

**TECHNISCHE
UNIVERSITÄT
DRESDEN**

Stochastical Dynamics of Open Quantum Systems

Diplomarbeit
zur Erlangung des wissenschaftlichen Grades
Diplom-Physiker

vorgelegt von

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geboren am 18.10.1987 in Dresden

Institut für Theoretische Physik
der Technischen Universität Dresden
2013

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Zusammenfassung

Abstract

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1. Introduction

1.1. Classical Open Systems

1.2. Open Quantum Systems

1.3. Established Results and Subject of this Work

1.4. Outline

2. Non-Markovian Quantum State Diffusion

The description of open quantum systems in terms of diffusive stochastic differential equations has a long tradition [1]. At first, seen merely as a tool to unravel a given master equation of Lindblad type, it was realized later that they possess a strong microscopical foundation in terms of continuous measurements or memoryless quantum environments [2].

Its non-Markovian generalization, the non-Markovian quantum state diffusion (NMQSD), took a quite similar path, which we roughly follow in this chapter: Although first discovered as an unravelling for the Feynman-Vernon influence functional in terms of stochastic propagators [Str96], the corresponding non-Markovian stochastic Schrödinger equation (NMSSE) was derived based upon the standard open-system-model, which we **recall** in Sect. 2.1. Following the lines of Diósi, Strunz and Gisin [DS97, DGS98, SDG99] we derive both a linear and numerically superior non-linear version of the NMSSE in Sect. 2.2 and 2.3 respectively.

Section 2.4 is concerned with the question if our NMSSE has a physical interpretation or is just merely a computational tool. **Anschließend** we drop the requirement of zero initial temperature used in the **vorherig** sections. This chapter is **abgeschlossen** by the treatment of an analytically soluble two-level system at zero temperature.

Most of the material covered can be found in reference [Str01], which we follow loosely.

2.1. The Microscopical Model

It is the foremost goal of this work to obtain a dynamical equation for an open quantum system. Nevertheless we introduce a full model of system and its environment first, the bosonic and non-relativistic standard model of an open quantum system extensively studied for example in the book of Weiss [Wei99]. There are three reasons for such a microscopical approach: On one hand this serves the purpose to better understand the physical origin of macroscopical properties used to characterize the bath later on. But more important, starting with a closed quantum system is the only strategy allowing us to derive the NMSSE from first principles, namely the Schrödinger equation. A last argument in favor of the microscopic approach, PRODUCT STATES, ENTANGLEMENT, etc. —we will not dwell on this any further.

As a starting point we consider an environment consisting of a finite number N of uncoupled harmonic oscillators¹. A Generalization to an infinite number can be carried out formally along the same lines, replacing sums by infinite series or even integrals; a different approach within our framework is presented later. The dynamics of both system and environment are then described by a unitary time evolution with the Hamiltonian

$$H_{\text{tot}} = H \otimes \mathbf{I} + \mathbf{I} \otimes H_{\text{env}} + H_{\text{int}}, \quad (2.1)$$

where H and H_{env} are the free Hamiltonians of the system and the bath respectively.² The latter is a sum over independent harmonic oscillators $H_{\text{env}} = \sum_{\lambda} \omega_{\lambda} a_{\lambda}^{\dagger} a_{\lambda}$ expressed in bosonic ladder operators a_{λ} and a_{λ}^{\dagger} of the λ^{th} mode with frequency ω_{λ} . Treating a finite number of independent reservoirs poses no further difficulties and therefore is not elaborated in this section.

For the interaction between environment and system we confine ourselves to the case of linear coupling

$$H_{\text{int}} = \sum_{\lambda} g_{\lambda}^* L \otimes a_{\lambda}^{\dagger} + g_{\lambda} L^{\dagger} \otimes a_{\lambda}. \quad (2.2)$$

¹We use “environment”, “reservoir” and “bath” interchangeably, although the later two suggest a large size compared to the system.

²For some models like the damped harmonic oscillator [CL83] an additional renormalization term arises from the interaction. Nevertheless such a contribution is best attributed to H since it only acts on the system’s Hilbert space.

Here L denotes the coupling operator in the system's Hilbert space and $g_\lambda \in \mathbb{C}$ the coupling strength of the λ^{th} mode. Since in typical examples the coupling of an individual bath mode scales inversely with the environment size [Wei99], the linear coupling in (2.2) seems reasonable for macroscopic large environments. But our framework also incorporates small environments—even to the extreme of a single harmonic oscillator—with strong coupling as well. For such cases the linearity needs to be imposed as another assumption of the model.

Beside the Hamiltonian another important influence on the system's subdynamics is the initial state, specifically the initial entanglement between system and bath. Throughout this work we only consider product initial conditions, where the bath is in the vacuum state with respect to all a_λ

$$|\Psi_0\rangle = |\psi_0\rangle \bigotimes_{\lambda} |0_\lambda\rangle. \quad (2.3)$$

Such a choice is not as restrictive as it seems on first glance: In Sect. 2.5 we show how a thermal bath state can be mapped to (2.3). However whether the NMSSE is applicable to initially entangled states is a question of current research.

To absorb the free dynamics of the environment in time dependent creation and annihilation operators, we switch to the interaction picture with respect to H_{env} . Since the bath operators only obtain an additional phase $e^{\pm i\omega_\lambda t}$, the transformed Hamiltonian from Eq. (2.4) reads³

$$H_{\text{tot}}(t) = H \otimes \mathbf{I} + \sum_{\lambda} \left(g_\lambda^* e^{i\omega_\lambda t} L \otimes a_\lambda^\dagger + g_\lambda e^{-i\omega_\lambda t} L^\dagger \otimes a_\lambda \right). \quad (2.4)$$

Our choice of unentangled initial conditions with a vacuum bath state ensures that the reduced density operator remains unaffected under the change of time-evolution picture.

It is instructive to rewrite the last equation using the operator valued force

$$B(t) = \sum_{\lambda} g_\lambda a_\lambda e^{-i\omega_\lambda t}. \quad (2.5)$$

³We refrain from introducing another label to distinguish between time-evolution pictures—in what follows we always work in the interaction picture.

The total Hamiltonian then reads $H_{\text{tot}}(t) = H \otimes \mathbf{I} + L \otimes B(t)^\dagger + L^\dagger \otimes B(t)$. Already from this equation it can be seen, that the complete action of the environment on the system is encoded in the operators $B(t)$. An important—and within our model the only—characteristic of them is the correlation function $\alpha(t-s) = \langle (B(t) + B(t)^\dagger)(B(s) + B(s)^\dagger) \rangle_\rho$ for an arbitrary bath state ρ . For a thermal state at temperature T , the correlation function can be calculated analytically [FHS10]

$$\alpha_T(t-s) = \sum_\lambda |g_\lambda|^2 \left(\coth \frac{\omega_\lambda}{2T} \cos \omega_\lambda(t-s) - i \sin \omega_\lambda(t-s) \right). \quad (2.6)$$

Introducing the spectral density $J(\omega) = \sum_\lambda |g_\lambda|^2 \delta(\omega - \omega_\lambda)$ and taking the limit $T \rightarrow 0$, the above equation can be rephrased as

$$\alpha(t-s) = \langle B(t)B(s)^\dagger \rangle_0 = \int_0^\infty J(\omega) e^{-i\omega(t-s)} d\omega. \quad (2.7)$$

In other words, the correlation function is simply given as one-sided Fourier transform of the spectral density. Of course this connection between response function and power spectrum—and its general form for $T \neq 0$ —is well known as fluctuation-dissipation relation. Since a genuine physical spectral density is real, we require admissible correlation function to be hermitian $\alpha(-t) = \alpha(t)^*$.

2.2. Linear NMSSE

The linear non-Markovian stochastic Schrödinger equation derived in this section is an equivalent reformulation of the interaction-picture Schrödinger equation

$$\partial_t |\Psi_t\rangle = -iH_{\text{tot}}(t) |\Psi_t\rangle, \quad |\Psi_0\rangle = |\psi_0\rangle \otimes |0\rangle, \quad (2.8)$$

corresponding to the model of the last section: Expressing the bath degrees of freedom in the Bargmann Hilbert space of anti-holomorphic functions [Bar61] provides a representation that is well suited for a Monte-Carlo treatment. To this end we introduce the unnormalized coherent state $|z_\lambda\rangle = \exp(z_\lambda a_\lambda^\dagger) |0_\lambda\rangle$ for each mode with

resolution of the identity for the environment

$$\mathbf{I} = \int \frac{e^{-|\mathbf{z}|^2}}{\pi^N} |\mathbf{z}\rangle\langle\mathbf{z}| d^{2N}z, \quad (2.9)$$

Here we employ the shorthand notation $|\mathbf{z}\rangle = \bigotimes_{\lambda} |z_{\lambda}\rangle$ and the “volume” integration measure for N complex numbers $d^{2N}z = \prod_{\lambda} d\Re z_{\lambda} d\Im z_{\lambda}$. Throughout this work the finite bath is often replaced by a continuum of oscillators; therefore we simply write $\mu(dz) = \pi^{-N} \exp(-|\mathbf{z}|^2) d^{2N}z$ to drop an explicit reference to N .

Equation (2.9) allows us to express the full state in a time-independent environment basis

$$|\Psi_t\rangle = \int |\psi_t(\mathbf{z}^*)\rangle \otimes |\mathbf{z}\rangle \mu(dz).$$

For the following derivation it is crucial to notice that the Bargmann transform $\mathbf{z} \mapsto \psi_t(\mathbf{z}^*)$ is an anti-holomorphic function with values in the system’s Hilbert space \mathcal{H}_{sys} . Naturally it is equivalent to any other representation of the full state $|\Psi_t\rangle$. As the coherent states are not orthogonal, but rather satisfy $\langle \mathbf{w} | \mathbf{z} \rangle = \exp(\sum_{\lambda} w_{\lambda}^* z_{\lambda})$, the reduced density operator, obtained by tracing over the bath degrees of freedom, reads

$$\rho(t) = \text{Tr}_{\text{env}} |\Psi_t\rangle\langle\Psi_t| = \int |\psi_t(\mathbf{z}^*)\rangle\langle\psi_t(\mathbf{z}^*)| \mu(dz). \quad (2.10)$$

After fixing the kinematic structure, the next step is to rewrite the dynamical equation: The representation of the ladder operators follow from the usual rules $\langle \mathbf{z} | a_{\lambda}^{\dagger} = z_{\lambda}^* \langle \mathbf{z} |$ and $\langle \mathbf{z} | a_{\lambda} = \partial_{z_{\lambda}^*} \langle \mathbf{z} |$. These expressions applied to Eq. (2.8) give us the system-bath Schrödinger equation in the transformed space

$$\partial_t \psi_t(\mathbf{z}^*) = -iH\psi_t(\mathbf{z}^*) - iL \sum_{\lambda} g_{\lambda}^* e^{-i\omega_{\lambda}t} z_{\lambda}^* \psi_t(\mathbf{z}^*) - iL^{\dagger} \sum_{\lambda} g_{\lambda} e^{i\omega_{\lambda}t} \frac{\partial \psi_t}{\partial z_{\lambda}}(\mathbf{z}^*). \quad (2.11)$$

Introducing an effective driving process like in Eq. (2.5)

$$Z_t^*(\mathbf{z}^*) = -i \sum_{\lambda} g_{\lambda}^* e^{i\omega_{\lambda}t} z_{\lambda}^* \quad (2.12)$$

allows us to combine the effect of the first bath-interaction term into a single multiplication operator—or process for reasons explained in the next paragraph. A similar conversion works for the second term as well with the help of the functional chain

rule $\frac{\partial}{\partial z_\lambda^*} = \int \frac{\partial Z_s^*}{\partial z_\lambda^*} \frac{\delta}{\delta Z_s^*} ds$. Combined our new equation of motion—the non-Markovian stochastic Schrödinger equation—reads

$$\partial_t \psi_t = -iH\psi_t + LZ_t^* \psi_t - L^\dagger \int_0^t \alpha(t-s) \frac{\delta \psi_t}{\delta Z_s^*} ds. \quad (2.13)$$

As we shown in Sect. 2.4.1, the integral boundaries arise due to the initial conditions (2.3); a less formal argumentation goes as follows: By construction of our processes (2.12) an initial state $|\psi_0\rangle \otimes |0\rangle$ translates to an initial $\psi_0(Z^*)$ that is completely independent of any noise. Then causality implies that ψ_t can only depend on Z_s^* for $0 \leq s \leq t$.

Up to this point we have merely rewritten the original Schrödinger equation (2.8) to an equivalent form: The original system-bath product Hilbert space $\mathcal{H}_{\text{sys}} \otimes \mathcal{H}_{\text{env}}$ is replaced by a Hilbert space of \mathcal{H}_{sys} -valued functions. A different attitude is quite fruitful, especially with a numerical solution of the NMSSE in mind: Equation (2.27) can be rewritten as $\rho_t = \mathbb{E}(|\psi_t\rangle\langle\psi_t|)$, where \mathbb{E} denotes the average over $\mu(dz) = \pi^{-N} \exp(-|z|^2) d^{2N}z$. Put differently the reduced density matrix ρ_t arises by averaging over the stochastic pure state projectors $|\psi_t(z^*)\rangle\langle\psi_t(z^*)|$ with Gaussian weight $\mu(dz)$. Hence we regard Eq. (2.13) as a stochastic differential equation for individual realisations $\psi_t(z^*)$. We refer to the later either as system state relative to $|z\rangle$ or, in the spirit of the stochastic Schrödinger equations emerging from continuous measurement theory [Car93], as quantum trajectory.

