its founding fathers, such as Niels Bohr, believed that it would not be applicable in a larger domain where systems are strongly coupled to complex environments. [57, 58]

Advancement of experimental techniques and equipments in recent decades, however, showed that quantum theory does apply in a broader range. It can well describe behaviors of *open* quantum systems, as large as a few microns, that are strongly coupled to their complex *environments* (e.g. the phase of the Cooper pairs, in SQUIDs, that is coupled to phonons, radiation field, normal electrons, nuclear spins, etc.) [1, 2, 7, 6, 26, 8, 24, 59].

In handling complex environments coupled to open quantum systems, the *effects* that the environments exert on the principal systems are of the main interest, not the the behaviors of the environments in their own right. As a result, theorists attempt to model complex environments by mapping them onto simpler ones that are better tractable and have the same *effects* on the principal systems. Two of these simple models that are now well established in the literature are the oscillator bath and spin bath models [1, 2, 57, 60, 53, 50, 3, 61, 62, 52, 22].

The oscillator bath model consists of a set of non-interacting simple harmonic oscillators that are individually coupled to the principal system. Caldeira and Leggett [1, p. 439] showed that at *absolute zero temperature*, any arbitrary environment whose each degree of freedom is only weakly perturbed, by the principal system, can be mapped onto an oscillator bath.

It is important to note that although each degree of freedom of the *environment* is weakly perturbed in this model, the *principal system* can be strongly perturbed by the overall effects of all the oscillators. We use the term *weak coupling limit* to refer to the case that each degree of freedom of the environment is weakly perturbed, and the term *strong coupling limit* when they are strongly perturbed. However, in both cases the principal system can be weakly or strongly perturbed.

The oscillator bath model has been extensively used in the literature to model finite temperature environments [2, 63, 64, 65, 66, 52, 22] . Such usage, e.g. to model phonons, electrons, magnons, spinons, holons, etc. at low energies and temperatures, are permissible as long as it is supported by a detailed microscopic theory (e.g. such as [23] for quasiparticles in Josephson junctions). However, in general there is no proof, to our knowledge, that a quite arbitrary environment at a nonzero temperature can be mapped onto the oscillator bath model, even in the weak coupling limit.

The spin bath model consists of microscopic spins that are independently coupled to the principal system. In real scenarios these spins interact with each other, although weakly [3, 12]. The spin bath model can be studied at both weak and strong coupling limits and there are practical cases associated to each of them [60, 53, 50, 3, 61, 62, 12, 67, 68, 11, 13, 30, 69].

Caldeira, Neto and de Carvalho [51] demonstrated that effects of a spin bath model with non-interacting spins and in the weak coupling limit can be obtained through the method that one uses for the oscillator bath model if one modifies the spectral density function suitably [51]. In this sense they showed that the noninteracting spin bath model maps onto the oscillator bath model in the weak coupling limit.

Weiss reached the same conclusion by use of the fluctuation-dissipation theorem for each degree of freedom of the environment, which is permissible in the weak coupling limit (See Sec. 3.5 and 6.1 of [22]).

However, despite this success in mapping noninteracting spin bath onto oscillator bath in the weak coupling limit, the scheme ccould not be extended trivially to the case of self interacting spin bath, mainly due to the difficulty of calculating spin correlation function for each spin of the bath in a self interacting bath. If one wishes to use the scheme of the oscillator bath model to solve such problems, one may need to resort to the original mapping of Caldeira and Leggett [1] for zero temperature or, possibly, the mapping of this Letter for finite temperatures. In either case, however, one observes that a straightforward relation

between the spectral density function of the spin bath and that of the oscillator bath cannot be achieved.

Prokof'ev and Stamp presented an independent solution for the spin bath model in both weak and strong coupling limits. For an interacting spin bath that is coupled to an effectively two-state system, in the strong coupling limit, they found that under most conditions the principal system undergoes an incoherent relaxation with relaxation rate that decreases by the increase of bias energy between the two states of the principal system. They stated that this effect is in contrast to what an oscillator bath would have: 'All of this is in complete contrast to how inelastic tunneling works in the presence of an oscillator bath; there the relaxation rate typically increases as one moves away from resonance' (i.e. as the bias energy increases) [3]. They, hence, concluded that the spin bath model in the strong coupling limit has completely different effects on principal systems and is not comparable with the oscillator bath model [60, 53, 50, 3, 61, 62].

In this Letter, we, however, show, as our second objective, that a Caldeira-Leggett oscillator bath can simulate the effect of an spin bath in the strong coupling limit of the spin bath. We demonstrate that an oscillator bath can produce an incoherent relaxation for the system that Prokof'ev and Stamp considered with a relaxation rate that decreases as one moves away from resonance. By choosing an appropriate spectral density function for the oscillator bath we obtain a relaxation rate that is even quantitatively comparable with that of the spin bath model.

7.2 Simulation of Spin bath by oscillator bath in strong coupling limit

In the second part of this Letter, we show how a Caldeira-Leggett oscillator bath can simulate the effect of the Prokof'ev-Stamp spin bath that is in a strong coupling limit.

For two state principal system, the Prokof'ev-Stamp spin bath Hamiltonian is [3]

$$\begin{aligned} H = & -\frac{\Delta}{2}\left\{\hat{\tau}_- \cos\left[\Phi - i\sum_{k=1}^{N}\vec{\alpha}_k\cdot\hat{\vec{\sigma}}_k\right] + \text{H.c.}\right\} \\ & -\frac{\xi}{2}\hat{\tau}_z + \hat{\tau}_z\sum_{k=1}^{N}\vec{\omega}_k^{\parallel}\cdot\hat{\vec{\sigma}}_k + \sum_{k=1}^{N}\vec{\omega}_k^{\perp}\cdot\hat{\vec{\sigma}}_k \\ & + \sum_{k,k'=1}^{N}\sum_{\alpha,\beta=1}^{3} V_{kk'}^{\alpha\beta}\hat{\sigma}_k^{\alpha}\hat{\sigma}_{k'}^{\beta} \end{aligned} \tag{7.1}$$

where $\hat{\vec{\tau}}$, $\hat{\vec{\sigma}}_k$ are Pauli operators of the principal system and the kth spin of the spin bath, respectively, Δ

is the tunnel splitting of the two state system, ξ is a bias energy, the last term is the self Hamiltonian of the spin bath, and Φ, $\vec{\alpha}_k$, $\vec{\omega}_k^{\parallel}$ and $\vec{\omega}_k^{\perp}$ are the parameters of the model that depend on the high energy Hamiltonian of the principal system (See Ref. [3])

Hamiltonian Eq. 7.1 is the result of truncation of the high energy Hamiltonian of a single molecule magnet (such as Fe8, Mn12, $\cdots$) and its surrounding nuclear spins, defects, etc. to a low energy effective Hamiltonian which represents the single molecule magnet by a two level system [60, 53, 50, 3, 61, 62]. The single molecule magnet forms a double well energy landscape with a very small tunnel splitting Δ [12]. In these problems, the latter is typically the smallest energy scale included in the Hamiltonian (For Fe8 for example $\Delta \sim 0.1\ \mu K$, $V_{k,k'}^{\alpha\beta} \sim 1\ \mu K$, $\xi \sim 0.1\ mK$, and the width of the fluctuating field $\xi_0 \sim 10\ mK$) [60, 53, 50, 3, 61, 62, 12, 67, 68, 11, 69]. .

In the strong coupling limit, the solution of this model under most conditions is an incoherent relaxation of the principal system with relaxation rate [60]

$$\Gamma(\xi) \sim \frac{\Delta^2}{\xi_0} e^{-\left|\frac{\xi}{\xi_0}\right|} \tag{7.2}$$

where ξ_0 is the width of resonance window over which the bias field of the bath fluctuates. ξ_0^{-1} is also roughly equal to the time scale of this fluctuation [60, 3].

