The Dissipative Two-State System

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Abstract

The effect of decoherence in a two-state system (TSS) coupled to its environment hampers the use of the TSSs as qubits in quantum computation. The search for novel ways of realizing this coupling which could provide us with a better control of the environment, and hence of decoherence, is an important and actual problem in condensed matter.

Motivated by recent experiments in the group of H.Mooij (Delft), we investigate the behavior of a TSS, when it is indirectly coupled to a harmonic bath, via an anharmonic oscillator. This is done after we reproduce literature results for direct coupling to the bath and indirect coupling to the bath, via a harmonic oscillator. The total influence of the intermediate anharmonic oscillator and the bath is given by the effective spectral function. We find the approximate form of this function, and use this result to calculate some physical quantities of the system. For example the asymptotic equilibrium value of the expectation value of the state of the TSS is calculated. This result is compared with the analogous time dependent expectation value in the case of direct coupling, and the equilibrium value of this for the case of coupling via a harmonic oscillator. Finally, we reproduce results for the partition function of the TSS directly coupled to the bath and suggest analogous calculations for the TSS indirectly coupled to the bath.
A Solution of the equation of motion of a quartic anharmonic oscillator 66

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1 Introduction

The quantum mechanical two-state system (TSS) is an important subject in theoretical physics. It is also called spin-boson model, two-level system (TLS), or qubit. The two-state system is the simplest model for studying the effects of an environment, in terms of constructive and destructive quantum interference. It allows in a rather extensive way to investigate decoherence and damping on the quantum system imposed by a phenomenological environment. This investigation has applications in various fields of physics and even chemistry. For example the electron transfer between two fixed sites in a biomolecule can be modelled in this way.

The properties of a TSS embedded in an environment are also important for quantum information processing. Unlike an ordinary bit, a qubit does not take a fixed value 0 or 1, but it takes values 0 and 1 with certain probabilities. Because a probability can take all real values between 0 and 1, a qubit contains much more information than a classical bit. The ability to perform a number of operations (for example the controlled NOT gate) on qubits is an essential requirement for building a quantum computer. A quantum computer would allow us to perform important calculations which are absolutely impossible to do now, like the decomposition of large numbers in prime factors.

If the decoherence and damping of the TSS in contact with an environment can be understood better, these ideas can also be applied to more difficult systems, like a n – state system in contact with an environment.

The phenomenological environment can be considered to be analogous to a heat bath in thermodynamics. As proposed by Caldeira and Leggett [1], the environment can modelled by a bath of harmonic oscillators. We then speak of a harmonic bath. The influence of the bath on the TSS is fully enclosed in the so-called spectral density $J(\omega)$. Imposing the number of harmonic oscillators in the bath to be practically infinite, $J(\omega)$ can be seen as a continuous function.

If the system is equally damped for all frequencies, which is the case in many physical systems, $J(\omega)$ should be chosen proportional to $\omega$. This gives problems for very large $\omega$, so we use a cut-off frequency or multiply $J(\omega)$ with a decaying exponential. The form $J(\omega) \propto \omega$ is called an Ohmic spectral density, where the more general case $J(\omega) \propto \omega^s$ is called sub-ohmic or super-ohmic for $0 < s < 1$ or $s > 1$, respectively. Another interesting limit is provided by $J(\omega) \propto \delta(\omega - \Omega)$, the case of a single environmental mode at the specific frequency $\Omega$. The case of a TSS coupled to an Ohmic environment has been extensively treated in the literature (see [2] for a review).

An interesting alternative model is that of a TSS in a bath described by a spectral density which is peaked at a specific characteristic frequency $\Omega$, but behaves Ohmically for small $\omega$. This model received growing interest in the
context of quantum computation studies using condensed matter systems. In a superconducting flux qubit device, the two directions of the magnetic flux form the two states of the TSS. The device is read out by a dc-SQUID, which couples inductively to the qubit superconducting loop. At bias currents well below the critical current $I_c$, the SQUID can be modelled as an inductor $L_J$. An additional shunt capacitance $C_s$ creates an on-ship environment which improves the resolution of the dc-SQUID. The resulting impedance of this dc-SQUID environment is the same as that of a circuit of three parallel linked components: an inductance $L_J$, a capacitance $C_s$, and a resistance $R_l$.

Quite interestingly, the previous model is equivalent to a TSS coupled to a single harmonic oscillator, which is itself coupled to an harmonic bath with Ohmic spectral function. This model for a structured environment was proposed by Garg, Onuchic and Ambegaokar [3]. In this model the TSS experiences an effective $J(\omega)$ that has the shape of a Lorentzian: it is Ohmic for small $\omega$, then peaks around some value of $\omega$, and falls off for large $\omega$.

This last model can be used to describe solid state realizations of a qubit coupled to a resonator. Among the various schemes of measurement, entanglement generation, quantum information transfer and interaction with a non-classical state, there are promising candidates for realizing qubits in a possibly scalable supercomputer. The experiments by Mooij et al [4] are an example of these measurements. Some fundamental quantum physical properties like macroscopic quantum coherence, Rabi oscillations, and Ramsey interference, can also be observed.

This thesis deals with a novel theoretical model, which is a generalization of the previous structured environment. The question we want to answer is: are there fundamental changes in the physical properties of the TSS if the intermediate oscillator, which is itself coupled to the harmonic bath, is anharmonic instead of being harmonic? This configuration is motivated by recent experiments in the group of H.Mooij in Delft, which investigates new types of coupling to the environment that could possibly reduce the effects of decoherence and allow for more control of the bath. In order to answer this question, we first have to understand how to include dissipation in quantum-mechanics, and in particular how to include it in the quantum mechanical description of a two-state system.

But before tackling these more complex questions we start in chapter 2 by demonstrating the importance and relevance of a two-state system through the concept of a SQUID. In chapter 3, it is explained how a system could dissipate energy when coupled to a large reservoir of harmonic oscillators. Some important quantities are introduced. In chapter 4, the simplest model, with a direct coordinate-coordinate coupling between the two-state system and the reservoir is worked out. Most interesting in this thesis are the models where the two-state system and the reservoir are coupled via an intermediate harmonic oscillator (in
chapter 5) and via an intermediate anharmonic oscillator (in chapter 6), and the derivation of an effective spectral function in these cases. In chapter 7 the probability of being in one particular state of the two-state system is calculated for the conventional direct coupling to the bath. In chapters 8 and 9, the equilibrium value of this probability is evaluated for coupling to the bath via a harmonic or via an anharmonic oscillator, respectively. The partition function for the simplest model of direct coupling is calculated in chapter 10. As an outlook, further work could be done on the derivation of the partition function for indirect coupling via the intermediate harmonic or anharmonic oscillator.

In this thesis, chapters 2, 3, and 4 consist of a well known theory, which can be considered as text book material. The chapters 5, 7, 8 and 10 are for the largest part worked out versions of respectively section 2 of [3], section 4 of [2], section 4 of [5], and appendix A of [2]. Finally, the chapters 6 and 9 are completely filled with original work.

2 A SQUID as a Two-State System

This chapter is concerned with the rf-SQUID (Superconducting Quantum Interference Device), which is an example of a possible configuration for the two-state system in practice [6]. A SQUID is based on the Josepson effect [7], thus we start by first explaining the latter.

2.1 The Josephson Effect

We consider an arrangement of two superconductors 1 and 2, separated by a non-superconducting material of thickness \( d \). For \( d \) large, the two superconductors can be considered as decoupled. For \( d \) small enough, the wave-functions \( \psi_1 \) and \( \psi_2 \) of the two superconducting pieces can be described by the coupled Schrödinger equations

\[
\begin{align*}
    i\hbar \dot{\psi}_1 &= E_1 \psi_1 + \Delta \psi_2, \\
    i\hbar \dot{\psi}_2 &= E_2 \psi_2 + \Delta^* \psi_1,
\end{align*}
\]

(1a) (1b)

where \( E_1 \) and \( E_2 \) are the energies in the superconductors and \( \Delta \) is the overlap between the two wave-functions. This configuration is known as a Josephson Junction. We choose \( \Delta \in \mathbb{R} \) and insert \( \psi_1 = \sqrt{n_1} e^{i\phi_1} \) and \( \psi_2 = \sqrt{n_2} e^{i\phi_2} \). This is quite useful, because for the macroscopic eigenfunctions we have \( |\psi_i|^2 = n_i \) where \( n_i \) is the order parameter. The real and imaginary parts of Eqs. (1a) and (1b) yield four equations

\[
\begin{align*}
-\hbar \dot{\phi}_1 &= \Delta \sqrt{\frac{n_2}{n_1}} \cos(\phi_1 - \phi_2) + E_1, \\
-\hbar \dot{\phi}_2 &= \Delta \sqrt{\frac{n_1}{n_2}} \cos(\phi_1 - \phi_2) + E_2,
\end{align*}
\]

(2a) (2b)
\dot{n}_1 = \frac{2\Delta}{\hbar} \sqrt{n_1 n_2} \sin(\phi_1 - \phi_2), \quad (2c)
\dot{n}_2 = \frac{2\Delta}{\hbar} \sqrt{n_1 n_2} \sin(\phi_1 - \phi_2). \quad (2d)

Assuming now \( n_1 \approx n_2 \), writing \( \sqrt{n_1 n_2} = n_s \), and using Eqs. (2a) - (2d) we obtain
\dot{\phi}_1 - \dot{\phi}_2 = \frac{E_2 - E_1}{\hbar}, \quad (3a)
-\dot{n}_1 = \dot{n}_2 = \frac{2n_s \Delta}{\hbar} \sin(\phi_1 - \phi_2). \quad (3b)

If we now realize that \( \dot{n}_2 \) equals the current \( i \) through the junction, and denoting the phase-difference \( \phi_1 - \phi_2 \) as \( \varphi \), we find
\[ i = i_0 \sin \varphi, \quad (4a) \]
\[ \dot{\varphi} = \frac{2eV}{\hbar}, \quad (4b) \]
where \( i_0 \equiv 2n_s \Delta / \hbar \) and \( 2eV = E_2 - E_1 \). Eqs. (4a) and (4b) now show that when there is a phase-difference between the two superconductors, a constant current flows between them, without a measurable voltage. The current is caused by transport of the so-called Cooper-pairs. This effect is possible up to a maximal current \( i_0 \), and is called the Josephson effect. Above the value \( i_0 \), a voltage will develop between the two superconductors.

### 2.2 The SQUID

A possible configuration for a SQUID is a superconducting ring closed by a Josephson junction, see Fig. 1. Suppose we apply an external magnetic field \( H_x \) perpendicular to the plane of the ring. The flux in the ring, caused by \( H_x \), is denoted by \( \phi_x \). From elementary electrodynamics we know that for the total flux \( \phi \)
\[ \phi - \phi_x = Li, \quad (5) \]
where \( L \) is the self-inductance of the ring and \( i \) is the total current through the ring. Now we analyze in detail the total current through the ring, which has 3 components:

- **Josephson current**: the current component due to tunneling of Cooper pairs through the ring is given by \( i_s = i_0 \sin \varphi \)

Normal current: if \( V \) denotes the voltage across the junction and \( R \) is the normal resistance of the material in its normal phase, from Ohm’s law we have \( i_n = V/R \).
Polarization current: for the finite junction capacitance $C$ we have the polarization current $i_c = CV$.

The total current is thus given by

$$i = i_0 \sin \varphi + \frac{V}{R} + CV.$$ (6)

Insertion of (6) in (5) yields

$$\varphi - \varphi_x = i_0 \sin \varphi + \frac{V}{R} + CV.$$ (7)

Furthermore, we know that the flux inside the superconducting ring is quantized in units of $\phi_0 = \frac{hc}{2e}$. From the quantization relation we obtain

$$\phi + \frac{\phi_n}{2\pi} = n\phi_0,$$ (8)

for $n$ an integer, and thus

$$\sin \varphi = \sin \left(\frac{2\pi \phi}{\phi_0}\right).$$ (9)

Insertion of (9) and the relation $V = -\dot{\phi}$ in (7) results into the following equation of motion for a particle with "coordinate" $\phi$

$$C\ddot{\phi} + \frac{\phi}{R} + U'(\phi) = 0,$$ (10)
where the potential $U(\phi)$ is given by

$$U(\phi) = \frac{(\phi - \phi_x)^2}{2L} - \frac{i_0 \phi_0}{2\pi} \cos \left( \frac{2\pi \phi}{\phi_0} \right). \quad (11)$$

Observe that (10) is nothing else than the Langevin equation, proving that we are dealing with a dissipative system. The configuration as mentioned above is called the rf-SQUID. The ring is usually part of a circuit, in the sense that two points on the ring are contacted with an external source. If we consider Eq. (10), we see that the rf-SQUID can be seen as a circuit consisting of three elements linked parallel to a source. The three elements are a capacitor $C$, a resistance $R$, and a nonlinear element. These elements correspond in this order to the terms on the LHS of Eq. (10). In the so-called dc-SQUID, the ring contains two Josephson junctions. If the ring is divided in two parts by the contacts with the external source, these parts contain one junction each. The dc-SQUID is better for high precision-measurements because its noise is smaller.

To render clear the behavior of the SQUID, we consider a rf-SQUID and the potential $U(\phi)$ in Eq. (11). The first and second derivative of $U(\phi)$ equal

$$U'(\phi) = \frac{\phi - \phi_x}{L} + i_0 \sin \left( \frac{2\pi \phi}{\phi_0} \right), \quad (12)$$

$$U''(\phi) = \frac{1}{L} + \frac{2\pi i_0}{\phi_0} \cos \left( \frac{2\pi \phi}{\phi_0} \right). \quad (13)$$

The potential has a (local) extremum if $U'(\phi) = 0$. This can only happen for $|\phi - \phi_x| \leq i_0 L$. Because the minima of $\sin \left( \frac{2\pi \phi}{\phi_0} \right)$ are separated by a distance $\phi_0$, and because $\phi - \phi_x$ only switch sign once, the number of minima of the potential is bounded and could be tuned to be exactly two. This is not even necessary for our goal, because the minima close to $\phi_x$ are deeper than the others.

For $\phi_x = 0$ there is one central and global deepest minimum at $\phi = 0$ accompanied by two equal local minima on its left and right (only if $i_0 L$ is large enough). This potential is plotted in Fig. 2a. For $\phi_x = \phi_0/2$ there are two equally deep minima at $\phi = \phi_0/2 \pm \phi_m$, where $\phi_m$ is slightly smaller than $\phi_0/2$. A picture of this can be found in Fig. 2b. Most interesting is the case where $0 < \phi_x < \phi_0/2$. Then there is a deepest minimum for $\phi$ slightly larger than 0, and a second minimum for $\phi$ slightly smaller then $\phi_0$. An example of this situation can be found in Fig. 3. These two minima could be used to form a two state system, where it should be prevented that other minima also play a role.
Figure 2: (a) Potential $U(\phi)$ for $\phi_x = 0$; (b) Potential $U(\phi)$ for $\phi_x = \phi_0/2$.

Figure 3: Potential $U(\phi)$ for $\phi_x = \phi_0/4$. 
3 General Theory

In this chapter the concept of a dissipative system is introduced, such that in a later stage the dissipative two-state system can be treated. We also introduce some important quantities which are going to be used later. This chapter is mainly based on the references [1], [2], and [8].

3.1 Dissipative Systems

The canonical quantization procedure, used to go from classical to quantum mechanics, is based on the fact that we study closed systems, where the energy is conserved. Around 1980 people wondered how to include dissipation (loss of energy) in the quantum mechanical description of systems. The final aim was to establish the quantum theory of a system which is described in the classical limit by the Langevin equation

\[ M \ddot{q} + \eta \dot{q} + V'(q) = f(t) \]  

(14)

where

\[ \langle f(t) \rangle = 0 \]

\[ \langle f(t) f(t') \rangle = 2 \eta k_B T \delta(t - t') \].

This equation describes a particle with coordinate \( q \), immersed in a viscous environment. The quantity \( M \) is the mass of the particle, \( \eta \) is the friction coefficient, \( V'(q) \) is an external potential and \( f(t) \) is a fluctuating force.

The idea of Caldeira and Leggett [1] was to study a larger system, that contains two subsystems. The energy loss of one subsystem can then be compensated by an increase of the energy of the other subsystem. The first subsystem is the system of interest, the second one is a large reservoir. This reservoir must be large for the following reason. The system of interest can lose energy to the reservoir, which changes the quantum state of the reservoir. By making the reservoir large, only the quantum state of a small part of the reservoir is changed, so we can ignore the possibility of the system of interest being influenced by the actual state where the reservoir is in. This reservoir could be seen as the environment of the original system. Let’s discuss how it could be modelled.

3.2 Reservoir of Harmonic Oscillators

We first observe that the environment is relatively large compared to the system. This implies that the environment is only weakly perturbed by the dynamics of the system. If we now model the environment by a collection of harmonic oscillators (with arbitrary frequencies) that is large enough, then the chance that double excitations of the harmonic oscillators occur is negligible. From this assumption it can be seen that modelling the environment by a reservoir of harmonic oscillators is not more restrictive than assuming that the coupling
between system and environment is linear. This assumption seems to be reasonable. More details on this subject can be found in Appendix C of [1]. The Hamiltonian of the environment can thus be written as

\[ H_E = \sum_j \left( \frac{p_j^2}{2m_j} + \frac{1}{2}m_j\omega_j^2x_j^2 \right). \]  (15)

In general, a system (particle) with coordinate \( q \) and conjugate momentum \( p \), could be linearly coupled to a set of harmonic oscillators with coordinates \( x_j \) and conjugate momenta \( p_j \) by means of four extra terms in the Hamiltonian:

\[ p \sum_j F_j x_j + q \sum_j G_j p_j + p \sum_j H_j p_j + q \sum_j K_j x_j. \]  (16)

If we impose that \( q \) and \( x_j \) have opposite behavior under time reversal, the last two terms of (16) must be left out. We know that \( p \) and \( p_j \) are proportional to \( \dot{q} \) and \( \dot{q}_j \) respectively. Hence (16) can be rewritten as

\[ \dot{q} \sum_j \tilde{F}_j x_j + q \sum_j \tilde{G}_j \dot{x}_j. \]  (17)

By adding a total time derivative \(-\frac{d}{dt} \left\{ q \sum_j \tilde{F}_j x_j \right\}\) to the Hamiltonian (this is equivalent to subtracting this term from the Lagrangian, which is allowed) the first term of (17) is replaced by a term \(-q \sum_j \tilde{F}_j \dot{x}_j\). Now only a coupling between \( q \) and the \( \dot{x}_j \) is left. By performing a canonical transformation on \( x_j \) and \( \dot{x}_j \), this coupling can be transformed into a coordinate-coordinate coupling. Here, the new coordinates \( \tilde{x}_\alpha \) do have the same behavior under time reversal as \( q \). Details of the previous procedure can be found in chapter IIb of [8]. The interaction can thus be modelled by a term

\[ H_{\text{int}} = q \sum_j c_j x_j. \]  (18)

Therefore, if \( H_s \) describes the Hamiltonian of the system of interest, which is linearly coupled to a bath of harmonic oscillators, then the total Hamiltonian reads

\[ H_{\text{tot}} = H_S + H_{\text{int}} + H_E. \]  (19)

### 3.3 Spectral Function

We could ask ourselves, if a system with coordinate \( q \) is linearly coupled to a bath of harmonic oscillators with coordinates \( x_j \), how is the dynamics of the system then influenced by the bath? We assume the \( j \)-th harmonic oscillator has mass \( m_j \), frequency \( \omega_j \) and is coupled to the system with strength \( c_j \). Then
the answer is quite elegant: the influence is given only in terms of the spectral function of the bath.

\[ J(\omega) \equiv \frac{\pi}{2} \sum_j \frac{c_j^2}{m_j\omega_j} \delta(\omega - \omega_j). \] (20)

This can, for example, be seen from [9]. Let us consider for the system plus bath the specific Hamiltonian

\[ H = \frac{p^2}{2M} + U(q) + \sum_j \left[ \frac{p_j^2}{2m_j} + \frac{1}{2}m_j\omega_j^2 \left( x_j + \frac{c_j}{m_j\omega_j}q \right)^2 \right], \] (21)

where the term proportional to \( q^2 \) is added, but can be compensated by a term in the potential \( U(q) \). Defining \( U'(q) \equiv dU/dq \) and using dots for time derivatives, the classical equations of motion read

\[ M\ddot{q} = -U'(q) - \sum_j c_j x_j - q \sum_j \frac{c_j^2}{m_j\omega_j}, \] (22a)

\[ m_j\ddot{x}_j = -m_j\omega_j^2 x_j - c_j q. \] (22b)

These equations of motion can be Fourier transformed. Denoting the Fourier transforms of \( q(t) \), \( x_j(t) \) and \( U'(q) \) as respectively \( q(z) \), \( x_j(z) \) and \( U'_z(q) \), we can find an equation for \( q(z) \), that involves \( U'_z(q) \) and \( J(z) \), but is independent of the \( x_j(z) \). Hence all the dependence of the dynamics of the system on the bath, can be "summarized" by the spectral function \( J \). Note that this is only true for a coordinate \( q \) that is directly and linearly coupled to the bath. If the coupling is different, for example via another single harmonic oscillator, then a different "effective spectral function" can be derived. This will become clearer at the moment when we actually calculate it.

For the case of direct, linear coupling to the bath, the equation for \( q(z) \) could be transformed back from Fourier-space to ordinary space, into a differential equation for \( q(t) \). Then a pleasant property of \( J(\omega) \) comes up: for any function \( f(s,t) \) we have

\[ \sum_j \frac{c_j^2}{m_j\omega_j} \int_0^\infty J(\omega) f(\omega, t) \, d\omega = \frac{2}{\pi} \int_0^\infty J(\omega) f(\omega, t). \] (23)

If the number of harmonic oscillators of the bath is large, the spectral function can be seen as a continuous function. The relation between \( J(\omega) \) and \( \omega \) can then be modelled by a power law. This results in

\[ J(\omega) = \eta \omega^s, \] (24)
where $s$ and $\eta$ are constants. The different values of $s$ correspond to a different behavior

$$
0 < s < 1 \quad \rightarrow \quad \text{subohmic regime},
$$

$$
s = 1 \quad \rightarrow \quad \text{ohmic case},
$$

$$
s > 1 \quad \rightarrow \quad \text{superohmic regime}.
$$

If we inspect in more detail the differential equation for $q(t)$ and insert the ohmic spectral function $J(\omega) = \eta \omega$ into it, then something interesting happens: the Langevin equation (14) is found. This was exactly our aim, when introducing the concept of a reservoir to model a dissipative system! In the ohmic case, the constant $\eta$ naturally acquires the meaning of friction coefficient.