In this approach the driving force Z_t^* is implemented as classical stochastic process defined by the concrete version (2.12) and the underlying probability measure μ . It is a complex Gaussian process uniquely characterized by its expectation value and covariances

$$\mathbb{E} Z_t = 0, \quad \mathbb{E} Z_t Z_s = 0, \quad \text{and} \quad \mathbb{E} Z_t Z_s^* = \alpha(t-s), \quad (2.14)$$

where α is the zero-temperature correlation function (2.7) for $J(\omega) = \sum_\lambda |g_\lambda|^2 \delta(\omega - \omega_\lambda)$. By virtue of the initial conditions, ψ_t depends on z only through the driving process; thus we can drop the coherent state labels and simply write $\psi_t(Z^*)$ denoting the trajectory corresponding to a realisation $Z^*(z^*)$. It is this alternative point of view that makes the NMSSE-approach so powerful: The entire influence of the environment is encoded in a complex function α , which acts both as correlation function for the

driving noise Z^* and as memory kernel for the damping term. A generalization to an arbitrary number of bath-oscillators is now straightforward: simply replacing the correlation function allows a unified description of arbitrary harmonic environments.

Except in the limit $\alpha(t) \sim \delta(t)$, elaborated in the next paragraph, the driving process Z_t^* is correlated for different times. This non-Markovian behavior, which makes a complete understanding of the dynamics highly desirable for application but also considerably harder, shows up in the equation of motion (2.13) as well. The damping term contains the functional derivative over the whole timespan and therefore takes the complete history of $\psi_t(Z^*)$ into account. In its own right the derivative is just as problematic: Since its computation requires not only the single realisation Z^* , but in some sense all adjacent ones as well, it seems questionable to regard the NMSSE (2.13) as a genuine stochastic differential equation [GW02]. Even from the purely pragmatic point of view both kinds of non-local behavior complicate a direct numerical simulation of the NMSSE, if not making it completely impracticable. Nevertheless there are two quite distinct solutions as shown in Sect. 2.2.2 and Chap. 3.

2.2.1. Markov Limit

The best understood open systems are Markovian. Based upon two physical assumptions, namely

weak coupling of the system to the reservoir and

memoryless environment, that is the time evolution is completely time-local,

it is possible to derive a general form of a master equation governing the reduced dynamics [Lin76]. Of course, the NMSSE is much more general. It is only in the standard Markovian limit $\alpha(t) = \gamma\delta(t)$ we can expect to obtain an equation of motion that describes a reduced time evolution without memory. A rescaling of the coupling operator L allows us to set $\gamma = 1$ without loss of generality.

The vacuum initial conditions $\frac{\delta\psi_0}{\delta Z_s^*} = 0$ with $s \in \mathbb{R}$ imply for an arbitrary bath correlation function

$$\frac{\delta\psi_t}{\delta Z_t^*} = \frac{1}{2} L\psi_t \quad (t > 0) \quad (2.15)$$

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as we show now. It is clear from its derivation that the NMSSE describes a unitary, time-dependent evolution. Therefore it can be solved formally using the Dyson series

$$\psi_t(Z^*) = \sum_{n=0}^{\infty} (-i)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{n-1}} dt_n H_{\text{tot}}(t_1) \dots H_{\text{tot}}(t_n) \psi_0, \quad (2.16)$$

where $H_{\text{tot}}(t)$ is the reformulation of (2.4) given by

$$-iH_{\text{tot}}(t) = -iH + LZ_t^* - L^\dagger \int_{-\infty}^{\infty} ds \alpha(t-s) \frac{\delta}{\delta Z_s^*}.$$

Throughout this work we often use the shorthand notation $\mathcal{D}_t = \int ds \alpha(t-s) \frac{\delta}{\delta Z_s^*}$ for the last term.

Applying the functional derivative $\frac{\delta}{\delta Z_s^*}$ to $H_{\text{tot}}(t)$ gives a single contribution $i\delta(t-s)L$, since both H and \mathcal{D}_t are independent of the noise. This allows us to calculate $\frac{\delta\psi_t}{\delta Z_t^*}$ order by order in Eq. (2.16)—the derivative of ψ_0 vanishes as imposed by the initial conditions. We obtain for the term with a n -fold time-integral neglecting a constant phase

$$\int_0^t dt_1 \dots \int_0^{t_{n-1}} dt_n \left(\delta(t_1 - t) L H_{\text{tot}}(t_2) \dots H_{\text{tot}}(t_n) + \dots + \delta(t_n - t) H_{\text{tot}}(t_1) \dots L \right).$$

We notice that the i^{th} summand contributes only if $t_i = t$: For $i = 1$ this is exactly the integral boundary while for $i = 2, \dots, n$ the integral boundary reaches t only for $t_1 = t$. As the latter condition has vanishing weight under the t_1 integral we eventually find Eq. (2.15).

Let us return to the Markov limit of our NMSSE. By virtue of the singular correlation function $\alpha = \delta$ the time-nonlocal damping operator reduces to a time-local form $\mathcal{D}_t = \frac{\delta}{\delta Z_t^*}$ as it is expected from a memoryless environment. Combined with Eq. (2.15) this leads to simple stochastic differential equation

$$\partial_t \psi_t(Z^*) = -iH\psi_t(Z^*) + LZ_t^* \psi_t(Z^*) - \frac{1}{2} L^\dagger L \psi_t(Z^*),$$

driven by a complex White Noise Z_t with $\mathbb{E}Z_t Z_s^* = \delta(t - s)$. In a formally exact fashion the equation above should be written as

$$d\psi_t = (-iH\psi_t - \frac{1}{2}L^\dagger L\psi_t) dt + L\psi_t d\xi_t^* \quad (2.17)$$

with a standard complex Brownian motion ξ_t . It is well known that such stochastic differential equations are problematic as ξ_t is not differentiable with respect to time. To define the solution ψ_t uniquely we need to specify an appropriate interpretation of the stochastic differential equation [Øks03, p. 36]: We imagine the Brownian motion as a limit of stochastic processes $\xi_t^{(n)} \rightarrow \xi_t$, such that $\xi_t^{(n)}$ are continuously differentiable with respect to time. Replacing the Brownian motion in Eq. (2.17) by $\xi_t^{(n)}$ transforms it into a deterministic differential equation. The limit of corresponding solutions $\psi_t^{(n)}$ coincides with ψ_t only if we understand Eq. (2.17) in the Stratonovich sense. However, in our case the Itô- and Stratonovich form agree since $\mathbb{E}Z_t Z_s = 0$ [G⁺85].

2.2.2. Convolutionless Formulation

As a cure for the non-locality issues, Diósi, Gisin, and Strunz [DGS98] proposed the powerful O -Operator substitution: It is based on the additional assumption, that one may replace the functional derivative by a system operator O , which only depends on the realisation of Z^* itself

$$\frac{\delta\psi_t(Z^*)}{\delta Z_s^*} = O(t, s, Z^*)\psi_t(Z^*). \quad (2.18)$$

Besides getting rid of the derivative, this substitution enables us to derive a convolutionless form of our NMSSE (2.13)

$$\partial_t \psi_t = -iH\psi_t(Z^*) + LZ_t^* \psi_t(Z^*) - L^\dagger \bar{O}(t, Z^*)\psi_t(Z^*) \quad (2.19)$$

with the time-local operator

$$\bar{O}(t, Z^*) := \int_0^t \alpha(t-s)O(t, s, Z^*) ds. \quad (2.20)$$

Conclusively Eq. (2.19) turns into a genuine stochastic differential equation for the trajectory $\psi_t(Z^*)$, but in the much smaller Hilbert space of the system. This makes it exceptionally well suited for dealing with infinite sized environments numerically, provided the \bar{O} -operator is known. Depending on the validity of the O -substitution the corresponding convolutionless NMSSE (2.19) might be as accurate as the original microscopic equation of motion (2.8).

For a few simple models—for example the dissipative two level system presented in ?? or its higher dimensional generalizations [JZYY12]—an exact analytic expression for O is known. In these rare cases one proceeds as follows [DGS98]: From the consistency condition

$$\partial_t \frac{\delta \psi_t(Z^*)}{\delta Z_s^*} = \frac{\delta}{\delta Z_s^*} \partial_t \psi_t(Z^*) \quad (2.21)$$

and the initial condition familiar from Sect. 2.2.1

$$O(s, s, Z^*) = L \quad (2.22)$$

we derive an equation of motion for $O(t, s, Z^*)$. It still contains the functional derivative, but is converted to a system of coupled, deterministic equations using a power series ansatz

$$O(t, s, Z^*) = \sum_{n=0}^{\infty} \int_0^t \dots \int_0^t O_n(t, s, \nu_1, \dots, \nu_n) d\nu_1 \dots \nu_n. \quad (2.23)$$

Nevertheless most treatments rely on approximation schemes, for example a perturbation expansion for small coupling parameter or almost-Markovian environments [YDGS99]. Also a closely related hierarchy of O -operators provides an efficient numerical algorithm similar in concept to the main result of this work [1].

2.2.3. Equivalent Master Equations

In the previous section we have introduced a convolutionless formulation primarily to simplify the treatment of the NMSSE. But the O -operator substitution is also essential clarify the connection to the master equations commonly used in the theory of open quantum systems. The latter are formulated in terms of reduced density operators, which we recover from the trajectories by averaging over the pure states projectors $P_t = |\psi_t(Z^*)\rangle\langle\psi_t(Z^*)|$. For certain systems this can be done analytically

in order to derive a equivalent master equation.

As a simple example we focus on models with a Z^* independent \bar{O} -operator such as the two-level system presented in ???. We follow the lines of Yu et al. [YDGS99, YDGS00], who also treat the general case using the functional expansion ???. The pure states projectors' equations of motion,

$$\partial_t P_t = -i[H, P_t] + Z_t^* L P_t - L^\dagger \bar{O}(t) P_t + Z_t P_t L^\dagger - P_t \bar{O}(t)^\dagger L, \quad (2.24)$$

yield a closed evolution equation for ρ_t after averaging over the bath degrees of freedom only if we can restate the terms containing Z_t^* in a noise-independent manner. This can be done with the help of Novikov's formula [Nov65]

$$\mathbb{E}(Z_t P_t) = \mathbb{E}\left(\int ds \alpha(t-s) \frac{\delta}{\delta Z_s^*} P_t\right). \quad (2.25)$$

A formal proof is provided in ??, but the main idea is simple: Under a Gaussian integral $\int d^2 z \exp(-|z|^2) \dots$ the multiplication by z can be rewritten as a derivation ∂_{z^*} . Partial integration yields a result similar to Eq. (2.25).

The right hand side of Novikov's formula is simplified further using the O -operator substitution. Since $|\psi_t\rangle$ is analytical in Z^* and accordingly $\langle\psi_t|$ analytical in Z_t , the derivative is further simplified to

$$\frac{\delta}{\delta Z_s^*} \left(|\psi_t(Z^*)\rangle \langle\psi_t(Z^*)| \right) = \left(\frac{\delta}{\delta Z_s^*} |\psi_t(Z^*)\rangle \right) \langle\psi_t(z)| = O(t, s) |\psi_t(Z^*)\rangle \langle\psi_t(Z^*)|.$$

Averaging over the equations of motion for the pure state projectors (2.24) finally gives the master equation for the reduced density matrix ρ_t

$$\partial_t \rho_t = -i[H, \rho_t] + [L, \rho_t \bar{O}(t)^\dagger] + [\bar{O}(t) \rho_t, L^\dagger]. \quad (2.26)$$

This expression closely resembles the well known Lindblad master equation ??? for Markovian open quantum systems, but involves time-dependent Lindbladians. As elaborated in Sect. 2.2.1 the \bar{O} -operator reduces to $\bar{O}(t) = \frac{\gamma}{2} L$ in the Markovian limit—thus our NMSSE produces the correct limit.

2.3. Nonlinear NMSSE

In the last section we emphasize that the non-Markovian stochastic Schrödinger equation (2.13) can be interpreted either as a system-environment Schrödinger equation or as a stochastic differential equation for the trajectories $\psi_t(Z^*)$. In the theory of Markovian systems the latter is often referred to as an unravelling of the corresponding master equation [?]. Either way in a numerical treatment the reduced density operator of the open system is determined by a Monte-Carlo evaluation of the partial trace (or stochastic average)

$$\rho_t = \mathbb{E}(|\psi_t\rangle\langle\psi_t|) = \int |\psi_t(\mathbf{z})\rangle\langle\psi_t(\mathbf{z})| \mu(d\mathbf{z}). \quad (2.27)$$

The fineness of such a scheme is drastically reduced if there are few highly peaked contributions [DS11]. As shown in a numerical investigation [?] the NMSSE shows exactly this behavior: for most trajectories the norm $\langle\psi_t(Z^*)|\psi_t(Z^*)\rangle$ goes to zero due to growing entanglement with the environment. To recover the unitarity of the closed time evolution $\mathbb{E}(\langle\psi_t|\psi_t\rangle) = \langle\Psi_t|\Psi_t\rangle = 1$ the few trajectories with significant contribution have to be taken into consideration. As we further elaborate in ?? this requires an insurmountable sample size for certain system parameters.

Seen purely as a stochastic tool to determine the reduced density operator the unravelling in Eq. (2.27) is not unique: we can perform any transformation under the integral which keeps its value fixed and obtain equally well relative states with a different measure. As it improves the behavior of the Monte-Carlo evaluation noticeably we perform a change of measure such that the average can be taken over normalized states. Such a procedure is well known from the theory of Markovian stochastic Schrödinger equation [?] and results in a nonlinear equation of motion—the same is true for our non-Markovian approach. Since the general case is treated later we focus on the convolutionless formulation of Eq. (2.19).