The simulator oscillator bath Hamiltonian of the above model is as follows

$$\begin{aligned} H &= -\frac{\Delta}{2}\ \hat{\tau}_x - \frac{\xi}{2}\ \hat{\tau}_z + \frac{\hat{\tau}_z}{2} \sum_i c_i\ \hat{x}_i \\ &+ \sum_i \frac{\hat{p}_i^2}{2m_i} + \frac{1}{2} m_i \omega_i^2 \hat{x}_i^2 \end{aligned} \tag{7.3}$$

This is the well known spin-boson Hamiltonian which couples simple harmonic oscillators to the principal two state system. The effect of this oscillator bath is encapsulated in its temperature T and spectral density function $J(\omega)$ [1, 2],

$$J(\omega) = \frac{\pi}{2} \sum_i \frac{c_i^2}{m_i \omega_i} \delta(\omega - \omega_i). \tag{7.4}$$

To simulate the effect of the spin bath we choose $T = 0$ and the spectral density function

$$J(\omega) = 2\pi\alpha\ \omega\ e^{-\omega/\xi_0} \tag{7.5}$$

with $\alpha = 1/2$. This is only for the sake of simulation. We disclaim an accurate physical content for it in this chapter. For a more accurate physical consideration please see chapter **??**. Nevertheless, we notice that

the memory time of the oscillator bath with the above spectral density function is of order ξ_0^{-1}, comparable with the time of fluctuation of the bias field in the spin bath. The width of the fluctuation of the bias field $\hat{\xi}_B = \frac{1}{2}\sum_i c_i \hat{x}_i$ is also of order ξ_0 at $T = 0$.

To see the latter one notes that

$$\begin{aligned} \langle \xi_B^2 \rangle &= \langle \frac{1}{4} \sum_{i,j} c_i c_j x_i x_j \rangle \\ &\simeq \langle \frac{1}{4} \sum_i c_i^2 x_i^2 \rangle \gtrsim \frac{1}{4} \sum_i c_i^2 \frac{\hbar}{m_i \omega_i} \end{aligned} \tag{7.6}$$

where we neglected the cross terms $\langle x_i x_j \rangle$ and used the ground state value of $\langle x_i^2 \rangle$. This is because at $T = 0$, almost all of the oscillators are not excited by the construction of Caldeira-Leggett oscillator bath model [1].

Since we have set $\hbar = 1$, the right hand side of Eq. 7.6 can be written in terms of the spectral density function Eq. 7.4 as

$$\frac{1}{4} \sum_i \frac{c_i^2}{m_i \omega_i} = \frac{1}{2\pi} \int_0^\infty J(\omega) d\omega \tag{7.7}$$

Combining Eq. 7.6 and Eq. 7.7 we obtain,

$$\frac{1}{2\pi} \int_0^\infty J(\omega) d\omega \lesssim \xi_0^2. \tag{7.8}$$

The choice of Eq. Eq. 7.5 gives

$$\frac{1}{2\pi} \int_0^\infty J(\omega) d\omega = \frac{1}{2} \xi_0^2. \tag{7.9}$$

which is compatible with Eq. 7.8. Thus, the spectral density of Eq. 7.5 produces a fluctuation of the bias field with the width of order ξ_0.

To solve the model one notes that $\Delta \ll \xi$, so one can do perturbation theory in Δ. This method of solving spin-boson Hamiltonian is known as the *golden-rule* and results in an *incoherent* relaxation of the principal system at rate [2]

$$\Gamma(\xi) = \Delta^2 \int_0^\infty dt \ \cos(\xi \ t) \ \cos(\frac{Q_1(t)}{\pi}) \ e^{-Q_2(t)/\pi} \tag{7.10}$$

where

$$Q_1(t) = \int_0^\infty \frac{J(\omega)}{\omega^2} \sin(\omega t) d\omega, \tag{7.11}$$

$$Q_2(t) = \int_0^\infty \frac{J(\omega)}{\omega^2} (1 - \cos(\omega t))\ d\omega \tag{7.12}$$

at $T = 0$. By use of $J(\omega)$ of Eq. Eq. 7.5 in Eqs. Eq. 7.11-Eq. 7.12, one obtains

$$Q_1(t) = 2\ \pi\alpha\ \tan^{-1} \xi_0 t \tag{7.13}$$

$$Q_2(t) = \alpha\ \pi\ \ln(1 + \xi_0^2 t^2). \tag{7.14}$$

Substituting these functions for $\alpha = 1/2$ into Eq. Eq. 7.10 and taking the integral we find the relaxation rate of the oscillator bath model

$$\Gamma(\xi) = \frac{\pi \Delta^2}{2\xi_0} e^{-\left|\frac{\xi}{\xi_0}\right|} \sim \frac{\Delta^2}{\xi_0} e^{-\left|\frac{\xi}{\xi_0}\right|} \tag{7.15}$$

This is comparable with the relaxation rate of the spin bath model Eq. 7.2. Therefore, we conclude that the oscillator bath model can simulate the effect of the spin bath model in the strong coupling limit of the spin bath. It can produce incoherent relaxation with a rate that decreases as the bias energy increases with the same functionality, as in the spin bath model.

7.3 Conclusion

In conclusion, we have shown in this chapter that an oscillator bath can simulate the effect of the spin bath in the strong coupling limit. This is the limit that has been thought to have strikingly different effects on principal systems.

Part III

SINGLE MOLECULE MAGNETS AND THE THEORY OF SPIN BATH: REVISITED

Chapter 8

Introduction

At the end of the twentieth century a new class of magnetic materials was discovered: Molecular magnets. Each molecule in these materials act like a giant spin when temperature is lowered sufficiently. The simplest model describing most of these single molecule magnets (SMMs) has a biaxial spin Hamiltonian as follows

$$H = -DS_z^2 + E(S_x^2 - S_y^2) \tag{8.1}$$

where $0 < |E| < D$ and sign of E can be positive or negative. In Hamiltonian 8.1, for $E > 0$, the easy axis is z, the medium axis is y, and the hard axis is x. This Hamiltonian is also called easy-plane-easy-axis Hamiltonian, referring to its easy plane, here YOZ, and its easy axis, x.

Fe8 and Mn12 are the most investigated SMMs [12]. Fe8 molecules with formula $([(tacn)_6Fe_8O_2(OH)_{12}]^{8+})$, where tacn, pronounced "tack-en", is the macrocyclic ligand $C_6H_{12}(NH)_3$, forms a crystal (Fig. 8.1). In each molecule of Fe8 there are eight ions of Fe^{3+} each with spin 5/2. These ions strongly interact with each other via intramolecule exchange interaction and below $400m$K they are locked into a fixed structure and act as a single giant spin with total spin of $S = 10$. The values of D, E, parameter $\Lambda = 2|E|/(D - |E|)$, Prokofev-Stamp action $A_0 = 2S/\sqrt{\Lambda}$ and Chudnovsky-Gunther action $S_E = S\ln[(1+\sqrt{\lambda})/(1-\sqrt{\lambda})]$, where $\lambda = \frac{D-E}{D+E}$, for Fe8 along with three other SMMs are tabulated in Table 8.1. We shall discuss the relevance of the last three quantities in the remaining of this chapter and next chapter.

Table 8.1: Parameters of biaxial Hamiltonian (8.1) of two major single molecule magnets Fe8, Mn12 along with Mn4's [12, 30]

SMM	S	D (K)	$\|E\|$ (K)	Λ	A_0	S_E
Fe8	10	0.295	0.056	0.47	29.1	23.4
Mn12	10	0.65	0	0	∞	∞
Mn4(S=9/2)	9/2	0.68	0.064	0.20	20.1	13.7
Mn4(S=8)	8	0.43	0.029	0.14	42.7	27.1

Similar argument apply for Mn12, Mn4(S=9/2) and Mn4(S=8). One can find their chemical formulas in Refs. [12, 30]. Infinite actions for Mn12 should not be surprising becuase the Hamiltonian (to quadratic

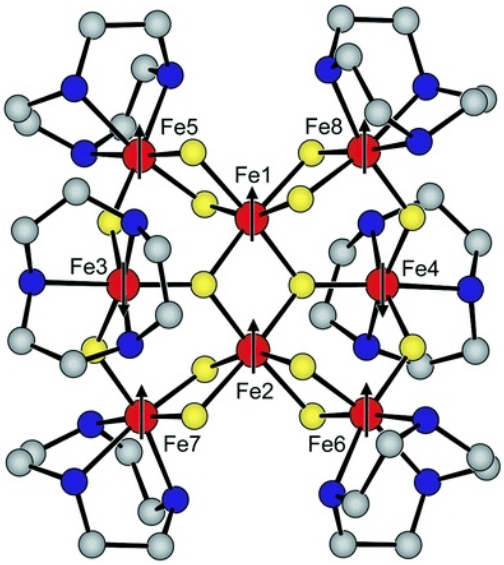

Figure 8.1: Fe8 molecular structure. Courtesy of Ref. [12]

order) commutes with S_z. To observe tunneling one usually applies magnetic field along the x or y axes [12]. For other three SMMs, one observes that the actions are finite but quite large. Thus, one can consider all of them to be in the WKB limit.