### 3.4 The Dynamical Reduced Density Operator

The density operator of a system plus bath is denoted by $\hat{\rho}(t)$. If $\{ |\psi^{(\alpha)}\rangle \}$ denotes the collection of states the composite system of system plus bath can be in, then the density operator is given by

$$
\hat{\rho}(t) = \sum_{\alpha} |\psi^{(\alpha)}\rangle P^{(\alpha)} \langle \psi^{(\alpha)} |. \tag{25}
$$

The density operator at time $t$ depends on the density operator at time zero as

$$
\hat{\rho}(t) = e^{-iHt/\hbar} \hat{\rho}(0) e^{iHt/\hbar}, \tag{26}
$$

where $H$ is the total Hamiltonian of system plus bath. In the position representation, we write the density operator as

$$
\hat{\rho}(q, \{x_{\alpha,i}\}; q_f, \{x_{\alpha,f}\}; t) = \langle q, \{x_{\alpha,i}\} | \hat{\rho}(t) | q_f, \{x_{\alpha,f}\} \rangle. \tag{27}
$$

For convenience, we now denote the bath variables as a vector $\mathbf{R}$ and use $x$ for the coordinate of the system. In this notation, the quantum mechanical propagator of system plus reservoir is given by

$$
K(x, \mathbf{R}, t; x', \mathbf{R}', 0) = \langle x, \mathbf{R} | e^{-iHt/\hbar} | x', \mathbf{R}' \rangle, \tag{28}
$$

which can be used to write the density operator

$$
\hat{\rho}(x, \mathbf{R}, y, \mathbf{Q}, t) = \int \int \int dx' dy' d\mathbf{R}' d\mathbf{Q}' \ K(x, \mathbf{R}, t; x', \mathbf{R}', 0) \ K^*(y, \mathbf{Q}, t; y', \mathbf{Q}', 0) \ \hat{\rho}(x', \mathbf{R}', y', \mathbf{Q}', 0). \tag{29}
$$

We assume a factorizable initial density operator, that is

$$
\hat{\rho}(x, \mathbf{R}, y, \mathbf{Q}, 0) = \hat{\rho}(x', \mathbf{R}', 0) \ \hat{\rho}(\mathbf{R}', \mathbf{Q}', 0). \tag{30}
$$
Now the (dynamical) reduced density operator $\tilde{\rho}(x, y, t)$ is defined as the reduced density operator, where the environmental variables $R$ are integrated out. From Eqs. (29) and (30) we find the following expression for $\tilde{\rho}(x, y, t)$

$$\tilde{\rho}(x, y, t) = \int dx' dy' J(x, y, t; x', y', 0) \tilde{\rho}(x', y', 0),$$

(31)

where

$$J(x, y, t; x', y', 0) \equiv \int \int dR' dQ' dR \{ K(x, R, t; x', R', 0) \times K^*(y, R, t; y', Q', 0) \tilde{\rho}(x', y', 0) \}.$$  

(32)

Here $\mathcal{J}$ can be seen as the super-propagator of the reduced density operator. This super-propagator can be calculated in more detail for the Hamiltonian (21). We can write down the corresponding Lagrangian

$$L = L_S + L_I + L_R + L_{CT},$$

(33)

where the Lagrangian for system, interaction, reservoir, and counter-term are respectively given by

$$L_S = \frac{1}{2} \dot{q}^2 - U(q),$$

(34a)

$$L_I = - \sum_j c_j q_j q_j,$$

(34b)

$$L_R = \sum_j \frac{1}{2} m_j \dot{q}_j^2 - \sum_j \frac{1}{2} m_j \omega_j^2 q_j^2,$$

(34c)

$$L_{CT} = - \sum_j \frac{1}{2} c_j^2 m_j \omega_j^2 q_j^2.$$  

(34d)

The corresponding parts of the action are denoted by $S_S$, $S_I$, $S_R$, and $S_{CT}$. The total action is denoted by $S$. Further, $S_0$ is defined as $S_S + S_{CT}$. We will use the functional integral representation for the total propagator

$$K(x, R, t; x', R', 0) = \int_x^x \int_{R'}^{R} \mathcal{D}x(t') \mathcal{D}R(t') \exp \left\{ \frac{i}{\hbar} S [x(t'), R(t')] \right\}.$$  

(35)

Inserting (35) into (32), we find the following expression for the super-propagator $\mathcal{J}$, in terms of the so called influence functional $\mathcal{F}$

$$\mathcal{J}(x, y, t; x', y', 0) = \int_{x'}^{x} \int_{y'}^{y} \mathcal{D}x(t') \mathcal{D}y(t') \exp \left\{ \frac{i}{\hbar} S_0 [x(t')] \right\} \times \exp \left\{ - \frac{i}{\hbar} S_0 [y(t')] \right\} \mathcal{F}[x(t'), y(t')].$$  

(36)
where $F$ is defined as
\[
F[x(t'), y(t')] \equiv \int \int dR'dQ'dR \rho_R(R', Q', 0) \int_{Q'}^R \int_R^R dR(t')dQ(t')
\]
\[
\times \exp \frac{i}{\hbar} \left[ S_I[x(t'), \mathbf{R}(t')] - S_I[y(t'), \mathbf{Q}(t')] + S_R[\mathbf{R}(t')] - S_R[\mathbf{Q}(t')] \right]. \tag{37}
\]
We will assume that at initial time the environmental harmonic oscillators are all in thermal equilibrium at temperature $T$. Then the density operator of the environment at $t = 0$ can be written as a product over the density operators of the individual harmonic oscillators that are in thermal equilibrium at $t = 0$. The density operator of one harmonic oscillator in thermal equilibrium was derived in [10] and it reads
\[
\rho_R(\mathbf{R}', \mathbf{Q}', 0) = \prod_k \rho^{(k)}_R(R'_k, Q'_k, 0)
= \prod_k \sqrt{\frac{m_k \omega_k}{2\pi \hbar \sinh (\hbar \omega_k / k_B T)}} \exp \left\{ -\frac{m_k \omega_k}{2\hbar \sinh (\hbar \omega_k / k_B T)} \right\} \times \left[ (R'_k^2 + Q'_k^2) \cosh (\hbar \omega_k / k_B T) - 2R'_kQ'_k \right]. \tag{38}
\]
The double path-integral in (37) can be expressed in terms of $K^{(k)}_{RI}$, the propagator of the $k$th environmental oscillator, when an external force $c_k x(t)$ acts on it. The double path-integral equals
\[
\int_{R'}^R \int_{Q'}^R dR(t')dQ(t') \exp \frac{i}{\hbar} \left\{ \ldots \right\} = \prod_k K^{(k)}_{RI}(R_k, R'_k) K^{(k)}_{RI}(R_k, Q'_k), \tag{39}
\]
where
\[
K^{(k)}_{RI}(R_k, R'_k) = \sqrt{\frac{m_k \omega_k}{2\pi \hbar \sin \omega_k t}} \exp \left\{ \frac{i}{\hbar} S^{(k)}_{cl}(R_k, R'_k) \right\}, \tag{40}
\]
\[
S^{(k)}_{cl}(R_k, R'_k) = \frac{m_k \omega_k}{2 \sin \omega_k t} \left[ (R_k^2 + R'_k^2) \cos \omega_k t - 2R_k R'_k \right.
+ \frac{2c_k R_k}{m_k \omega_k} \int_0^t x(t') \sin \omega_k t' dt' + \frac{2c_k R'_k}{m_k \omega_k} \int_0^t x(t') \sin \omega_k (t - t') dt' \left. - \frac{2c_k^2}{m_k \omega_k^2} \int_0^t dt' \int_0^t dt'' x(t') x(t'') \sin \omega_k (t - t') \sin \omega_k t'' \right]. \tag{41}
\]
After substituting Eqs. (38) and (39) into Eq. (37), and performing some Gaussian integrals, we obtain
\[
F[x(t'), y(t')] = \exp \left\{ -\frac{i}{\hbar} \int_{t_0}^{t'} \int_{t_0}^{t'} d\tau d\sigma \alpha_I(\tau - \sigma)[x(\tau) - y(\tau)] \cdot [x(\sigma) + y(\sigma)] \right\}
\times \exp \left\{ -\frac{i}{\hbar} \int_{t_0}^{t'} \int_{t_0}^{t'} d\tau d\sigma \alpha_R(\tau - \sigma)[x(\tau) - y(\tau)] \cdot [x(\sigma) - y(\sigma)] \right\}, \tag{42}
\]
where the functions $\alpha_I$ and $\alpha_R$ are defined as
\[
\alpha_I(\tau - \sigma) \equiv - \sum_k \frac{c_k^2}{2m_k \omega_k} \sin[\omega_k(\tau - \sigma)],
\]
\[
\alpha_R(\tau - \sigma) \equiv \sum_k \frac{c_k^2}{2m_k \omega_k} \coth\left(\frac{\hbar \omega_k}{2k_B T}\right) \cos[\omega_k(\tau - \sigma)].
\] (43)

Using the spectral function $J(\omega)$ these functions can be rewritten as
\[
\alpha_I(\tau - \sigma) = -\frac{1}{\pi} \int_{0}^{\infty} d\omega J(\omega) \sin[\omega(\tau - \sigma)],
\]
\[
\alpha_R(\tau - \sigma) = \frac{1}{\pi} \int_{0}^{\infty} d\omega J(\omega) \coth\left(\frac{\hbar \omega}{2k_B T}\right) \cos[\omega(\tau - \sigma)].
\] (44)

The result (42) can be substituted into Eq. (36), yielding
\[
J(x, y, t; x', y', 0) = \int_{x'}^{x} \int_{y'}^{y} Dx(t') Dy(t') \exp\left\{ i \frac{\hbar}{\beta} S_0[x(t')] - i \frac{\hbar}{\beta} S_0[y(t')]\right\}
\]
\[
+ \frac{i}{\hbar} \int_{t_0}^{t'} \int_{t_0}^{\tau} \alpha_I(\tau - \sigma)[x(\tau) - y(\tau)][x(\sigma) + y(\sigma)] d\sigma d\tau
\]
\[
- \frac{1}{\hbar} \int_{t_0}^{t'} \int_{t_0}^{\tau} \alpha_R(\tau - \sigma)[x(\tau) - y(\tau)][x(\sigma) - y(\sigma)] d\sigma d\tau\right\}.
\] (45)

The result in Eqs. (36) and (37) was initially derived in [11]. The actual form of Eq. (45) was obtained in [2].

3.5 The Equilibrium Reduced Density Operator

We are mainly interested in the density operator in equilibrium. A way to consider this, would be to take the limit $t \to \infty$ in Eq. (45). However, it is easier to consider the density operator in terms of the system variables only. The density operator for the composite system in equilibrium reads
\[
\rho(q_i, \{x_{\alpha,i}\}; q_f, \{x_{\alpha,f}\}; \beta) = \langle q_i, \{x_{\alpha,i}\} | e^{-\beta H} | q_f, \{x_{\alpha,f}\}\rangle.
\] (46)

It can be observed that this expression equals the expression for $\rho(q_i, \{x_{\alpha,i}\}; q_f, \{x_{\alpha,f}\}; t)$, if the identification $\beta = (i/\hbar) t$ is made.

The reduced density operator in equilibrium is now defined as the density operator in equilibrium, where the bath variables are traced out. In other words, it is given by
\[
\rho(q_i, q_f; \beta) = \prod_{\alpha} \left( \int_{-\infty}^{\infty} dx_{\alpha,i} \right) \rho(q_i, \{x_{\alpha,i}\}; q_f, \{x_{\alpha,f}\}; \beta).
\]
Assuming the initial position \( q_i \) and the final position \( q_f \) to be equal, \( q_i = q_f = q \), we can use the Euclidean action to find the following expression for \( \rho(q, q; \beta) \):

\[
\rho(q, q; \beta) = \prod_{\alpha} \left( \int_{-\infty}^{\infty} dx_{\alpha i} \right) \rho(q, \{ x_{\alpha i} \}; q, \{ x_{\alpha i} \}; \beta)
\]

\[
= \prod_{\alpha} \left( \int_{-\infty}^{\infty} dx_{\alpha i} \right) \int_{q(0)=q}^{q(\beta h)=q} \mathcal{D}q(\tau) \prod_{\alpha} \int_{x_{\alpha}(0)=x_{\alpha i}}^{x_{\alpha}(\beta h)=x_{\alpha i}} \mathcal{D}x_{\alpha}(\tau)
\]

\[
\times \exp\left\{-S_E[q(\tau), \{ x_{\alpha}(\tau) \}] / \hbar \right\}
\]

\[
= K_0(\beta) \int_{q(0)=q}^{q(\beta h)=q} \mathcal{D}q(\tau) \exp\left\{-S_{eff}[q(\tau)] / \hbar \right\}.
\]  

Here, \( S_{eff}(\tau) \) is defined in the following way [12]

\[
S_{eff}[q(\tau)] \equiv \int_0^{\beta h} \left[ \frac{1}{2} M \dot{q}^2 + V(q) \right] d\tau + \frac{1}{2} \int_{-\infty}^{\infty} d\tau' \int_0^{\beta h} d\tau \alpha(\tau - \tau') [q(\tau) - q(\tau')]^2,
\]

\[
\alpha(\tau - \tau') \equiv \frac{1}{2\pi} \int_0^{\infty} J(\omega) \exp(-\omega |\tau - \tau'|) d\omega = \alpha(\tau - \tau').
\]  

(48)

In the expression for \( S_{eff}[q(\tau)] \) we continue \( q(\tau) \) outside the region \([0, \beta h]\), by the prescription \( q(\tau + \beta h) \equiv q(\tau) \). For an ohmic spectral density, we can easily obtain

\[
\alpha(\tau - \tau') = \frac{n}{2\pi} \frac{1}{(\tau - \tau')^2}.
\]

The prefactor \( K_0(\beta) \) equals (as can be seen in (4.17) of [1])

\[
K_0(\beta) = \prod_{\alpha} \frac{1}{2 \sinh(\omega_{\alpha} \hbar / 2)}.
\]  

(49)

Hence, in the limit \( \omega_{\alpha} \hbar \beta \gg 1 \) this implies

\[
K_0(\beta) = \prod_{\alpha} \frac{1}{\exp(\omega_{\alpha} \hbar \beta / 2) - \exp(-\omega_{\alpha} \hbar \beta / 2)}
\]

\[
\approx \prod_{\alpha} \frac{1}{\exp(\omega_{\alpha} \hbar \beta / 2)}
\]

\[
= \exp(-\beta \sum_{\alpha} \hbar \omega_{\alpha} / 2).
\]  

(50)

Thus, in this limit the prefactor can be seen as a shift of the total energy.

These results will be used later, when we calculate the partition function \( Z(\beta) \), which can be written in terms of the reduced density operator \( \rho(q, q; \beta) \) as

\[
Z(\beta) = \int \rho(q, q; \beta) dq.
\]  

(51)
4 The Model

In the previous chapters some preliminary concepts were treated, and the two-state system was already mentioned. In this chapter a rigorous treatment of the two-state system makes it possible to define our basic model, which is a dissipative two-state system [2]. This is the simplest model we are going to discuss and some variations of it are treated in the next two chapters.

4.1 The Two-State System

In this first part we consider a system that consists of a continuous degree of freedom \( q \), in the presence of a potential \( V(q) \) that has two minima at \( \pm \frac{1}{2}q_0 \). Furthermore, around these minima, the potential is very accurately represented by quadratic functions, such that for small oscillations the system acts like a harmonic oscillator with center \( \pm \frac{1}{2}q_0 \), and frequency \( \omega_{\pm} \). These frequencies \( \omega_{\pm} \) are both taken to be of order \( \omega_0 \). We also assume that the potential barrier \( V_0 \) between the two minima is sufficiently high (\( V_0 \gg \hbar \omega_0 \)). To be precise, we define the quantity \( \epsilon \) such that \( V(\pm \frac{1}{2}q_0) = \pm \frac{1}{2} \epsilon \). This can be done by shifting the whole potential up or down. This way of realizing a two-state system, using a continuous coordinate (that not necessarily needs to be geometrical), is called a truncated two-state system. This contrasts with an intrinsic two state system, that can take only two states (for example the spin of an electron).

For the moment, we consider our two-state system to be completely isolated from the environment. Then the system can be described by the Hamiltonian

\[
H = -\frac{1}{2} \hbar \Delta \sigma_x + \frac{1}{2} \epsilon \sigma_z,
\]

(52)
where the Pauli matrices $\sigma_x$, $\sigma_y$ and $\sigma_z$ are given by

$$
\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \tag{53}
$$

In Eq. (52) the basis is chosen such that the eigenstates $\pm 1$ of $\sigma_z$ correspond to the system being localized in the well around $\pm q_0/2$. The meaning of the "tunneling" matrix element $\hbar \Delta / 2$ is that it brings the system from the one state to the other. In general we will consider $\hbar \Delta < \epsilon$ or even $\hbar \Delta \ll \epsilon$. The actual value of $\Delta$ will become clear when we calculate the partition function $Z(\beta)$ of the system.

### 4.2 Coupling to a Reservoir of Harmonic Oscillators

A completely isolated two-state system is not very likely to occur. Almost always, there will be some interaction with the environment. We will model this interaction by a term $\sigma_z \Omega$ in the Hamiltonian, where $\Omega$ is an operator acting on the environment. Because the three Pauli-matrices together with the $2 \times 2$ identity matrix span the space of $2 \times 2$ matrices, we should expect a coupling term of the kind $I \cdot \Omega + \sigma_x \Omega_x + \sigma_y \Omega_y + \sigma_z \Omega_z$ in the Hamiltonian. The coupling to the identity matrix is needless, because it would equally affect both eigenstates, and hence leave the two-state system, as a whole, invariant.

We take $\psi_+$ and $\psi_-$ to be the eigenstates (up to order $\Delta/\omega_0$) in respectively the right and the left well. Because $\sigma_x$ and $\sigma_y$ have only non-diagonal nonzero elements, they change $\psi_\pm$ to $\psi_{\mp}$. Thus any interaction proportional to $\sigma_x$ and $\sigma_y$ will also be proportional to the overlap of $\psi_+$ and $\psi_-$. This will be of order $\hbar \Delta$ and hence relatively small. Only the interaction containing $\sigma_z$ is directly proportional to $\psi_+$ and $\psi_-$. Therefore we will consider a Hamiltonian of the form

$$
H = -\frac{1}{2} \hbar \Delta \sigma_x + \frac{1}{2} \epsilon \sigma_z + \sigma_z \Omega + H_E. \tag{54}
$$

Here, $\Omega$ is an operator on the environment and $H_E$ is the Hamiltonian of the environment, if it would not be connected to the two-state system. We consider an environment composed of harmonic oscillators, so the expression for $H_E$ can be found in Eq. (15). From the general expression (18) for the interaction term, we find for our case

$$
\Omega = \frac{1}{2} q_0 \sum_j c_j x_j. \tag{55}
$$

Finally, collecting Eqs. (15), (54) and (55) we obtain the total Hamiltonian, which is called "spin-boson Hamiltonian" in the literature, given by

$$
H_{SB} = -\frac{1}{2} \hbar \Delta \sigma_x + \frac{1}{2} \epsilon \sigma_z + \frac{1}{2} q_0 \sigma_z \sum_j c_j x_j + \sum_j \left( \frac{p_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 x_j^2 \right). \tag{56}
$$
This Hamiltonian describes a two-level system linearly coupled to a bath of harmonic oscillators. The properties of the bath are fully incorporated in the spectral function

\[ J(\omega) = \frac{\pi}{2} \sum_j \frac{c_j^2}{m_j \omega_j} \delta(\omega - \omega_j), \] (57)

which in the ohmic case becomes

\[ J(\omega) = \begin{cases} \omega \text{ if } \omega \leq \Omega, \\ 0 \text{ otherwise.} \end{cases} \] (58)

5 Coupling to the Bath via an Intermediate Harmonic Oscillator

In the previous chapter we considered a (truncated) two-state system that was directly coupled to a bath of harmonic oscillators. This means that the coupling term in the Hamiltonian depends both, on the two-state system and on the bath of harmonic oscillators. In this section we will look at an intrinsic two-state system that is coupled to one single harmonic oscillator. This harmonic oscillator is itself coupled to a bath of harmonic oscillators. One could say that the two-state system is coupled to the bath via a harmonic oscillator. In the treatment of this model, we follow the approach of [3]. For deriving a so-called effective spectral density, we use a prescription from [8].

5.1 Description of the Model

The model of coupling via an intermediate harmonic oscillator describes the electron transfer in biomolecules. In this case, an electron can travel between two localized sites. These sites can be in the same molecule or in two different molecules. In most chemical processes involving electron transfer, distances between the localized sites are small. This leads to short tunneling times, and the electron travels almost freely between the sites. In some biological processes, however, the sites are separated by larger distances, such that a small matrix-element determines the probability of "tunneling" between the sites. It’s now time to present the model more explicitly.

We consider the donor-site and the acceptor-site of the electron as a two-state system where these two states are identified with the eigenvalues +1 and −1 of \(\sigma_z\), respectively. We assume the two corresponding positions to be +q₀ and −q₀. The possibility of tunneling is associated with a matrix-element, that we define as \(\hbar \Delta/2\). This is why we include a term \(\hbar \Delta \sigma_z/2\) in the combined Hamiltonian.

For the single harmonic oscillator, with continuous coordinate y and frequency \(\Omega\), we include in the combined Hamiltonian the standard kinetic term \(p_y^2/2M\).
The potential energy term for this single harmonic oscillator can be combined with the coupling to the two-state system. Because the potential must be harmonic in any case, and the coupling can be taken to be proportional to $\sigma_z$, this can be done by defining the potential

$$V(y; \sigma_z) = \frac{1}{2} M \Omega^2 (y + y_0 \sigma_z)^2 + \frac{1}{2} \epsilon \sigma_z.$$  

(59)

For the bath variables $x_\alpha$ we include in the Hamiltonian the kinetic terms $p_\alpha^2 / 2m_\alpha$ and the potential terms $m_\alpha \omega_\alpha^2 x_\alpha^2 / 2$. The coupling between the single harmonic oscillator and those of the bath is again a linear coordinate-coordinate coupling, thus the terms $c_\alpha x_\alpha y$ are added.

Finally, to render the minimum of the potential energy of the oscillator bath equal to zero, we add terms $c_\alpha^2 y^2 / 2m_\alpha \omega_\alpha^2$ to the Hamiltonian for electron transfer, which thus takes the form

$$H_{ET} = \frac{\hbar}{2} \sigma_z + \frac{p_y^2}{2M} + V(y; \sigma_z) + \sum_\alpha \left[ \frac{p_\alpha^2}{2m_\alpha} + \frac{1}{2} m_\alpha \omega_\alpha^2 \left( x_\alpha + \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2} y \right)^2 \right].$$  

(60)

Considering the potential in Eq. (59), we define the crossing point $y^*$ as being the point $-y_0 < y^* < y_0$ that satisfies $V(y^*, -) = V(y^*, +)$. Then, most of the tunneling occurs within a length of order $l_{LZ}$, where $l_{LZ}$, the Landau-Zener length, is defined to be

$$l_{LZ} = \frac{\hbar}{|F_+ - F_-|},$$

$$F_\pm = \left[ \frac{\partial V(y; \pm)}{\partial y} \right]_{y=y^*}. $$

(61)

If the temperature is high enough, the friction is relatively small and the tunneling electron will pass the Landau-Zener region in one time. In this case the probability of tunneling is a small quantity, which could be calculated. If the temperature is low, the friction is relatively so large that the electron can stay in the Landau-Zener region and make many transitions after each other. In this case, coherence between the states will get lost in due time. These qualitative considerations could help us to understand what is happening in the system.