Of course it is trivial to rewrite Eq. (2.27) as an average over normalized states

$$\rho_t = \int \frac{d^{2N}z}{\pi^N} e^{-|z|^2} \langle\psi_t(\mathbf{z})|\psi_t(\mathbf{z})\rangle \frac{|\psi_t(\mathbf{z})\rangle\langle\psi_t(\mathbf{z})|}{\langle\psi_t(\mathbf{z})|\psi_t(\mathbf{z})\rangle},$$

now expressed with a time dependent density function. To highlight the physical

significance of the norm we notice that the latter is just the Q- or Husimi-function⁴ of the bath given by [Sch11]

$$Q_t(\mathbf{z}, \mathbf{z}^*) = \frac{e^{-|\mathbf{z}|^2}}{\pi^N} \langle \mathbf{z} | \text{Tr}_{\text{sys}} (|\Psi_t\rangle\langle\Psi_t|) | \mathbf{z} \rangle = \frac{e^{-|\mathbf{z}|^2}}{\pi^N} \langle \psi_t(\mathbf{z}) | \psi_t(\mathbf{z}) \rangle. \quad (2.28)$$

Expressed in terms of Q_t the reduced density operator reads

$$\rho_t = \int Q_t(\mathbf{z}, \mathbf{z}^*) \frac{|\psi_t(\mathbf{z})\rangle\langle\psi_t(\mathbf{z})|}{\langle\psi_t(\mathbf{z})|\psi_t(\mathbf{z})\rangle} d^{2N}\mathbf{z}. \quad (2.29)$$

Due to being non-negative and normalized to unity $\int Q(\mathbf{z}, \mathbf{z}^*) d\mathbf{z} = 1$ the Husimi-function can be regarded as the (quasi)-probability distribution on phase space of the bath degrees of freedom: Since a coherent state $|z\rangle$ resembles a wave packet localized around $z = (q + ip)/\sqrt{2}$, there is a well defined correspondence between coherent state labels z and the canonical variables (q, p) . Hence the norm of $\psi_t(\mathbf{z})$ simply determines the probability to find the bath oscillators in the coherent state $|\mathbf{z}\rangle$.

We can now incorporate the dynamics of the environment in a comoving coherent state basis for our trajectories $\psi_t(\mathbf{z})$. Making use of the microscopic Hamiltonian (2.11) and the analyticity $\partial_{z_\lambda} |\psi_t(\mathbf{z})\rangle$ gives the time evolution of the Husimi-function

$$\partial_t Q_z(\mathbf{z}, \mathbf{z}^*) = - \sum_{\lambda} \partial_{z_\lambda}^* (ig_\lambda e^{-i\omega_\lambda t} \langle L^\dagger \rangle_t Q_t(\mathbf{z}, \mathbf{z}^*)) - \text{c.c.} \quad (2.30)$$

It is obvious that the equation of motion above contains the full back-reaction of the system due to the quantum average⁵

$$\langle L^\dagger \rangle_t = \frac{\langle \psi_t(\mathbf{z}) | L^\dagger | \psi_t(\mathbf{z}) \rangle}{\langle \psi_t(\mathbf{z}) | \psi_t(\mathbf{z}) \rangle}.$$

Remarkably Eq. (2.30) can be solved with the method of characteristics since it has exactly the form of a (complex) Liouville equation. The corresponding characteristic

⁴We point out that the Husimi function is usually defined in terms of normalized coherent states. Hence the additional factor $\exp(-|z|^2)$ for each oscillator in our notation.

⁵We do not indicate its (non-holomorphic) dependence on \mathbf{z}^* explicitly because our main goal is not the solution of ???. Instead Q_t is only used to derive normalized versions of our NMSSE-trajectories.

curves are described by

$$\dot{z}_\lambda^*(t) = ig_\lambda e^{-i\omega_\lambda t} \langle L^\dagger \rangle_t. \quad (2.31)$$

We denote the corresponding flow by ϕ_t ; hence by the usual abuse of notation $z_\lambda^*(t) = \phi_{\lambda,t}^*(z_\lambda^*)$ with initial conditions $z_\lambda^*(0) = \phi_{\lambda,0}^*(z_\lambda^*) = z_\lambda^*$. Equation (2.31) tells us that if the full state at time t is $|\psi_t(\mathbf{z})\rangle \otimes |\mathbf{z}\rangle$ and therefore the Husimi-function is localised around $\mathbf{z} = (\mathbf{q} + i\mathbf{p})/\sqrt{2}$ then the dominant contribution at $t + \Delta t$ comes from the coherent state $|\mathbf{z} + \dot{\mathbf{z}}\Delta t\rangle$. For this reason we should use system states relative to $|\mathbf{z}(t)\rangle$ instead in order to avoid vanishing contributions of single trajectories to ρ_t .

By construction the flow ϕ_t also gives as a solution to Eq. (2.30) for the Husimi-function

$$Q_t(\mathbf{z}, \mathbf{z}^*) = \int Q_0(\mathbf{z}_0, \mathbf{z}_0^*) \delta(\mathbf{z} - \phi_t(\mathbf{z}_0)) d^{2N} z_0$$

where $\delta(\mathbf{z} - \mathbf{z}') = \prod_\lambda \delta(\Re(z_\lambda - z'_\lambda)) \delta(\Im(z_\lambda - z'_\lambda))$. Since at the beginning our total state is given by the product $|\Psi_t\rangle = |\psi_0\rangle \otimes |\mathbf{z}\rangle$ the initial condition for the Husimi-function reads $Q_0(\mathbf{z}, \mathbf{z}^*) = \pi^{-N} e^{-|\mathbf{z}|^2}$ as seen from Eq. (2.28). With $\psi'_t = \psi_t \circ \phi_t$ we can rewrite Eq. (2.27) for the reduced density matrix as⁶

$$\rho_t = \int \frac{d^{2N} z}{\pi^N} e^{-|\mathbf{z}|^2} \frac{|\psi_t(\phi_t^*(\mathbf{z}^*))\rangle \langle \psi_t(\phi_t^*(\mathbf{z}^*))|}{\langle \psi_t(\phi_t^*(\mathbf{z}^*)) | \psi_t(\phi_t^*(\mathbf{z}^*)) \rangle} = \mathbb{E} \left(\frac{|\tilde{\psi}_t\rangle \langle \tilde{\psi}_t|}{\langle \tilde{\psi}_t | \tilde{\psi}_t \rangle} \right). \quad (2.32)$$

By its definition $\tilde{\psi}_t(\mathbf{z}^*)$ is just the relative state of $|\Psi_t\rangle$ belonging to the coherent state $|\phi_t(\mathbf{z}^*)\rangle$. Put differently these are exactly the expansion coefficients of the full system-bath pure state in the comoving environmental basis. It is quite remarkable that a closed equation of motion for the $\tilde{\psi}_t$ can be derived starting with

$$\partial_t(\psi_t \circ \phi_t^*) = \partial_t \psi_t \circ \phi_t^* + \sum_\lambda (\partial_{z_\lambda^*} \psi_t \circ \phi_t^*) \cdot (\partial_t \phi_{t,\lambda}^*). \quad (2.33)$$

For the first term we can use the evolution equation (2.19) of the fixed-basis relative states ψ_t with the O -operator substitution in place. Replacing the coherent state labels by their comoving counterparts leads us to a shifted process: The integral form

⁶Instead of directly introducing the normalized states $\tilde{\psi}_t$ as done by Strunz [Str01] we explicitly define the interim state ψ'_t to compare with the corresponding result for the hierarchy in Sect. 3.2.4.

of Eq. (2.31)

$$\phi_{t,\lambda}^*(z_\lambda^*) = z_\lambda^* + ig_\lambda \int_0^t \exp(-i\omega_\lambda s) \langle L^\dagger \rangle_s ds \quad (2.34)$$

plugged into the microscopic version of the process (2.12) yields the shifted stochastic driving as

$$\tilde{Z}_t^*(z^*) := Z_t^*(\phi_t^*(z^*)) = Z_t^*(z^*) + \int_0^t \alpha(t-s)^* \langle L^\dagger \rangle_s ds. \quad (2.35)$$

Since the O -operator substitution ensures that the equations of motion for ψ_t are local with respect to Z^* , the comoving dynamics just amount to replacing Z_t^* by \tilde{Z}_t^* in the first addend of (2.33).

The second addend, due to the intrinsic time dependence of the shifted coherent states, is treated on the same footing: It is just the well-known functional derivative term from the NMSSE as Eq. (2.31) reveals:

$$\begin{aligned} \sum_\lambda \frac{\partial \phi_{t,\lambda}^*}{\partial t}(z_\lambda^*) \cdot \frac{\partial \psi_t}{\partial z_\lambda^*}(\phi_t^*(z^*)) &= i \sum_\lambda g_\lambda e^{-i\omega_\lambda t} \langle L^\dagger \rangle_t \frac{\partial \psi_t}{\partial z_\lambda^*}(\phi_t^*(z^*)) \\ &= \langle L^\dagger \rangle_t \int_0^t \alpha(t-s) \frac{\delta \psi_t}{\delta Z_s^*}(\phi_t^*(z^*)) ds \\ &= \langle L^\dagger \rangle_t \bar{O}(t, \tilde{Z}^*) \tilde{\psi}_t(\tilde{Z}^*), \end{aligned}$$

where the last line reflects the definition of the \bar{O} -operator in Eqs. (2.18) and (2.20). Both terms of (2.33) combined yield the desired closed equation for $\tilde{\psi}_t$

$$\partial_t \tilde{\psi}_t = -iH \tilde{\psi}_t + L \tilde{Z}_t^* \tilde{\psi}_t - (L^\dagger - \langle L^\dagger \rangle_t) \bar{O}(t, \tilde{Z}^*) \tilde{\psi}_t. \quad (2.36)$$

We want to recall that $\tilde{\psi}_t$ was introduced to allow averaging over normalized states in Eq. (2.32). It does not imply $\tilde{\psi}_t(\tilde{Z}^*)$ being normalized for all times, which would be favourable for interpreting the NMSSE as an stochastic equation for genuine pure system states. It is quite remarkable that an extended version of Eq. (2.36) exists that even preserves normalization of single realizations: By considering the trajectories $|\psi'_t\rangle = |\tilde{\psi}_t\rangle / \sqrt{\langle \tilde{\psi}_t | \tilde{\psi}_t \rangle}$ it is straightforward to derive the corresponding equation of

motion [?]

$$\begin{aligned} \partial_t \psi'_t = & -iH\psi'_t + (L - \langle L \rangle_t) \tilde{Z}_t^* \psi'_t \\ & - \left((L^\dagger - \langle L^\dagger \rangle_t) \bar{O}(t, \tilde{Z}^*) - \langle (L^\dagger - \langle L^\dagger \rangle_t) \bar{O}(t, \tilde{Z}^*) \rangle \right) \psi'_t \end{aligned} \quad (2.37)$$

As mentioned in the motivation the nonlinear equations should be given precedence over the linear version when it comes to Monte-Carlo simulation. They allow us to compute the density matrix as an average over realizations with same order of magnitude while restoring the reference measure (in the microscopic model) to the well known, time-independent Gaussian weight (2.9). Therefore generating realizations of the shifted processes \tilde{Z}_t^* is within the scope of general methods, as long as the expectation value $\langle L^\dagger \rangle_s$ for all times $0 \leq s \leq t$ is known. This is the only contribution to the nonlinear NMSSE which is explicitly time-nonlocal; but since it involves only the average value there is no storage problem for numerical application. Of course these comments neglect the question how to obtain the \bar{O} -operator, which contains all non-Markovian feedback of the environment. For to application to any realistic physical system—except a few exactly solvable ones—this is actually the critical part in the implementation.

2.4. Interpretation of NMSSE

In Sect. 2.2 we propose two different schemes how to interpret our linear non-Markovian stochastic Schrödinger equation (2.13) from a formal point of view: On one hand we treat it like a simple reformulation of the microscopic Schrödinger equation (2.11). Alternatively the convolutionless formulation (2.19) resembles more closely the common stochastic differential equations.

In the Markovian regime the latter perspective is often favoured. . .

It remained an open question for a long time whether the non-Markovian stochastic Schrödinger equation allows

2.4.1. Linear NMSSE as Schrödinger Equation

2.4.2. A Time-Nonlocal Picture

2.5. Finite Temperature Theory

Until now we were only concerned with the temperature zero theory, which was defined by an initial product state with the environment in the vacuum state $|\Psi_0\rangle = |\psi_0\rangle \otimes |0\rangle$. It translates into our NMSSE-framework as the demand of vanishing functional derivatives at the time $t = 0$

$$\frac{\delta\psi_0}{\delta Z_s^*} = 0 \quad (s \in \mathbb{R}).$$

This allows us to restrict the integral domain for the derivatives in Eq. (2.13) and is therefore crucial for the O -operator substitution. Without the upper limit t for the integral the hierarchical equations of motion presented in Sect. 3.2 fail as well. In order to treat non-zero temperature systems with the non-Markovian stochastic Schrödinger equation we devise two methods that map to the vacuum initial conditions of the zero-temperature case.

To start off we assume a product initial state, but this time with a Gibbs state $\rho(\beta) = \frac{e^{-\beta H_{\text{env}}}}{Z}$ on the environment's side

$$\rho_0 = |\psi_0\rangle\langle\psi_0| \otimes \rho(\beta) \tag{2.38}$$

with the bath partition function $Z = \text{Tr}_{\text{exp}}[-\beta H_{\text{env}}]$ at inverse temperature $\beta = 1/k_B T$. This choice amounts to the following experimental setting: At $t_{\text{init}} \rightarrow -\infty$ the environment is brought into contact with an even larger heat bath at given temperature, while the coupling to the system is switched off. The environment is allowed to thermalize until $t = 0$ when the super-bath is removed and the system-coupling is instantly tuned as given by Eq. (2.2). The choice of a pure state projector for the system in Eq. (2.38) is merely for convenience; in contrast we cannot drop its product form. Therefore a initial thermal state of the system and bath with respect to H_{tot} cannot be treated with the NMSSE due to entanglement.

2.5.1. Thermo Field Method

Thermo field dynamics was introduced as a real-time approach to quantum fields at finite temperature [?]. It is favored over other methods in application to the NMSSE since it does not change the equation of motion [DGS98] as shown below. In the course of this section we follow the slightly more detailed accounts of Yu and Strunz [Yu04, Str01].