In general, one may need to add fourth order (quartic) terms to Hamiltonian of Eq. 8.1 for a complete study of SMMs. However, those terms are orders of magnitude smaller than the quadratic terms and in many theoretical studies of SMMs are not neglected [70, 71, 72, 73, 74, 75, 76, 50, 53].

The SMMs in their crystals interact with other SMMs and with nuclear spins, defects, phonons, etc.. Prokofev, Stamp and compony (PS) developed a theory, called the theory of spin bath, to solve the problem of the dynamics of giant spins in interaction with their surrounding microscopic spins [53, 61, 62, 77, 50, 60, 3]. The theory assumes a biaxial Hamiltonian for the giant spin (Eq. (1.3) of [50, p. 2903])

$$H_{PS} = \frac{1}{S}[-K_{\|}S_z^2 + K_{\perp}S_y^2] \tag{8.2}$$

and adds to it interaction Hamiltonian of the giant spin with microscopic spins (spin bath) and also the self Hamiltonian of the spin bath to form the total high energy Hamiltonian of the system plus environment (the spin bath). Since in low temperatures only the ground state doublet of the giant spin is of the main importance, PS truncate the resultant Hamiltonian to find the effective Hamiltonian of the two level system and the spin bath.

The whole literature of the spin bath theory [53, 61, 62, 77, 60, 3] is based on the validity of this truncation

scheme. Tupitsyn, Prokofev and Stamp (TPS) describe this truncation method in details in Ref. [50]. There are two assumptions that they make for Hamiltonian of Eq. 8.2 and truncate the high energy Hamiltonian assuming validity of these two:

$$S \gg 1 \tag{8.3}$$

$$\Lambda = K_{\perp}/K_{\parallel} \gg 1 \tag{8.4}$$

[50, p. 2909]. The first limit ($S \gg 1$) is relevant to some SMMs, however, the most important SMMs, Fe8 and Mn12, for purpose of quantum tunneling do not satisfy the second one ($\Lambda \gg 1$). In fact $\Lambda < 1$ for a large class of SMMs. For $E < 0$ one can write Eq. 8.1 in the form of Eq. 8.2 as follows,

$$H = -(D - |E|)S_z^2 + 2|E|S_y^2 \tag{8.5}$$

where we added $|E|S.S = |E|S(S+1)$ to the Hamiltonian. From Eq. 8.5 one observes that

$$\Lambda = \frac{2|E|}{D - |E|} \tag{8.6}$$

If $E > 0$ one can rotate the coordinate around the z-axis by 90 degrees to relabel x and y axis and obtain the same result. The values of Λ for the four major SMMs are tabulated in the fourth column of Table 8.1. As one sees $\Lambda < 1$ for all four SMMs. Thus, the second assumption that TPS make exclude the interesting examples of SMMs. To further insure this point we quote a passage from book [12] on Molecular Nanomagnets. Gatteschi, Sessoli, and Villain on discussion of diabolic points write [12, p. 208]:

"*For $H_z = 0$, the Hamiltonian (6.81) reduces to $\mathcal{H} = -D'S_z^2 + BS_x^2 + g\mu_B H_x S_x$. This Hamiltonian is easy to study in the limit $0 < D' \ll B$ (Weigert 1994). This case has perhaps no important experimental applications, but it is interesting because the number of diabolic points for given values of B and D can be determined by simple, analytical argument.*"

The limit $0 < D' \ll B$ in the Hamiltonian in the quotation is the same as $\Lambda \gg 1$. As mentioned in the quotation this limit has perhaps no important experimental applications. In the theory of the spin bath the truncation problem is solved in the limit $K_{\perp}/K_{\parallel} \gg 1$ "so that the tunneling amplitude is appreciable" [50, p. 2909]. However, experiments later showed that in fact tunnel splittings in molecular magnets are very small [11]. For Fe8, for instance, the tunnel splitting in the ground state is $\Delta_{Fe_8} = 0.1\ \mu\text{K} \simeq 2$ kHz, which is six orders of magnitude smaller than a tunnel splitting in superconducting flux qubits $\Delta_{\text{flux qubit}} \sim 1 - 5$ GHz [24].

The results of the theory of spin bath depend on the assumptions about S and Λ. This means that for molecular magnets in the regime $\Lambda < 1$ one has to repeat the truncation procedure to obtain the effective Hamiltonian. In chapter 9 we show that even the equation for the tunnel splitting of a bare giant spin in the theory of spin bath depends on the assumption of $\Lambda \gg 1$, let alone the rest of the truncation. We use coherent spin bath integral technique to obtain the tunnel splitting and compare our result with the literature.

In chapter 10 we use the method of chapter 3 to truncate the biaxial Hamiltonian for a half odd integer spin in the regime of $\Lambda < 1$ and compare our effective Hamiltonian with that of the theory of spin bath. We show our results differ from those of the spin bath theory.

Finally, in chapter 11 we return to the regime of $\Lambda \gg 1$ of the spin bath theory and compare the effect of topological decoherence terms, which are byproducts of truncation procedure in the theory of spin bath [3], and transition to higher excited states. We find that topological decoherence terms are of the same importance as the terms that cause transition to higher excited states, beyond the two lowest lying levels. The transition terms are always truncated in the truncation procedure. We conclude that in a consistent picture if one wants to include the decoherence terms in the effective Hamiltonian, one needs to include two more states to the picture and instead of a two level system, truncate the high energy Hamiltonian to a four level system.

Chapter 9

Coherent state path integral calculation of tunnel splitting in single molecule magnets

9.1 Introduction

In this chapter we calculate tunnel splitting in single molecule magnets with biaxial Hamiltonians. We use coherent path integral method for calculating the tunnel splitting. Van Hemmen and Süto Gunther1995 solved this problem by use of WKB method. Chudnovsky and Gunther[78] employed the path integral technique and solved the equation of motion for the classical paths to calculate the tunnel splitting. We instead use imaginary angles to find the splitting without solving the equation of motion for instantons.

This chapter can be regarded as a continuation of the work of Garg in Ref. [70] who employed imaginary angles to find the values of quenching fields when a field along the hard axis is applied. Garg calculated the imaginary part of the action for the instantons to study their topological effects. We calculate the real part as well to find the tunnel splitting completely.

In handling the real part of the action we find that the two paths that Garg [70] considered as the classical paths give rise to an unphysical result. There are two other paths that serve as the classical paths and one should calculate their actions. This finding can help one to solve a more general problem of a giant spin in presence of a magnetic field with component along the medium and hard axes.

9.2 Instanton calculation of tunnel splitting

The instanton method seems peculiar in the sense that it uses imaginary time. In some spin Hamiltonian problems use of the instanton method can become even more strange as one has to utilize imaginary angles as well. Is this method reliable in multidimensional problems such as the spin Hamiltonian problems? In this chapter we calculate tunnel splitting in single molecule magnets with biaxial Hamiltonian and use imaginary angles. We show that our result agree with WKB result and other path integral results which did not use imaginary angles.

To begin with, we orient the coordinate system such that the hard axis is along the z-axis, the easy axis is along the x-axis and the medium axis is along y. The biaxial Hamiltonian can be written as

$$H = k_1 S_z^2 + k_2 S_y^2 \tag{9.1}$$

where $k_1 > k_2 > 0$.

We use the coherent-state path integral method to find the splitting. It is ideal for this technique to not orient the easy axis along the z-axis, because the coherent state along the south pole on the z-axis is not well defined. The best choice is to orient the easy axis along the x-axis as we do here. Below we briefly review the path integral technique.