### 5.2 Derivation of the Effective Spectral Density

The two-state system and reaction coordinate (the coordinate of the intermediate harmonic oscillator) are only influenced by the bath through the spectral density

$$J_0(\omega) = \frac{\pi}{2} \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha} \delta(\omega - \omega_\alpha).$$

(62)

For a dissipative term proportional to the velocity in the equation of motion, $J_0(\omega)$ acquires an ohmic form with a high-frequency cutoff $\Lambda$ and is given by

$$J_0(\omega) = \eta \omega \exp(-\omega/\Lambda).$$

(63)
Instead of this spectral density we now want to find an effective spectral density which describes how the two-state system is influenced by the reaction coordinate and bath together. This $J_{\text{eff}}(\omega)$ is the same as the one that controls the dynamics of a continuous variable $q$ moving in a potential $U(q)$ coupled to the reaction coordinate and the bath in the same way as the two-state system is coupled. We thus consider the general Hamiltonian

$$\begin{aligned}
H_q &= \frac{p_q^2}{2\mu} + U(q) + \frac{1}{2M} p_y^2 + \frac{1}{2} M \Omega^2 (y + q)^2 \\
&+ \sum_\alpha \left[ \frac{p_\alpha^2}{2m_\alpha} + \frac{1}{2} m_\alpha \omega_\alpha^2 \left( x_\alpha + \frac{c_\alpha}{m_\alpha \omega_\alpha^2} y \right)^2 \right],
\end{aligned} \quad (64)$$

where $p_q$ is the momentum conjugate to $q$. Defining $U'(q) \equiv dU/dq$ and using dots for time derivatives, the classical equations of motion are

$$\begin{aligned}
\mu \ddot{q} &= -U'(q) - M \Omega^2 (y + q), \\
M \ddot{y} &= -M \Omega^2 (y + q) - \sum_\alpha c_\alpha x_\alpha - y \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2}, \\
m_\alpha \ddot{x}_\alpha &= -m_\alpha \omega_\alpha^2 x_\alpha - c_\alpha y.
\end{aligned} \quad (65)$$

We denote the Fourier transform of $q$, $y$, $x_\alpha$, and $U'(q)$ as $q(z)$, $y(z)$, $x_\alpha(z)$ and $U'(q)$. In the definition of these Fourier transforms we demand the imaginary part of $z$ to be strictly negative. This will be required later, when we perform contour integrations. Using the Fourier transforms, Eqs. (65) can be written as

$$\begin{aligned}
\left( -\mu z^2 + M \Omega^2 \right) q(z) + M \Omega^2 y(z) &= -U'(q), \\
\left( -M z^2 + M \Omega^2 + \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2} \right) y(z) + \sum_\alpha c_\alpha x_\alpha(z) &= -M \Omega^2 q(z), \\
x_\alpha(z) &= -\frac{c_\alpha}{m_\alpha \left( \omega_\alpha^2 - z^2 \right)} y(z).
\end{aligned} \quad (66)$$

Inserting now Eq. (66c) into Eq. (66b), we obtain

$$\begin{aligned}
\left[ -M z^2 + M \Omega^2 - z^2 \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2 \left( \omega_\alpha^2 - z^2 \right)} \right] y(z) &= -M \Omega^2 q(z). \quad (67)
\end{aligned}$$

Using the notation

$$L(z) = -z^2 \left[ M + \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2 \left( \omega_\alpha^2 - z^2 \right)} \right],$$

this can be written as

$$y(z) = -M \Omega^2 \frac{L(z)}{M \Omega^2 + L(z)} q(z). \quad (68)$$
Inserting Eq. (68) into Eq. (66a), we obtain
\[
-\mu z^2 + \frac{M\Omega^2 (M\Omega^2 + L(z))}{M\Omega^2 + L(z)} - \frac{(M\Omega^2)^2}{M\Omega^2 + L(z)} q(z) = -U'_z(q). \tag{69}
\]
Introducing the function \(K(z)\), this can be written as:
\[
K(z)q(z) = -\mu z^2 + \frac{M\Omega^2 \cdot L(z)}{M\Omega^2 + L(z)} q(z) = -U'_z(q). \tag{70}
\]
It is convenient to evaluate the function \(L(z)\) in more detail, using Eqs. (62) and (63),
\[
L(z) = -z^2 \left[ M + \sum_\alpha \frac{c^2_\alpha}{m_\alpha \omega^2_\alpha (\omega^2_\alpha - z^2)} \right] \\
= -z^2 \left[ M + \frac{2}{\pi} \int_0^{\infty} \frac{d\omega'}{\omega'} \left( \frac{1}{\omega' (\omega'^2 - z^2)} \sum_\alpha \frac{c^2_\alpha}{m_\alpha \omega_\alpha} \delta(\omega' - \omega_\alpha) \right) \right] \\
= -z^2 \left[ M + \frac{2}{\pi} \int_0^{\infty} \frac{J_0(\omega')}{\omega' (\omega'^2 - z^2)} d\omega' \right] \\
= -z^2 \left[ M + \frac{2\eta}{\pi} \int_0^{\infty} \frac{\exp(-\omega'/\Lambda)}{\omega'^2 - z^2} d\omega' \right]. \tag{71}
\]
Taking the cutoff to be infinity leaves us with an integral over \(1/(\omega'^2 - z^2)\) which has poles at \(\omega' = \pm z\). These poles are real, but from the assumption that \(\text{Im}(z) < 0\) (in the definition of the Fourier transform) they can be shifted to \(\omega' = \pm (z + i\epsilon)\). Closing the contour with a semicircle in the upper halfplane with radius going to infinity gives no contribution to the integral. We have a residue at \(\omega' = -z + i\epsilon\) that equals \(-1/(2z)\). Hence
\[
L(z) = -Mz^2 - \frac{\eta z^2}{\pi} \int_{-\infty}^{\infty} \frac{1}{(\omega'^2 - z^2)} d\omega' \\
= -Mz^2 + \frac{\eta z^2}{\pi} \cdot 2\pi i \cdot \frac{1}{2z} = -Mz^2 + i\eta z. \tag{72}
\]
The Leggett prescription for obtaining \(J_{\text{eff}}(\omega)\) is derived in [8], and can be formulated as
\[
J_{\text{eff}}(\omega) = \lim_{\epsilon \to 0^+} \text{Im}[K(\omega - i\epsilon)]. \tag{73}
\]
In this case we can just take
\[ J_{\text{eff}}(\omega) = \text{Im}[K(\omega)] \]
\[ = \text{Im} \left[ -\mu \omega^2 + \frac{M \Omega^2 (-M \omega^2 + i \eta \omega)}{M \Omega^2 - M \omega^2 + i \eta \omega} \right] \]
\[ = \text{Im} \left[ -\mu \omega^2 + \frac{M \Omega^2 (-M \omega^2 + i \eta \omega)(M \Omega^2 - M \omega^2 - i \eta \omega)}{(M \Omega^2 - M \omega^2)^2 + (\eta \omega)^2} \right] \]
\[ = \frac{\eta \omega M^2 \Omega^4}{M^2 (\Omega^2 - \omega^2)^2 + \eta^2 \omega^2} \]
\[ = \frac{\eta \omega \Omega^4}{(\Omega^2 - \omega^2)^2 + 4 \gamma^2 \omega^2}. \] (74)

In the last step the definition \( \gamma \equiv \eta / 2M \) was used. The result in Eq. (74) was derived in [3]. In Fig. 5b this effective spectral density is plotted for some typical values of the parameters. The usual ohmic spectral density with high-frequency cutoff \( \Lambda \) is also plotted in Fig. 5a for comparison.

\[ J_{\text{eff}}(\omega) \]
\[ J_{0}(\omega) \]
\[ J_{\text{eff}}(\omega) \]

Figure 5: (a) the ohmic spectral density for \( \eta = 2 \) and exponential cutoff \( \Lambda = 0.5 \); (b) the effective spectral density for \( \eta = 5, \Omega = 1 \) and \( \gamma = 0.2 \).

6 Coupling to the Bath via an Anharmonic Oscillator

In this chapter we repeat the procedure that was applied in the previous chapter, when we considered the coupling of a two-state system to a bath of harmonic oscillators via a harmonic oscillator. Now we consider an anharmonic oscillator instead of the (single) harmonic oscillator. The aim is to obtain an effective spectral density that has a better form, and is easier to control, than the one we
found in Eq. (74), in the case of a harmonic oscillator. This chapter contains only new calculations.

6.1 The Model

In order to describe an anharmonic oscillator, we add an extra term \((\zeta/4)y^4\) to the Hamiltonian (64). In principle, the constant \(\zeta\) should be positive, to prevent that the potential for \(y\) develops a second minimum. The Hamiltonian then reads

\[
H_q = \frac{p_q^2}{2\mu} + U(q) + \frac{1}{2M}p_y^2 + \frac{1}{2}M\Omega^2(y + q)^2 + \frac{\zeta}{4}y^4 \\
+ \sum_{\alpha} \left[ \frac{p_{\alpha}^2}{2m_\alpha} + \frac{1}{2}m_\alpha\omega_\alpha^2 \left( x_\alpha + \frac{c_\alpha}{m_\alpha\omega_\alpha^2} y \right)^2 \right],
\]

(75)

where again \(p_q\) is the momentum conjugate to \(q\).

6.2 Derivation of the Effective Spectral Density

Defining \(U'(q) = dU/dq\) and using dots for time derivatives, the classical equations of motion that can be found from (75) are

\[
\mu\ddot{q} = -U'(q) - M\Omega^2(y + q),
\]

(76a)

\[
M\ddot{y} + \left( M\Omega^2 + \sum_{\alpha} \frac{c_\alpha^2}{m_\alpha\omega_\alpha^2} \right) y + \zeta y^3 = -M\Omega^2q - \sum_{\alpha} c_\alpha x_\alpha,
\]

(76b)

\[
m_\alpha \ddot{x}_\alpha = -m_\alpha\omega_\alpha^2 x_\alpha - c_\alpha y.
\]

(76c)

Compared to Eq. (65), only the second equation of motion (76b) is different. If we set the RHS of (76b) to zero and define

\[
A = \Omega^2 + \frac{1}{M} \sum_{\alpha} \frac{c_\alpha^2}{m_\alpha\omega_\alpha^2},
\]

\[
B = \frac{\zeta}{M},
\]

the homogeneous nonlinear equation to be solved reads

\[
M \left[ \ddot{y}(t) + Ay(t) + By^3(t) \right] = 0.
\]

(77)

This equation may, in good approximation, be solved by

\[
y(t) = a \left[ \sin(\omega\sqrt{At}) + \frac{\lambda}{32} \sin(3\omega\sqrt{At}) \right],
\]

(78)
where \( a \) can be chosen freely, and
\[
\begin{align*}
\omega &= \sqrt{1 + \frac{3B}{4A}} a, \\
\bar{\lambda} &= \frac{(a^2 B)/A}{1 + (3aB)/(4A)}.
\end{align*}
\]
This result is derived in Appendix A. Even if the RHS of Eq. (76b) is some periodic function of time (we would then have a forced Duffing oscillator), the formal solution for the equation is still an infinite sum of sines:
\[
y(t) = \sum_{n=0}^{\infty} a_n \sin [(2n+1)\tilde{\omega}t],
\]
(79)
as shown in [13]. Therefore, it is reasonable to expect that the solution will look like (78), even for the original RHS of (76b). This can be used to write
\[
y(t) \approx a \sin(\omega\sqrt{A}t) + \frac{a\bar{\lambda}}{32} \sin(3\omega\sqrt{A}t).
\]
(80)
Approximately, we also have
\[
\ddot{y}(t) = -A\omega^2 \sin(\omega\sqrt{A}t) - \frac{9aA\omega^2 \bar{\lambda}}{32} \sin(3\omega\sqrt{A}t)
\]
\[
= -A\omega^2 \left[ a \sin(\omega\sqrt{A}t) + \frac{a\bar{\lambda}}{32} \sin(3\omega\sqrt{A}t) \right]
\]
\[
- \frac{8A\omega^2 \bar{\lambda}}{32} \left[ a \sin(3\omega\sqrt{A}t) + \frac{a\bar{\lambda}}{32} \sin(9\omega\sqrt{A}t) \right]
\]
\[
\approx -A\omega^2 \ddot{y}(t) - \frac{8A\omega^2 \bar{\lambda}}{32} \dddot{y}(3t),
\]
(81)
where the last term of order \( \sin(9\omega\sqrt{A}t) \) has been neglected. This procedure implies linearization of the LHS of Eq. (76b) by making the replacement
\[
M \left[ \ddot{y}(t) + Ay(t) + By^3(t) \right] \implies M \left[ \ddot{y}(t) + \tilde{A}(a)y(t) + \tilde{B}(a)y(3t) \right].
\]
(82)
Here, we have defined
\[
\tilde{A}(a) \equiv A\omega^2 = A + \frac{3}{4} Ba,
\]
\[
\tilde{B}(a) \equiv \frac{8A\omega^2 \bar{\lambda}}{32} = \frac{1}{4} Ba^2.
\]
(83)
Thus, the second equation of motion (76b) is replaced by the linearized equation
\[
M \ddot{y}(t) + \left( M\Omega^2 + \sum_{\alpha} \frac{c_{\alpha}^2}{m_{\alpha} \omega_{\alpha}^2} + \frac{3}{4} \zeta a \right) \dddot{y}(t) + \frac{1}{4} \xi a^2 \dddot{y}(3t) = -M\Omega^2 q(t) - \sum_{\alpha} c_{\alpha} x_{\alpha}(t).
\]
We denote the Fourier transform of $q$, $y$, $x_\alpha$ and $U'_z(q)$ as $q(z)$, $y(z)$, $x_\alpha(z)$ and $U'_z(q)$. In the definition of these Fourier transforms we require the imaginary part of $z$ to be strictly negative. The Fourier transformed equations of motion are now

\[
(-\mu z^2 + M\Omega^2)q(z) + M\Omega^2y(z) = -U'_z(q), \tag{84a}
\]

\[
\left(-Mz^2 + M\Omega^2 + \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2} + \frac{3}{4} \zeta a^2 y \left(\frac{z}{\sqrt{3}}\right)\right) y(z) + \frac{1}{4} \zeta a^2 y \left(\frac{z}{\sqrt{3}}\right) + \sum_\alpha c_\alpha x_\alpha(z) = -M\Omega^2 q(z), \tag{84b}
\]

\[
x_\alpha(z) = -\frac{c_\alpha}{m_\alpha (\omega_\alpha^2 - z^2)} y(z). \tag{84c}
\]

Inserting now Eq. (84c) into Eq. (84b), we obtain

\[
-M\Omega^2 q(z) = f(z) y(z) + \frac{1}{4} \zeta a^2 y \left(\frac{z}{\sqrt{3}}\right),
\]

\[
f(z) = -Mz^2 + M\Omega^2 - z^2 \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2 (\omega_\alpha^2 - z^2)} + \frac{3}{4} \zeta a. \tag{85}
\]

Now we use the expected form of $y(t)$ and hence of $y(z)$ and $y(z/3)$. From Eq. (80), that is

\[
y(t) \approx a \sin(\omega \sqrt{A} t) + \frac{a \lambda}{32} \sin(3\omega \sqrt{A} t), \tag{86}
\]

which, in terms of delta functions, may be rewritten as

\[
y(z) \approx a \left[ i \delta(z - \omega \sqrt{A}) - i \delta(z + \omega \sqrt{A}) \right]
+ \frac{a \lambda}{32} \left[ i \delta(z - 3\omega \sqrt{A}) - i \delta(z + 3\omega \sqrt{A}) \right], \tag{87}
\]

\[
y \left(\frac{z}{3}\right) \approx a \left[ i \delta(z - 3\omega \sqrt{A}) - i \delta(z + 3\omega \sqrt{A}) \right]
+ \frac{a \lambda}{32} \left[ i \delta(z - 9\omega \sqrt{A}) - i \delta(z + 9\omega \sqrt{A}) \right]. \tag{88}
\]

This now suggests that for $z \pm \omega \sqrt{A} = 0$ we have $y(z/3) = 0 \cdot y(z)$ and for $z \pm 3\omega \sqrt{A} = 0$ we have $y(z/3) = 32/\lambda \cdot y(z)$. Now we take

\[
y(z/3) = 32/\lambda \cdot b(z) y(z), \tag{89}
\]

for a real function $b(z)$ that takes values 1 at $z = \pm 3\omega \sqrt{A} = \pm 3\sqrt{A + 3aB/4}$ and 0 at $z = \pm \omega \sqrt{A} = \pm \sqrt{A + 3aB/4}$. Let us propose an ansatz for the
function $b(z)$. We suppose it is a double Gaussian and that the width of each peak depends on the non-linearity $\zeta$:

$$b(z, \zeta) = \exp \left[ -\frac{2}{\zeta(A + 3aB/4)} \left( |z| - 3\sqrt{A + 3aB/4} \right)^2 \right], \quad (90)$$

as plotted in Fig. (6). Note that for $\zeta = 0$, $b(z, \zeta)$ does not need to take the value 1 at $\omega = \pm 3\sqrt{A + 3aB/4}$, as can be seen from Eqs. (87) and (88). For large $\zeta$ the demand that $b(z, \zeta)$ is approximately 0 at $\omega = \pm \sqrt{A + 3aB/4}$, is violated. This is not a real problem because in this limit the perturbative approach we have chosen will break down anyway.

Inserting now Eq. (89) into Eq. (85), we obtain

$$y(z) = -\frac{M \Omega^2 z^2}{M \Omega^2 - z^2 \sum_{\alpha} \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2 (\omega_\alpha^2 - z^2)} + \frac{3}{4} \sqrt{a + \frac{8a^2}{\lambda}} b(z, \zeta)} q(z)$$

$$= \frac{-M \Omega^2}{M \Omega^2 + L(z) + \frac{3}{4} \sqrt{a + \frac{8a^2}{\lambda}} b(z, \zeta)} q(z). \quad (91)$$

Substitution of $y(z)$ in Eq. (84a) yields

$$K(z)q(z) \equiv \left\{ -\mu z^2 + \frac{M \Omega^2}{M \Omega^2 + L(z) + \frac{3}{4} \sqrt{a + \frac{8a^2}{\lambda}} b(z, \zeta)} \right\} q(z)$$

$$= -U'(q). \quad (92)$$

In Eq. (72) we calculated that

$$L(z) = -M z^2 + i\eta z.$$
The Leggett prescription for obtaining $J_{\text{eff}}(\omega)$ is as follows

$$J_{\text{eff}}(\omega) = \lim_{\epsilon \to 0^+} \text{Im}[K(\omega - i\epsilon)].$$  \hfill (93)

In our case we can just take

$$J_{\text{eff}}(\omega) = \text{Im}[K(\omega)]$$

$$= \im\left\{ \frac{M\Omega^2}{M\Omega^2 + \zeta(32a^2 b(\omega, \zeta)/\bar{\lambda} + 3a)/4 - M\omega^2 + i\eta\omega}\right\}$$

$$= \frac{\eta\omega M^2\Omega^4}{\eta\omega M^4} \frac{\Omega^2 + \zeta(32a^2 b(\omega, \zeta)/\bar{\lambda} + 3a)/(4M) - \omega^2}{\omega^2 + \zeta^2\omega^2}.$$  \hfill (94)

In the calculation we used the fact that $b$ is a real function and in the last step the definitions $\gamma \equiv \eta/2M$ and $\Delta(\omega, \zeta) \equiv \zeta(32a^2 b(\omega, \zeta)/\bar{\lambda} + 3a)/(4M)$ were used. We recall that $a$ was the typical amplitude of the intermediate harmonic oscillator $y(t)$. In the definition of $\Delta(\omega, \zeta)$ above, $a$ is encountered, as well as the other parameter $\zeta$ that characterizes the nonlinearity.

A further inspection of $\Delta(\omega, \zeta)$ seems needed. After the insertion of

$$\bar{\lambda} = \frac{a^2 B/A}{1 + (3aB)/(4A)},$$

we find

$$\Delta(\omega, \zeta) = \frac{\zeta}{4M} \left[ 32a^2 \left(1 + \frac{3aB}{4A}\right) b(\omega, \zeta) + 3a \right]$$

$$= \frac{\zeta}{4M} \left[ \left( \frac{32A}{B} + 24a \right) b(\omega, \zeta) + 3a \right].$$  \hfill (95)

In the limit $\zeta \to 0$ the $J_{\text{eff}}$ of Eq. (94) should equal the $J_{\text{eff}}$ in the harmonic case, as can be found in Eq. (74). This directly implies that

$$\lim_{\zeta \to 0} \Delta(\omega, \zeta) = 0.$$  \hfill (96)

After the insertion of $B = \zeta/M$, we have

$$\Delta(\omega, \zeta) = 8ab(\omega, \zeta) + \frac{6\zeta a}{M} b(\omega, \zeta) + \frac{3\zeta a}{4M}.$$  

The function $b(\omega, \zeta)$ has the property

$$\lim_{\zeta \to 0} b(\omega, \zeta) = 0, \quad \text{for all } \omega,$$  \hfill (97)
and thus the property demanded in Eq. (96) is satisfied.

Looking at the effective spectral density in Eq. (94), most important is the behavior of the term inside the square brackets in the denominator. Only if this is nearly zero, a peak arises. If it is large, $J_{\text{eff}}$ is small. We can write:

$$[\Omega^2 + \Delta(\omega, \zeta) - \omega^2] = (\Omega^2 + \Delta - \omega^2) + [\Delta(\omega, \zeta) - \Delta] \propto b(\omega, \zeta)$$

The function $\Omega^2 + \Delta - \omega^2$ is nearly zero around $\omega = \sqrt{\Omega^2 + \Delta} \equiv \omega_a$, and

Figure 7: effective spectral density for $\zeta = 1$, $\omega_a = 1$ and (a) $3\omega_b = 0.8$; (b) $3\omega_b = 1.7$; (c) $3\omega_b = 2.5$; (d) $3\omega_b = 3.3$.

$b(\omega, \zeta)$ is only nonzero (and positive) in a relatively sharp region around $\omega = 3\sqrt{A + 3aB/4} \equiv 3\omega_b$. Therefore, four types of behavior can be identified (see Fig. 7):

1) for $3\omega_b \lesssim \omega_a$, the $J_{\text{eff}}(\omega)$ looks like the one we saw before, but is only reduced to nearly zero in a region around $\omega = 3\omega_b$, see Fig. 7a.
2) for $\omega_a \lesssim 3\omega_b \lesssim 2\omega_a$, the effects 1 and 3 are mixed, see Fig. 7b.
3) for $2\omega_a \leq 3\omega_b \leq \omega_c$, $J_{\text{eff}}(\omega)$ also looks like the one we saw before, but two small peaks are added directly left and right of $\omega = 3\omega_b$, see Fig. 7c.