The main idea is to introduce a second fictitious bath of oscillators, which is independent from the physical environment and does not interact with the system. Expressing its degrees of freedom in ladder operators b_λ and b_λ^\dagger gives us the new Hamiltonian in the Schrödinger picture

$$H_{\text{tot}} = H \otimes \mathbf{I} + \sum_{\lambda} (g_{\lambda}^* L \otimes a_{\lambda}^{\dagger} + g_{\lambda} L^{\dagger} \otimes a_{\lambda}) + \mathbf{I} \otimes \sum_{\lambda} \omega_{\lambda} (a_{\lambda}^{\dagger} a_{\lambda} - b_{\lambda}^{\dagger} b_{\lambda}). \quad (2.39)$$

Although this Hamiltonian is not bounded from below due to negative frequencies of the fictitious oscillators, there are no stability problems since they do not interact with the physical degrees of freedom. For the same reason the reduced dynamics obtained from Eq. (2.39) are identical to the original microscopical model (2.4). Therefore both yield equal reduced density matrices for our system provided we choose an initial state that reproduces Eq. (2.38) upon tracing of the unphysical degrees of freedom. Since these are given by a product the choice of a total initial state is independent from the system and is equivalent to demand

$$\text{Tr}_{\text{b}} \tilde{\rho} = \rho(\beta) \quad (2.40)$$

for the density matrix of both environments. Here Tr_{b} denotes the partial trace with respect to the fictitious degrees of freedom.

Remarkably a solution $\tilde{\rho}$ of Eq. (2.40) is given by the pure state projector on a vacuum state with respect to new annihilation operators A, B . They are connected to the old ladder operators by a temperature dependent Bogoliubov transformation

$$\begin{aligned} A_{\lambda} &= \sqrt{\bar{n}_{\lambda} + 1} a_{\lambda} + \sqrt{\bar{n}_{\lambda}} b_{\lambda}^{\dagger} \\ B_{\lambda} &= \sqrt{\bar{n}_{\lambda}} a_{\lambda}^{\dagger} + \sqrt{\bar{n}_{\lambda} + 1} b_{\lambda}, \end{aligned}$$

with $\bar{n}_{\lambda} = (\exp(\beta\omega_{\lambda}) - 1)^{-1}$ denoting the mean thermal occupation number of the

(physical) oscillator mode λ . An extensive but elementary calculation reveals that $|0_{AB}\rangle\langle 0_{AB}|$ with $|0_{AB}\rangle = |0_A\rangle \otimes |0_B\rangle$ defined by $A_\lambda|0_{AB}\rangle = B_\lambda|0_{AB}\rangle = 0$ satisfies Eq. (2.40).

The doubling in degrees of freedom ensures that the reduced density matrix obtained from an initial pure state $|\tilde{\Psi}_0\rangle = |\psi_0\rangle \otimes |0_{AB}\rangle$ in the enlarged Hilbert space coincides with the original one lacking unphysical bath oscillators. Expressed in these new coordinates the total Hamiltonian (2.39) reads

$$\begin{aligned} H_{\text{tot}} = & H \otimes \mathbf{I} + \sum_{\lambda} \sqrt{\bar{n}_{\lambda} + 1} \left(g_{\lambda}^* L \otimes A_{\lambda}^{\dagger} + g_{\lambda} L^{\dagger} \otimes A_{\lambda} \right) \\ & + \sum_{\lambda} \sqrt{\bar{n}_{\lambda}} \left(g_{\lambda} L^{\dagger} \otimes B_{\lambda}^{\dagger} + g_{\lambda}^* L \otimes B_{\lambda} \right) \\ & + \mathbf{I} \otimes \sum_{\lambda} \omega_{\lambda} \left(A_{\lambda}^{\dagger} A_{\lambda} - B_{\lambda}^{\dagger} B_{\lambda} \right). \end{aligned} \quad (2.41)$$

Our new Hamiltonian is identical to the zero-temperature model except for the system coupling to two separate oscillator baths instead of one; therefore we need two independent processes Z_t^* and W_t^* for a stochastic version of Eq. (2.41) in general:

$$\begin{aligned} \partial_t \psi_t = & -iH\psi_t + LZ_t^* \psi_t - L^{\dagger} \int_0^t \alpha_1(t-s) \frac{\delta \psi_t}{\delta Z_s^*} ds \\ & + L^{\dagger} W_t^* \psi_t - L \int_0^t \alpha_2(t-s) \frac{\delta \psi_t}{\delta W_s^*} ds. \end{aligned} \quad (2.42)$$

All effects of the original thermal initial state are now encoded in the correlation functions

$$\alpha_1(t) = \sum_{\lambda} (\bar{n}_{\lambda} + 1) |g_{\lambda}|^2 e^{-i\omega_{\lambda} t} \quad \text{and} \quad \alpha_2(t) = \sum_{\lambda} \bar{n}_{\lambda} |g_{\lambda}|^2 e^{i\omega_{\lambda} t}$$

for Z_t^* and W_t^* respectively. Since both are Gaussian, their independence is equivalent to the vanishing of all their mutual correlations $\mathbb{E}(Z_t^* W_s) = \mathbb{E}(Z_t W_s) = 0$.

As we doubled the bath degrees of freedom merely to cope with a thermal initial state it is quite natural that the zero-temperature result from ?? with a single driving process is recovered in the limit $T \rightarrow 0$.

The thermo field approach to our non-Markovian Schrödinger equation turns out to be especially simple in the case of self-adjoint coupling operators L^{\dagger} : From Eq. (2.42)

we see how both driving processes Z_t^* and W_t^* can be combined into a single one, which we will denote by Z_t^* again. Since we took them to be mutually independent we find for our new sum process using $2\bar{n}_\lambda + 1 = \coth \frac{\beta\omega_\lambda}{2}$:

$$\mathbb{E}(Z_t Z_s^*) = \sum_\lambda \left(|g_\lambda|^2 \coth \frac{\beta\omega_\lambda}{2} \cos \omega_\lambda(t-s) - i \sin \omega_\lambda(t-s) \right). \quad (2.43)$$

Consequently the finite temperature NMSSE takes the form identical to zero temperature theory. They only differ in how the correlation function is obtained from a microscopical model or rather from a spectral density. It is not surprising that our combined correlation function (2.43) agrees with the result of Feynman and Vernon [FV63] derived in the path integral formalism for quantum Brownian motion. But our approach is much more general since it can tackle any kind of open quantum system with linear coupling.

2.5.2. unitary noise

As shown in the last section we can treat classical thermal noise on the same footing as quantum noise under certain circumstances just by using a modified correlation function (2.43). It is worth noticing how the influence of thermal fluctuations modify only the real part of α , a feature that explicitly distinguishes noisy classical perturbations [FHS10]. Therefore it is quite instructive to present a different method for treating non-zero temperature within the non-Markovian quantum state diffusion.

We start off by expanding the thermal bath state in a coherent state basis [WM08]

$$\rho(\beta) = FILLIN$$

which is quite reminiscent of the expansion for a pure state projector that lead to our stochastic Schrödinger equation. The corresponding pure initial states are a product involving all environmental oscillators

$$|\Psi_0(\xi)\rangle = e^{-\frac{|\xi|^2}{2}} |\psi_0\rangle \bigotimes_{\lambda} |\xi_\lambda\rangle$$

where the additional prefactor is usually absorbed by using normalized coherent states. A simple shift for the creation and annihilation operators $A_\lambda^\dagger = a_\lambda^\dagger - \xi_\lambda^*$

and $A_\lambda = a_\lambda - \xi_\lambda$ respectively maps the environmental part of the initial state above onto the vacuum. Therefore we can apply our zero-temperature derivation to the total Hamiltonian expressed in A and A_λ^\dagger . The resulting NMSSE reads

$$\partial_t \psi_t(Z^*, \xi) = \left(-iH + L\xi_t^* + L^\dagger \xi_t + LZ_t^* - L^\dagger \int_0^t \alpha(t-s) \frac{\delta}{\delta Z_s^*} ds \right) \psi_t(Z^*, \xi, \xi^*) \quad (2.44)$$

with a classical driving process $\xi_t = \sum_\lambda g_\lambda \xi_\lambda e^{-i\omega_\lambda t}$ and its familiar quantum counterpart Z_t^* . The former's properties are once again fixed by its correlations

$$\mathbb{E} \xi_t = 0, \quad \mathbb{E} \xi_t \xi_s = 0, \quad \text{and} \quad \mathbb{E} \xi_t \xi_s^* = 2 \sum_\lambda \bar{n}_\lambda |g_\lambda|^2 \cos \omega_\lambda(t-s).$$

Recovering the reduced density matrix not only requires an average over Z^* but also over all realizations of the thermal noise process ξ_t . Since all thermal occupation numbers \bar{n}_λ tend to zero for $T \rightarrow 0$, we obtain the zero temperature limit simply by setting $\xi_t = 0$. This amounts to the trivial decomposition $\rho(T=0) = |\mathbf{0}\rangle\langle\mathbf{0}|$ of the zero temperature environmental state.

2.6. Dissipative Two-level System

$$\partial_t \psi_t = -i\frac{\omega}{2} \sigma_z \psi_t + c\sigma_- Z_t^* \psi_t - c\sigma_+ \int_0^t \alpha(t-s) \frac{\delta \psi_t}{\delta Z_s^*} ds \quad (2.45)$$

2.6.1. O-Operator Method

As elaborated in Sect. 2.2.2 we can simplify the NMSSE (2.45) by replacing the functional derivative with an operator $O(t, s, Z^*)$. We try to solve the consistency condition (2.21) by a noise-independent ansatz

$$O(t, s) = cf(t, s)\sigma_-, \quad (2.46)$$

hence all non-Markovian feedback from the environment is now encoded in the function $f(t, s)$ to be determined. Plugging this ansatz into the evolution equation for O yields

$$\partial_t cf(t, s)\sigma_- = \left[-i\frac{\omega}{2} \sigma_z - c^2 F(t) \sigma_+ \sigma_-, cf(t, s)\sigma_- \right] \quad (2.47)$$

with a shorthand notation $F(t) := \int_0^t \alpha(t-s)f(t,s) ds$. From its definition (2.20) we see that F is also the prefactor for the integrated operator $\bar{O}(t) = cF(t)\sigma_-$. Since the operator algebra in Eq. (2.47) closes, our ansatz solves the equation of motion for O provided f evolves according to

$$\partial_t f(t,s) = (i\omega + c^2 F(t)) f(t,s) \quad (s \leq t).$$

Appropriate initial conditions follow trivially from Eq. (2.22); they read $f(s,s) = 1$. In the special case of an exponential bath correlation function $\alpha(t) = g e^{-\gamma|t| - i\Omega t}$ we can also derive a differential equation that is closed in F

$$\partial_t F(t) = g + (i(\omega - \Omega) - \gamma)F(t) + c^2 F(t)^2.$$

Such correlation functions play a major role in the subsequent work.

As shown in Sect. 2.2.3, a convolutionless NMSEE with noise independent O -operator can be transformed into a master equation without any approximation. For the model under consideration it turns out to closely resemble a Lindblad-type equation except for time-dependent coefficients

$$\partial_t \rho_t = -i\frac{\omega}{2}[\sigma_z, \rho_t] + c^2[\sigma_-, \rho_t \sigma_+] + c^2 F(t)[\sigma_- \rho_t, \sigma_+].$$

2.6.2. Noise-Expansion Method

In this section we propose a different method for solving Eq. (2.45): It is based on the expansion discussed in ??, which allows us to express the quantum trajectories $\psi_t(Z^*)$ in a functional Taylor series with respect to the noise process. Due to the particular coupling structure of the model we can neglect all terms higher than linear order in Z_t^* . As further elaborated in Sect. A.1, our NMSSE (2.45) reduces to a \mathbb{C} -valued integro-differential equation

$$\dot{\psi}^+(t) = -i\frac{\omega}{2}\psi^+(t) - c^2 \int_0^t \alpha(t-s)e^{i\frac{\omega}{2}(t-s)}\psi^+(s) ds, \quad (2.48)$$

which is nevertheless quite involved—even from a numerical point of view. The situation is noticeably simpler for an exponential correlation function for which we calculate an analytic solution in the appendix as well.

But even without an explicit solution for $\psi^+(t)$ we may still discover some illuminating consequences concerning the O -operator from the last section. With $\psi^-(t) = \psi^-(0) \exp(i\omega t/2)$ the full quantum trajectory reads

$$\psi_t(Z^*) = \begin{pmatrix} \psi^+(t) \\ \psi^-(t) \end{pmatrix} + c \int_0^t \begin{pmatrix} 0 \\ e^{i\frac{\omega}{2}(t-s)} \psi^+(s) \end{pmatrix} Z_s^* ds. \quad (2.49)$$

This allows us to calculate the functional derivative with respect to the driving process explicitly; for $0 \leq s \leq t$ we find⁷

$$\frac{\delta \psi_t(Z^*)}{\delta Z_s^*} = c \begin{pmatrix} 0 \\ e^{i\frac{\omega}{2}(t-s)} \psi^+(s) \end{pmatrix}$$

which agrees with our ansatz (2.46) in case we choose

$$f(t, s) = \frac{\psi^+(s)}{\psi^+(t)} e^{-i\frac{\omega}{2}(s-t)}. \quad (2.50)$$

A similar structure for the O -operator has been obtained by Strunz [Str01] within a Heisenberg-operator method.

⁷For $s = t$ there is no additional prefactor $\frac{1}{2}$ from integrating a δ -function localized at the upper integral boundary as explained in the footnote on page 53.