For any Hamiltonian H, the operator e^{-HT} can be expanded in terms of its energyeigen states $|i\rangle$ and eigenvalues E_i as follows

$$e^{-HT} = \sum_i e^{-E_i T} |i\rangle\langle i| \tag{9.2}$$

Here T is a quantity with dimension of inverse energy and otherwise of no physical significance (it is unrelated to inverse of actual temperature of the environment β). Consider two states $|\mathbf{n}_1\rangle$ and $|\mathbf{n}_2\rangle$. For the matrix element

$$\langle \mathbf{n}_1 | e^{-HT} | \mathbf{n}_2 \rangle = \sum_i e^{-E_i T} \langle \mathbf{n}_1 | i \rangle \langle i | \mathbf{n}_2 \rangle \tag{9.3}$$

the largest terms in the expansion in the limit of $T \to \infty$ are the terms corresponding to the ground state and possibly the first excited state, if the energy of the latter is close enough to the ground state energy. Other terms are exponentially smaller. Thus,

$$\langle \mathbf{n}_1 | e^{-HT} | \mathbf{n}_2 \rangle \simeq e^{-E_0 T} \langle \mathbf{n}_1 | 0 \rangle \langle 0 | \mathbf{n}_2 \rangle + e^{-E_1 T} \langle \mathbf{n}_1 | 1 \rangle \langle 1 | \mathbf{n}_2 \rangle \tag{9.4}$$

If $|\mathbf{n}_1\rangle$ and $|\mathbf{n}_2\rangle$ are chosen such that, and $|0\rangle$, $|1\rangle$ allow such choice,

$$\langle \mathbf{n}_1 | 0 \rangle \langle 0 | \mathbf{n}_2 \rangle = -\langle \mathbf{n}_1 | 1 \rangle \langle 1 | \mathbf{n}_2 \rangle \tag{9.5}$$

then Eq. 9.4 becomes

$$\langle \mathbf{n}_1 | e^{-HT} | \mathbf{n}_2 \rangle \simeq 2 \; e^{-(E_0+E_1)T/2} \; \langle \mathbf{n}_1 | 0 \rangle \langle 0 | \mathbf{n}_2 \rangle \sinh[\frac{(E_1 - E_0)T}{2}] \tag{9.6}$$

Thus one can read off the energy splitting between the ground state and first excited state $\Delta E = E_1 - E_0$, if one knows how to calculate $\langle \mathbf{n}_1 | e^{-HT} | \mathbf{n}_2 \rangle$. This is typically done by use of path integral technique. For spin Hamiltonians, the relevant path integral is the coherent-state path integral and state $|\mathbf{n}_1\rangle$ and $|\mathbf{n}_2\rangle$ are

chosen to be the coherent states. The matrix element is equal to

$$\langle \mathbf{n}_1 | e^{-HT} | \mathbf{n}_2 \rangle = \int_{\mathbf{n}_1}^{\mathbf{n}_2} D\mathbf{n}(\tau)\ e^{-S_E[\mathbf{n}(\tau)]} \tag{9.7}$$

where the Euclidean action $S_E[\mathbf{n}(\tau)]$ is

$$S_E[\mathbf{n}(\tau)] = \int_{-T/2}^{T/2} [-iS(1-\cos\theta)\dot{\phi}(\tau) + E(\mathbf{n}(\tau))] d\tau. \tag{9.8}$$

Here the semiclassical energy $E(\mathbf{n})$ is

$$E(\mathbf{n}) = \langle \mathbf{n} | H | \mathbf{n} \rangle. \tag{9.9}$$

The path integral of Eq. 9.7 is dominated by the paths which have the largest Re $e^{-S_E[\mathbf{n}(\tau)]}$. These paths can be found by condition $\delta S_E[\mathbf{n}(\tau)] = 0$ that lead to [70]

$$iS\frac{d\mathbf{n}}{d\tau} = -\mathbf{n} \times \frac{\partial E}{\partial \mathbf{n}} \tag{9.10}$$

$$\frac{dE}{d\mathbf{n}} = 0 \tag{9.11}$$

The second equation implies that the semiclassical energy is conserved on the optimum action path (the classical path or the instanton). This alone can be used to find the action on the classical path.

The semiclassical energy of Hamiltonian of Eq. 9.1 can be written as [70]

$$E(\theta, \phi) = K_1 \cos^2\theta + K_2 \sin^2\theta \sin^2\phi \tag{9.12}$$

where $K_1 = S^2 k_1$, $K_2 = S^2 k_2$. $E(\theta, \phi)$ is nonnegative. Thus, its minimum value 0 minimizes the action 9.8. Therefore, the energy of the instanton is zero and the action on the classical path reduces to

$$S_E[\mathbf{n}(\tau)] = \int_{-T/2}^{T/2} [-iS(1-\cos\theta)\dot{\phi}(\tau)] d\tau = -iS \int_{\phi(-T/2)}^{\phi(T/2)} (1-\cos\theta) d\phi \tag{9.13}$$

The simplicity of the problem of coherent state path integral in the above situation is that one does not have to solve the classical equation and find the classical path in order to evaluate the above integral. If one can find $\cos\theta$ as a function of ϕ on the classical path, one can calculate the integral. This can be easily done by setting

$$E(\theta, \phi) = 0 \tag{9.14}$$

and solving for $\cos\theta$. The result is

$$\cos\theta_{\pm} = \frac{\pm i\lambda^{1/2}\sin\phi}{(1-\lambda\sin^2\phi)^{1/2}} \tag{9.15}$$

where

$$\lambda = K_2/K_1 = k_2/k_1 < 1. \tag{9.16}$$

$\cos\theta_+$ has a negative real part action $\mathrm{Re}S_E$ and is rejected on the physical ground. $\cos\theta_-$ has a positive real part action, and hence negative $-\mathrm{Re}S_E$ in e^{-S_E}, with, as a result, a well behaved fluctuation determinant and we accept it as the physical solution. Garg [70] used $\cos\theta_+$ instead and did not include $\cos\theta_-$ paths. In calculation of the imaginary part of the action this choice does not matter. But for the real part that we do here it makes a difference. On physical ground we reject $\cos\theta_+$ term. We will see that this choice give a correct WKB result while the other one would give a different tunnel splitting.

Since the starting point and final point of the instanton are coherent state $|\mathbf{n}_1\rangle = |\theta_1, \phi_1\rangle$ and $|\mathbf{n}_2\rangle = |\theta_2, \phi_2\rangle$ with real values of θ_i , ϕ_i , the instanton should start from and end at real-valued spherical angles. The only real values that satisfy

$$E(\theta, \phi) = 0 \tag{9.17}$$

are $\theta = \pi/2$, $\phi = 0, \pm\pi$. Therefore, the classical action integral becomes

$$S_E = -iS\int_0^{\pm\pi}(1-\cos\theta_-)d\phi \tag{9.18}$$

where $\cos\theta_-$ is given by either (9.15).

The integral over $\cos\theta_-$ is

$$\int_0^{\pm\pi}\frac{-i\lambda^{1/2}\sin\phi}{(1-\lambda\sin^2\phi)^{1/2}}d\phi = i\ \ln\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}} \tag{9.19}$$

Therefore the action becomes

$$S_E = \mp iS\pi - S\ln\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}} \tag{9.20}$$

The contributions of these two terms in the path integral are

$$D\ Te^{-i\pi S+S\ln\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}}} + D\ Te^{+i\pi S+S\ln\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}}} = 2DT\cos\pi S\left(\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}}\right)^S \tag{9.21}$$

where D is the contribution of fluctuations around the classical path apart from the zero mode which gives rise to T. These are contributions of single instantons. One can add contributions of multiple instantons to

find an expression similar to Eq. 9.6 with sinh function and energy splitting equal to

$$\Delta E = 4D\ |cos\pi S|\ \left(\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}}\right)^{S} = 4D\ |\cos\pi S|\ e^{-S\ln\frac{1+\sqrt{\lambda}}{1-\sqrt{\lambda}}} \tag{9.22}$$

This completes our derivation of tunnel splitting using imaginary angles. We note that the $\cos\theta_+$ path would give rise to a an action $S\ln\frac{1+\sqrt{\lambda}}{1-\sqrt{\lambda}}$ which is positive and does not describe the minimal path.