4) for $3\omega_b \geq \omega_c$, $J_{\text{eff}}(\omega)$ also looks like the one we saw before, but one small peak is added, centered at $\omega = 3\omega_b$, see Fig. 7d.

The value of $\omega_c$ depends on the constant $[\Delta(\omega, \zeta) - \Delta]/b(\omega, \zeta)$. Using the ex-

pressions for $A$, $B$, and $\Delta$, the values of $\omega_a$ and $\omega_b$ can be calculated to be

$$\omega_a = \sqrt{\Omega^2 + \frac{3}{4M} a \zeta}, \quad (99a)$$

$$\omega_b = \sqrt{\Omega^2 + \frac{1}{M} \sum_{\alpha} \frac{e_{\alpha}^2}{m_{\alpha} \omega_{\alpha}^2} + \frac{3}{4M} a \zeta}. \quad (99b)$$

From Eqs. (99a) and (99b), we see that $\omega_a < \omega_b$. This is why the types of behavior 1 and 2 are excluded. Notice that although we have plotted behavior 3 for $\omega_a < \omega_b$, in principle, we could still find that $\omega_c = 5\omega_a$, for instance,
which could allow for $3\omega_b = 4\omega_a$, satisfying the constraint $\omega_a < \omega_b$. However, as we will see in the following, this case does not occur for the physical range of parameters. For behavior 4, we plot $J_{\text{eff}}$ as a function of both $\omega$ and $\zeta$ in Fig. 8. The fact only types 3 and 4 can occur, suggests the definition of the functions

$$J_1(\omega) \equiv \frac{\eta \omega \Omega^4}{[\Omega^2 + \Delta - \omega^2]^2 + 4\gamma^2 \omega^2},$$

$$J_2(\omega) \equiv \frac{\eta (3\omega_b) \Omega^4}{[\Omega^2 + \Delta(\omega, \zeta) - (3\omega_b)^2]^2 + 4\gamma^2 (3\omega_b)^2}.$$  \hspace{1cm} (100) \hspace{1cm} (101)

Now we have $J_1 \approx J_{\text{eff}}$ for $\omega \lesssim 3\omega_a$ and $J_1 \approx 0$ for larger $\omega$. Further $J_2 \approx J_{\text{eff}}$ for $\omega \gtrsim 3\omega_b$ and $J_2 \approx 0$ for smaller $\omega$. The fact that $\omega_a < \omega_b$ now implies that for all $\omega$

$$J_{\text{eff}}(\omega) \approx J_1(\omega) + J_2(\omega).$$  \hspace{1cm} (102)

Of these two, $J_1$ has one peak centered at $\omega_a$, and $J_2$ has one or two small peaks centered at $3\omega_b$. The functions $J_1$ and $J_2$ are plotted in Fig. 9 (a) and (b) respectively, for the values $\zeta = 1$, $\omega_a = 1$, and $3\omega_b = 3.3$. These pictures can thus be compared with Fig. 7 (d). Examination of the function $J_2$ shows that it has two peaks for $[(3\omega_b)^2 - (\Omega^2 + \Delta)] < [\Delta(\omega, \zeta) - \Delta]/b(\omega, \zeta)$. For all $\zeta > 0$, this implies $9\omega_b^2 - \omega_a^2 < 8\omega_b^2$ and thus $\omega_b^2 < \omega_a^2$. This however contradicts the fact that $\omega_a < \omega_b$. So obviously the critical value $\omega_c$ defined before takes the value $3\omega_a$ and only the type of behavior 4 occurs. An example of this case can be found in Fig. 8 and Fig. 9 (a) and (b). The maximum amplitude of the $J_2$ peak is about 10% of the maximal value of $J_1$, in consistency with our perturbative approach.

For large $\zeta$, the scenario depicted in Fig. 7 (c) could reappear. In addition, as $\zeta$ increases, the behavior evolves towards Fig. 7 (b), i.e., the intermediate
peak softens and approaches the higher one. However, we expect that in this limit our perturbative approach for the non-linearity should break down and therefore we refrain from drawing conclusions in this limit.

7 The Dynamics of the System

In the last three chapters we focussed on (effective) spectral density functions in three cases: direct coupling and coupling via (an)harmonic oscillator. In this chapter we calculate $P(t)$, the expectation value of the position of the two-state system as a function of time. This will be done in the case of direct coupling, where we follow closely the chapter 4 of [2]. The result is worked out in detail in the so called noninteracting-blip approximation (NIBA), discussed [2]. In the next two chapters the equilibrium value of $P(t)$, $P_\infty$, is calculated for the two cases of indirect coupling.

7.1 Formal Expression for $P(t)$

We consider a two-state system, that at time $t = -\infty$ is uncoupled from the reservoir of harmonic oscillators. At time $t_0 < 0$ the interaction between system and reservoir is "switched on", and the system is hold in the state $x(t) = x_i$ until $t = 0$. At time $t = 0$ this constraint is removed and for $t > 0$, $x(t)$ can take the values $\pm q_0/2$. Without loss of generality, we can take $x_i = +q_0/2$.

The reduced density operator $\rho(x', y, t)$ can be found using the influence-functional method. After determining $p(t) = \rho(q_0/2, q_0/2, t)$, the probability of being at $q_0/2$ at time $t$, we can then use $p(t)$ to calculate $P(t)$, the expectation value of $\sigma_z$ at time $t$. The quantities $P(t)$ and $p(t)$ are connected by

$$P(t) = 2p(t) - 1. \quad (103)$$

We thus calculate

$$\rho(x, y, t) = \int \int dx' dy' J(x, y, t; x', y', t_0) \rho(x', y', t_0) = J(x, y, t; x_i, x_i, t_0), \quad (104)$$

where we used the fact that $\rho(x', y', t_0) = \delta(x' - x_i) \cdot \delta(y' - x_i)$. The RHS of Eq. (104) can now be expressed in terms of the influence functional $\mathcal{F}$, that is a functional of two paths $x(t)$ and $y(t)$ (in real time), that satisfy the required initial conditions for $t < 0$,

$$J(x, y, t; x_i, x_i, t_0) = \int_{x_i}^{x} \int_{x_i}^{y} Dx(\tau) Dy(\tau) \mathcal{F}[x(\tau), y(\tau)] \times \exp \left\{ \frac{i}{\hbar} \{ S_0[x(\tau)] - S_0[y(\tau)] \} \right\}. \quad (105)$$
In this context, the variable $\tau$ is used as an integration variable, it should not be confused with imaginary time. If we define the functions $\alpha_I(\tau - \sigma)$ and $\alpha_R(\tau - \sigma)$ as

$$
\alpha_I(\tau - \sigma) = -\frac{1}{\pi} \int_0^\infty d\omega J(\omega) \sin(\omega(\tau - \sigma)),
$$

$$
\alpha_R(\tau - \sigma) = \frac{1}{\pi} \int_0^\infty d\omega J(\omega) \cos(\omega(\tau - \sigma)) \coth\left(\frac{\beta \hbar \omega}{2}\right),
$$

then the influence functional can be written as

$$
\mathcal{F}[x(t), y(t)] = \exp\left\{\frac{i}{\hbar} \int_{t_0}^t \int_{t_0}^{\tau} d\tau d\sigma \left\{\alpha_I(\tau - \sigma)[x(\tau) - y(\tau)] [x(\sigma) + y(\sigma)]\right\}\right\}
\times \exp\left\{-\frac{1}{\hbar} \int_{t_0}^t \int_{t_0}^{\tau} d\tau d\sigma \left\{\alpha_R(\tau - \sigma)[x(\tau) - y(\tau)] [x(\sigma) - y(\sigma)]\right\}\right\}.
$$

(106)

The expressions (104)-(106) were derived in Eqs. (31)-(45), but can also be found in [2].

The two paths $x(t)$ and $y(t)$ in the two-state space $\{-q_0/2, q_0/2\}$, can also be seen as one path in four-state space. The four possible states for the pair $(x(t), y(t))$ are

$$A \equiv (q_0/2, q_0/2), \quad B \equiv (q_0/2, -q_0/2), \quad C \equiv (-q_0/2, q_0/2), \quad D \equiv (-q_0/2, -q_0/2).$$

For later use we define an alternative way to describe the path in four-state space by two paths in two-state space

$$\xi(t) = [x(t) - y(t)]/q_0,$$

$$\chi(t) = [x(t) + y(t)]/q_0.$$

In Eq. (105), the factor $\exp(i/\hbar)S_0[x(\tau)]$ governs the system following a path $x(\tau)$, in the absence of the environment. If we look back to the Hamiltonian defined in Eqs. (59) and (60), we see that for a period of time $dt$, the amplitude to stay in $\pm q_0/2$ equals $\exp(\mp i(\epsilon/2\hbar)dt)$. The amplitude to switch between states, equals $i(\Delta/2)dt$, up to first order in $dt$. For the path $y(\tau)$, the exponent in Eq. (106) comes with a minus sign. This results in an extra minus sign in the exponents of the amplitudes to stay, and in a minus sign in the amplitudes to switch. From the amplitudes for the paths $x(\tau)$ and $y(\tau)$, we easily see that for the four-state space, the amplitude to stay in the same state is $\exp[-i(\epsilon/\hbar)\xi(t)dt]$. The amplitude to switch states, equals $i\lambda(\Delta/2)dt$. Here

$$\lambda = \begin{cases} 
-1 & \text{for } A \rightleftharpoons B \text{ and } C \rightleftharpoons D \\
0 & \text{for } A \rightleftharpoons D \text{ and } B \rightleftharpoons C \\
+1 & \text{for } A \rightleftharpoons C \text{ and } B \rightleftharpoons D
\end{cases}.$$
We want to calculate the RHS of Eq. (105), with \(x = y = x_i = q_0/2\) to find \(p(t) = \rho(q_0/2, q_0/2, t)\). Defining the functional \(\tilde{F}\) as

\[
\tilde{F}[\chi[\tau], \xi[\tau]] = \mathcal{F}\left[\frac{q_0}{2}(\chi[\tau] + \xi[\tau]), \frac{q_0}{2}(\chi[\tau] - \xi[\tau])\right]
\times \exp\left[-i(\epsilon/\hbar)\int_0^t \xi[\tau]d\tau\right],
\]

the quantity \(p(t)\) can be calculated as the functional integral of \(\tilde{F}\) over all possible paths \(\chi[\tau]\) and \(\xi[\tau]\), that satisfy the boundary conditions. From Eq. (106) and the definition of the paths \(\xi[\tau]\) and \(\chi[\tau]\) it can be seen that

\[
\tilde{F}[\chi[\tau], \xi[\tau]] = \exp\left\{-\frac{q_0^2}{\hbar} \int_{t_0}^t \int_{t_0}^\tau d\tau d\sigma \left[i\alpha_I(\tau - \sigma)\chi(\sigma) + \alpha_R(\tau - \sigma)\xi(\sigma)\xi(\sigma)\right]\right\}
\times \exp\left[-i(\epsilon/\hbar)\int_0^t \xi[\tau]d\tau\right].
\]

Because only paths that start and end in \(+q_0/2\) are considered, all these paths contain \(2n\) transitions, for \(n\) integer. This is caused by the amplitudes of \(A \leftrightarrow D\) and \(B \leftrightarrow C\), which equal zero if terms of order \(dt^2\) and higher are neglected. Denoting the transition times as \(t_1, \ldots, t_{2n}\), then the path is in the state \(A\) or \(D\) on the intervals \(t_{2j} < t < t_{2j+1}\) and in the state \(B\) or \(C\) on the intervals \(t_{2j-1} < t < t_{2j}\). The \(2n\) transitions cause a factor \((-1)^n(\Delta/2)^{2n}\) due to the factors \(\pm i\Delta/2\) from the bare amplitude of the system.

The intervals \(t_{2j} < t < t_{2j+1}\) and \(t_{2j-1} < t < t_{2j}\) are characterized, respectively, by \(\eta_j\) and \(\zeta_j\), where

\[
\eta_j = \begin{cases} +1 & \text{for state } A \\ -1 & \text{for state } D \end{cases}, \quad \zeta_j = \begin{cases} +1 & \text{for state } B \\ -1 & \text{for state } C \end{cases}.
\]

From the boundary conditions we have \(\eta_0 = \eta_n = +1\), thus the path in four-state space can be expressed in terms of \(\{t_m\}\) for \(1 \leq m \leq n\), \(\{\zeta_i\}\) for \(1 \leq i \leq n\) and \(\{\eta_i\}\) for \(1 \leq i \leq n - 1\). This can now be used to find an expression for \(p(t)\)

\[
p(t) = \sum_{n=0}^\infty \int \mathcal{D}\xi[\tau] \mathcal{D}\chi[\tau] \tilde{F}_n[\chi[\tau], \xi[\tau]],
\]

\[
\tilde{F}_n[\chi[\tau], \xi[\tau]] \equiv \left.\left.\tilde{F}[\chi[\tau], \xi[\tau]]\right|_{\text{number of transitions equals } 2n}\right).
\]

Hence

\[
p(t) = 1 + \frac{1}{2} \sum_{n=0}^\infty (-1)^n \Delta^{2n} K_n(t),
\]
where
\[ K_n(t) \equiv 2^{-(2n-1)} \sum_{\{\xi_i, \eta_i\}} \int_0^t dt_{2n} \int_0^{t_{2n-1}} dt_{2n-1} \cdots \int_0^{t_2} dt_1 \times \tilde{F}_n (t_1, \ldots, t_{2n}; \xi_1, \ldots, \xi_n; \eta_1, \ldots, \eta_{n-1}). \] (111)

From Eq. (103) and the definition \( K_0(t) \equiv 1 \) we find
\[ P(t) = 2p(t) - 1 \]
\[ = \sum_{n=0}^\infty (-1)^n \Delta^{2n} K_n(t). \] (112)

Now it is time to consider more explicitly the form of \( \tilde{F}_n \). For this goal we substitute in Eq. (108) the expressions
\[ \chi(\tau) \equiv \sum_{j=0}^n \eta_j [\theta(\tau - t_{2j}) - \theta(\tau - t_{2j+1})], \]
\[ \xi(\tau) \equiv \sum_{j=1}^n \zeta_j [\theta(\tau - t_{2j-1}) - \theta(\tau - t_{2j})]. \] (113)

For \( j > k \), we can write
\[ \int_{t_{2j-1}}^{t_{2j}} \int_{t_{2k-1}}^{t_{2k}} d\tau d\sigma \]
\[ = \frac{1}{2} \left[ \int_{t_{2k-1}}^{t_{2j}} \int_{t_{2k-1}}^{t_{2k}} + \int_{t_{2k}}^{t_{2j-1}} \int_{t_{2k}}^{t_{2k-1}} - \int_{t_{2k}}^{t_{2j}} \int_{t_{2k}}^{t_{2k-1}} - \int_{t_{2k-1}}^{t_{2j-1}} \int_{t_{2k-1}}^{t_{2k-1}} \right] d\tau d\sigma. \]

It can be observed that the second integrals of \( \sin(-\omega s) \) and \( \cos(-\omega s) \) with respect to \( s \), from 0 to \( t \) are given by \( \sin(\omega t)/\omega^2 \) and \( [1-\cos(\omega t)]/\omega^2 \) respectively. Considering the functions \( \alpha_T \) and \( \alpha_R \), it is thus convenient to define
\[ Q_1(t) \equiv \int_0^\infty \frac{J(\omega)}{\omega^2} \sin(\omega t) d\omega, \] (114)
\[ Q_2(t) \equiv \int_0^\infty \frac{J(\omega)}{\omega^2} [1 - \cos(\omega t)] \coth \left( \frac{\beta \hbar \omega}{2} \right) d\omega. \] (115)

This enables us to write the factor, coming from the term proportional to \( \xi(\tau) \xi(\sigma) \) in the exponent in Eq. (108), as
\[ \exp -\frac{\alpha^2}{\pi \hbar} \left( \sum_{j=1}^n S_j(t_{2j}, t_{2j-1}) + \sum_{k=1}^n \sum_{j=k+1}^n \zeta_j \zeta_k \Lambda_{j,k}(t_{2j}, t_{2j-1}, t_{2k}, t_{2k-1}) \right), \] (116)
where we have defined
\[ S_j(t_{2j}, t_{2j-1}) \equiv Q_2(t_{2j} - t_{2j-1}), \]
\[ \Lambda_{j,k}(t_{2j}, t_{2j-1}, t_{2k}, t_{2k-1}) \equiv Q_2(t_{2j} - t_{2k-1}) + Q_2(t_{2j-1} - t_{2k}) - Q_2(t_{2j} - t_{2k}) - Q_2(t_{2j-1} - t_{2k-1}). \] (117)
The imaginary part of the exponent in Eq. (108) can be written as

\[ i \left( -\frac{\epsilon}{\hbar} \sum_{j=1}^{n} \zeta_j \cdot (t_{2j} - t_{2j-1}) + \frac{q_0^2}{\pi \hbar} \sum_{k=0}^{n-1} \sum_{j=k+1}^{n} \eta_k \zeta_j X_{jk}(t_{2j}, t_{2j-1}, t_{2k+1}, t_{2k}) \right), \quad (118) \]

where we have defined

\[ X_{j,k}(t_{2j}, t_{2j-1}, t_{2k+1}, t_{2k}) \equiv Q_1(t_{2j} - t_{2k+1}) + Q_1(t_{2j-1} - t_{2k}) - Q_1(t_{2j} - t_{2k}) - Q_1(t_{2j-1} - t_{2k+1}). \quad (119) \]

This imaginary part can be split up in two parts. The first part contains all terms where \( k \neq 0 \). Performing the summation over the values \( \pm 1 \), that \( \eta_k \) can take for \( k \neq 0 \), we arrive at

\[ \sum_{\{ \eta_k \}} \exp \left\{ i \frac{q_0^2}{\pi \hbar} \sum_{k=1}^{n-1} \sum_{j=k+1}^{n} \eta_k \zeta_j X_{jk} \right\} = \prod_{k=1}^{n-1} \left[ \exp \left\{ i \frac{q_0^2}{\pi \hbar} \sum_{j=k+1}^{n} \zeta_j X_{jk} \right\} + \exp \left\{ -i \frac{q_0^2}{\pi \hbar} \sum_{j=k+1}^{n} \zeta_j X_{jk} \right\} \right] = 2^{n-1} \prod_{k=1}^{n-1} \cos \left( \frac{q_0^2}{\pi \hbar} \sum_{j=k+1}^{n} \zeta_j X_{jk} \right). \quad (120) \]

The second part is all what is left, and it can be shown to be invariant under simultaneous reversal of the signs of all the \( \zeta_j \) for \( 1 \leq j \leq n \). This thus gives a factor

\[ \frac{1}{2} \sum_{l=\pm 1} \exp \left\{ l \sum_{j=1}^{n} \zeta_j \left[ (t_{2j} - t_{2j-1}) \frac{\epsilon}{\hbar} - \frac{q_0^2}{\pi \hbar} X_{j0} \right] \right\} = \cos \left\{ \sum_{j=1}^{n} \zeta_j \left[ (t_{2j} - t_{2j-1}) \frac{\epsilon}{\hbar} - \frac{q_0^2}{\pi \hbar} X_{j0} \right] \right\}. \quad (121) \]

Observing that the summation over the \( \eta_j \) is already performed, we can gather the results from Eqs. (116) - (121) and find the worked-out version of Eq. (111)

\[ K_n(t) = 2^{-n} \sum_{\{ \zeta_j \}} \int_0^t dt_{2n} \int_0^{t_{2n-1}} ... \int_0^{t_2} dt_1 F_n(t_1, \ldots, t_{2n}; \zeta_1, ..., \zeta_n; \epsilon), \]

\[ F_n(\{ t_m, \zeta_i \}, \epsilon) = F_n^{(a)}(\{ t_m \}) F_n^{(b)}(\{ t_m, \zeta_i \}) F_n^{(c)}(\{ t_m, \zeta_i \}) F_n^{(d)}(\{ t_m, \zeta_i \}, \epsilon). \quad (122) \]

The four factors in Eq. (122) are given by

\[ F_n^{(a)} \equiv \exp \left\{ -\frac{q_0^2}{\pi \hbar} \sum_{j=1}^{n} S_j \right\}, \quad (123a) \]

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\[ F_n^{(b)} \equiv \exp \left\{ -\frac{q_0^2}{\pi \hbar} \sum_{k=1}^{n} \sum_{j=k+1}^{n} \zeta_j \zeta_k \Lambda_{j,k} \right\}, \]  
\[ (123b) \]

\[ F_n^{(c)} \equiv \prod_{k=1}^{n-1} \cos \left( \frac{q_0^2}{\pi \hbar} \sum_{j=k+1}^{n} \zeta_j X_{j,k} \right), \]  
\[ (123c) \]

\[ F_n^{(d)} \equiv \cos \left\{ \sum_{j=1}^{n} \zeta_j \left[ (t_{2j} - t_{2j-1}) \frac{\epsilon}{\hbar} - \frac{q_0^2}{\pi \hbar} X_{j,0} \right] \right\}. \]  
\[ (123d) \]

7.2 Interpretation of the Formal Result

The expression in Eqs. (122) and (123) seems hard to interpret. To make a start, we represent a path in four-state space by a line along the time-axis if the system is in state A or D. These parts of the path are called "sojourns". If the system is in state B or C we draw a line above the time-axis, that is parallel to the axis. These parts are called "blips". The other relevant information that is left, is formed by the labels \( \zeta_j \). These are added separately. The "j-th blip" is the period \( t_{2j-1} < t < t_{2j} \), and the "j-th sojourn" is the period \( t_{2j} < t < t_{2j+1} \). Thus, by definition, the j-th blip precedes the j-th sojourn.

The four factors in Eq. (122) can now be seen as "interactions" between blips, and between blips and sojourns. These interactions are called "repulsive" if they tend to decrease \( F_n \), and are called "attractive" if they tend to increase \( F_n \). The term \( F_n^{(a)} \) can be seen as the self-interaction of the blips, while \( F_n^{(b)} \) can be seen as the interaction between different blips. Because \( \sum_{j=1}^{n} S_j \) is always positive, \( F_n^{(a)} \) is always repulsive. Because \( \sum_{j=1}^{n} S_j + \sum_{j=1}^{n} \sum_{k=1}^{j-1} \zeta_j \zeta_k \Lambda_{j,k} \) is also positive, the product \( F_n^{(a)} F_n^{(b)} \) is always repulsive. The term \( F_n^{(c)} \) represents for all blips the interaction with the previous sojourns. Finally, the term

Figure 10: graphical representation of a path in four-state space
1.3 The Noninteracting-blip Approximation

The Noninteracting-blip approximation (NIBA) is a simplification of the formal result in Eqs. (122) and (123), based on two prescriptions:

(I) set \( X_{jk} = 0 \) for \( k \neq j - 1 \) and \( X_{j,j-1} = Q_1(t_{2j} - t_{2j-1}) \),

(II) set \( \Lambda_{jk} \) equal to zero for all \( j \) and \( k \).