3. Numerical treatment

3.1. Hierarchical Equations of Motion

3.2. Stochastic Hierarchical Equations of Motion

In this section we present one of the main results of this work, namely a numerical method to solve the non-Markovian stochastic Schrödinger equation

$$\partial\psi_t = -ih\psi_t + LZ_t^*\psi_t - L^\dagger \int_0^t \alpha(t-s) \frac{\delta\psi_t}{\delta Z_s^*} ds \quad (3.1)$$

without making use of the O -operator substitution. Therefore we need to conceive a different way to deal with the nonlocality of the functional derivative with respect to time and the noise-process, as it prevents us from employing the common techniques for dealing with stochastic Schrödinger equations in the Markovian regime [?].

It turns out that for certain correlation functions the linear NMSSE (3.1) is formally equivalent to an infinite hierarchy of completely local stochastic differential equations. Although we borrow the main idea from the hierarchical equations of motion presented in the last section, we need to deal with some peculiarities of our NMSSE first.

3.2.1. Time derivation

The basic idea is to absorb the action of the functional derivative on $\psi_t(Z^*)$ into an auxiliary stochastic pure state

$$\psi_t^{(1)}(Z^*) := \int_0^t \alpha(t-s) \frac{\delta\psi_t(Z^*)}{\delta Z_s^*} ds. \quad (3.2)$$

We recall that the integral boundaries arise only after we apply the derivative on states $\psi_t(Z^*)$ that satisfy the additional condition $\delta\psi_t(Z^*)/\delta Z_s^* = 0$ for $s < 0$ and

3. Numerical treatment

$s > t$. Therefore we may write Eq. (3.2) more concise as

$$\psi_t^{(1)}(Z^*) = \left(\int \alpha(t-s) \frac{\delta}{\delta Z_s^*} ds \right) \psi_t(Z^*) =: \mathcal{D}_t \psi_t(Z^*)$$

with the integrated functional derivation operator \mathcal{D}_t .

The first step toward deriving an equation of motion for $\psi_t^{(1)}(Z^*)$ is answering the question under which conditions a simple expression for $\dot{\mathcal{D}}_t$ exists. Since the latter is given by $\dot{\mathcal{D}}_t = \int \dot{\alpha}(t-s) \delta / \delta Z_s^* ds$ we assume an exponential bath correlation function

$$\alpha(t) = g e^{-\gamma|t| - i\Omega t} = g e^{-i\Omega t} (\Theta(t)e^{-\gamma t} + \Theta(-t)e^{\gamma t}). \quad (3.3)$$

with the Heaviside function Θ . As the singular terms in the time derivative cancel we obtain

$$\dot{\alpha}(t) = g e^{-i\Omega t} ((-\gamma - i\Omega)\Theta(t)e^{-\gamma t} + (\gamma - i\Omega)\Theta(-t)e^{\gamma t}).$$

Therefore we cannot exploit $\dot{\mathcal{D}}_t \sim \mathcal{D}_t$ on the level of operators in general. Even the vanishing of the functional derivative $\delta\phi_t(Z^*)/\delta Z_s^*$ for $s > t$ with some arbitrary stochastic state $\phi_t(Z^*)$ does not imply $\dot{\mathcal{D}}_t\phi_t(Z^*) \sim \mathcal{D}_t\phi_t(Z^*)$ as the following example shows: Take $\phi_t(Z^*) = \varphi \cdot (Z_t^* + Z_{t'}^*)$ for some noise independent system state φ and $0 < t' < t$. It clearly satisfies the required boundary conditions, but

$$\dot{\mathcal{D}}_t\phi_t(Z^*) = (\dot{\alpha}(0) + \dot{\alpha}(t-t'))\varphi = \left(-2i\Omega - (\gamma + i\Omega)e^{-(\gamma+i\Omega)(t-t')} \right) \varphi$$

which is not proportional to

$$\mathcal{D}_t\phi_t(Z^*) = (\alpha(0) + \alpha(t-t'))\varphi = g(1 + \exp(-\gamma + i\Omega)(t-t'))\varphi.$$

We see that the problematic first summand arises due to singular behavior of $\delta\phi_t(Z^*)/\delta Z_s^*$ for $s = t$.

Such problems do not occur once we restrict $\dot{\mathcal{D}}_t$ to solutions of our NMSSE (3.1) with vacuum initial conditions. Indeed we see from ?? that the functional derivative of $\psi_t(Z^*)$ at the upper boundary is regular and therefore has vanishing weight under the integral. Hence we obtain for the time derivative of our integrated derivation operator

$$\dot{\mathcal{D}}_t\psi_t(Z^*) = -(\gamma + i\Omega)\mathcal{D}_t\psi_t(Z^*) \quad (3.4)$$

In the remaining work we use the shorthand notation $w = \gamma + i\Omega$.

3.2.2. Linear Hierarchy

We now return to our non-Markovian stochastic Schrödinger equation; with the auxiliary stochastic state (3.2) it can be written as

$$\partial_t \psi_t(Z^*) = -iH\psi_t(Z^*) + LZ_t^* \psi_t(Z^*) - L^\dagger \psi_t^{(1)}(Z^*).$$

Hence if we can derive a tractable equation of motion for $\psi_t^{(1)}(Z^*)$ we reduce the problems with a functional derivative to propagating a coupled system of simpler equations. With the result from the last section Eq. (3.4) and the original NMSSE (3.1) we find

$$\begin{aligned} \partial_t(\mathcal{D}_t \psi_t) &= -w\mathcal{D}_t \psi_t + \mathcal{D}_t(-iH + LZ_t^* - L^\dagger \mathcal{D}_t) \psi_t \\ &= (-iH - w + LZ_t^*) \psi_t^{(1)} + [\mathcal{D}_t, Z_t^*] L \psi_t - L^\dagger \mathcal{D}_t \psi_t^{(1)}, \end{aligned} \quad (3.5)$$

where we use that \mathcal{D}_t commutes with all system operators. It is not surprising that the functional derivative reappears in the equation for $\psi_t^{(1)}$; therefore we need to introduce another auxiliary state. This scheme leads to an infinite hierarchy of auxiliary states defined by

$$\psi_t^{(k)} := \mathcal{D}_t \psi_t^{(k-1)} = \mathcal{D}_t^k \psi_t. \quad (3.6)$$

Expressed in the new auxiliary states and with $[\mathcal{D}_t, Z_s^*] = \alpha(t-s)$ Eq. (3.5) reads

$$\partial_t \psi_t^{(1)} = (-iH - w + LZ_t^*) \psi_t^{(1)} + \alpha(0) L \psi_t^{(0)} - L^\dagger \psi_t^{(2)}.$$

Along these lines it is straightforward to derive the full hierarchy of equations of motions for all $\psi_t^{(k)}$. Since the commutator $[\mathcal{D}_t, Z_s^*]$ is a \mathbb{C} -number each auxiliary state only couples to the order directly above and below

$$\partial_t \psi_t^{(k)} = (-iH - kw + LZ_t^*) \psi_t^{(k)} + k\alpha(0) \psi_t^{(k-1)} - L^\dagger \psi_t^{(k+1)}. \quad (3.7)$$

The vacuum initial condition for the true quantum trajectory $\delta\psi_0/\delta Z_s^* = 0$ requires that all auxiliary states vanish at $t = 0$.

Of course the infinite hierarchy is even more intricate to solve than the original non-

3. Numerical treatment

Markovian stochastic Schrödinger equation; therefore we need to truncate at some finite order. It is quite remarkable that this can be done in a self-consistent manner, which even approximately incorporates all truncated orders into the remaining equations. We start by transforming ?? into an equivalent integral equation. Notice that

$$\begin{aligned} \psi_t^{(k+1)} = & \int_0^t e^{-(k+1)w(t-s)} T_+ e^{\int_s^t -iH + LZ_u^* du} \\ & \left((k+1)\alpha(0)L\psi_s^{(k)} - L^\dagger \psi_s^{(k+2)} \right) ds \end{aligned} \quad (3.8)$$

formally satisfies the corresponding equation of motion together with the required initial condition.

3.2.3. Nonlinear Hierarchy

We mention in Sect. 2.3 how the scaling of our Monte-Carlo sampling with the number of realizations improves drastically we use a nonlinear version. This can be achieved within our microscopical using the comoving coherent states defined in Eq. (2.34). Up to a certain extend this method can also be employed for our hierarchical equations of motion.

As a function of the coherent state labels \mathbf{z}^* instead of the process Z^* we define comoving auxiliary states by

$$\tilde{\psi}_t^{(k)}(\mathbf{z}^*) := (\psi_t^{(k)} \circ \phi_t)(\mathbf{z}^*) = \psi_t^{(k)}(\phi_t(\mathbf{z}^*)).$$

Then the same steps that give Eq. (2.36) lead to a similar result

$$\begin{aligned} \partial_t \tilde{\psi}_t^{(k)}(\mathbf{z}^*) = & \left(-iH - kw + LZ_t^* + L \int_0^t \alpha(t-s)^* \langle L^\dagger \rangle_s ds \right) \tilde{\psi}_t^{(k)}(\mathbf{z}^*) \\ & + k\alpha(0)L\tilde{\psi}_t^{(k-1)} - (L^\dagger - \langle L^\dagger \rangle_t)\tilde{\psi}_t^{(k+1)}(\mathbf{z}^*), \end{aligned} \quad (3.9)$$

with the normalized expectation value taken with respect to the true quantum trajectory—or put differently with respect to the zeroth order auxiliary state

$$\langle L^\dagger \rangle_s = \frac{\langle \tilde{\psi}_t^{(0)} | L^\dagger | \tilde{\psi}_t^{(0)} \rangle}{\langle \tilde{\psi}_t^{(0)} | \tilde{\psi}_t^{(0)} \rangle}.$$

Notice that the exponential correlation function necessary for the hierarchy also simplifies the treatment of the memory-term in Eq. (3.9). Indeed we can derive a simple and closed evolution equation for it.

But one caveat remains: for the convolutionless formulation we can go one step further and even derive an equation for normalized pure state trajectories (2.37). The same does not hold true for the hierarchical equations of motion.

3.2.4. Multimodes

Of course most physically interesting systems cannot be modeled with only a single exponential bath mode. We now present the changes necessary to accommodate a more general environmental structure given by a finite number of exponential modes coupling to the system with arbitrary operators. As the crucial points do not depend on the choice of linear or nonlinear version we are only concerned with the former in this section.

The linear non-Markovian stochastic Schrödinger equation for a finite number N of independent environments may be derived along the same lines presented in Sect. 2.2. It reads

$$\partial_t \psi_t = -iH\psi_t + \sum_{j=1}^N L_j Z_{j,t}^* \psi_t - \sum_{j=1}^N L_j^\dagger \int_0^t \alpha_j(t-s) \frac{\delta \psi_t}{\delta Z_{j,t}^*} ds$$

with independent noise processes satisfying

$$\mathbb{E} Z_{i,t} = 0, \quad \mathbb{E} Z_{i,t} Z_{j,s} = 0, \quad \text{and} \quad \mathbb{E} Z_{i,t} Z_{j,s}^* = \delta_{ij} \alpha_i(t-s).$$

Remember that our hierarchical equations of motion rely on an exponential bath correlation function; therefore we need to introduce several derivation operators $\mathcal{D}_{j,t}$, one for each process. Just as in the single-mode case, the auxiliary states are obtained by successively applying $\mathcal{D}_{i,t}$ on the quantum trajectory $\psi_t(Z^*)$

$$\psi_t^{(k_1, \dots, k_N)} := \mathcal{D}_{1,t}^{k_1} \dots \mathcal{D}_{N,t}^{k_N} \psi_t. \quad (3.10)$$

Note that all $\mathcal{D}_{j,t}$ mutually commute. Consequently Eq. (3.10) is the most general form how a functional derivative may occur in the derivation of the corresponding hierarchy. Hereinafter we use the shorthand notation $\psi_t^{(\mathbf{k})}$ for the auxiliary state

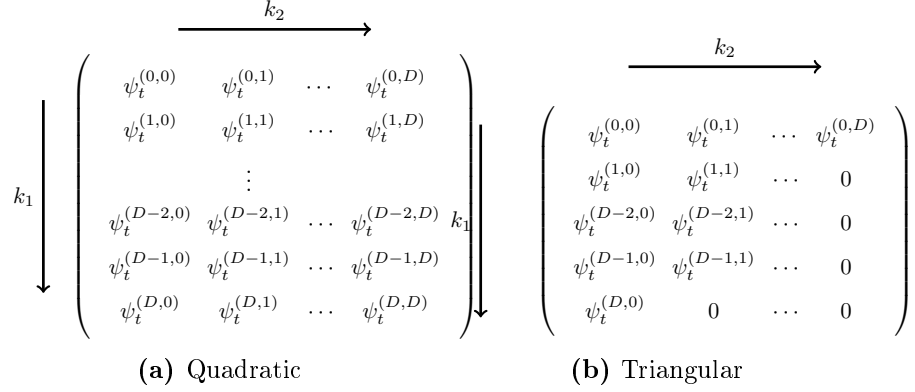


Figure 3.1.: Comparison of the two truncation schemes in the special case of $N = 2$ processes with order D .

defined above. Similar to (3.7) its equation of motion reads

$$\partial_t \psi_t^{(\mathbf{k})} = (-iH - \mathbf{k} \cdot \mathbf{w} + \sum_j L_j Z_{j,t}^*) \psi_t^{(\mathbf{k})} + \sum_j k_j \alpha_j(0) \psi_t^{(\mathbf{k} - \mathbf{e}_j)} - \sum_j L_j^\dagger \psi_t^{(\mathbf{k} + \mathbf{e}_j)}, \quad (3.11)$$

where \mathbf{e}_j denotes the j -th unit vector in \mathbb{R}^N and $\mathbf{k} \cdot \mathbf{w} = \sum_j k_j w_j$ is the euclidean scalar product.¹

When it comes to truncating the hierarchy (3.11) the most obvious strategy is simply to cut off each mode separately at given order D . In other words the truncation condition reads $0 \leq k_j \leq D$ for all $j = 1, \dots, N$; any auxiliary state not satisfying it is set to zero. We refer to this scheme as “cubic truncation scheme” since the shape of all states under consideration resembles an N -cube (see also Fig. 3.1a). Clearly the number of auxiliary states scales exponentially like $(D + 1)^N$, which makes the treatment of physically interesting systems with N in the order of 100 absolutely impossible.