9.3 Comparison with the literature

Let us now compare the above result with the literature. Chudnovsky and Gunther in Ref. [78, p. 662-Eq. (14)] obtained a similar result, without the factor of $\cos\pi S$, by solving the equation of motion for the classical path and calculating the action of the classical path. Later, Loss, DiVincenzo, and Grinstein [79, p. 3233-Eq. (6)] found the factor of $\cos\pi S$ by including the factor of $iS\dot{\phi}$ in the Euclidean Lagrangian. Van Hemmen and Süto in [80, p. 27] compared their WKB result with the result of Chudnovsky and Gunther[78], similar to Eq. 9.22, and showed that their results agree in the limit that is sensible to tunneling (One can work out readily and observe that the limit they refer to, in the last paragraph of [80, p. 27], is $\Lambda < 1$ in the language of TPS).

We can compare the tunnel splitting of Eq. 9.22 with the one that Tupitsyn, Prokofev and Stamp found in Ref. [50] by writing TPS Hamiltonian

$$H_{TPS} = \frac{1}{S}[-K_{\|}S_z^2 + K_{\perp}S_y^2] \tag{9.23}$$

in the form of Hamiltonian of Eq. 9.1. To this end, we add a constant $K_{\|}(S+1) = \frac{1}{S}K_{\|}S.S$ to the above Hamiltonian to get

$$H_{TPS} = \frac{1}{S}[(K_{\perp}+K_{\|})S_y^2 + K_{\|}S_x^2] \tag{9.24}$$

Now we relabel the axes by the cyclic rotation $x \to y \to z \to x$ to obtain

$$H_{TPS} = \frac{1}{S}[(K_{\perp}+K_{\|})S_z^2 + K_{\|}S_y^2] \tag{9.25}$$

This is the same as Hamiltonian of Eq. 9.1 if one sets

$$k_1 = \frac{K_{\perp}+K_{\|}}{S} \tag{9.26}$$

$$k_2 = \frac{K_{\|}}{S} \tag{9.27}$$

For this system, according to Eq. 9.22 the tunnel splitting is

$$\Delta E = 4D\ |\cos \pi S| \exp\left(-S \ln \frac{1+\sqrt{K_{\parallel}/(K_{\perp}+K_{\parallel})}}{1-\sqrt{K_{\parallel}/(K_{\perp}+K_{\parallel})}}\right) \tag{9.28}$$

In the limit of $\Lambda = K_{\perp}/K_{\parallel} \gg 1$, which is the limit in which TPS solved the truncation problem, the factor in the square root becomes very small and one can approximate the above expression by

$$\Delta E = 4D\ |\cos \pi S| \exp\left(-2S\sqrt{K_{\parallel}/K_{\perp}}\right) = 4D\ |\cos \pi S| \exp\left(-2S/\sqrt{\Lambda}\right) \tag{9.29}$$

Eq. 9.29 agrees with the result of TPS for a bare system with biaxial Hamiltonian (9.23) [50, p. 2909]. A similar comparison is done by Chudnovsky in Ref. [80, p. 80-81].

In general, however, Eqs. 9.28 and 9.29 are not equivalent. They are only equivalent in the limit of $\Lambda \gg 1$. This indicates that the TPS results in Ref. [50] for the truncation problem depend on the condition $\Lambda \gg 1$. If this is not satisfied, which isn't for many single molecule magnets, TPS effective Hamiltonian can give an incorrect result. This is indeed the case as we show in the next chapter. In it I truncate the biaxial Hamiltonian for half-odd-integer-spin in presence of a spin environment by use of method of chapter 3 and compare the result with that of the theory of spin bath. I show there is a strike difference between the two results, suggesting that the theory of spin bath results are only valid in the limit of $\Lambda \gg 1$ that is not relevant to many SMMs.

9.4 Conclusion

In conclusion, in this chapter we calculated tunnel splitting of a giant spin with a biaxial Hamiltonian by the use of imaginary angles in coherent path integral technique. Our result match the literature that use other methods in calculating the splitting. In the limit of $\Lambda \gg 1$ our result also match with the ones of the spin bath theory. We conclude that the assumption of $\Lambda \gg 1$ plays a key role in the spin bath theory and for molecular magnets whose $\Lambda < 1$ one needs to solve the problem of truncation and calculation of the spin bath again. We do the truncation problem for a case of half integer spin in the next chapter.

Chapter 10

Truncation in presence of environment

10.1 Introduction

In this chapter we use the results of chapter 3 to solve the truncation problem for half-odd-integer-spin single molecule magnets in presence of a spin environment. We compare our results with those of Tupitsyn, Prokofev and Stamp [50] and show that they differ by orders of magnitudes. We suspect that the difference is because the TPS result is inapplicable in the limit of $\Lambda < 1$ that is relevant to molecular magnets.

Although tunneling of magnetization is theoretically predicted to be suppressed in half-odd-integer-spins at zero magnetic field [79], experiments have always witnessed tunneling of magnetization in single-molecule magnets (SMM) with half-odd-integer spins [30, 12]. It is now believed that the tunneling is due to interaction of SMM with nuclear spins and other molecular spins in the sample [30]. One could use Prokofev-Stamp spin bath theory to justify this effect, at least qualitatively, if the theory was formulated for the limits relevant to real SMMs. However, as we discussed in the previous chapter this is not the case. Thus, one has to truncate the high energy Hamiltonian of half-integer SMM and spin bath for right limits and from that find the reason behind the tunneling. We shall do so in this chapter.

10.2 Truncation procedure

Suppose the giant spin has the biaxial Hamiltonian

$$H_S = -DS_z^2 + E(S_x^2 - S_y^2), \tag{10.1}$$

which is a generic Hamiltonian for SMMs to second order. The spin may interact with the microscopic spins of the spin bath through various types of interactions. Sum of all these interactions can be written in the following from

$$H_{int} = \sum_{k;\ \alpha,\beta} \omega^k_{\alpha\beta} S_\alpha \sigma^k_\beta \tag{10.2}$$

where $\alpha, \beta = x, y, z$; $\boldsymbol{\sigma}^k$ is the spin operator of the spin k of the bath, and $\omega^k_{\alpha\beta}$ are coupling coefficients. The self interaction of the microscopic spins also can be written in general as

$$H_{env} = \sum_{kl;\ \alpha,\beta} V^{kl}_{\alpha\beta} \sigma^k_\alpha \sigma^l_\beta \tag{10.3}$$

Thus, the total Hamiltonian of the system plus the bath is

$$H = H_S + H_{int} + H_{env} \tag{10.4}$$

To truncate this Hamiltonian we use the truncation method described in chapter 3. Since the ground state of half-odd-integer-spin is degenerate, by Kramers theorem or destructive interference of instantons [79], we use Eq. 3.11. Applying Eq. 3.11 and evaluating the submatrix elements of S_x, S_y, S_z numerically in the basis of the ground state $|0\rangle$ and first excited state $|1\rangle$ we obtain

$$H_{\text{eff}} = \sum_{k;\ \alpha,\beta} \omega^k_{\alpha\beta} c_\alpha \hat{\tau}_\alpha \sigma^k_\beta + \sum_{kl;\ \alpha,\beta} V^{kl}_{\alpha\beta} \sigma^k_\alpha \sigma^l_\beta \tag{10.5}$$

where τ_α are Pauli matrices describing spin-1/2 operator of the two level system, as in chapter 3, and c_α are coefficients that are obtained numrically. The values of c_α depend on parameters of Hamiltonian Eq. 10.1. For example for Mn_4 (S = 9/2), with $S = 9/2$, $D = 0.68$ K, and $E = 0.064$ K they are

$$c_x = 7.7 \times 10^{-5}, \qquad c_y = 8.5 \times 10^{-5}, \qquad c_z = 4.49671 \approx S \tag{10.6}$$

c_z is nearly $S = 9/2$. There is a way to understand this. For a degenerate state any linear combination of eigenstates is an eigenstate. We chose to use the localized states in either wells of Hamiltonian of Eq. 10.1 as the basis of the truncation in our numerical calculation of Eq. 10.6. Thus,

$$|0\rangle \approx |S\rangle, \tag{10.7}$$

$$|1\rangle \approx |-S\rangle \tag{10.8}$$

where states $|m = \pm S\rangle$ on the right hand side of the above equations are eigenstates of S_z. In this basis the submatrix elements of operator S_z are

$$(S_z)_{\{|0\rangle,|1\rangle\}} \approx \begin{pmatrix} S & 0 \\ 0 & -S \end{pmatrix} = S\ \tau_z \tag{10.9}$$

To understand values of c_x and c_y one should note that Eqs. (10.7-10.8) are only approximation and the ground state doublet are not exactly $|\pm S\rangle$,

$$|0\rangle \neq |S\rangle, \tag{10.10}$$

$$|1\rangle \neq |-S\rangle, \tag{10.11}$$

they rather have tails under the barrier. Therefore, one expects that the submatrix elements of S_x and S_y to be nonzero, although they can be exponentially small similar to tunnel splitting Δ_0 in absence of interference effects of the instantons. The tunnel splitting Δ in SMM problems can be written in general as $\Delta = \Delta_0 \cos \pi S$ [79]. For half-odd integer S, Δ is zero. However, by varying magnetic field along the hard axis one can find Δ_0 approximately [70]. For Mn4 (S=9/2) I obtained $\Delta_0 \sim 3 \times 10^{-5}$ K, which is exponentially small compared to $D \sim 0.7$ K.