Using a spectral density \( J(\omega) \sim \omega^s \), prescription (I) can be made plausible by noticing that in general the length of sojourns is longer than the length of blips. This is caused by the fact that the self interaction of blips is repulsive, as can be seen from \( F_n^{(a)} \). This should be exploited in different ways for different values of \( s \), as can been seen from [2]. For \( J(\omega) \sim \omega^s \), prescription (II) is valid if we are in one of the following situations:

(i) the coupling is extremely weak,

(ii) \( s > 1 \) (super-ohmic case) and zero temperature,

(iii) \( s > 2 \) and finite temperature,

(iv) "Golden-rule" limit.

More on this Golden-rule limit can be found in [2].

The result of the prescriptions (I) and (II) is that, in the unbiased case \( \epsilon = 0 \), the Eq. (123) reduces to

\[
F_n^{(a)} = \prod_{j=1}^{n} \exp \left( -\frac{q_0^2}{\pi h} Q_2(t_{2j} - t_{2j-1}) \right), \quad (124a)
\]

\[
F_n^{(b)} = 1, \quad (124b)
\]

\[
F_n^{(c)} = \prod_{j=2}^{n} \cos \left( \frac{q_0^2}{\pi h} Q_1(t_{2j} - t_{2j-1}) \right), \quad (124c)
\]

\[
F_n^{(d)} = \cos \left( \frac{q_0^2}{\pi h} Q_1(t_2 - t_1) \right). \quad (124d)
\]

Here the symmetry of the cosine is used to leave out the factors \( \zeta_j \), that take values \( \pm 1 \). These results can be inserted in Eq. (122) to find

\[
F_n (\{t_m, \zeta_i\}, \epsilon) = F_n (t_1, t_2, \ldots t_{2n}) = \prod_{j=1}^{n} \cos \left( \frac{q_0^2}{\pi h} Q_1(t_{2j} - t_{2j-1}) \right) \exp \left( -\frac{q_0^2}{\pi h} Q_2(t_{2j} - t_{2j-1}) \right). \quad (125)
\]
If we now insert this in the expression for $K_n$, the sum over the $\zeta_j$ cancels the factor $2^{-n}$. From Eq. (112) we now see that

$$P(t) = \sum_{n=0}^{\infty} \int_0^t dt_{2n} \int_0^{t_{2n}} dt_{2n-1} \ldots \int_0^{t_2} dt_1 \prod_{j=1}^{n} f(t_{2j} - t_{2j-1}),$$

$$f(t) = \Delta^2 \cos \left( \frac{q^2}{\pi \hbar} Q_1(t) \right) \exp \left( -\frac{q^2}{\pi \hbar} Q_2(t) \right). \quad (126)$$

We now denote the Laplace transform of $P(t)$ by $P(\lambda)$, and the Laplace transform of $f(t)$ by $f(\lambda)$. Then it holds that

$$P(\lambda) = \int_0^\infty e^{-\lambda t} P(t) dt$$
$$= \sum_{n=0}^{\infty} (-1)^n \int_0^\infty dt \int_0^t dt_{2n} \int_0^{t_{2n}} dt_{2n-1} \ldots \int_0^{t_2} dt_1 e^{-\lambda t} \prod_{j=1}^{n} f(t_{2j} - t_{2j-1})$$
$$= \sum_{n=0}^{\infty} (-1)^n \int_0^\infty dt \int_0^\infty dt_1 \int_0^\infty dt_2 \ldots \int_0^\infty dt_{2n} e^{-\lambda(t_1 + t_2 + \ldots + t_{2n})} \prod_{j=1}^{n} f(t_{2j}),$$

where we have redefined the integration variables. Further calculation (using the theory of geometric series) gives

$$P(\lambda) = \sum_{n=0}^{\infty} (-1)^n \left( \int_0^\infty e^{-\lambda t} dt \right)^n \left( \int_0^\infty e^{-\lambda t} f(t) dt \right)^n$$
$$= \frac{1}{\lambda} \sum_{n=0}^{\infty} \left( -\frac{f(\lambda)}{\lambda} \right)^n$$
$$= \frac{1}{\lambda} \left[ 1 - \left( -\frac{f(\lambda)}{\lambda} \right) \right]^{-1}$$
$$= \left[ \lambda + f(\lambda) \right]^{-1}. \quad (127)$$

The Laplace transform $f(\lambda)$ is given by

$$f(\lambda) \equiv \Delta^2 \int_0^\infty \cos \left( \frac{q^2}{\pi \hbar} Q_1(t) \right) \exp \left( -\lambda - \frac{q^2}{\pi \hbar} Q_2(t) \right). \quad (128)$$

In principle, $f(\lambda)$ can be calculated for any form of $J(\omega)$. Using $f(\lambda)$, $P(t)$ can be found to equal

$$P(t) = \frac{1}{2\pi i} \int_C e^{\lambda t} P(\lambda) d\lambda$$
$$= \frac{1}{2\pi i} \int_C e^{\lambda t} \left[ \lambda + f(\lambda) \right]^{-1} d\lambda. \quad (129)$$

Here, $C$ is the so-called Bromwich contour. This is any contour from $-i\infty$ to $i\infty$ lying on the right of the singularities of $P(\lambda)$.
8 Asymptotic Behavior of the Two-State System Coupled to the Bath via a Harmonic Oscillator

In this chapter we will derive $P_\infty$, the asymptotic time-averaged population of the two-state system, in the noninteracting-blip approximation (NIBA). $P_\infty$ will be calculated for the case the two-state system is coupled to the bath via an harmonic oscillator and in the next chapter this is repeated for the case the two-state system is coupled to the bath via an anharmonic oscillator. In principle, $J_\infty$ could be obtained by taking the limit $t \to \infty$ in $P(t)$, but this would be quite cumbersome. In this chapter we will use another method: we use the $J_{\text{eff}}$ found by Garg et al in [3] (see Eq. (74) in chapter 5) and reproduce the calculation in [5]. In the next chapter we use the $J_{\text{eff}}$ that was found in Eq. (94).

8.1 Calculation of $P_\infty$

In [14] it was derived that in the NIBA, and in the limit of driving frequencies $\Omega_d$ much larger than the tunneling amplitude $\Delta$, one finds

$$P_\infty = \frac{k^-_0(0)}{k^+_0(0)}.$$  \hfill (130)

Here,

$$k^-_0(0) = \Delta^2 \int_0^\infty dt \ h^-(t) \sin(\epsilon_0 t) J_0 \left( \frac{2s}{\Omega_d} \sin \left( \frac{\Omega_d t}{2} \right) \right),$$

$$k^+_0(0) = \Delta^2 \int_0^\infty dt \ h^+(t) \cos(\epsilon_0 t) J_0 \left( \frac{2s}{\Omega_d} \sin \left( \frac{\Omega_d t}{2} \right) \right),$$ \hfill (131)

with $J_0$ the zeroth order Bessel function, which is given by

$$J_0(x) = \sum_{l=0}^{\infty} \frac{(-1)^l}{(l!)^2} \left( \frac{x}{2} \right)^{2l}.$$  \hfill (132)

Further, $h^+$ and $h^-$ are defined by

$$h^+(t) = e^{-Q'(t)} \cos [Q''(t)],$$

$$h^-(t) = e^{-Q'(t)} \sin [Q''(t)],$$ \hfill (133)

and $Q'(t)$ and $Q''(t)$ are the real and imaginary parts of the bath correlation function

$$Q(t) = \int_0^\infty d\omega \ J(\omega) \frac{\cosh(\omega/2) - \cosh[\omega(\beta/2 - it)]}{\sinh(\omega/2)}.$$  \hfill (134)
If we denote the frequency of the intermediate harmonic oscillator by $\Omega$, then

$$J_{\text{eff}}(\omega) = \frac{\eta \omega \Omega^4}{(\Omega^2 - \omega^2)^2 + 4\gamma^2 \omega^2}. $$

Now the integrand of (133) can be seen as the product of the two terms

$$I_1(\omega) \equiv \frac{J(\omega)}{\omega^2} = \frac{\eta \Omega^4}{\omega \left[ (\Omega^2 - \omega^2)^2 + 4\gamma^2 \omega^2 \right]}, $$

$$I_2(\omega, t) \equiv \frac{\cosh(\omega \beta/2) - \cosh(\omega(\beta/2 - it))}{\sinh(\omega \beta/2)}. $$

Because both terms are antisymmetric in $\omega$, we can write

$$Q(t) = \frac{1}{2} \int_{-\infty}^{\infty} d\omega \ I_1(\omega) I_2(\omega, t). $$

To perform contour integration, we investigate the poles in the upper half of the complex plane. $I_1$ has poles at $\omega_{\pm} = \pm \Omega + i\gamma$ where we have defined $\Omega^2 = \Omega^2 - \gamma^2$. $I_2$ has poles at $\nu_n = 2\pi in/\beta$. For these poles we have

$$\text{Res}(I_1, \omega_{\pm}) = -\eta \frac{\Omega^2 - \gamma^2}{8 \Omega\gamma}, $$

$$I_2(\nu_{\pm}) = \frac{\cosh(\beta/2(\pm\Omega + i\gamma)) - \cosh(\beta/2 - it)(\pm\Omega + i\gamma))}{\sinh(\omega \beta/2(\pm\Omega + i\gamma))}, $$

$$\text{Res}(I_2, \nu_n) = -\frac{2}{\beta} \left[ e^{-\nu_n t} - 1 \right], $$

$$I_1(\nu_n) = \frac{\eta \Omega^4}{\nu_n \left[ (\Omega^2 - \nu_n^2)^2 + 4\gamma^2 \nu_n^2 \right]}.

Now $Q(t)$ can be found as

$$Q(t) = \pi i \sum_{\alpha = \pm} \text{Res}(I_1, \omega_\alpha) I_2(\omega_\alpha) + \sum_{n=1}^{\infty} \text{Res}(I_2, \nu_n) I_1(\nu_n), $$

where it should be observed that the point $\omega = 0$ is a pole for both $I_1$ and $I_2$, but the residue for $I_1 I_2$ at this point is zero. Working out Eq. (138), we obtain the result

$$Q'(t) = Q'_1(t) - e^{-\gamma t} \left[ B_1 \cos(\Omega t) + B_2 \sin(\Omega t) \right], $$

$$Q''(t) = A_1 - e^{-\gamma t} \left[ A_1 \cos(\Omega t) + A_2 \sin(\Omega t) \right], $$

where

$$Q'_1(t) = B_1 + \frac{\sinh(\beta \Omega)/(\Omega + \sin(\beta \gamma)/\gamma)}{\cosh(\beta \Omega) + \cos(\beta \gamma)} \frac{\pi \eta \Omega^2 t}{4} - \frac{\pi \eta \Omega^4}{\beta} \sum_{n=1}^{\infty} (e^{-\nu_n t} - 1)/\nu_n + t \frac{\Omega^2 + \nu_n^2}{(\Omega^2 + \nu_n^2)^2 - 4\gamma^2 \nu_n^2}. $$
and

\[
\begin{align*}
A_1 &= \frac{\pi \eta}{2}, \\
A_2 &= -\frac{\pi \eta}{4} \frac{\Omega^2 - \gamma^2}{\Omega \gamma}, \\
B_1 &= \frac{A_2 \sinh(\beta \Omega) - A_1 \sin(\beta \gamma)}{\cosh(\beta \Omega) + \cos(\beta \gamma)}, \\
B_2 &= \frac{A_1 \sinh(\beta \Omega) - A_2 \sin(\beta \gamma)}{\cosh(\beta \Omega) + \cos(\beta \gamma)}.
\end{align*}
\] (141)

Now the functions \( h^+ \) and \( h^- \) can be written in a more convenient form. Because the functions \( \cos[Q''(t)] \), \( \sin[Q''(t)] \) and \( \exp[-Q'(t) + Q'_1(t)] \) oscillate with frequency \( \Omega \), they can thus be expanded as

\[
\begin{align*}
\cos[Q''(t)] &= \sum_{n=-\infty}^{\infty} [D_n(t) \cos(n \Omega t) + E_n(t) \sin(n \Omega t)], \quad (142a) \\
\sin[Q''(t)] &= \sum_{n=-\infty}^{\infty} [F_n(t) \cos(n \Omega t) + G_n(t) \sin(n \Omega t)], \quad (142b) \\
\exp[-Q'(t) + Q'_1(t)] &= \sum_{n=-\infty}^{\infty} [H_n(t) \cos(n \Omega t) + K_n(t) \sin(n \Omega t)]. \quad (142c)
\end{align*}
\]

First we define the quantities

\[
\begin{align*}
A &= e^{-\gamma t} \sqrt{A_1^2 + A_2^2}, \\
\sin X &= A_2 / \sqrt{A_1^2 + A_2^2}, \\
B &= e^{-\gamma t} \sqrt{B_1^2 + B_2^2}, \\
\sin Y &= B_2 / \sqrt{B_1^2 + B_2^2},
\end{align*}
\] (143)

so we can write

\[
\begin{align*}
\cos[Q''(t)] + i \sin[Q''(t)] &= \exp[iQ''(t)] \\
&= \exp[iA_1 - iA \cos X \cos(\Omega t) - iA \sin X \sin(\Omega t)] \\
&= \exp(iA_1) \exp[i(-A) \cos(X - \Omega t)] \quad (144)
\end{align*}
\]

\[
\begin{align*}
\exp[-Q'(t) + Q'_1(t)] &= \exp[B \cos Y \cos(\Omega t) + B \sin Y \sin(\Omega t)] \\
&= \exp[B \cos(Y - \Omega t)] \quad (145)
\end{align*}
\]
For \( m \) an integer, the Bessel function \( J_m \) and the modified Bessel function \( I_m \) are defined by

\[
J_m(x) = \sum_{l=0}^{\infty} \frac{(-1)^l}{l! (m+l)!} \left( \frac{x}{2} \right)^{2l+m},
\]
\[
I_m(x) = \sum_{l=0}^{\infty} \frac{1}{l! (m+l)!} \left( \frac{x}{2} \right)^{2l+m}.
\]

Useful are now the Jacobi-Anger identity:

\[
e^{iz \cos \phi} = \sum_{n=-\infty}^{\infty} i^n J_n(z) e^{i n \phi}, \quad (146)
\]

and the analogous identity for the modified Bessel function:

\[
e^{z \cos \phi} = \sum_{n=-\infty}^{\infty} I_n(z) \cos(n \phi). \quad (147)
\]

They can be used to find

\[
\cos[Q''(t)] + i \sin[Q''(t)] = \exp(iA_1) \sum_{n=-\infty}^{\infty} i^n J_n(-A) \]
\[
\times [\cos(nX - n\tilde{\Omega}t) + i \sin(nX - n\tilde{\Omega}t)]
\]
\[
= [\cos A_1 + i \sin A_1] \sum_{n=-\infty}^{\infty} (-i)^n J_n(A) \]
\[
\times [(\cos nX \cos n\tilde{\Omega}t + \sin nX \sin n\tilde{\Omega}t)
+ i (\sin nX \cos n\tilde{\Omega}t - \cos nX \sin n\tilde{\Omega}t)] \quad (148)
\]

\[
\exp[-Q'(t) + Q'_1(t)] = \sum_{n=-\infty}^{\infty} I_n(B) \cos(nY - n\tilde{\Omega}t)
\]
\[
= \sum_{n=-\infty}^{\infty} I_n(B)[\cos nY \cos n\tilde{\Omega}t + \sin nY \sin n\tilde{\Omega}t] \quad (149)
\]

After the definition of

\[
a_n = \cos(n\pi/2) = \begin{cases} 
0 & \text{for } n \text{ odd}, \\
(-1)^{n/2} & \text{for } n \text{ even},
\end{cases}
\]
\[
b_n = \sin(n\pi/2) = \begin{cases} 
0 & \text{for } n \text{ even}, \\
(-1)^{(n-1)/2} & \text{for } n \text{ odd},
\end{cases}
\]
and separation of real and imaginary part in Eq. (148), we find the (time-dependent) coefficients from Eqs. (142a) - (142c) to be

\[ D_m = \left[ a_m \cos(A_1) + b_m \sin(A_1) \right] J_m(A) \cos(mX), \]

\[ E_m = \left[ a_m \cos(A_1) + b_m \sin(A_1) \right] J_m(A) \sin(mX), \]

\[ F_m = \left[ a_m \sin(A_1) + b_m \cos(A_1) \right] J_m(A) \cos(mX), \]

\[ G_m = \left[ a_m \sin(A_1) + b_m \cos(A_1) \right] J_m(A) \sin(mX), \]

\[ H_m = I_m(B) \cos(mY), \]

\[ K_m = I_m(B) \sin(mY). \]

(151)

In the same way the function \( J_0[(2s/\Omega_d) \sin(\Omega_d t/2)] \) is periodic with period \( \Omega_d t \), and can be expanded using the following identity from [15]

\[ J_0(z \sin \alpha) = \sum_{k=-\infty}^{\infty} J_k^2 \left( \frac{z}{2} \right) \cos(2k\alpha). \]

(152)

The result is

\[ J_0[(2s/\Omega_d) \sin(\Omega_d t/2)] = \sum_{m=-\infty}^{\infty} L_m \cos(m\Omega_d t), \]

(153)

where

\[ L_m = J_m^2 \left( \frac{s}{\Omega_d} \right). \]

(154)

At this point, we are ready to write the integrand of \( k_0^\pm(0) \) in Eq. (131) in a more convenient way, using the expansions in Eqs. (142) and (153). For the coefficients in Eq. (151), it is useful to express

\[ a_n = \text{Re}(i^n), \]

\[ b_n = \text{Im}(i^n). \]

(155)

After the use of the identity [16]

\[ J_n(x + iy) = \sum_{m=-\infty}^{\infty} i^{n-m} I_{n-m}(y) J_m(x), \]

(156)

one of the three sums in the integrand disappears. After interchanging the resting sums and the integral, and some rewriting, we find

\[ k_0^\pm(0) = \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \Delta^2 \left[ \int_0^\infty dt e^{-Q'_1(t)} f_{mn}^\pm(t) \right]. \]

(157)

Here,

\[ f_{mn}^+(t) = \text{Re} \left[ e_{mn}^+(t) \cos(\epsilon_{mn}t) + e_{mn}^-(t) \sin(\epsilon_{mn}t) \right], \]

\[ f_{mn}^-(t) = \text{Im} \left[ e_{mn}^-(t) \cos(\epsilon_{mn}t) - e_{mn}^+(t) \sin(\epsilon_{mn}t) \right]. \]

(158)
\[ c_{mn}^+(t) = J_n^2 \left( \frac{S}{\Omega} \right) J_m \left( e^{-\gamma t} \omega_1 \right) \cos(m\phi)(-i)^m e^{-iA_1}, \]
\[ c_{mn}^-(t) = J_n^2 \left( \frac{S}{\Omega} \right) J_m \left( e^{-\gamma t} \omega_1 \right) \sin(m\phi)(-i)^m e^{-iA_1}. \] (159)

In these expressions the following definitions were used
\[ \epsilon_{mn} = \epsilon_0 - m\Omega - n\Omega_d, \]
\[ \omega_1 = \sqrt{(A_1 - iB_1)^2 + (A_2 - iB_2)^2}, \]
\[ \tan \phi = -\frac{A_2 - iB_2}{A_1 - iB_1}. \] (160)

The quantities \( \omega_1 \) and \( \phi \) can be expressed in terms of \( A, B, X, \) and \( Y \), but look more elegantly if they are worked out in terms of \( A_1, A_2, B_1, \) and \( B_2 \).

Now we consider the limit \( \gamma/\Omega \ll 1 \), so \( \bar{\Omega} \approx \Omega \). Furthermore, for \( T \) not too large (and thus \( \beta \) not too small) this implies that
\[ \cos(\beta \gamma) \ll \cosh(\beta \Omega), \]
\[ \sin(\beta \gamma) \ll \sinh(\beta \Omega). \] (161)

From
\[ \tan \phi = -\frac{A_2 - iB_2}{A_1 - iB_1} \approx i \tanh \left( \frac{\beta \Omega}{2} \right), \] (162)

it directly follows that
\[ \tan m\phi \approx i \tanh \left( \frac{m \beta \Omega}{2} \right). \] (163)

Inserting Eq. (163) into Eq. (159) yields
\[ i \tanh \left( \frac{m \beta \Omega}{2} \right) c_{mn}^+(t) = c_{mn}^-(t). \] (164)

If the integrand of Eq. (131) is not damped too fast, and for a certain \( m \) and \( n \) it holds that \( \epsilon_{mn} = 0 \), we expect the sum in Eq. (157) to be dominated by the coefficient of \( \cos \epsilon_{mn} \). This implies for \( m \) and \( n \) that
\[ f_{mn}^+(t) \approx \Re \left[ c_{mn}^+(t) \right], \]
\[ f_{mn}^-(t) \approx \tanh \left( \frac{m \beta \Omega}{2} \right) \Re \left[ c_{mn}^+(t) \right]. \] (165)

For this specific \( m \) integer it now holds that
\[ P_\infty = \tanh \left( \frac{m \beta \Omega}{2} \right). \] (166)
9 Asymptotic Behavior of the Two-State System Coupled to the Bath via an Anharmonic Oscillator

In this chapter we will derive $P_\infty$, the asymptotic time-averaged population of the two-state system, in the noninteracting-blip approximation (NIBA), for the case when the two-state system is coupled to the bath via an anharmonic oscillator. Parts of the derivation of $P_\infty$ are similar to the derivation for the case of coupling via a harmonic oscillator, in the previous chapter. This chapter contains only original calculations.

9.1 Calculation of $P_\infty$

For the calculation of $P_\infty$, we take the value of $J_{\text{eff}}$ that was found in Eq. (94) and make use of the representation in Eq. (102). Because this spectral density consists of two separated peaks and the shape of the second peak (the lower one) was found after some assumptions, it is not much weaker to assume that both peaks have the same shape, in the sense that

$$J_{\text{eff}}(\omega) = J_h(\omega) + J_a(\omega), \quad (167)$$

where the harmonic and anharmonic part of $J_{\text{eff}}$ are given by

$$J_h(\omega) = \frac{\eta_h \omega \Omega_h^4}{(\Omega_h^2 - \omega^2)^2 + 4\gamma^2\omega^2}, \quad (168)$$

$$J_a(\omega) = \frac{\eta_a \omega \Omega_a^4}{(\Omega_a^2 - \omega^2)^2 + 4\gamma^2\omega^2}. \quad (169)$$

The values $\eta_h$ and $\Omega_a$ can be chosen such that the second peak has the same center and height as before. We already know that $3\Omega_h \lesssim \Omega_a$.