Examining Eq. (3.11) closer we notice that the term responsible for suppression of the \mathbf{k} -th order auxiliary state is $\exp(-\mathbf{k} \cdot \mathbf{w})$. Instead of treating each mode individually we use a condition better suited for the product $\mathbf{k} \cdot \mathbf{w}$, namely $0 \leq |\mathbf{k}| \leq D$ with $|\mathbf{k}| = \sum_j k_j$. For $N = 2$ the corresponding states form a triangular shape as shown in Fig. 3.1b, hence the name “triangular truncation”. The appropriate

¹Although \mathbf{w} is complex in general no complex conjugation occurs at any time.

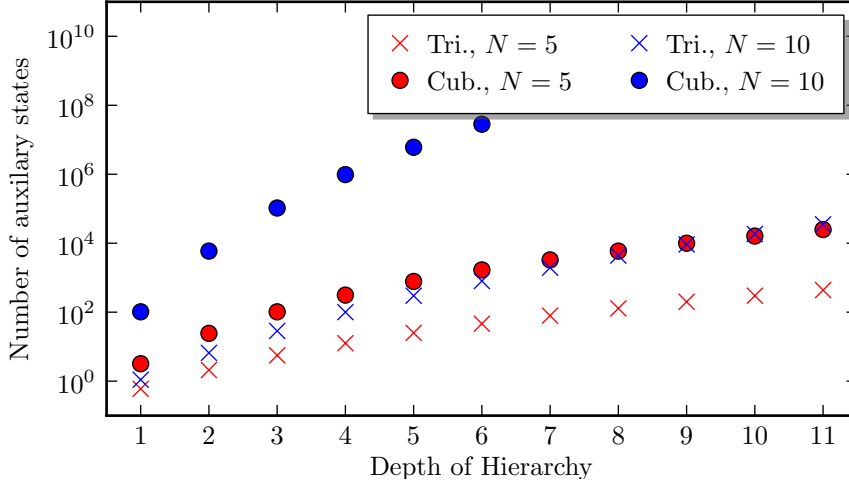


Figure 3.2.: Comparison. . .

generalization to arbitrary N is a simplex where the number of elements is given by

$$\sum_{d=0}^D \binom{d+N-1}{N-1} = \frac{(D+N)!}{D!N!}$$

showing a much softer scaling compared to the cubic scheme. In Fig. 3.2 we display the number of auxiliary states required for a given number of modes and truncation order D . Clearly the triangular truncation is far superior although for small N both methods are still feasible. The difference is more pronounced for larger N required in the study of realistic systems. As an example take the Fenna-Matthews-Olson complex further investigated in ?? : In a simplified model we couple mutually independent environments with 13 exponential terms to each of its seven sites for a total of 91 modes. This yields an insurmountable number of about 10^{30} auxiliary states for the cubic truncation in first order hierarchy, compared to only 5151 using the triangular scheme.

3.3. Correlation Function Expansion

Applicability of our hierarchical equations of motion to any physically interesting system depends predominantly on the ability to express the relevant bath correlation

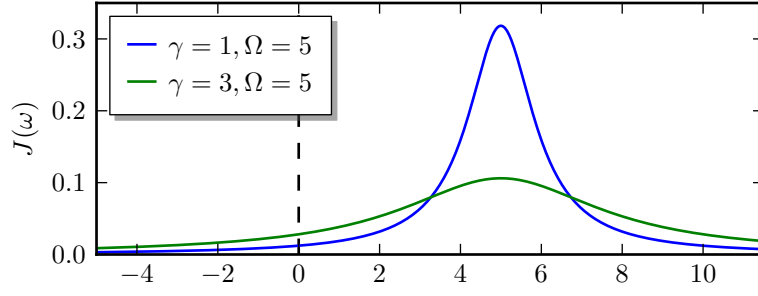


Figure 3.3.: Lorentzian spectral densities (see Eq. (3.13) for notation).

function

$$\alpha(t) = \int_0^\infty J(\omega) \left(\coth \frac{\beta\omega}{2} \cos \omega(t-s) - i \sin \omega(t-s) \right) ds \quad (3.12)$$

as a sum of exponentials like (3.3). Such exponential functions arise as Fourier transforms of a Lorentzian spectral density

$$J(\omega) = \frac{1}{\pi} \frac{\gamma}{(\omega - \Omega)^2 + \gamma^2}. \quad (3.13)$$

Hence they can be obtained from Eq. (3.12) in the zero-temperature limit provided we extend the integral domain to include arbitrary negative frequencies as well. This unphysical assumption is a good approximation to the exact case without negative frequencies only for certain parameters as Fig. 3.3 shows: For $\gamma \ll \Omega$ the Lorentzian density J is concentrated mostly on the positive semi-axis and the negative frequency contribution to Eq. (3.12) can be neglected.

A more systematic way to obtain the desired bath correlation function in the case $T > 0$ was proposed by Meier and Tannor [MT99]. They employ anti-symmetrized Lorentzian distributions $\tilde{J}(\omega) := J(\omega) - J(-\omega)$ to include the negative frequencies without any approximation. Indeed, since \tilde{J} , \coth , and \sin are anti-symmetric and \cos is symmetric with respect to reflection at the origin we have

$$\int_0^\infty \tilde{J}(\omega)(\dots) d\omega = \frac{1}{2} \int_{-\infty}^\infty \tilde{J}(\omega)(\dots) d\omega.$$

Of course the same holds true for any anti-symmetric spectral density. However, the bath correlation function does not have the form necessary due to the additional \coth for $T > 0$. This can only be accomplished approximately by expanding the latter

and evaluating Eq. (3.12) using the residue theorem. We focus on the real part of Eq. (3.12) given by

$$a(t) = \frac{1}{2} \int_{-\infty}^{\infty} J(\omega) \coth \frac{\beta\omega}{2} \cos \omega t = \frac{1}{2} \int_{-\infty}^{\infty} J(\omega) \coth \frac{\beta\omega}{2} e^{i\omega t}, \quad (3.14)$$

since it encodes all thermal effects. Calculating the corresponding imaginary part is straightforward.

3.3.1. Matsubara Spectrum Decomposition

A commonly used expansion scheme for the hyperbolic cotangens is the Matsubara spectrum decomposition [Mah00]

$$\coth \left(\frac{\beta\omega}{2} \right) = \frac{2}{\beta} \sum_{n=-\infty}^{\infty} \frac{1}{i\omega_n - \omega}, \quad (3.15)$$

with the Matsubara frequencies $\omega_n = 2\pi n/\beta$. A short proof and remarks on the convergence of the series above is given in Sect. B.1. Let us parametrize the anti-symmetrized Lorentzian by

$$\tilde{J}(\omega) = \frac{P\omega}{((\omega - \Omega)^2 + \gamma^2)((\omega + \Omega)^2 + \gamma^2)} \quad (3.16)$$

Leaving aside the degenerate case $\gamma = 0$ there are four simple poles of \tilde{J} given by $\omega = i(\pm\Omega \pm i\gamma)$. The symmetry with respect to complex conjugation is due to $\tilde{J}(\omega)$ being real for $\omega \in \mathbb{R}$ whereas point symmetry with respect to the origin carries over from \tilde{J} . As the singularities of \tilde{J} and \coth are distinct and do not possess an accumulation point the integral (3.14) can be expressed as a sum over individual poles

$$a(t) = \sum_{\omega=\pm\Omega+i\gamma} \text{Res } J(\omega) \coth \frac{\beta\omega}{2} e^{i\omega t} + \sum_{\omega_n>0} \text{Res } \coth \frac{\beta\omega}{2} (i\omega_n) J(i\omega_n) e^{-\omega_n t} \quad (3.17)$$

For $t > 0$ we close the integration contour in the upper complex half-plane; hence only poles with non-negative imaginary part need to be included. Therefore the factor $\exp(-\omega_n t)$ goes to zero as $n \rightarrow \infty$ and we can safely truncate the sum at some finite n_0 . This leads exactly to the desired sum-of-exponentials form for the bath correlation function with in general complex parameters; we will discuss the connected

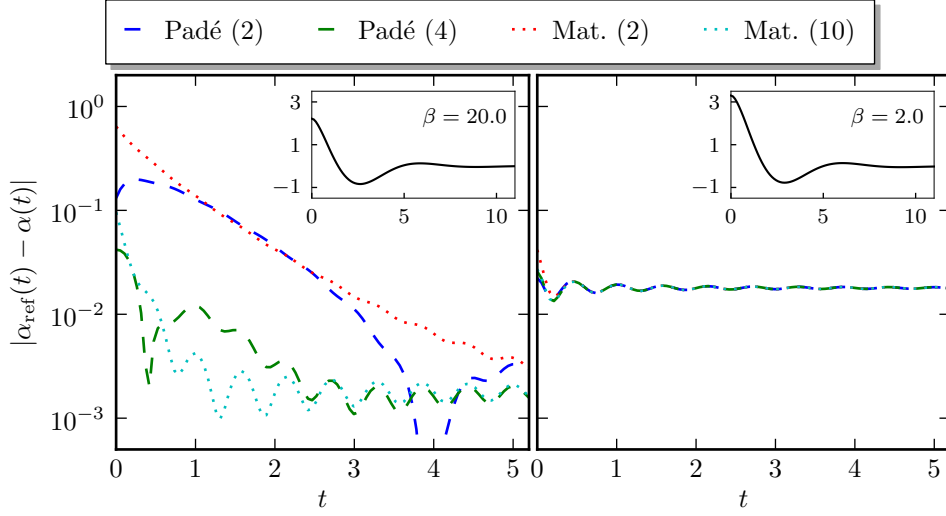


Figure 3.4.: Comparisson. . .

problems in. . .

3.3.2. Padé Spectrum Decomposition

Although the Matsubara spectrum decomposition provides the desired form for the bath correlation function it is worthwhile to consider alternative schemes—especially since the computational effort of our hierarchical equations of motion depends crucially on the numbers of exponential modes under consideration. Only recently Hu et al. proposed an expansion based on the Padé approximant [HXY10, HLJ⁺11], which is vastly superior to the Matsubara expansion in terms of convergence speed. For further details on the theory of Padé approximants we refer to the book by Baker and Graves-Morris [BGM96].

To start off we recall that the hyperbolic cotangens has a simple pole at $z = 0$ with $\text{Res coth}(0) = 1$; therefore we have the convergent Taylor series

$$f(z) := \coth z - \frac{1}{z} = \lim_{N \rightarrow \infty} z \sum_{k=0}^{2N-1} a_k z^{2k} = \lim_{N \rightarrow \infty} z f_{2N-1}(z) \quad (3.18)$$

where we use that \coth as well as $z \mapsto 1/z$ are anti-symmetric function; consequently

all even terms in the Taylor series vanish. By definition of the coefficients a_k we have $f(z) - z f_{2N-1}(z) = \mathcal{O}(z^{4N+1})$ for $|z| < \pi$. In fact the radius of convergence cannot be increased any further since there are additional singularities of f at $z = \pm i\pi$ and the approximating functions f_N are analytical.

The main idea in using a Padé approximant is to incorporate these poles into the approximating functions. The simplest choice is the class of rational functions, that is functions of the form $f_{M,N}(z) = P_M(z)/Q_N(z)$ with polynomials P_M and Q_N of degree less than M and N respectively. We refer to these as $[M, N]$ -approximants. In order to simplify the following treatment we only consider approximants of class $[N-1, N]$, which have proven to be especially suitable for Bose-Einstein and Fermi-Dirac distribution function [HLJ⁺11]. Let us also introduce the shorthand notation $x = z^2$. In order to replace f_N in Eq. (3.18) with a $[N-1, N]$ Padé approximant $f_{N-1,N}$, the latter needs to fulfill the interpolation condition

$$f_{2N-1}(x) = \sum_k^{2N-1} a_k x^k = \frac{P_{N-1}(x)}{Q_N(x)} + \mathcal{O}(x^{2N})$$

by the usual abuse of notation $f_{2N-1}(z) = f_{2N-1}(x)$. Unless $Q_N(0) = 0$ this is equivalent to $Q_N(x)f_{2N-1}(x) = P_{N-1}(x) + \mathcal{O}(x^{2N})$, which always has a unique solution as shown by comparing coefficients on both sides. Using the Padé approximant we can rewrite Eq. (3.18) as

$$\coth(z) = \frac{1}{z} + z \frac{P_{N-1}(z^2)}{Q_N(z^2)} + \mathcal{O}(z^{4N+1}) \quad (3.19)$$

Although P_{N-1} and Q_N are of degree $N-1$ and N respectively there are only $2N$ free parameters because both are only fixed up to a common prefactor. Therefore $f_{N-1,N}$ and f_{2N-1} have the same number of coefficients to be determined; details on the numerical implementation can be found in...