We finally note that the precision of our numerical calculations of c_α is about 10^{-15}, so nonzero values of c_x and c_y are not results of round off error.

Now we can use the effective Hamiltonian Eq. 10.5 to find the reason of tunneling of half-odd-integer giant spins.

10.3 Why do half-odd-integer single-molecule magnets tunnel?

Suppose the SMM starts in the localized state $|0\rangle \approx |S\rangle$. By definition of the Pauli matrices in chapter 3

$$\tau_x|0\rangle = |1\rangle \qquad \tau_y|1\rangle = -i\ |0\rangle. \tag{10.12}$$

Two terms in the effective Hamiltonian (10.5) have these operators

$$\tau_x \sum_{k;\ \beta} \omega^k_{x\beta} c_x \sigma^k_\beta \qquad \tau_y \sum_{k;\ \beta} \omega^k_{y\beta} c_y \sigma^k_\beta \tag{10.13}$$

Thus, these terms cause the SMM to tunnel through the barrier and go to the other localized state $|1\rangle$. This is the reason behind tunneling of half-odd-integer spin SMMs in experiments (For example Wernsdorfer et. al. report tunneling of Mn4 (S=9/2) in [30]). In the process of tunneling, the microscopic spins involved, as the mediator, can flip with the molecular spin or can stay fixed. In both cases the giant spin can flip. One can also use the above argument and replace the microscopic spins by the molecular spins of other molecules in the crystal to argue that molecular interactions can also contribute to the tunneling process.

Next we compare our results with those of Tupitsyn, Prokofev and Stamp [50]

10.4 Comparison with Prokofev-Stamp spin bath theory

Tupitsyn, Prokofev and Stamp in Sec. 4 of [50, p.2916-2918] write their biaxial Hamiltonian 8.2 in units of $K_\perp$ as follows

$$H = \frac{1}{S}[-S_z^2 + \Lambda S_y^2 + \frac{\omega_0}{2}\vec{S}.\vec{\sigma}] \tag{10.14}$$

plus a Zeeman term for external magnetic field that we set to zero since we are going to study the case of zero external magnetic field here. Above, $\Lambda = K_\perp/K_\parallel$ as in chapter 8. For half-odd-integer giant spin TPS obtain effective Hamiltonian [50, p. 2918]

$$H_{\text{eff,TPS}} \approx (-1)^{2S+1}\{2\Delta_0\ \alpha\ \tau_y\sigma_x - 2\ \Delta_0\frac{\alpha}{\sqrt{\Lambda}}\tau_x\sigma_y\} + \frac{\omega_0}{2}\tau_z\sigma_z \tag{10.15}$$

where $\alpha = \pi\omega_0/2\Omega_0$ and $\Omega_0 = 2(K_\parallel K_\perp)^{1/2}$ [50, p. 2909]. In unit of $K_\parallel$, one has $\Omega_0 = 2\sqrt{\Lambda}$. Ω_0 is, according to TPS [50, p. 2904], the bounce frequency of the instantons and is roughly equal to the energy gap between the lowest doublet of states and the higher excited states.

To compare with TPS results we start with Hamiltonian of Eq. 10.14 and truncate it using the method of chapter 3, as described in previous sections of this chapter. We obtain

$$H_{\text{eff}} = \frac{\omega_0}{2}\left(c'_x\tau_x\sigma_x + c'_y\tau_y\sigma_y + c'_z\tau_z\sigma_z\right) \tag{10.16}$$

where for example for $S = 9/2$ and $\Lambda = 0.2$ the coefficients are

$$c'_x = 1.9\times10^{-5} \quad c'_y = 1.7\times10^{-5} \quad c'_z = 0.9992 \tag{10.17}$$

In comparison with Eq. 10.15, the coefficient of the term that involves τ_z matches pretty well. However, in equation (10.16), τ_x is coupled to σ_x and τ_y is coupled to σ_y while in Eq. 10.15 one has $\tau_y\sigma_x$ and $\tau_x\sigma_y$ couplings. If one associates this to different definitions of coordinate systems, one still sees that the coefficients of these two terms do not match in Eqs. (10.15) and (10.16). The ratio of the coefficient of the σ_x term to the coefficient of σ_y term in Eq. 10.15 is $\sqrt{\Lambda}$, which for $\Lambda = 0.2$ this number is $\sqrt{\Lambda} = 0.44$. However, the ratio of the coefficient of the σ_x term to the coefficient of σ_y term in Eq. 10.15 is $c'_x/c'_y = 1.11$ that is larger than $\sqrt{\Lambda} = 0.44$. Even $c'_y/c'_x = 0.89$ is twice as large as $\sqrt{\Lambda}$. Thus, the coefficients do not match.

The above discussion was irrespective to the value of Δ_0. If one uses the formula of TPS to calculate Δ_0 one observes strike difference between Eqs. (10.15) and (10.16). TPS state on page 2909 of [50] that

$$|\Delta_0| = \sqrt{2\ \mathrm{Re}A_0\ /\pi}\ \Omega_0\ e^{-A_0} \tag{10.18}$$

where

$$A_0 = 2S(K_\parallel/K_\perp)^{1/2} \tag{10.19}$$

$$\Omega_0 = 2(K_\parallel K_\perp)^{1/2}. \tag{10.20}$$

A_0 is the action of instantons on the semiclassical trajectories, neglecting the interference effects. In units of $K_\parallel$, Δ_0 becomes

$$|\Delta_0| = \sqrt{\frac{4S}{\pi\Lambda^{1/2}}}\ 2\sqrt{\Lambda}\ e^{-2S/\Lambda^{1/2}} \tag{10.21}$$

where the action became $A_0 = 2S/\Lambda^{1/2}$ as noted on page 2921 of [50]. The conclusion of TPS is that as long as $A_0 > 1$ their formulas are rather accurate [50, p. 2923]. For $S = 9/2$ and $\Lambda = 0.2$, $A_0 = 20.1$. Thus, one expects that Eq. 10.15 to match with truncation of Eq. 10.16, which is based on the standard method of truncation described in chapter 3. However, from Eq. (10.21) one obtains for $S = 9/2$ and $\Lambda = 0.2$,

$$\Delta_0 = 8.6 \times 10^{-9}. \tag{10.22}$$

which is very smaller than a bare tunneling one would expect for Mn4. Substituting this into Eq. 10.15 and using the definition of $\alpha = \pi\omega_0/2\Omega_0 = 1.72\ \omega_0$ one obtains numerical coefficients of TPS effective Hamiltonian

$$H_{\mathrm{eff,TPS}} \approx \frac{\omega_0}{2}\{2.9 \times 10^{-8}\ \tau_y\sigma_x - 6.5 \times 10^{-8}\tau_x\sigma_y + \tau_z\sigma_z\} \tag{10.23}$$

The coefficients of the first two terms are three orders of magnitude smaller than those of Eq. 10.16. We believe this much discrepancy is due to an error in the formula of TPS for Δ_0 Eq. 10.18 (i.e. Eqs. (3.9-3.12) of Ref. [50]). We shall discuss this point in more details in the next chapter.

10.5 Conclusion

In conclusion, in this chapter we truncated the high energy Hamiltonian of half-odd-integer-spin single molecule magnets in interaction with their spin bath environment and showed that the interaction terms in the truncated Hamiltonian cause the SMM to tunnel. We compared the result of our truncation with that of the spin bath theory [50] and showed that our results differ from that.