From the definition of $Q(t)$ in Eq. (133) it can be seen that

$$Q(t) = Q_h(t) + Q_a(t), \quad (170)$$

where

$$Q_h(t) = \int_0^\infty d\omega \frac{J_h(\omega)}{\omega^2} \frac{\cosh(\omega\beta/2) - \cosh[\omega(\beta/2 - it)]}{\sinh(\omega\beta/2)},$$

$$Q_a(t) = \int_0^\infty d\omega \frac{J_a(\omega)}{\omega^2} \frac{\cosh(\omega\beta/2) - \cosh[\omega(\beta/2 - it)]}{\sinh(\omega\beta/2)}. \quad (171)$$

Indicating real and imaginary part by, respectively, one and two primes, yields

$$Q'(t) = Q_h'(t) + Q_a'(t),$$

$$Q''(t) = Q_h''(t) + Q_a''(t).$$
The quantities that depended on $\Omega$ and $\eta$ in the previous chapter can now be defined similarly for $\Omega$ and $\eta$ replaced by $\Omega_b$ and $\eta_b$ or $\Omega_a$ and $\eta_a$. These quantities should then be labelled with an $b$ or an $a$. As an example we can define

$$\Omega_b^2 = \Omega_b^2 - \gamma^2,$$

$$\Omega_a^2 = \Omega_a^2 - \gamma^2.$$

This labelling can also be done for all quantities involved in the expansions of $\cos[Q''(t)]$, $\sin[Q''(t)]$, and $\exp[-Q'(t) + Q'_1(t)]$. Starting from

$$\begin{align*}
\cos[Q''(t)] \exp[-Q'(t)] &= \cos[Q''_0(t)] \exp[-Q'_b(t)] \cos[Q''_a(t)] \exp[-Q'_a(t)] \\
&= \sin[Q''_b(t)] \sin[Q''_a(t)] \exp[-Q''_b(t)] \exp[-Q''_a(t)] ,
\end{align*}$$

$$\begin{align*}
\sin[Q''(t)] \exp[-Q'(t)] &= \cos[Q''_b(t)] \exp[-Q'_a(t)] \sin[Q''_a(t)] \exp[-Q''_a(t)] \\
&= \cos[Q''_b(t)] \exp[-Q'_b(t)] \cos[Q''_a(t)] \exp[-Q''_a(t)] .
\end{align*}$$

(172)

some results of the previous chapter can be used again, that is

$$\begin{align*}
\Delta^2 \int_0^\infty dt \ e^{-Q'_1(t)} \sum_{n=-\infty}^{\infty} e^{-Q'(t) + Q'_1(t)} \cos[Q''(t)] \cos(\epsilon_0 t) \cos(n \Omega_d t) \\
&= \Delta^2 \int_0^\infty dt \ e^{-Q'_1(t)} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \left\{ \text{Re} \left[ e^{-m \Omega(t)} \right] \cos[(\epsilon_0 - m \bar{\Omega}) t] \cos(n \Omega_d t) \\
&+ \text{Re} \left[ e^{-m \Omega(t)} \right] \sin[(\epsilon_0 - m \bar{\Omega}) t] \cos(n \Omega_d t) \right\} .
\end{align*}$$

(173)

This equality holds because the terms proportional to $\sin(n \Omega_d t)$ must cancel each other. The equivalent of Eq. (173) for $\cos[Q''(t)]$ replaced by $\sin[Q''(t)]$ and $\cos(\epsilon_0 t)$ replaced by $\sin(\epsilon_0 t)$ is easy to find

$$\begin{align*}
\Delta^2 \int_0^\infty dt \ e^{-Q'_1(t)} \sum_{n=-\infty}^{\infty} e^{-Q'(t) + Q'_1(t)} \sin[Q''(t)] \sin(\epsilon_0 t) \cos(n \Omega_d t) \\
&= \Delta^2 \int_0^\infty dt \ e^{-Q'_1(t)} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \left\{ \text{Im} \left[ e^{-m \Omega(t)} \right] \cos[(\epsilon_0 - m \bar{\Omega}) t] \cos(n \Omega_d t) \\
&- \text{Im} \left[ e^{-m \Omega(t)} \right] \sin[(\epsilon_0 - m \bar{\Omega}) t] \cos(n \Omega_d t) \right\} .
\end{align*}$$

(174)

Finally the equivalents of Eqs. (173) and (174) for $\cos[Q''(t)]$ replaced by $\sin[Q''(t)]$, or vice versa, can be found by replacing $c_{mn}^+(t)$ by $d_{mn}^+(t)$, where

$$\begin{align*}
d_{mn}^+(t) &= J_2^0 \left( \frac{s}{\Omega} \right) J_2 (e^{-\gamma t} \omega_1) \cos(m \phi)(-i)^{m+1} e^{iA_1} ,
\end{align*}$$

$$\begin{align*}
d_{mn}^-(t) &= J_2^0 \left( \frac{s}{\Omega} \right) J_2 (e^{-\gamma t} \omega_1) \sin(m \phi)(-i)^{m+1} e^{iA_1} .
\end{align*}$$

(175)
Now we start the calculation by
\[
\Delta^2 \int_0^\infty dt \left\{ e^{-Q_{s_1}(t) - Q_{a}(t)} \cos \left[ Q''_{a}(t) \right] \right. \\
\times \sum_{n=-\infty}^{\infty} e^{-Q_{a}(t) + Q'_{s_1}(t)} \cos \left[ Q''_{a}(t) \right] \cos(\epsilon_0 t) \cos(n\Omega dt) \left. \right\}
\]
\[
= \Delta^2 \int_0^\infty dt \left\{ e^{-Q_{s_1}(t) - Q_{a}(t)} \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \right. \\
\left[ \text{Re} \left[ c_{h m n}^+ (t) \right] e^{-Q_{s_1}(t) + Q'_{s_1}(t)} \cos \left[ Q''_{a}(t) \right] \cos(\epsilon_0 t) \cos(n\Omega dt) \right] \\
+ \text{Re} \left[ c_{h m n}^- (t) \right] e^{-Q_{s_1}(t) + Q'_{s_1}(t)} \cos \left[ Q''_{a}(t) \right] \sin(\epsilon_0 t) \cos(n\Omega dt) \right\}
\]
\[
= \Delta^2 \int_0^\infty dt \left\{ e^{-Q_{s_1}(t) - Q_{a}(t)} \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \right. \\
\left[ \text{Re} \left[ c_{h m n}^+ (t) \right] \text{Re} \left[ c_{a l n}^+ \right] + \text{Re} \left[ c_{h m n}^- (t) \right] \text{Im} \left[ d_{a l n}^- \right] \right] \cos(\epsilon_{l m n} t) \\
+ \left[ \text{Re} \left[ c_{h m n}^+ (t) \right] \text{Re} \left[ c_{a l n}^- \right] - \text{Re} \left[ c_{h m n}^- (t) \right] \text{Im} \left[ d_{a l n}^+ \right] \right] \sin(\epsilon_{l m n} t) \right\},
\]
(176)

where \( \epsilon_{l m n} \) is defined as \( \epsilon_0 - l\Omega_h - m\Omega_h - n\Omega_d \). Analogously we have
\[
\Delta^2 \int_0^\infty dt \sum_{n=-\infty}^{\infty} e^{-Q_{s_1}(t) - Q_{a}(t)} \sin \left[ Q''_{a}(t) \right] \cos(\epsilon_0 t) \cos(n\Omega dt) \\
= \Delta^2 \int_0^\infty dt \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \right. \\
\left[ \text{Re} \left[ d_{h m n}^+ (t) \right] \text{Re} \left[ d_{a l n}^+ \right] + \text{Re} \left[ d_{h m n}^- (t) \right] \text{Im} \left[ c_{a l n}^- \right] \right] \cos(\epsilon_{l m n} t) \\
+ \left[ \text{Re} \left[ d_{h m n}^+ (t) \right] \text{Re} \left[ d_{a l n}^- \right] - \text{Re} \left[ d_{h m n}^- (t) \right] \text{Im} \left[ c_{a l n}^+ \right] \right] \sin(\epsilon_{l m n} t) \right\},
\]
(177)

\[
\Delta^2 \int_0^\infty dt \sum_{n=-\infty}^{\infty} e^{-Q_{s_1}(t) - Q_{a}(t)} \sin \left[ Q''_{a}(t) \right] \cos(\epsilon_0 t) \cos(n\Omega dt) \\
= \Delta^2 \int_0^\infty dt \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \right. \\
\left[ \text{Im} \left[ c_{h m n}^- (t) \right] \text{Re} \left[ c_{a l n}^+ \right] - \text{Im} \left[ c_{h m n}^+ (t) \right] \text{Im} \left[ d_{a l n}^- \right] \right] \cos(\epsilon_{l m n} t) \\
+ \left[ \text{Im} \left[ c_{h m n}^- (t) \right] \text{Re} \left[ c_{a l n}^- \right] + \text{Im} \left[ c_{h m n}^+ (t) \right] \text{Im} \left[ d_{a l n}^+ \right] \right] \sin(\epsilon_{l m n} t) \right\},
\]
(178)
\[ \Delta^2 \int_0^\infty dt \sum_{n=-\infty}^{\infty} e^{-Q'_h(t)-Q'_a(t)} \sin [Q''_h(t)] \cos [Q''_a(t)] \cos(\epsilon_0 t) \cos(\Omega t) dt \]

\[ = \Delta^2 \int_0^\infty dt e^{-Q'_h(t)-Q'_a(t)} \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \left\{ \left[ \text{Im}(d_{h,lm}^+) \text{Im}(c_{a,ln}^-) - \text{Im}(d_{a,lm}^-) \text{Re}(d_{a,ln}^+) \right] \cos(\epsilon_{lmn} t) \\
+ \left[ \text{Im}(d_{h,lm}^-) \text{Re}(d_{a,ln}^-) + \text{Im}(d_{h,lm}^+) \text{Im}(c_{a,ln}^+ \right] \sin(\epsilon_{lmn} t) \right\}, \quad (179) \]

From these expressions, \( k_0^+ \) and \( k_0^- \) can be found by adding Eqs. (176) and (177), respectively Eqs. (178) and (179). The result can be written as

\[ k_{0}^\pm(0) = \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \Delta^2 \int_0^\infty dt e^{-Q'_h(t)-Q'_a(t)} f_{lmn}^\pm(t). \quad (180) \]

Here,

\[ f_{lmn}^+(t) = \left[ \text{Re}(c_{h,lm}^+) \text{Re}(c_{a,ln}^+) + \text{Re}(c_{h,lm}^-) \text{Im}(d_{a,ln}^-) \\
+ \text{Re}(d_{h,lm}^+) \text{Re}(d_{a,ln}^+) + \text{Re}(d_{h,lm}^-) \text{Im}(c_{a,ln}^+) \right] \cos(\epsilon_{lmn} t) \\
+ \left[ \text{Re}(c_{h,lm}^+) \text{Re}(c_{a,ln}^-) - \text{Re}(c_{h,lm}^-) \text{Im}(d_{a,ln}^-) \\
+ \text{Re}(d_{h,lm}^+) \text{Re}(d_{a,ln}^-) - \text{Re}(d_{h,lm}^-) \text{Im}(c_{a,ln}^+) \right] \sin(\epsilon_{lmn} t), \]

\[ f_{lmn}^-(t) = \left[ \text{Im}(c_{h,lm}^-) \text{Re}(c_{a,ln}^-) - \text{Im}(c_{h,lm}^+) \text{Im}(d_{a,ln}^-) \\
+ \text{Im}(d_{h,lm}^-) \text{Im}(c_{a,ln}^-) - \text{Im}(d_{h,lm}^+) \text{Re}(d_{a,ln}^-) \right] \cos(\epsilon_{lmn} t) \\
+ \left[ \text{Im}(c_{h,lm}^-) \text{Re}(c_{a,ln}^-) + \text{Im}(c_{h,lm}^+) \text{Im}(d_{a,ln}^-) \\
+ \text{Im}(d_{h,lm}^-) \text{Im}(c_{a,ln}^-) + \text{Im}(d_{h,lm}^+) \text{Re}(d_{a,ln}^-) \right] \sin(\epsilon_{lmn} t). \quad (181) \]

This result seems complicated, but now we investigate the limit \( \gamma/\Omega_h \ll 1 \) and \( \gamma/\Omega_a \ll 1 \). Furthermore, for \( T \) not too large

\[ \tan m\phi_h \approx i \tanh \left( \frac{m\beta\Omega_h}{2} \right), \quad (182a) \]

\[ \tan m\phi_a \approx i \tanh \left( \frac{m\beta\Omega_a}{2} \right). \quad (182b) \]

Inserting Eqs. (182a) and (182b) into Eq. (159) yields

\[ \text{Im}[c_{h,lm}^- (t)] \approx \tanh \left( \frac{m\beta\Omega_h}{2} \right) \text{Re}[c_{h,lm}^+(t)], \]

\[ \text{Im}[c_{a,lm}^- (t)] \approx \tanh \left( \frac{m\beta\Omega_a}{2} \right) \text{Re}[c_{a,lm}^+(t)], \quad (183) \]
and also
\[ \text{Re}[c_{h \, mn}^-(t)] \approx - \tanh \left( \frac{m \beta \Omega_h}{2} \right) \text{Im}[c_{h \, mn}^+(t)], \]
\[ \text{Re}[c_{a \, mn}^-(t)] \approx - \tanh \left( \frac{m \beta \Omega_a}{2} \right) \text{Im}[c_{a \, mn}^+(t)]. \tag{184} \]

Other useful relations are given by
\[ \text{Im}[d_{mn}^+(t)] = \text{Re}[c_{mn}^+(t)], \]
\[ \text{Re}[d_{mn}^+(t)] = \text{Im}[c_{mn}^+(t)], \]
\[ \text{Im}[d_{mn}^-(t)] = \text{Re}[c_{mn}^-(t)], \]
\[ \text{Re}[d_{mn}^-(t)] = \text{Im}[c_{mn}^-(t)]. \tag{185} \]

If the integrand of Eq. (131) is not damped too fast, and for certain \( l, m, \) and \( n \) it holds that \( \epsilon_{lmn} = 0 \), we expect the sum in Eq. (180) to be dominated by the coefficient of \( \cos \epsilon_{lmn} \),
\[ f_{lmn}^+(t) \approx \left[ 1 + \tanh \left( \frac{m \beta \Omega_h}{2} \right) \tanh \left( \frac{l \beta \Omega_a}{2} \right) \right] \times \left[ \text{Re}(c_{h \, mn}^+) \text{Re}(c_{a \, ln}^+) + \text{Im}(c_{h \, mn}^+) \text{Im}(c_{a \, ln}^+) \right], \]
\[ f_{lmn}^-(t) \approx \left[ \tanh \left( \frac{m \beta \Omega_h}{2} \right) + \tanh \left( \frac{l \beta \Omega_a}{2} \right) \right] \times \left[ \text{Re}(c_{h \, mn}^+) \text{Re}(c_{a \, ln}^+) + \text{Im}(c_{h \, mn}^+) \text{Im}(c_{a \, ln}^+) \right]. \tag{186} \]

For this specific \( l, m \in \mathbb{Z} \) it now holds that
\[ P_\infty = \frac{\tanh \left( \frac{m \beta \Omega_h}{2} \right) + \tanh \left( \frac{l \beta \Omega_a}{2} \right)}{1 + \tanh \left( \frac{m \beta \Omega_h}{2} \right) \tanh \left( \frac{l \beta \Omega_a}{2} \right)} = \tanh \left( \frac{m \beta \Omega_h + l \beta \Omega_a}{2} \right). \tag{187} \]

It is interesting to observe that the additive property of \( J(\omega) = J_1(\omega) + J_2(\omega) \) results into an additive behavior in the argument of the \( \tanh \).

10 Derivation of the Partition Function

In this chapter, we will derive the partition function \( Z(\beta) \) of the two-state system for the case when it is directly coupled to a bath of harmonic oscillators. This model was described in detail in chapter 4. The derivation is a worked out version of Appendix A of [2]. This thesis only contains the derivation of the partition function in the case of direct coupling, the derivation in the cases of indirect coupling could be a subject for future work.
10.1 General Expression for the Partition Function

In the derivation, we use the splitting of the spectral density $J_0(\omega)$ into a low-frequency part $J(\omega)$ and a high-frequency $J'(\omega)$, in the following way

$$
J_0(\omega) \equiv J(\omega) + J'(\omega)
$$

$$
J(\omega) \equiv e^{-\omega/\omega_c} J_0(\omega)
$$

$$
J'(\omega) \equiv (1 - e^{-\omega/\omega_c}) J_0(\omega).
$$

(188)

In first instance, we set the coupling constants $C_j$ for the low-frequency oscillators in the reservoir to zero. This implies that $J_0(\omega) = J'(\omega)$.

The partition function $Z(\beta)$ can be expressed in terms of the reduced density operator $\rho(q, q; \beta)$ by

$$
Z(\beta) = \int \rho(q, q; \beta) dq.
$$

(189)

The reduced density operator, in terms of the potential and the spectral density, was calculated in Eqs. (47) - (50). The result was

$$
\rho(q, q; \beta) \propto \int_{q(0)=q}^{q(\beta\hbar) = q} \mathcal{D}q(\tau) \exp\{-S_{eff}[q(\tau)]/\hbar\}.
$$

Here, $S_{eff}(\tau)$ was defined in the following way

$$
S_{eff}[q(\tau)] \equiv \int_0^{\beta\hbar} \left[\frac{1}{2} M q^2 + V(q)\right] d\tau + \frac{1}{2} \int_{-\infty}^{\infty} d\tau' \int_0^{\beta\hbar} d\tau' \alpha(\tau - \tau') [q(\tau) - q(\tau')]^2,
$$

$$
\alpha(\tau - \tau') \equiv \frac{1}{2\pi} \int_0^{\infty} J(\omega) \exp(-\omega|\tau - \tau'|) d\omega = \alpha(\tau' - \tau).
$$

The evaluation of $\rho(q, q; \beta)$ can be done by considering (near)-classical paths in

$$
V_1(q)
$$

Figure 11: Example of an inverted potential. The thick line corresponds to the values of $q$ that contribute.

an inverted potential $V_i(q) \equiv -V(q)$. Here, the paths which include tunneling
between $\pm q_0/2$ must also be taken into account (see [2], Appendix A). We note that $\rho(q, q; \beta)$ is only large near $\pm q_0/2$, the local maxima of the inverted potential $V_i(q)$. We define the zero point position $q_{zp}^{\pm}$ as $(q_{zp}^{\pm})^2 \equiv \langle q^2 \rangle$, for a particle of mass $M$, moving in the harmonic potential that approximates $V(q)$ near $\pm q_0/2$. Now we can state that $\rho(q, q; \beta)$ is only contributing in $Z(\beta)$ for $|q \mp q_0/2| \lesssim q_{zp}^{\pm}$.

We thus consider $q$ in one of these regions. The majority of near-classical paths that start and end at $q$ are of the same type. Paths of this type start in $q$ at imaginary time zero, will have just enough kinetic energy to reach the nearby maximum of $V_i(q)$, stay at that point or make transitions to the other maximum of $V_i(q)$ and back, and return from the nearby maximum to $q$ at imaginary time $h\beta$.

For $q$ near $\pm q_0/2$, $\rho(q, q)$ (we leave the dependence on $\beta$ out for the moment) will only differ from $\rho(\pm q_0/2, \pm q_0/2)$ by two small pieces of the path, from $q$ to $\pm q_0/2$ and vice versa. For a harmonic oscillator centered around $q = 0$ we have

$$\rho(q, q) \sim \exp(-q^2/q_{zp}^2). \quad (190)$$

This result is obtained in Appendix B of [1]. Hence for a harmonic oscillator centered at $\pm q_0/2$ we find

$$\rho(q, q) \sim \exp(-(q \mp q_0/2)^2/q_{zp}^{\pm^2}) \quad (191)$$

This result implies that for our problem, for $q$ near $\pm q_0/2$ it holds that

$$\rho(q, q) \sim \exp(-(q \mp q_0/2)^2/q_{zp}^{\pm^2}) \cdot \rho^{\pm}$$
$$\rho^{\pm} \equiv \rho(\pm q_0/2, \pm q_0/2). \quad (192)$$
We are now able to calculate the partition function

\[
Z(\beta) = \int_{-\infty}^{\infty} \rho(q, \beta) \, dq \\
\approx \int_{+q_0/2 + q_p^+}^{+q_0/2 - q_p^+} dq \, \exp \left[ -\left( q - q_0/2 \right)^2 / \left( q_{zp}^+ \right)^2 \right] \cdot \rho^+ \\
+ \int_{-q_0/2 + q_p^-}^{-q_0/2 - q_p^-} dq \, \exp \left[ -\left( q + q_0/2 \right)^2 / \left( q_{zp}^- \right)^2 \right] \cdot \rho^- \\
= \rho^+ \cdot \int_{-q_p^+}^{+q_p^+} dq \, \exp \left[ -q^2 / \left( q_{zp}^+ \right)^2 \right] \\
+ \rho^- \cdot \int_{-q_p^-}^{+q_p^-} dq \, \exp \left[ -q^2 / \left( q_{zp}^- \right)^2 \right] \\
\approx \rho^+ \cdot \int_{-\infty}^{\infty} dq \, \exp \left[ -q^2 / \left( q_{zp}^+ \right)^2 \right] \\
+ \rho^- \cdot \int_{-\infty}^{\infty} dq \, \exp \left[ -q^2 / \left( q_{zp}^- \right)^2 \right] \\
= \sqrt{\pi} \left[ \left( q_{zp}^+ \right)^\rho^+ + \left( q_{zp}^- \right)^\rho^- \right]. \tag{193}
\]

However, it is not necessary to calculate \( q_{zp}^\pm \), because it will turn out that \( \rho^\pm \) is proportional to \( \left( q_{zp}^\pm \right)^{-1} \).