It is quite remarkable that the Padé approximant not only converges on larger subset of \mathbb{C} than the power series in general, but also provides a superior sum-over-poles decomposition compared to the Matsubara expansion Eq. (3.15). It turns out that the roots of $Q_N(x)$ are mutually distinct and negative [HXY10]; therefore we denote them by ξ_i^2 ($i = 1, \dots, N$). Replacing the analytic approximant f_N in

Eq. (3.18) by $f_{N-1,N}$ and expanding the latter in terms of partial fractions gives

$$\coth\left(\frac{\beta\omega}{2}\right) = \frac{2}{\beta\omega} + \frac{2}{\beta} \sum_{j=1}^N \left(\frac{\eta_j}{\omega + i\xi_j} + \frac{\eta_j}{\omega - i\xi_j} \right) + \mathcal{O}(\omega^{4N+1}) \quad (3.20)$$

which agrees with Eq. (3.15) for $N \rightarrow \infty$. It is only for a finite number of summands that the Padé expansion comes to fruition. As an example take $N = 5$, then we find

$$\begin{aligned} \left(\frac{\beta\xi_j}{2\pi j} \right)_j &= (1.00, 1.00, 1.01, 1.18, 2.69) \\ \left(\frac{\beta\eta_j}{2} \right)_j &= (1.00, 1.00, 1.11, 2.80, 26.59), \end{aligned}$$

showing a noticeable different behavior than the Matsubara poles and residues given by $\beta\xi_j/2\pi j = 1$ and $\beta\eta_j/2 = 1$ respectively. Also recall from Eq. (3.17) that the suppressing term for the sum over poles is given by $\exp^{-\xi_j t}$; hence the super-linear growth of the Padé poles ensure that less summands and thus less exponential modes in our hierarchical equations are needed. In conclusion the Padé spectrum decomposition (3.20) can be interpreted as an optimally corrected truncated Matsubara expansion, where neglected terms are included by adjusting the final poles and residues.

3.4. Spin-Boson Model

The Spin-Boson model is a well known example in the fields of solid state physics and quantum optics for a dissipative system [1]. Despite being comparatively simple and traceable it shows many characteristics like that can be found in more realistic systems as well. . . . Therefore it is well suited to investigate the properties of a general open quantum system and their dependence on the environmental parameters. It can also be used to approximately describe systems with a continuous degree of freedom confined by a double well potential [LCD⁺87]. Examples for the latter are the motion of defects in some crystalline solids or the motion of the magnetic flux trapped in a superconducting qubit [CL83]. In this section we use it to further investigate our hierarchy and the dependence on numerical parameters like its depth or the number of realizations.

In general the model is given by a two-level system with the free time evolution described by the Hamiltonian

$$H = -\frac{1}{2}\Delta\sigma_x + \frac{1}{2}\epsilon\sigma_z,$$

coupled linearly to a bath of harmonic oscillators by $L = c\sigma_z$. Here σ_i denotes the Pauli matrices.

3.4.1. Sample Size

Since our hierarchical equations of motion are based upon a stochastic differential equation, it is crucial to assess their reliability in dependence on the sample size. In this section we emphasize in particular the superiority of the nonlinear version (3.9) over the linear hierarchy (3.7). For a systematic investigation we study the Spin-Boson model using two quite distinct sets of parameters, which we refer to as weakly and strongly coupled, although other parameters as the memory time of the bath γ^{-1} have to be taken into account as well. The former choice of parameters can even be treated using an improved perturbation scheme [GHZ10, HZ08]. Nevertheless the nonlinear equations of motion constitute a noticeable improvement in convergence speed even in these almost trivial parameter regimes.

In Fig. 3.5 we plot the expectation value of the spin-operator in z -direction for an initial eigenstate corresponding to the eigenvalue $s_z = +\frac{1}{2}$. In the case of the weakly coupled system at the top there seems to be almost no difference between the linear version on the left and the nonlinear version on the right. Nevertheless for very few realizations the latter is still in better agreement with the more accurate results. Especially for large times the dampening on the right hand side is more pronounced as comparing the graph for a sample size of ten reveals.

When it comes to the strongly coupled parameter set at the bottom the picture changes dramatically: First notice that the sample size has to be increased by an order of magnitude to obtain a reliable result. Notwithstanding the linear version does display any visible change for the different sample sizes shown; hence we cannot expect convergence for even larger numbers of realizations. Even qualitative features as the vanishing of $\langle\sigma_z\rangle$ for $t \rightarrow \infty$ cannot be reproduced in this scheme. It is only

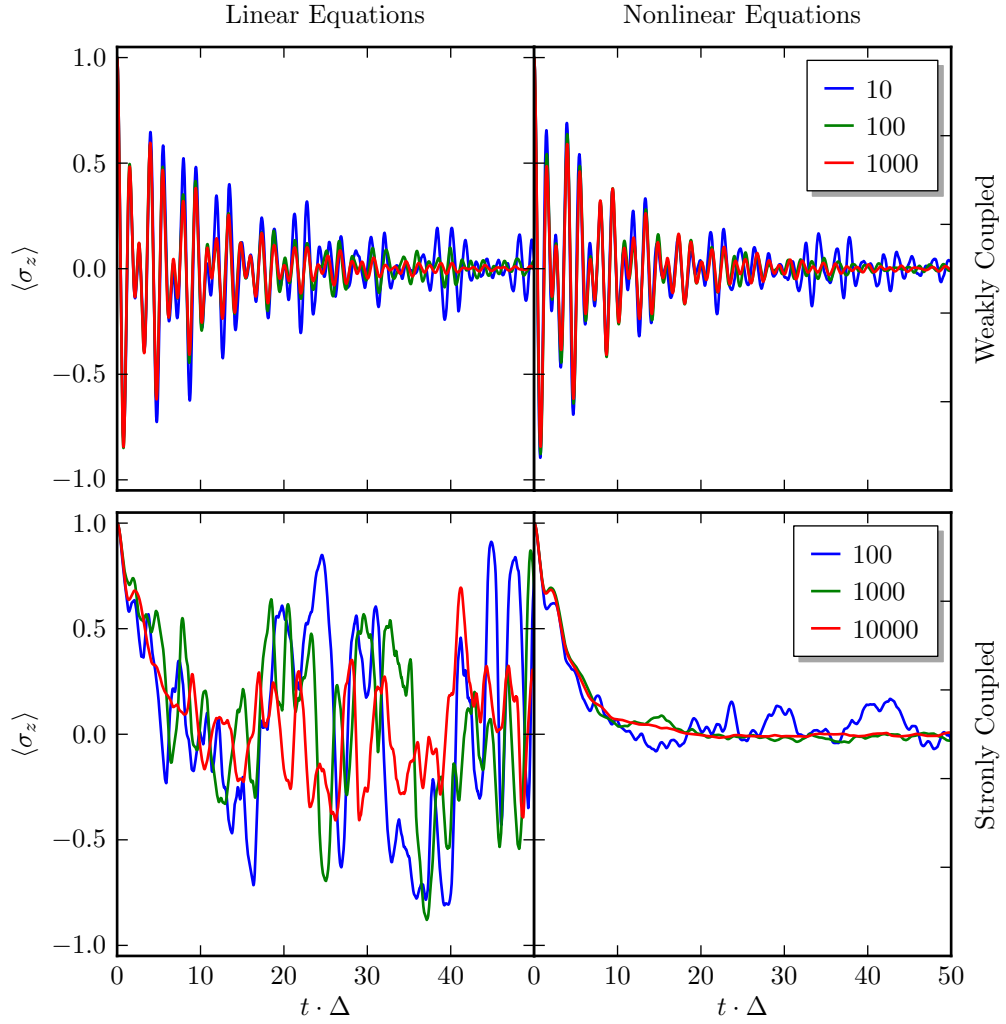


Figure 3.5.: Expectation values for the σ_z -operator calculated using the linear (left) and nonlinear (right) hierarchical equations for two sets of parameters. On the top we display a weakly coupled system given by parameters... In contrast the bottom pictures show a strongly coupled system with...

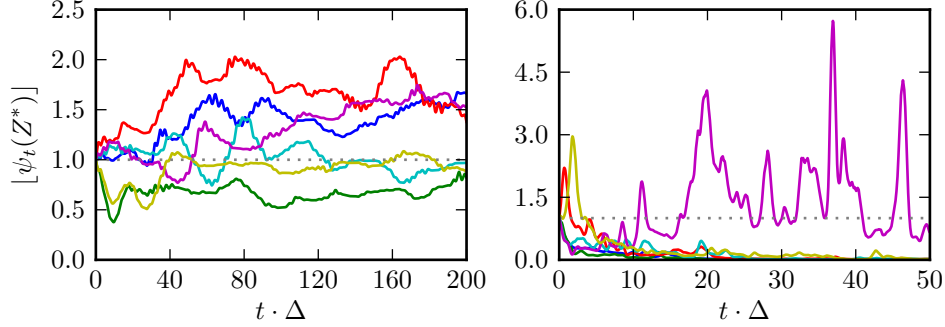


Figure 3.6.: Contributions of single trajectories to the reduced density operator for the linear equations. Left: weakly; right: strongly For definition see Eq. (3.22). . . Parameters are the same as in Fig. 3.5 Dotted line = nonlinear.

for very small times that both sides agree up to a certain extent. In contrast the nonlinear version improves steadily with growing sample size and provides a good approximation with only minor fluctuations already at 1000 trajectories.

To understand this behavior recall that we have introduced the nonlinear equations of motion in order to achieve an average over single contributions of the same order of magnitude. As already mentioned in Sect. 2.3 individual trajectories of the linear version violate this requirement in general. To get a better assessment of different contributions over time we have to rescale the norm as follows: First we calculate the reduced density operator ρ_t by the usual Monte-Carlo average

$$\rho_t = \sum_{n=1}^N |\psi_t(Z_n^*)\rangle \langle \psi_t(Z_n^*)| \quad (3.21)$$

over a large number of noise process realizations Z_n^* . Afterwards we obtain a genuine, normalized density matrix by rescaling with $(\text{Tr } \rho_t)^{-1}$. Therefore we can assess the contribution of a single trajectory $\psi_t(Z_n^*)$ to Eq. (3.21) by $[\psi_t(Z_n^*)]/N$ with

$$[\psi_t(Z_n^*)] = \sqrt{\frac{\langle \psi_t(Z_n^*), \psi_t(Z_n^*) \rangle}{\text{Tr } \rho_t}}. \quad (3.22)$$

Since the states in Eq. (3.21) are normalized for the nonlinear equation, we have for its realizations $[\psi_t(Z_n^*)] = 1$.

We display the contribution using the linear version in ??; clearly we do not obtain a constant contribution for both sets of parameters. But for the weakly coupled system on the left all trajectories remain roughly the same order of magnitude. In contrast the contributions on the right hand side belonging to the strongly coupled system all vanish for large t , but also show pronounced peaks for a few trajectories.

In conclusion. . .

3.4.2. Hierarchy Depth and Cutoff

Apart from the sample size there is an even more significant factor limiting the applicability of our hierarchical equations of motion: As seen from Fig. 3.2 the number of auxiliary states required—and with it the computational expense—grows faster than linear with the truncation depth. Before investigating the latter more thoroughly we demonstrate the effectiveness of using an appropriate terminator instead of just truncating the hierarchy. For that matter the Spin-Boson model is well suited since contrary to the Dissipative two-level system presented in Sect. 2.6 it does not terminate at any finite order.

4. Application

4.1. Basic Model

4.1.1. The Aggregat Hamiltonian

In the following chapter we treat molecular aggregates with a size in the order of magnitude from a few up to a hundred molecules. Let us consider the latter composed of electrons and point-like nuclei quantum mechanically described by canonical-conjugated pairs of operators (p_j, q_j) and (P_j, Q_j) respectively. The corresponding Hamiltonian is given by

$$H_{\text{mol}} = T_{\text{el}} + T_{\text{nuc}} + V_{\text{el-el}} + V_{\text{nuc-nuc}} + V_{\text{el-nuc}} \quad (4.1)$$

with the kinetic energies T and appropriate Coulomb interactions V . We drop possible contributions from internal spin degrees of freedom since they induce only negligible corrections for the systems under consideration.

The vast difference in masses of electrons and nuclei allows us to separate the dynamics of both into two individual parts using the Born-Oppenheimer approximation: As electrons move on a much faster time scale they can respond to any changes in the nuclear arrangement almost instantaneously. This amounts to including the motion of nuclei mediated by the Coulomb potential $V_{\text{el-nuc}}$ only adiabatically when calculating the electron dynamics from Eq. (4.1). Therefor we can reorganize the summands in Eq. (4.1) more appropriately to

$$H_{\text{mol}} = H_{\text{el}}(\mathbf{Q}) + T_{\text{nuc}} + V_{\text{nuc-nuc}}, \quad (4.2)$$

where the notation $H_{\text{el}} = T_{\text{el}} + V_{\text{el-el}} + V_{\text{el-nuc}}(\mathbf{Q})$ indicates that we regard the electronic Hamiltonian to depend only parametrically on the nuclear coordinates \mathbf{Q} . For the processes under consideration only the valence electrons need to be taken into

4. Application

account explicitly; others are included to the nucleon-part without further notice.

The same reasoning applies to the complete Hamiltonian of the aggregat, which besides contributions of the form (4.1) for each individual molecule contains intermolecular interactions between electrons and nuclei in all possible combinations. Therefore it can rephrased similarly to Eq. (4.2)

$$H_{\text{agg}} = H_{\text{el}}(\mathbf{Q}) + T_{\text{vib}} + V_{\text{vib-vib}}, \quad (4.3)$$

Here we use the more general notion of vibrational degrees of freedom, which not only comprises the intra- and intermolecular nuclear coordinates, but also possible environmental degrees of freedom not belonging to the aggregat. These appear for example when studying molecular compounds immersed in a liquid solvent.