Chapter 11

Topological decoherence and transition to higher excited states

11.1 Introduction

In this chapter we compare the probability amplitude of transition to higher excited states, beyond the ground state doublet, and the probability amplitude of tunneling only due to the topological terms. We restrict ourselves to the limit $\Lambda \gg 1$ that Tupitsyn, Prokofev and Stamp considered in Ref. [50]. The result of this chapter is that these two probabilities are comparable for times $t \leq \Delta_0/\sqrt{S}$. This time scale is quite relevant to fast sweep experiments that are usually performed to measure the tunnel splitting, as we shall discuss in this chapter.

The self Hamiltonian of giant spin , as in Ref. [50], is

$$H_{TPS} = \frac{1}{S}[-K_{\|}S_z^2 + K_{\perp}S_y^2] \tag{11.1}$$

Two of the main assumptions of Ref. [50], in practice, for truncating this Hamiltonian when it interacts with nuclear spins with couplings ω_k are: (1) $\Lambda = K_{\perp}/K_{\|} \gg 1$ [50, p. 2909]. (2) $\omega_k/\Omega_0 \ll 1$, where Ω_0 is the energy gap between the lowest doublet of states and higher excited states. The second assumption is made to suppress transition to higher excited states. On this point TPS write: "In this paper we concentrate on the weak coupling regime $\omega_k/\Omega_0 \ll 1$ when the idea of an effective Hamiltonian is meaningful (we recall that by definition H_{eff} is operating in the low-energy subspace, and that a consistent solution requires $\omega_k \ll \Omega_0$" [50, pp. 2912-2913].

As a result of assumption (2), one would expect to not have terms of order ω_k/Ω_0 in the effective Hamiltonian H_{eff} of the two level system. However, one sees that all the topological terms that TPS incorporated in H_{eff} are of this order.

As a concrete example we shall study here the case of half-odd-integer-spin in interaction with one nuclear spin and in absence of external magnetic field as in Sec. 4 of [50]. This case is ideal for our study because one can isolate the effects of topological terms. Without these terms (if one considers the TPS truncation as

a complete theory in the limit $\Lambda \gg 1$) the central spin cannot tunnel. It is only the topological terms that allow the central spin to tunnel. TPS effective Hamiltonian for zero magnetic field, half-odd-integer spin in interaction with one nuclear spin is (Eq. (4.13) on p. 2918 of [50], we set $\psi \propto H_y = 0$. ψ is given in Eq. (4.4) of the reference and is proportional to external magnetic field along the y-axis)

$$H_{\text{eff}} = (-1)^{2S+1}\{2\Delta_0\alpha\ \tau_y\ \sigma_x - 2\Delta_0\frac{\alpha}{\sqrt{\Lambda}}\ \tau_x\ \sigma_y\} + \frac{\omega_0}{2}\ \tau_z\ \sigma_z \tag{11.2}$$

where

$$\alpha = \frac{\pi\omega_0}{2\Omega_0} \tag{11.3}$$

$\vec{\sigma}$ and $\vec{\tau}$ are Pauli operators of the nuclear spin and the central spin or the truncated giant spin. The localized state of the giant spin in either wells are the eigenstates of τ_z. This Hamiltonian is the truncation of (Eq. (4.1) on p. 2916 of Ref. [50])

$$H = \frac{1}{S}\left[-S_z^2 + \Lambda S_y^2 + \frac{\omega_0}{2}\vec{S}.\vec{\sigma}\right]. \tag{11.4}$$

The topological terms in the effective Hamiltonian (11.2) are the ones in the curly brackets. As one observes they are at most of order of $2\Delta_0\alpha$. Δ_0 is the tunnel splitting that it would be there if the destructive interference effects of the instantons did not exist. It is exponentially small. α is of order of ω_0/Ω_0 and is assumed to be small by assumption (2) to suppress transitions to higher excited states. Including a term of order $2\Delta_0\alpha$ in the effective Hamiltonian can be worrisome in two aspects: (a) It seems that the probability amplitude for tunneling through the barrier due to topological terms is comparable to probability amplitude of transition to higher excited state which one neglects to achieve the effective Hamiltonian (11.2). (b) The tunneling time that the topological terms can cause tunneling is of order $\frac{\Omega_0/\omega_0}{\Delta_0}$ which is much larger than Δ_0^{-1} and it is not obvious that it is relevant to experiments.

We study the first aspect quantitatively here. In the next section we calculate the transition probability amplitude from ground state to second excited state of the Hamiltonian of Eq. 11.4.

11.2 Transition probability of excitation to higher states beyond low lying levels

Since the spin is half-odd-integer the ground state of the bare biaxial Hamiltonian is degenerate. One can consider the ground states as the localized states in either wells

$$|0\rangle \approx |S\rangle \tag{11.5}$$

$$|1\rangle \approx |-S\rangle \tag{11.6}$$

The coupling term to the nuclear spin ω_0 is much smaller than energy gap Ω_0. Thus, one can treat the third term in Eq. 11.4 as a perturbation on the first two terms when one is interested in the physics of the system in one well and not the tunneling effect. Then one can use the perturbation theory to evaluate the transition amplitude between the ground state and the second excited state

$$|2\rangle \approx |S-1\rangle. \tag{11.7}$$

An important observation to make is that the matrix element of S_x between state $|0\rangle$ and $|2\rangle$ is

$$\langle 2|S_x|0\rangle \approx \sqrt{\frac{S}{2}} \tag{11.8}$$

This can be understood in virtue of Eqs. (11.5) and (11.7). I checked this numerically for exact ground state $|0\rangle$ and second excited state $|2\rangle$ for different values of Λ. In fact the exact value of $\langle 2|S_x|0\rangle$ is somewhat larger than $\sqrt{\frac{S}{2}}$ and exact value of $|\langle 2|S_y|0\rangle|$ is somewhat smaller than $\sqrt{\frac{S}{2}}$ (Eq. 11.8 is more general than the case of half odd integer and applies to full integer spins with a similar biaxial Hamiltonian as well. One can readily see this by noting that in this case $|0\rangle \approx (|S\rangle + |-S\rangle)/\sqrt{2}$ and $|2\rangle \approx (|S-1\rangle + |-S+1\rangle)/\sqrt{2}$. We confirmed this point numerically too.).

If the central spin starts in the ground state at $t = 0$ and the nuclear spin in $|+\rangle$, the eigenstate of σ_x, then the probability of being found in the second excited state according to perturbation theory (Sakurai Sec. 5.6, Eq. (5.6.22) [81] is

$$|c(t)|^2 = \sum_{n=\pm} \frac{4\left|\langle 2,n|\frac{1}{S}\frac{\omega_0}{2}\vec{S}.\vec{\sigma}|0,+\rangle\right|^2}{\Omega_0^2} \sin^2\Omega_0 t/2 \sim (\frac{\sqrt{2}\omega_0}{\sqrt{S}\Omega_0})^2 \sin^2\Omega_0 t/2 \tag{11.9}$$

Ω_0 is large compare to other energy scale in the problem. Thus, $\sin^2\Omega_0 t/2$ causes the probability to quickly oscillate between zero and $(\frac{\sqrt{2}\omega_0}{\sqrt{S}\Omega_0})^2$. Therefore, the probability amplitude oscillates between zero and

$$c_{max} \sim \frac{\sqrt{2}\omega_0}{\sqrt{S}\Omega_0} \tag{11.10}$$

We wish to compare this probability amplitude with the probability amplitude of tunneling due to the topological terms in 11.2. The latter can be regarded as a measure of the effect of topological terms. We calculate this probability amplitude in the next section.