### 10.2 Paths that Stay in One Well

We first consider the trivial paths \( q(\tau) = +q_0/2 \) and \( q(\tau) = -q_0/2 \) for all \( \tau \). Defining \( U \equiv h\beta \), the effective action for these paths equals

\[
S_{cl}[q(\tau)] = \int_{0}^{U} V[q(\tau)] \, d\tau = \int_{0}^{U} \pm \frac{\epsilon}{2} \, d\tau = \pm \frac{U}{2}. \tag{194}
\]

This can be seen from our expression (190) for the effective action, and realizing that for a constant path we have \( \dot{q}(\tau) = 0 \) and \( q(\tau) = q(\tau') = 0 \) for all \( \tau, \tau' \). Now \( \rho^\pm \) will differ from \( \exp(-S_{cl}/h) = \exp(\mp \epsilon U/2h) \) by a multiplicative factor caused by small fluctuations around the classical path. This multiplicative factor, can be determined if we write

\[
q(\tau) = -\frac{q_0}{2} = \sum_n q_n e^{i\omega_n \tau} \quad \omega_n = \frac{\pi n}{h\beta}, \tag{195}
\]

and use the Fourier transform \( \alpha(\omega) \) of \( \alpha(\tau - \tau') \)

\[
\alpha(\tau - \tau') = \frac{1}{2\pi} \int_{-\infty}^{\infty} \alpha(\omega) e^{i\omega(\tau - \tau')} \, d\omega, \\
\alpha(\omega) = \int_{-\infty}^{\infty} \alpha(\tau - \tau') e^{-i\omega(\tau - \tau')} \, d(\tau - \tau'). \tag{196}
\]

54
This directly implies

\[
\Pi(\omega) = -\frac{2}{M} [\alpha(\omega) - \alpha(0)] \\
= -\frac{2}{M} \int_{-\infty}^{\infty} \alpha(\tau - \tau') \left( e^{i\omega(\tau - \tau')} - 1 \right) d(\tau - \tau').
\]  

(197)

Near \( \pm q_0/2 \) the potential is well estimated by the quadratic function

\[
V(q) = \pm \frac{\epsilon}{2} + \frac{1}{2} M \omega_{\pm}^2 (q \mp q_0/2)^2.
\]  

(198)

Inserting now Eqs. (195) - (198) into the expression for the effective action, we obtain that

\[
S_{\text{eff}}[q(\tau)] = \int_{0}^{\beta} \frac{1}{2} M \left( \frac{d}{d\tau} \sum_n q_n e^{i\omega_n \tau} \right) \left( \frac{d}{d\tau} \sum_m q_m e^{i\omega_m \tau} \right) d\tau \\
+ \int_{0}^{\beta} \left[ \pm \frac{\epsilon}{2} + \frac{1}{2} M \omega_{\pm}^2 \left( \sum_n q_n e^{i\omega_n \tau} \right) \left( \sum_m q_m e^{i\omega_m \tau} \right) \right] d\tau \\
+ \frac{1}{2} \int_{-\infty}^{\infty} \int_{0}^{\beta} \alpha(\tau - \tau') \left( \sum_n q_n e^{i\omega_n (\tau - \tau')} - e^{i\omega_n (\tau - \tau')} \right) \\
\times \left( \sum_m q_m e^{i\omega_m (\tau - \tau')} - e^{i\omega_m (\tau - \tau')} \right) d\tau d\tau' \\
= \pm \frac{\epsilon U}{2} + \sum_{m,n} \left\{ \frac{1}{2} M q_n q_m \left[ -\omega_n \omega_m + (\omega_{\pm})^2 \right] \int_{0}^{\beta} e^{i\tau (\omega_n + \omega_m)} d\tau \right\} \\
+ \sum_{m,n} \frac{1}{2} q_n q_m \int_{-\infty}^{\infty} \int_{0}^{\beta} \alpha(t) e^{i\tau (\omega_n + \omega_m)} \\
\times (1 - e^{i\omega_n (\tau - \tau')})(1 - e^{i\omega_m (\tau - \tau')}) d\tau d\tau'.
\]  

(199)

In the RHS of Eq. (199) the integration variables can be transformed as

\[
r \equiv \tau, \quad s \equiv \tau' - \tau,
\]  

(200)
thus leading to

\[
S_{eff}[q(\tau)] = \pm \frac{\epsilon U}{2} + \sum_{n} \left\{ \frac{1}{2} MU (q_n)^2 \left[ (\omega_n)^2 + (\omega_\pm)^2 \right] \right\} \\
+ \frac{1}{2} \int_{-\infty}^{\infty} \int_{0}^{\beta h} e^{i r (\omega_n + \omega_m)} dr \\
\times \alpha(s) \left[ (1 - e^{i \omega_n s}) + (1 + e^{i \omega_n s}) \right] ds \\
= \pm \frac{\epsilon U}{2} + \sum_{n} \left\{ \frac{1}{2} MU (q_n)^2 \left[ (\omega_n)^2 + (\omega_\pm)^2 \right] \right\} \\
+ \frac{1}{2} \int_{-\infty}^{\infty} \frac{2}{M} \alpha(s) (e^{i \omega_n s} - 1) ds \\
= \pm \frac{\epsilon U}{2} + U \sum_{n} \left\{ \frac{1}{2} MU (q_n)^2 \left[ (\omega_n)^2 + (\omega_\pm)^2 + \Pi(\omega_n) \right] \right\}. \tag{201}
\]

Realizing that the quantities \((q_n)^2 U\) are small and dimensionless (as can be seen from the expression for the Fourier coefficients), we can obtain the prefactor in \(\rho^{\pm}\) for small fluctuations, using Gaussian integration over \(y_n \equiv q_n U^{1/2}\). In this Gaussian integral only small values of \(q_n\) (hence small fluctuations) will contribute.

\[
\int_{-\infty}^{\infty} dy \exp \left( x - \frac{\alpha n}{2} y^2 \right) = \left( \frac{\pi}{\alpha} \right)^{1/2} \exp x \\
\prod_{n=0}^{\infty} \left( \int_{-\infty}^{\infty} dy_n \right) \exp \left( x - \sum_{n} \frac{\alpha_n}{2} y_n^2 \right) = \prod_{n=0}^{\infty} \left( \int_{-\infty}^{\infty} \exp \left( \frac{\alpha_n}{2} y_n^2 \right) \right) \exp x \\
= \prod_{n=0}^{\infty} \left( \frac{\pi}{\alpha_n} \right)^{1/2} \exp x. \tag{202}
\]

This implies

\[
\rho^{\pm} = C^{\pm}(U) \cdot \exp \left[ \frac{-S_{cl}(\pm q_0/2)}{\hbar} \right] \\
C^{\pm}(U) = \text{const} \cdot \prod_{n=0}^{\infty} \left( M [ (\omega_\pm)^2 + (\omega_n)^2 + \Pi(\omega_n) ] \right)^{-1/2}, \tag{203}
\]

where the factors \(\pi^{1/2}\) are placed in the constant. Using appropriate normalisation, we have

\[
C^{\pm}(U) = (q_{zp}^{\pm})^{-1} \exp(-\Lambda^{\pm}(U)) \\
\Lambda^{\pm}(T) \equiv \text{const} \left\{ \frac{1}{2} \sum_{n=0}^{\infty} \ln[(\omega_\pm)^2 + (\omega_n)^2 + \Pi(\omega_n)] \right\}. \tag{204}
\]
We will first consider the function \( \Pi(\omega) \) in more detail. Following \[8\], we can use the expression (190) for \( \alpha(\tau - \tau') \) to obtain

\[
\alpha(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\tau \int_{0}^{\infty} d\omega' e^{-|\tau|} e^{-i\omega'\tau} j'(\omega')
\]

\[
= \frac{1}{2\pi} \int_{0}^{\infty} d\omega' J'(\omega') \int_{0}^{\infty} d\tau \left\{ e^{-\omega'-i\omega}\tau + e^{-(\omega'+i\omega)\tau} \right\}
\]

\[
= \frac{1}{2\pi} \int_{0}^{\infty} d\omega' J'(\omega') \left\{ \frac{1}{-\omega' - i\omega} - \frac{1}{-\omega' + i\omega} \right\}
\]

\[
= \frac{1}{\pi} \int_{0}^{\infty} \left\{ J'(\omega') \frac{\omega'}{(\omega')^2 + \omega^2} \right\} d\omega'.
\]

(205)

In the case of an ohmic spectrum, with low-frequency cutoff \( \omega_c \), \( J'(\omega) = \eta\omega \) implying that

\[
\Pi(\omega) = \frac{2}{M} \left[ \alpha(0) - \alpha(\omega) \right]
\]

\[
= \frac{2\eta}{\pi M} \int_{0}^{\infty} \left\{ \frac{(\omega')^2}{(\omega')^2 + \omega^2} - \frac{(\omega')^2}{(\omega')^2 + \omega^2} \right\} d\omega'
\]

\[
= \frac{2\eta}{\pi M} \int_{0}^{\infty} \frac{\omega^2}{(\omega')^2 + \omega^2} d\omega'
\]

\[
= \frac{2\eta}{\pi M} \left[ \omega \arctan(\omega) \right]_{0}^{\infty}
\]

\[
= \frac{\eta}{M} \arctan(\omega) = 2\gamma |\omega|.
\]

(206)

The quantity \( \gamma \) is defined as \( \gamma = \eta/2M \). Now we are not interested in \( \Lambda_+ \) and \( \Lambda_- \) themselves, as given by Eq. (204), but in their difference. We define this quantity in the following way

\[
\delta \Lambda = \Lambda_+ - \Lambda_-
\]

\[
= \frac{1}{2\pi} \int_{0}^{\infty} d\omega \ln \left\{ \frac{(\omega_+)^2 + \omega^2 + \Pi(\omega)}{(\omega_-)^2 + \omega^2 + \Pi(\omega)} \right\}.
\]

(207)

Because \( \Pi(\omega) \) is always larger or equal to zero, we see immediately that the integral in Eq. (207) is convergent. Putting now together the results (193), (204), and (207), we obtain that

\[
Z(\beta) = (q_z^+ \rho_+ + q_z^- \rho_-)
\]

\[
= (q_z^+ \exp \left( \frac{\epsilon U}{2\hbar} \right) \cdot C_+(U) + (q_z^- \exp \left( \frac{\epsilon U}{2\hbar} \right) \cdot C_-(U)
\]

\[
= \exp \left\{ \left( -\frac{\epsilon}{2\hbar} - \Lambda_+ \right) U \right\} + \exp \left\{ \left( \frac{\epsilon}{2\hbar} - \Lambda_- \right) U \right\}.
\]

(208)
Multiplying this expression with a factor $\frac{1}{2} \exp \left( \frac{\Lambda_+ + \Lambda_-}{2} \right) U$ does not change the meaning of the expression for $Z(\beta)$, hence

$$Z(\beta) = \exp \left[ \left( -\frac{\epsilon}{2\hbar} - \frac{1}{2} \delta \Lambda \right) U \right] + \exp \left[ \left( \frac{\epsilon}{2\hbar} + \frac{1}{2} \delta \Lambda \right) U \right] = \exp \left[ \left( -\frac{\tilde{\epsilon}}{2\hbar} \right) U \right] + \exp \left[ \left( \frac{\tilde{\epsilon}}{2\hbar} \right) U \right] = \cosh(\beta \tilde{\epsilon}/2).$$

Here, we have defined $\tilde{\epsilon} = \epsilon + \hbar \delta \Lambda$, which can be seen as a "renormalized energy splitting", because this partition function equals the one of a two-state system with localized states, but then with $\tilde{\epsilon}$ replaced by $\epsilon$. Up to now, we only considered paths that did not go from $q_0/2$ to $-q_0/2$ and vice versa. These paths that do go from $q_0/2$ to $-q_0/2$ (and vice versa) have to be included too. A transition between $q_0/2$ and $-q_0/2$ is called a bounce.

### 10.3 Paths that Contain Bounces

For the moment we consider the case $\epsilon < 0$, thus the case where $V(q_0/2) < V(-q_0/2)$. Then in the inverted potential $V_i$, the higher peak is at position $q_0/2$. As a consequence a path starting at $q_0/2$ with zero kinetic energy would not stop at $-q_0/2$, but would have enough kinetic energy ($-\epsilon$) to pass this point. This means that there is no classical path from $q_0/2$ to $-q_0/2$ that is stationary at $-q_0/2$. In order to construct a near-classical path for the unadapted potential, we change the inverted potential by some amount $\Delta V$ and find a classical path in this adapted inverted potential. We impose that the function $\Delta V$ satisfies:

1. $V_i(-q_0/2) + \Delta V(-q_0/2) = V_i(q_0/2)$
2. $\Delta V$ is continuous
3. $\Delta V$ is only nonzero in a relative small interval around $-q_0/2$
4. $\Delta V$ is harmonic around $-q_0/2$ (for convenience)

This set of conditions then lead to the following definition:

$$\Delta V = \left[ (-\epsilon) - \frac{1}{2} M(\omega_-)^2 \left( q + \frac{q_0}{2} \right) \right] \cdot \theta(\Delta q - |q + q_0/2|)$$

$$\Delta q = \left[ \frac{2(-\epsilon)}{M(\omega_-)^2} \right]^{1/2},$$

where it should be observed that $(-\epsilon)$ is positive. The quantity $\Delta q$ is chosen in such a way that $\Delta V$ is continuous at $-q_0/2 \pm \Delta q$.

The classical path in the adapted inverted potential that starts at $q_0/2$ and ends at $-q_0/2$ is called $q_{ncl}(\tau)$, because it is the "near classical path" in the unadapted potential. Now we consider small fluctuations around $q_{ncl}(\tau)$ and
\[ q(\tau) \equiv q_{ncl}(\tau) + \delta q(\tau) \]
\[ \delta q(-\infty) = \delta q(\infty) = 0. \]  \hfill (211)

Using again the actual potential for calculating the effective action, we obtain
\[ \delta S \equiv S_{eff}[q(\tau)] - S_{eff}[q_{ncl}(\tau)] = \delta S^{(1)} + \delta S^{(2)} + \text{h.o.t.} \]  \hfill (212)

Here \( \delta S^{(1)} \) contains terms linear in \( \delta q \) and \( \delta \dot{q} \). By h.o.t. we mean higher order terms (in this case all other terms of order two and higher in \( \delta q \) and \( \delta \dot{q} \)).

If we use the adapted potential \( V(q) - \Delta V(q) \), we can use the stationarity of the action to see that \( S_{eff}[q(\tau)] - S_{eff}[q_{ncl}(\tau)] \) is zero to linear order in \( \delta q \). Now, in the case of the actual potential (what we really want to consider), we have that \( \delta S^{(1)} \) equals the part of \( \int_0^{\beta \hbar} (\Delta V[q(\tau)] - \Delta V[q_{ncl}(\tau)]) d\tau \) that is linear in \( \delta q \). This gives
\[ \delta S^{(1)} + \text{h.o.t.} = \int_0^{\beta \hbar} (\Delta V[q(\tau)] - \Delta V[q_{ncl}(\tau)]) d\tau 
\]
\[ = -\frac{1}{2} M(\omega_-)^2 \int_0^{\beta \hbar} \left[ \left( q_{ncl}(\tau) + \delta q(\tau) + \frac{1}{2} q_0 \right)^2 - \left( q_{ncl}(\tau) + \frac{1}{2} q_0 \right)^2 \right] \]
\[ \times \theta(\Delta q - |q_{ncl}(\tau) + q_0/2|) d\tau \]
\[ = -\frac{1}{2} M(\omega_-)^2 \int_0^{\beta \hbar} \left[ 2 \left( q_{ncl}(\tau) + \frac{1}{2} q_0 \right) \delta q(\tau) + (\delta q(\tau))^2 \right] \]
\[ \times \theta(\Delta q - |q_{ncl}(\tau) + q_0/2|) d\tau. \]  \hfill (213)

Introducing the notation \( z_{ncl}(\tau) \equiv q_{ncl}(\tau) + q_0/2 \), Eq. (213) may be rewritten as
\[ \delta S^{(1)} = -M(\omega_-)^2 \int_0^{\beta \hbar} z_{ncl}(\tau) \delta q(\tau) \theta(\Delta q - |z_{ncl}(\tau)|) d\tau. \]  \hfill (214)

We first consider the contribution of \( \delta S^{(2)} \) to the partition function. This can be done by expressing \( \delta q \) in terms of an orthonormal basis of eigenvectors of the operator \( \delta / \delta \tau \). These eigenvectors are called \( q_n(\tau) \) for \( n \geq 0 \) in such a way that for the corresponding eigenvalues we have \( \lambda_0 = 0 \) and \( \lambda_n > 0 \) for \( n > 0 \). Observe that \( q_0 = \text{const.} \delta q_{ncl}(\tau)/d\tau \) is the "zero mode", which needs to be treated separately. Because we want to integrate over all paths, and these are expanded around the near classical path in terms of the coefficients \( c_n \) and \( c_m \) we can integrate over all these coefficients from \(-\infty\) to \(+\infty\). In the calculation
it will arise that the only contribution comes from \( c_n c_m \) for \( n = m \), and only for small values of \( c_n \) in the Gaussian integral

\[
\exp \left[ -\frac{1}{\hbar} \int d\tau \delta S^{(2)} \right] \propto B \cdot \prod_{n,m > 0} \left( \int_{-\infty}^{\infty} dc_n \int_{-\infty}^{\infty} dc_m \right) \\
\times \exp \left[ -\text{const} \cdot \left( \sum_{n>0} c_n \frac{dq_n(\tau)}{d\tau} \right) \left( \sum_{m>0} c_m q_m(\tau) \right) \right] \\
= B \cdot \prod_{n>0} \left( \int_{-\infty}^{\infty} dc_n \right) \exp \left[ -\text{const} \cdot \left( \sum_{n>0} \lambda_n(c_n)^2 \right) \right] \\
= \text{const} \cdot B \cdot \prod_{n>0} (\lambda_n)^{-1/2}. \tag{215}
\]

Here, \( B \) is the multiplicative contribution of the zero mode. We define the product over \( (\lambda_n)^{-1/2} \), as \( D_0 \). Because in fact the eigenvalues \( \lambda_n \) depend on the time \( t_1 \) of the center of the bounce, we have

\[
D_0(t_1) = \prod_{n>0} (\lambda_n(t_1))^{-1/2}. \tag{216}
\]

Using the same expansion for \( \delta q \), the contribution of the \( \delta S^{(1)} \) to the partition function can be seen to be the extra multiplicative factor \( F \) as defined below,

\[
F \equiv \exp \left[ \sum_{n>0} \frac{C_n}{\lambda_n \hbar} \right], \\
C_n = (M \omega_-)^2 \int_0^{\beta \hbar} z_{ncl}(\tau) q_n(\tau) \theta(\Delta q - |z_{ncl}(\tau)|) d\tau. \tag{217}
\]

This equation can be obtained by integrating \( \exp(-\delta S^{(1)}/\hbar) \) over small values of the expansion coefficients \( c_n \), because we only want to consider small fluctuations. In the contribution for \( \delta S^{(1)} \), the zero mode plays no role. In the contribution for \( \delta S^{(2)} \), the factor \( B \) can be found to equal the contribution for the zero mode in the case the path \( q_{ncl} \) would have been really classical. If we denote by \( B^* \) twice the kinetic energy integrated along \( q_{ncl} \) in the adapted potential, we arrive at

\[
B = \left( \frac{B^*}{2\pi \hbar} \right)^{1/2} U. \tag{218}
\]

Finally, we need to avoid overcounting of small fluctuations around the parts of the path where it is nearly stationary. To implement this step, the result obtained so far must be divided by \( C_+(t_1)C_-(U - t_1) \). Here, \( C_\pm \) are the same as in Eq. (204).
Gathering the results, we have that the multiplicative factor for a single bounce (also called single-instanton) equals

\[ I_0 = \left( \frac{D_0(t_1)}{C^+(t_1)C^-(U - t_1)} \right) F \left( \frac{B_2^*}{2\pi \hbar} \right)^{1/2} \exp \left( -\frac{S_{cl}}{\hbar} \right) \]

\[ = \frac{\Delta}{2} \left( (q_{zp}^+ + q_{zp}^-)^{1/2} \right). \tag{219} \]

The quantity \( \Delta \) is defined as below, in such a way that it plays the same role as the quantity \( \Delta \) in the "bare" tunneling element for the two-state problem.

\[ \Delta \equiv \left( \frac{2}{(q_{zp}^+ q_{zp}^-)^{1/2}} \right) \left( C^+(t_1)C^-(U - t_1) \right) F \left( \frac{B_2^*}{2\pi \hbar} \right)^{1/2} \exp \left( -\frac{S_{cl}}{\hbar} \right). \tag{220} \]

The corresponding expressions for a bounce from \(-q_0/2\) to \(q_0/2\) (this we call an anti-instanton) are easily obtained by interchanging \(C^+\) and \(C^-\). We will now proceed with the calculation of the partition function, by choosing the zero of energy at

\[ \frac{1}{2} \left[ V \left( \frac{1}{2} q_0 \right) + V \left( -\frac{1}{2} q_0 \right) + \frac{1}{2} \hbar (\Lambda_+ + \Lambda_-) \right]. \]

For a time \( dt \) spent at the well around \( \pm q_0/2 \), we have a factor \( \exp(\mp \epsilon/2\hbar) \) \( dt \) from the classical action and a factor \( (q_{zp}^\pm)^{-1} \exp(\mp \delta \Lambda/2) \) \( dt \) from the harmonic fluctuations. With \( \tilde{\epsilon} \) defined as \( \epsilon + \hbar \delta \Lambda \), this gives a total factor of \( (q_{zp}^\pm)^{-1} \exp(\mp \tilde{\epsilon}/2\hbar) \) \( dt \).

All the paths contributing to \( \rho^+ \) have an equal number \( n \) of instantons and anti-instantons and together they give a factor \( (\Delta/2)^{2n} \cdot (q_{zp}^+ q_{zp}^-)^n \). The \( n + 1 \) pieces in the "+" well and the \( n \) pieces in the "−" well contribute with \( (q_{zp}^+)^{-n+1}(q_{zp}^-)^{-n} \) times exponential factors that do depend on the time associated with the center of the bounce. Independent of \( n \), all these paths contain one factor \( q_{zp}^+ \) which will cancel the factor \( (q_{zp}^+)^{-1} \) in the expression (193) for the partition function. The same holds for the factor \( q_{zp}^- \) in \( \rho^- \) and the factor \( (q_{zp}^-)^{-1} \) in Eq. (193).

Taking into account the exponents depending on the bounce-times, we obtain the following form of \( Z(\beta) \)

\[ Z(\beta) = \sum_{n=0}^{\infty} S_n(\beta) \left( \frac{\Delta}{2} \right)^{2n}, \tag{221} \]
\[ S_0(\beta) \equiv 2 \cosh \left( \frac{\beta \epsilon}{2} \right), \]
\[ S_n(\beta) \equiv 2 \int_0^{\beta \hbar} dt_{2n} \int_0^{t_{2n-1}} dt_{2n-1} \ldots \int_0^{t_2} dt_1 \]
\[ \times \exp \left[ \frac{\epsilon}{2\hbar} \left( \sum_{j=0}^{n} (t_{2j+1} - t_{2j}) - \sum_{j=0}^{n-1} (t_{2j+2} - t_{2j+1}) \right) \right]. \] (222)

The last expression for \( S_n \) holds for all \( n > 0 \), under the boundary conditions \( t_{2n+1} = \beta \hbar \) and \( t_0 = 0 \).