11.3 Effect of topological terms on the tunneling probability

Since the central system starts in a localized state, its initial state in the two level system picture is $|\tau_z = +1\rangle$. The probability amplitude of tunneling to the other localized state $|\tau_z = -1\rangle$ is therefore

$$|d(t)|^2 = \sum_{n=\pm 1} \left|\langle \tau_z = -1, \sigma_x = n|e^{-iH_{\text{eff}}t}|\tau_z = +1, \sigma_x = +1\rangle\right|^2. \tag{11.11}$$

For times $t \ll (2\alpha\Delta_0)^{-1}$ we can keep terms to linear order in $2\alpha\Delta_0 t$ in the above expression to find the order of magnitude estimate of $|d(t)|$:

$$|d(t)| \sim 2\alpha\Delta_0 t + \mathcal{O}(\frac{2\alpha\Delta_0}{\omega_0}). \tag{11.12}$$

11.4 Comparison between the two probabilities

Comparing Eqs. (11.10) and (11.12), we observe that the tunneling probability amplitude due to topological terms are of order of probability amplitude of transition of the system to third excited state if

$$\Delta_0 t + \mathcal{O}(\frac{\Delta_0}{\omega_0}) \sim \frac{1}{\sqrt{S}}. \tag{11.13}$$

ω_0 is usually orders of magnitude larger than Δ_0. Thus, the largest of the two in the left hand side of the above equation for time $t \sim 1/(\Delta_0\sqrt{S})$ is the first one. Therefore, the condition becomes

$$\Delta_0 t \sim \frac{1}{\sqrt{S}} \tag{11.14}$$

This means that for

$$0 < t < \frac{1}{\Delta_0\sqrt{S}} \tag{11.15}$$

the transition probability to higher excited states is larger than the probability of tunneling due to topological terms. In other words, over such time scales the topological terms are immaterial if one wishes to use the

two level system (TLS) picture. Including them in the Hamiltonian is equivalent to breaking down the TLS picture.

11.5 Relevance to experiments

The time range of condition 11.15 is in fact quite relevant to many experiments. For example in order to measure tunnel splitting in SMMs, researchers conduct Landau Zener experiments [11, 30, 12]. The measurements are done in the fast sweep rate where the probability of flipping of the central spin is less than 4% (See e.g. Ref. [30], Fig. 2 and Eq. (2)). We recall from chapter 5 that the probability of not flipping is

$$P = e^{-\pi\Delta^2/2v} \tag{11.16}$$

Thus, in the fast sweep regime of such experiments

$$1 - e^{-\pi\Delta/2v} < 0.04. \tag{11.17}$$

This condition implies that

$$v > 38\Delta^2. \tag{11.18}$$

We also recall from chapter 5 that the transition time in Landau Zener problems is of order $t \sim \Delta/2v$. Over this time the tunneling can take place. For the fast sweep of inequality 11.18 this time is

$$t < \frac{1}{76\Delta}. \tag{11.19}$$

Comparing inequalities (11.19) and (11.15), we come to the conclusion that for

$$S < 5776 \tag{11.20}$$

the topological terms are as important as the transition terms to higher excited states in fast sweep experiments. Most, if not all, the SMMs satisfy condition (11.20).

11.6 Conclusion

In conclusion, we showed in this chapter that even in the regime of $\Lambda \gg 1$, the topological terms have the same significance as the transition terms to higher states for times $t < 1/\Delta_0\sqrt{S}$. We discussed that the time regime $t < 1/\Delta_0\sqrt{S}$ is relevant to sweep experiments that are conducted for measuring tunnel splitting in

molecular magnets. As a result, a consistent truncation of half-odd-integer should incorporate all four low lying levels simultaneously. The idea of the two level system is not accurate in this case.

For full integer spins since the gap is not quenched and gives rise to a term of order Δ in the effective Hamiltonian, which is much larger than topological terms of order $\alpha\Delta$, one can utilize the two level system picture. However, one can neglect topological terms because they are of order of transition terms to higher excited states.

Appendix A

A property of the reduced density matrix

In this appendix we prove the inequality Eq. 6.14, $|\rho_{10}| \leq \sqrt{\rho_{00}\rho_{11}}$, for any 2×2 reduced matrix.

Quite generally, the density matrix of the universe can be written in terms of a statistical mixture of pure states of the universe, $\rho_U = \sum_n p_n |\Psi_n\rangle\langle\Psi_n|$ where $|\Psi_n\rangle$ are pure states of the universe and p_n are their statistical probabilities ($p_n \geq 0$, $\sum_n p_n = 1$.)

One can choose basis $\{|i,\mu\rangle\}$ for the universe which is a direct product of a basis of the system $\{|i\rangle\}$, where $i = 0, 1$, and a basis of the bath $\{|\mu\rangle\}$. Then one can expand the pure states $|\Psi_n\rangle$ in terms of this basis, $|\Psi_n\rangle = \sum_{i,\mu} c^n_{i,\mu} |i,\mu\rangle$ and rewrite the universe density matrix as

$$\rho_U = \sum_{n;i,j;\mu,\nu} p_n \; c^n_{i,\mu} \; c^{n^*}_{j,\nu} \; |i,\mu\rangle\langle j,\nu| \tag{A.1}$$

Now the reduced density matrix elements are as follows,

$$\rho_{00} = \sum_{n;\mu} p_n \; \left|c^n_{0,\mu}\right|^2 \tag{A.2}$$

$$\rho_{11} = \sum_{n;\mu} p_n \; \left|c^n_{1,\mu}\right|^2 \tag{A.3}$$

$$\rho_{10} = \sum_{n;\mu} p_n \; c^n_{1,\mu} \; c^{n^*}_{0,\mu}. \tag{A.4}$$

$|\rho_{01}|$ satisfies the following inequality

$$|\rho_{01}| = \left|\sum_{n;\mu} p_n \; c^n_{1,\mu} \; c^{n^*}_{0,\mu}\right| \leq \sum_{n;\mu} p_n \; \left|c^n_{1,\mu}\right| \; \left|c^{n^*}_{0,\mu}\right|. \tag{A.5}$$

We define vectors $\vec{R}_i$

$$\vec{R}_i = (\sqrt{p_0} \; \left|c^0_{i,0}\right|, \sqrt{p_0}|c^0_{i,1}|, \cdots ; \sqrt{p_1} \; \left|c^1_{i,0}\right|, \sqrt{p_1}|c^1_{i,1}|, \cdots) \tag{A.6}$$

Then we observe that $\rho_{00} = \left|\vec{R}_0\right|^2$, $\rho_{11} = \left|\vec{R}_1\right|^2$, and, from inequality Eq. A.5,

$$|\rho_{01}| \leq \vec{R}_0.\vec{R}_1. \tag{A.7}$$

Since, $\vec{R}_0.\vec{R}_1 \leq \left|\vec{R}_0\right|\left|\vec{R}_1\right|$ we conclude that

$$|\rho_{10}| \leq \sqrt{\rho_{00}\rho_{11}}. \tag{A.8}$$

Appendix B

Construction of $\Phi(t,\omega)$

Φ is a random process with Gaussian distribution whose variance $\sigma^2(t)$ is time dependent. We demanded that the process be differentiable with respect to time on each sample path in order to have a well-defined magnetic field in Eqs. (6.27)-(6.30). Such a process exists, as long as $\sigma^2(t)$ is differentiable with respect to time [82]. There are many constructions for Φ, depending on what correlation function one would like for the random process. The simplest construction is the following [82]: Let Z be a standard Gaussian random variable with mean zero and variance equal to unity. By definition of random variables, Z is a function from a sample space Ω to the real line : $\omega \mapsto Z(\omega)$. Now let

$$\Phi(t,\omega) = \sigma(t)Z(\omega), \tag{B.1}$$

where $\sigma(t)$ is taken from Eq. (6.21). $\Phi(t,\omega)$ has the properties we wanted. For each t fixed, the distribution of the process is Gaussian because the distribution of Z is Gaussian. Also the mean and variance are

$$\langle \Phi(t,\omega) \rangle_\omega = \langle \sigma(t)Z(\omega) \rangle_\omega = \sigma(t) \langle Z(\omega) \rangle_\omega = 0 \tag{B.2}$$

$$\left\langle \Phi(t,\omega)^2 \right\rangle_\omega = \sigma^2(t) \left\langle Z(\omega)^2 \right\rangle_\omega = \sigma^2(t) \tag{B.3}$$

as expected. For each ω fixed, $\Phi(t,\omega)$ is differentiable with respect to t since $\sigma(t)$ is so. Finally, $\Phi(t_i,\omega) = 0$ for initial pure states because $\sigma(t_i) = 0$ for such states. In this case the right hand side of Eq. (6.21) is zero. For mixed states the right hand side of Eq. (6.21) is nonzero and $\sigma(t_i) \neq 0$. However, $\Phi(t_i,\omega)$ is not required in the classical model of Sec. 6.5 to be zero either. Thus the random phase Eq. B.1 works well for this case, too.

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