### 10.4 The Final Result

The functional form of \( Z(\beta) \) can be obtained by writing
\[ S_n(\beta) = \int_0^{\beta \hbar} dt_{2n} \int_0^{t_{2n-1}} dt_{2n-1} \exp \left\{ \frac{\epsilon}{2\hbar} (t_{2n+1} - t_{2n}) - (t_{2n} - t_{2n-1}) \right\} \]
\[ \times S_{n-1}(t_{n-1}/\hbar), \]
\[ = \int_0^{\beta \hbar} dt' \int_0^{t'} dt \exp \left[ \frac{\epsilon}{2\hbar} (\beta \hbar - 2t' + t) \right] \cdot S_{n-1}(t'/\hbar). \] (223)

Differentiating with respect to \((\beta \hbar)\) twice, we find
\[ \frac{d S_n(\beta)}{d(\beta \hbar)} = \frac{\epsilon}{2\hbar} \int_0^{\beta \hbar} dt' \int_0^{t'} dt \exp \left[ \frac{\epsilon}{2\hbar} (\beta \hbar - 2t' + t) \right] \cdot S_{n-1}(t'/\hbar) \]
\[ + \int_0^{\beta \hbar} dt \exp \left[ \frac{\epsilon}{2\hbar} (-\beta \hbar + t) \right] \cdot S_{n-1}(t/\hbar), \]
\[ \frac{d^2 S_n(\beta)}{d(\beta \hbar)^2} = \left( \frac{\epsilon}{2\hbar} \right)^2 \int_0^{\beta \hbar} dt' \int_0^{t'} dt \exp \left[ \frac{\epsilon}{2\hbar} (\beta \hbar - 2t' + t) \right] \cdot S_{n-1}(t'/\hbar) \]
\[ + \frac{\epsilon}{2\hbar} \int_0^{\beta \hbar} dt \exp \left[ \frac{\epsilon}{2\hbar} (-\beta \hbar + t) \right] \cdot S_{n-1}(t/\hbar) \]
\[ - \frac{\epsilon}{2\hbar} \int_0^{\beta \hbar} dt \exp \left[ \frac{\epsilon}{2\hbar} (-\beta \hbar + \beta \hbar) \right] \cdot S_{n-1}(\beta \hbar) \]
\[ + \exp \left[ \frac{\epsilon}{2\hbar} (-\beta \hbar + \beta \hbar) \right] \cdot S_{n-1}(\beta \hbar) \]
\[ = \left( \frac{\epsilon}{2\hbar} \right)^2 S_n(\beta) + S_{n-1}(\beta). \] (224)
This result can be used to obtain a differential equation for \( Z(\beta) \). In this procedure we will use that \( S_{-1}(\beta) \equiv 0 \).

\[
\frac{d^2 Z(\beta)}{d (\beta \hbar)^2} = \sum_{n=0}^{\infty} \frac{d^2 S_n(\beta)}{d (\beta \hbar)^2} \left( \frac{\Delta}{2} \right)^{2n} = \left( \frac{\epsilon}{2\hbar} \right)^2 \sum_{n=0}^{\infty} S_n(\beta) \left( \frac{\Delta}{2} \right)^{2n} + \sum_{m=0}^{\infty} S_{m-1}(\beta) \left( \frac{\Delta}{2} \right)^{2m} = \frac{1}{4} \left( \frac{\epsilon^2}{\hbar^2} + \Delta^2 \right) Z(\beta). \tag{225}
\]

To find the appropriate boundary conditions, we use Eqs. (221) and (222)

\[
S_0(0) = 2, \quad S_n(0) = \left. \frac{d S_0}{d\beta} \right|_{\beta=0} = \left. \frac{d S_n}{d\beta} \right|_{\beta=0} = 0,
\]

where again \( n > 0 \), to find that

\[
Z(0) = 2, \quad \left. \frac{d Z}{d\beta} \right|_{\beta=0} = 0.
\]

Using these boundary conditions, we then easily find the final expression for the partition function

\[
Z(\beta) = 2 \cosh \left[ \frac{\beta \hbar}{2} \left( \frac{\epsilon^2}{\hbar^2} + \Delta^2 \right)^{1/2} \right] = 2 \cosh \left[ \frac{\beta}{2} \left( \epsilon^2 + (\hbar \Delta)^2 \right)^{1/2} \right]. \tag{226}
\]

This answer should be corrected because we did not account for the low-frequency part of the bath. In [2], however, it was argued that this correction is negligibly small.

To conclude this chapter we will show that the partition function that we found equals the one of a two-state system with a tunneling matrix element \( \frac{1}{2} \hbar \Delta \). The latter is described by the Hamiltonian

\[
H = -\frac{1}{2} \hbar \Delta \sigma_x + \frac{1}{2} \epsilon \sigma_z, \tag{227}
\]

which can be promptly diagonalized with eigenvalues \( \pm (\epsilon^2 + (\hbar \Delta)^2)^{1/2} \). Because the partition function of a two-state system without tunneling (thus with Hamiltonian \( H = \epsilon \sigma_z / 2 \) ) equals \( \cosh [\beta \epsilon / 2] \), the partition function of a two-state system with tunneling equals Eq. (226).
11 Conclusions

In summary, we have investigated the behavior of a two-state system (TSS) coupled to its environment. We first discussed the possibility of creating a TSS using a Superconducting Quantum Interference Device (SQUID) in the presence of a perpendicular magnetic field. The magnetic flux inside the ring is quantized in units of the flux quantum $\phi_0 = \hbar c/(2e)$. By tuning the external flux $\phi_x$, we can control the effective potential, for instance, for $\phi_x = \phi_0/2$ we obtain an unbiased TSS.

The TSS is never isolated from its environment. Caldeira and Leggett [1] introduced the idea of coupling of the system of interest with a bath to account for dissipation. The Spin-Boson Hamiltonion, which describes a TSS in contact with a bath of harmonic oscillators was proposed by Legget et al. [2]. The bath is fully characterized by its spectral function, which is nothing but the imaginary part of the Fourier transform of the retarded dynamical susceptibility of the oscillators bath in the classical limit. The spectral function for direct coupling of an arbitrary system to a harmonic bath is called Ohmic if it is proportional to $\omega$, that is, if the system is equally damped for all frequencies. This gives problems for very large $\omega$, hence we use a cut-off frequency or multiply $J(\omega)$ with a decaying exponential. The model with Ohmic environment has in the classical limit the Langevin equation as equation of motion. Besides the direct coupling between the system of interest and the bath, other more elaborate models have been proposed such as coupling a system to an Ohmic bath via an intermediate harmonic oscillator (HO) [3]. This model yields an effective spectral function that has a Lorentzian shape: it is peaked around the characteristic frequency $\Omega$ of the intermediate HO, but behaves ohmically at small frequencies.

Here we consider a system coupled to an ohmic bath via an intermediate anharmonic oscillator (AHO). We find that the spectral function then consists of two peaks. The first peak is centered around $\Omega$, the characteristic frequency of the harmonic part of the intermediate AHO. The second peak is centered around a value slightly larger than $3\Omega$. Both peaks can approximately be represented by Lorentzian curves. The height and exact location of the center of the second peak are related, and both depend on the parameter characterizing the anharmonicity of the AHO. The height of the second peak is maximally about 10% of the height of the first peak.

We then conclude that a structured spectral function leads to a better control of the environment. Further, the nonlinearity leads to an additive behavior: $J(\omega) = J_1(\omega) + J_2(\omega)$, where $J_1$ and $J_2$ are ordinary Lorentzian spectral functions. This property may help to control decoherence in quantum computation.

Focussing on the TSS, we first reproduced some results obtained by Leggett et al. [2]. We have shown that the partition function of the TSS without tunneling is $Z(\beta) = \cosh[\beta\epsilon/2]$, whereas the partition function of the TSS with
tunneling equals the partition function above with $\epsilon$ replaced by the square root of $\epsilon^2 + \hbar^2 \Delta^2$.

Another interesting physical property is $P(t)$, the expectation value of the position of the TSS at time $t$ if the locations of the minima of the potential are denoted by $\pm 1$. For direct coupling with the Ohmic bath this quantity can be derived in the well known noninteracting-blip approximation (NIBA) [2]. For coupling to the bath via an intermediate HO, the equilibrium value of $P(t)$, which is denoted by $P_\infty$, can be calculated to be approximately $P_\infty = \tanh[m/\beta \Omega/2]$, for a certain integer $m$ [5]. The value of $m$ is found from the assumption that one mode dominates in an integral over a sum of Fourier-modes.

We calculated $P_\infty$ for coupling to the bath via an intermediate AHO. If the center of the second peak of the spectral function is denoted by $\Omega_a$, then $P_\infty$ approximately equals $\tanh[\beta(k \Omega + l \Omega_a)/2]$, for certain integers $k$ and $l$. As before, the values of $k$ and $l$ are found from the assumption that one mode dominates in an integral over a sum of Fourier-modes.

As a final remark, we can say that by changing the coupling between the TSS and the bath, we modify the physical quantities related to the system. For example, tuning the amount of anharmonicity in an intermediate AHO could be a useful tool in influencing $P_\infty$. 
A Solution of the equation of motion of a quartic anharmonic oscillator

If we couple the two-state system to the bath of harmonic oscillators via an anharmonic oscillator, we encounter the classical equation of motion (76b). If we set the RHS of (76b) equal to zero, it is convenient to know the solution of the following differential equation

\[ \ddot{y}(t) + Ay(t) + By^3(t) = 0, \]  
where \( A, B \geq 0 \). This last prescription is very important. The equation

\[ \ddot{y}(t) - Ay(t) - By^3(t) = 0, \]  
(can be solved by \( y(t) = \sqrt{A/B} \cot \left( \sqrt{A/2}t \right) \)). The equation

\[ \ddot{y}(t) + Ay(t) - By^3(t) = 0, \]  
(can be solved by \( y(t) = \sqrt{A/B} \coth \left( \sqrt{A/2}t \right) \)), by making the replacements \( \sin \rightarrow \sinh \) and \( \cos \rightarrow \cosh \) in the solution of Eq. (229). Finally, the equation

\[ \ddot{y}(t) + Ay(t) + By^3(t) = 0, \]  
(could be solved by making the substitution \( t \rightarrow it \) in the solution of Eq. (229). This, however results in the function \( y(t) = i\sqrt{A/B} \coth \left( \sqrt{A/2}t \right) \). This function is imaginary, while we are looking for a real solution \( y(t) \).

We will now look for solutions of

\[ \ddot{y}(t) + y(t) + \lambda y^3(t) = 0, \]  
(because in a solution of this equation for \( \lambda = B/A \) we can take \( t \rightarrow \sqrt{A}t \) and obtain a solution of Eq. (231). An exact solution for Eq. (232) turns out to exist, but the result is practically useless. From [17] it can be seen to be

\[ t = \frac{1}{(1 + 4\lambda E)^{1/4}} \left\{ K \left( \frac{a^2}{a^2 + b^2} \right) - F \left( \cos^{-1} \left( \frac{y(t)}{a} \right), \frac{a^2}{a^2 + b^2} \right) \right\}. \]  
(233)

Here \( F(\theta, \phi) \) is the elliptic integral of the first kind, that is

\[ F(\phi, k) = \int_0^\phi \frac{d\theta}{\sqrt{1 - k^2 \sin^2 \theta}} = \int_0^{\sin \phi} \frac{dt}{\sqrt{(1 - k^2 t^2) (1 - t^2)}}, \]  
(234)

and \( K(k) \) is defined as \( F(\pi/2, k) \). Further, \( E \) is the "constant of motion" given by

\[ E = \frac{1}{2} \dot{y}^2 + \frac{1}{2} y^2 + \frac{\lambda}{4} y^4, \]  
(235)
but in solving the differential equation (232) it is just an undetermined (free) constant. Further

\[ a^2 = \frac{1}{\lambda} \left( \sqrt{1 + 4\lambda E} - 1 \right), \quad b^2 = \frac{1}{\lambda} \left( \sqrt{1 + 4\lambda E} + 1 \right). \tag{236} \]

The most important conclusion that can be drawn from Eq. (233) is that the frequency of the motion \( \omega \) is constant

\[ \omega = \frac{\pi}{2} \frac{(1 + 4\lambda E)^{1/4}}{K [a^2 / (a^2 + b^2)]}. \tag{237} \]

The frequency \( \omega \) behaves as \( 1 + O(\lambda) \) for \( \lambda \ll 1 \) and as \( \lambda^{1/4} \) for \( \lambda \gg 1 \).

Now we look for an accurate approximation of the solution of Eq. (232). For this aim, the equation is rewritten as

\[ \ddot{y} + \omega_0^2 y = -\lambda \left( y^3 - \frac{3}{2} \langle y^2 \rangle y \right), \tag{238a} \]

\[ \omega_0^2 = 1 + \frac{3}{2} \lambda \langle y^2 \rangle. \tag{238b} \]

This rewriting is a trick to make the RHS of the differential equation small for all values of \( \lambda \), coming from the fact that for a simple harmonic function (sine or cosine) it holds that \( \langle y^4 \rangle = 3/2 \cdot \langle y^2 \rangle^2 \) because \( \langle \sin^4(t) \rangle = \langle \cos^4(t) \rangle = 3/8 \) and \( \langle \sin^2(t) \rangle = \langle \cos^2(t) \rangle = 1/2 \).

For a moment we take the RHS of Eq. (238a) to be zero, which results in the solution

\[ y_0 = a \cos(\omega_0 t). \tag{239} \]

Inserting this solution in Eqs. (238b) and (235) now gives

\[ \omega_0^2 = 1 + \frac{3}{4} \lambda a^2, \tag{240a} \]

\[ E = \frac{1}{2} \omega_0^2 a^2 \left( 1 - \frac{3 \lambda a^2}{16 \omega_0^2} \right). \tag{240b} \]

From Eqs. (240a) and (240b) we can find the scale factors \( \omega_0 \) and \( a \)

\[ a^2 = \frac{2E}{\omega_0^2} \left( 1 + \frac{3}{16} \lambda \right), \tag{241a} \]

\[ \omega_0^2 = \frac{1}{2} \left\{ 1 + \left[ 1 + 6\lambda E \left( 1 + \frac{3}{16} \lambda \right) \right]^{1/2} \right\}. \tag{241b} \]
Here the quantity $\tilde{\lambda}$ is the rescaled coupling constant as defined below, and it can be expressed in terms of $\lambda$ and $E$

$$
\tilde{\lambda} \equiv \frac{\lambda a^2}{\omega_0^2} = \frac{8E\lambda}{(1 + \sqrt{1 + 6\lambda E})^2}.
$$

(242)

From Eq. (241b) it can be seen that $\omega_0$ has the right behavior as function of $\lambda$, for $\lambda \ll 1$ and for $\lambda \gg 1$. Now we define the scaled variables $x = y/a$ and $\tau = \omega_0 t$ and rewrite Eq. (238a) in the form

$$
\frac{d^2x}{d\tau^2} + x = -\tilde{\lambda} \left( x^3 - \frac{3}{2} (x^2)x \right).
$$

(243)

To find an approximated solution of this equation, we use the Lindstedt-Poincare technique. More on this technique can be found in [18]. We write $x(t)$ as a $\tilde{\lambda}$-power series

$$
x(t) = \sum_{n=0}^{\infty} x_n(t) \tilde{\lambda}^n.
$$

(244)

The terms independent of $\tilde{\lambda}$ give the differential equation

$$
\frac{d^2x_0}{d\tau^2} + x_0 = 0,
$$

(245)

which can be solved by $x_0(\tau) = \sin \tau$. The terms linear in $\tilde{\lambda}$ give the differential equation

$$
\frac{d^2x_1}{d\tau^2} + x_1 = -\left( x_0^3 - \frac{3}{4} x_0 \right),
$$

(246)

which can be solved by $x_1(\tau) = \frac{1}{32} \sin(3\tau)$. Up to first order in $\tilde{\lambda}$ we have

$$
x(t) = \sin(\tau) + \frac{\tilde{\lambda}}{32} \sin(3\tau).
$$

(247)

From Eq. (242) it can be seen that even for $\lambda \to \infty$, $\tilde{\lambda}$ remains finite and approaches the value $4/3$. This implies that the convergence is fast for all $\lambda$. Up to order $\tilde{\lambda}$ we obtain

$$
\omega = \omega_0,
$$

(248a)

$$
y(t) = a \left[ \sin(\omega t) + \frac{\tilde{\lambda}}{32} \sin(3\omega t) \right].
$$

(248b)

In [Baner84] it was checked that this approximation has an accuracy better than 2% for all values of $\lambda$.

As a summary, the differential equation (231) can in good approximation be solved by

$$
y(t) = a \left[ \sin(\omega \sqrt{At}) + \frac{\tilde{\lambda}}{32} \sin(3\omega \sqrt{At}) \right],
$$

(249)
where $a$ can be chosen freely, and $\omega$ and $\bar{\lambda}$ are given by

\begin{align*}
\omega &= \sqrt{1 + \frac{3B}{4A}} a, \\
\bar{\lambda} &= \frac{(a^2 B)/A}{1 + (3aB)/(4A)}.
\end{align*}

(250)

For $B > 0$, compared with the well-known solution $y(t) = a \sin(\sqrt{A}t)$ for $B = 0$, the period of the solution shrinks, and an extra term is added. This extra term effectively flattens the solution a small amount at its extremal values. In figure 12, the solution is plotted for the cases $B = 0$ and $B > 0$.

![Graph showing the approximated solution for $A = 10$ and $B = 6$ (the one with smallest period) and for $A = 10$ and $B = 0$ (the other)](image)

Figure 12: the approximated solution for $A = 10$ and $B = 6$ (the one with smallest period) and for $A = 10$ and $B = 0$ (the other)

### B Alternative Calculation of the Effective Spectral Density in the Case of an Intermediate Anharmonic Oscillator

Instead of comparing the expected form of $y(z)$ and $y(z/3)$, as we did before, another attempt for deriving the effective spectral density in the case of coupling via an anharmonic oscillator, is presented below. In this derivation an assumption is made about term-wise equality in an equality between two sums. Probably this is quite rough, so that details in the spectral function are lost, as can be seen from the fact that the second smaller peak we saw before, does not occur now.

From the Leggett-prescription, which we used before, we know that there must be a function $K(z)$ such that

\[ K(z)q(z) = -U_z''(q). \]

(251)
Now Eq. (84a) directly implies that there should be a function $g(z)$ such that

$$g(z) = g(z)q(z),$$

(252)

where then $K(z)$ is given by

$$K(z) = -\mu z^2 + M\Omega^2 + M\Omega^2 g(z).$$

(253)

Using Eq. (85) and summing in a convenient way, we find for arbitrary $d_n(z)$ that

$$-M\Omega^2 \sum_n d_n(z)q \left( \frac{z}{3^n} \right) = \sum_n d_n(z)f \left( \frac{z}{3^n} \right) + \frac{1}{4}\zeta a^2 \sum_n d_n(z)q \left( \frac{z}{3^n+1} \right) \left( \frac{z}{3^n} \right),$$

(254)

If we take the equality to be valid term by term, we obtain from Eq. (252)

$$y \left( \frac{z}{3^n} \right) = g \left( \frac{z}{3^n} \right)q \left( \frac{z}{3^n} \right),$$

(255)

that for all $n$

$$-M\Omega^2 \frac{y \left( \frac{z}{3^n} \right)}{g \left( \frac{z}{3^n} \right)} = f \left( \frac{z}{3^n} \right) + \frac{1}{4}\zeta a^2 \frac{d_{n-1}(z)}{d_n(z)}.$$  

(256)

From this equality it can be inferred that the fraction $d_{n-1}(z)/d_n(z)$ should be a function of $z/3^n$, and further independent of $n$. This can be implemented in a trivial way by choosing $d_n = \text{constant}$. Choosing $n = 0$ in Eq.(256) we see that

$$g(z) = \frac{-M\Omega^2}{f(z) + \frac{1}{4}\zeta a^2},$$

(257)

Again using the notation

$$L(z) = -z^2 \left[ M + \sum_\alpha \frac{c_\alpha^2 \omega_\alpha^2}{m_\alpha (\omega_\alpha^2 - z^2)} \right],$$

this can be written as

$$g(z) = \frac{-M\Omega^2}{M\Omega^2 + \frac{1}{4}\zeta a^2 + \frac{1}{4}\zeta a + L(z)}.$$

(258)

Now, from Eq. (253) we find

$$K(z) = -\mu z^2 + M\Omega^2 - \frac{(M\Omega^2)^2}{M\Omega^2 + \frac{1}{4}\zeta (a^2 + 3a) + L(z)} \left( \frac{M\Omega^2 + \frac{1}{4}\zeta (a^2 + 3a) + L(z)}{M\Omega^2 + \frac{1}{4}\zeta (a^2 + 3a) + L(z)} \right).$$

(259)
In Eq. (72) we calculated that
\[ L(z) = -Mz^2 + i\eta z. \]

The Leggett prescription for obtaining \( J_{\text{eff}}(\omega) \) is as follows
\[
J_{\text{eff}}(\omega) = \lim_{\epsilon \to 0^+} \text{Im}[K(\omega - i\epsilon)].
\]

In our case we can just take
\[
J_{\text{eff}}(\omega) = \text{Im}[K(\omega)] = \text{Im}\left\{ \frac{M\Omega^2 [\zeta(a^2 + 3a)/4 - M\omega^2 + i\eta\omega]}{M\Omega^2 + \zeta(a^2 + 3a)/4 - M\omega^2 + i\eta\omega} \right\} = \frac{\eta\omega M^2 \Omega^4}{M^2 [(\Omega^2 + \zeta(a^2 + 3a)/(4M) - \omega^2)^2 + \eta^2 \omega^2]}
\]
\[
= \frac{\eta\omega \Omega^4}{[\Omega^2 + \Delta - \omega^2]^2 + 4\gamma^2 \omega^2}.
\]

In the last step the definitions \( \gamma \equiv \eta/2M \) and \( \Delta \equiv \zeta(a^2 + 3a)/(4M) \) were used.

We recall that \( a \) was the typical amplitude of the intermediate harmonic oscillator \( y(t) \). In the definition of \( \Delta \) above, \( a \) is encountered, as well as the other parameter \( \zeta \) that characterizes the nonlinearity.

The derivation of the effective spectral density in Eq. (261) involved some assumptions and simplifications, but gives an idea about the physics involved. We observe that the effective spectral density in Eq. (261) can be identified with the one in Eq. (74) in the case of coupling to the bath via a harmonic oscillator. This identification can be made by transforming
\[
\Omega_{\text{anh}} = \sqrt{\Omega^2 + \Delta},
\]
\[
\eta_{\text{anh}} = \frac{\eta\Omega^4}{(\Omega^2 + \Delta)^2}.
\]

After this transformation we have
\[
J_{\text{eff}}(\omega) = \frac{\eta\omega \Omega^4}{(\Omega^2 + \Delta - \omega^2)^2 + 4\gamma^2 \omega^2}
\]
\[
= \frac{\eta_{\text{anh}} \omega \Omega_{\text{anh}}^4}{(\Omega_{\text{anh}}^2 - \omega^2)^2 + 4\gamma^2 \omega^2}.
\]

The anharmonic part in the intermediate anharmonic oscillator has always the same sign as the harmonic part (\( \Omega \) and \( \zeta \) are both positive) and should make the coupling stronger. From the resulting inequality \( \Omega_{\text{anh}} \geq \Omega \) we know that the proportionality constant between \( \Delta \) and \( \zeta \) must be positive.
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