The Born-Oppenheimer approximation allows us to analyse the electronic separately from the vibrational part of Eq. (4.3) for a fixed coordinate vector \mathbf{Q} . We split up the former into contributions for each individual electron

$$H_{\text{el}} = \sum_m H_m^{(\text{el})} + \frac{1}{2} \sum_{m,n} U_{mn}^{(\text{el-el})},$$

where $H_m^{(\text{el})}$ contains the m^{th} electron's kinetic energy as well as its coupling to the vibrational degrees of freedom and U_{mn} is simply the Coulomb interaction between the m^{th} and n^{th} electron. The "free" Hamiltonians $H_m^{(\text{el})}$ define distinct electronic states by

$$H_m^{(\text{el})}(\mathbf{Q})\varphi_{ma}(q, \mathbf{Q}) = \epsilon_{ma}(\mathbf{Q})\varphi_{ma}(q, \mathbf{Q})$$

for each given environmental configuration \mathbf{Q} . The index m runs over all electrons under consideration and a is used to label the individual states, which we assume to be ordered by the corresponding energies. Similar to the Hartree-Fock method we build up an expansion basis for the total electronic state by a product ansatz

$$\phi_{\mathbf{a}}(\mathbf{q}, \mathbf{Q}) = \prod_m \varphi_{m,a_m}(q_m, \mathbf{Q}), \quad (4.4)$$

which in general needs to be anti-symmetrized to fulfill the Pauli exclusion principle.

If there is at most one valence electron per molecule we need to take into consideration, which is furthermore tightly bound, then the situation simplifies dramatically:

In this case the spreading of the single-electron states $|\varphi_{ma}\rangle = |m, a\rangle$ is small compared to the distance between two molecules; we can neglect the overlap $\langle m, a | n, b \rangle$ for different molecules $m \neq n$. Consequently ?? yields a complete basis for the electronic degrees of freedom. We also have the following representation for the Hamiltonian (4.3)

$$H_{\text{el}} = \sum_{m,a} \epsilon_{m,a} |m, a\rangle \langle m, a| + \frac{1}{2} \sum_{m,n,a,b,a',b'} U_{mn}(aa', bb') |m, a; n, b\rangle \langle m, a'; n, b'| \quad (4.5)$$

with the matrix elements of the Coulomb interaction¹

$$U_{mn}(aa', bb') = \langle m, a; n, b | U_{mn} | m, a'; n, b' \rangle.$$

Note that all terms in ?? still depend on vibrational coordinates. For example the matrix elements $U_{mn}(aa'; bb')$ is influenced by the distance between the m^{th} and n^{th} molecule, while the electronic eigenenergies $\epsilon_{m,a}$ primarily depend on the positions of other electrons belonging to the same molecule.

4.1.2. The Exciton Model

In order to describe the experimental setting described in the introduction we do not need to consider the complete electronic Hamiltonian (4.5): if only a single valence electron is initially in the lowest excited state S_1 above its ground state S_0 ² and if the various transition energies are in the same order of magnitude, then it is sufficient to take only the S_0 state $|m, 0\rangle$ as well as the first excited state $|m, 1\rangle$ for each molecule into account. Under these circumstances the matrix elements $U_{mn}(aa', bb')$ can be classified with respect to a few physical processes such as electrostatic interactions or charge-induced transitions. But most important is the resonant contribution $U_{mn}(01; 10)$ (and its inversion $U_{mn}(10; 01)$) describing a $S_0 \rightarrow S_1$ excitation for the m^{th} electron induced by a $S_0 \rightarrow S_1$ transition at the n^{th} molecule. In the following we neglect all but the last class of processes, which is frequently called Heitler-London approximation.

¹This does not include the exchange interaction, since we assume a vanishing mutual overlap for the electrons.

²Note that S_0 describes the lowest energy state of the valence electron with all other electrons of the molecule fixed, not to be confused with the atomic ground states.

4. Application

Restricting the allowed electronic states to the two lowest energy levels has a remarkable interpretation in terms of quasi-particles: The product

$$|m\rangle = |m, 1\rangle \prod_{n \neq m} |n, 0\rangle \quad (4.6)$$

describes an excited electron localized in the vicinity of the m^{th} molecule, which we refer to as an exciton of the electronic system. Due to the Heitler-London approximation our adiabatic Hamiltonian (4.5) conserves the number of excitons. Therefore an initial state $|m\rangle$ (or linear combinations thereof) always remains in the one-exciton Hilbert space $\mathcal{H}^{(1)}$. The interaction matrix elements

$$V_{mn} = V_{nm} = \langle m, 0; n, 1 | U_{mn} | m, 1; n, 0 \rangle$$

allow us to express the restriction of H_{el} to $\mathcal{H}^{(1)}$ as

$$H_{\text{el}}^{(1)}(\mathbf{Q}) = \sum_m \epsilon_m(\mathbf{Q}) |m\rangle \langle m| + \sum_{m,n} V_{mn}(\mathbf{Q}) |n\rangle \langle m|.$$

For the rest of this section we assume the V_{mn} to be independent of vibrational degrees of freedom; a more general treatment poses no further difficulties.

Up to this point we have neglected the dynamical evolution of the vibrational environment, which is essential in a complete description of a molecular aggregate. In common settings for the physical systems under consideration a harmonic approximation is sufficient to obtain a realistic model. There are two reasons for this: First of all most proteins disintegrate at temperatures much higher than room temperature; therefore thermal excitation only leads to small energy gains for each vibrational degree of freedom. The other mechanism for driving the environment is dissipation of the electronic system. But the latter is small compared to the vast number of vibrational degrees of freedom and energy typically spreads evenly across the environment. We can thusly assume that all Q_λ experience only a small displacement from their equilibrium positions, which we set to $Q_\lambda = 0$.

As a consequence both $V_{\text{vib-vib}}(\mathbf{Q})$ and $\epsilon_m(\mathbf{Q})$ can be expanded in a Taylor series neglecting all but the first non-trivial term. To alleviate notation we further assume that each vibrational degree of freedom only couples to one specific exciton. This

leads to exactly the microscopical model presented in Sect. 2.1: a bath of harmonic oscillators linearly coupled to the electronic system

$$H_{\text{agg}} = \sum_m \epsilon_m(0) |m\rangle\langle m| + \sum_{m,n} V_{mn} |m\rangle\langle n| + \sum_{m,\lambda} \omega_{m,\lambda} A_{m,\lambda}^\dagger A_{m,\lambda} \\ + \sum_{m,\lambda} g_{m,\lambda} |m\rangle\langle m| \otimes (A_{m,\lambda}^\dagger + A_{m,\lambda}),$$

where $A_{m,\lambda}/A_{m,\lambda}^\dagger$ are ladder operators corresponding to the λ^{th} vibrational mode coupling to the m^{th} exciton.

4.2. Transfer Dynamics

As a first exemplary application of our hierarchical equations of motion we study energy transfer in the Fenna-Matthews-Olson (FMO) complex found in low-light adapted green sulfur bacteria. This protein complex plays a crucial role in connecting the light harvesting antenna (chlorosome) to the photosynthetic reaction center, where the absorbed solar energy is converted to a charge gradient. The vast amount of literature on the subject is not only by virtue of its relatively small size—making it an ideal model system for numerical investigation—but particularly caused by its genuine “quantumness”. Only lately Engel et al. as well as Ishizaki-Fleming demonstrated that the FMO complex achieves its remarkable almost-unit efficiency by coherent exciton motion instead of classical hopping described by Förster theory [ECR⁺07,IF09].

The FMO protein complex is subdivided into three identical monomers, each comprising eight bacteriochlorophyll pigments (BChls). In contrast to the first seven BChls the eighth was only discovered in recent years due to its rather weak coupling to the remaining BChls and instability during the isolation procedure in experiments [TCABA09,SaBMhEAMR10]. As the main goal here is to show the applicability and reliability of our hierarchical equations of motion we ignore BChl number eight in what follows—this model has been treated thoroughly with a vast array of methods. For the same reason we also restrict our attention to an individual monomer: as shown by Ritschel et al. such a limitation is reasonable for the short time scales under consideration as the intermonomeric interaction strength is rather weak.

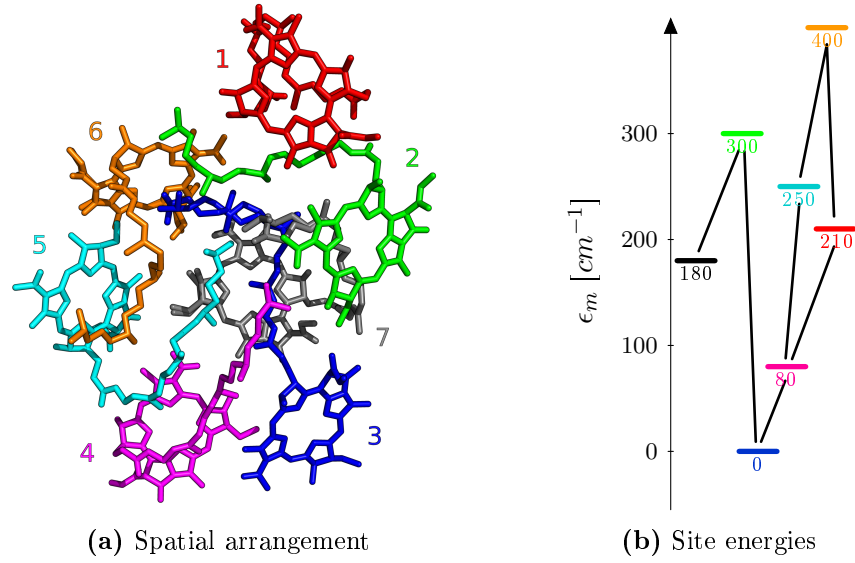


Figure 4.1.: Here all eight BChls. Created using PyMol, based on PDB entry 3ENI [Sch10, TCABA09]

In Fig. 4.1a we display the spatial structure and numbering of a monomer. The BChls 1 and 6 are situated in the vicinity of the baseplate and receive excitation energy captured by the chlorosomes, while BChl 3 acts as energy sink.

4.3. Absorption Spectra

4.3.1. NMSSE for Spectra

4.3.2. Results

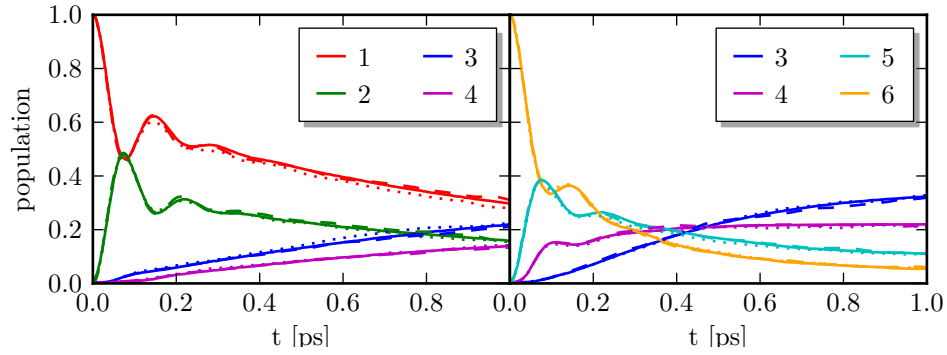


Figure 4.2.: Exciton transfer of the simplified FMO-monomer with seven BChls using our hierarchical equation of motion up to first (dotted line) and second order (dashed line). We place the initial excitation on both receiving BChls, that is the first (left) and the sixth molecule (right). For comparison the solid line shows the results of Ishizaki and Fleming [IF09], which were obtained in the density-matrix HEOM approach.

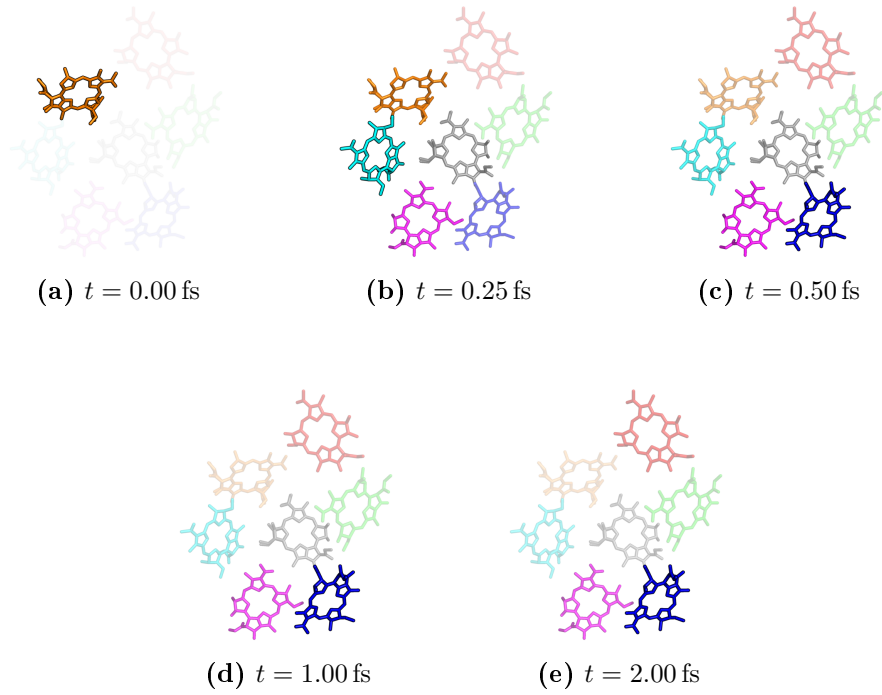


Figure 4.3.: Same as ?? with initial excitation on the sixth BChl (orange). Notice how the energy flow is not evenly distributed but shows a pronounced drift towards the energy sink located at the third BChl (blue). For the sake of clarity we do not show the full molecular structure.

5. Conclusions

A. Analytic Solution of Dissipative Two-level System

A.1. General Solution

Here we present in full detail the analytic solution of the dissipative two-level system introduced in Sect. 2.6. The relevant NMSSE can always be written in terms of a single process

$$\partial_t \psi_t = -i \frac{\omega}{2} \sigma_z \psi_t + c \sigma_- Z_t^* \psi_t - c \sigma_+ \int \alpha(t-s) \frac{\delta \psi_t}{\delta Z_s^*} ds. \quad (\text{A.1})$$

As already mentioned we employ an ansatz for the quantum trajectory at most linear in Z_s^*

$$\psi_t(Z^*) = \psi(t) + \int_0^t \psi_s(t) Z_s^* ds. \quad (\text{A.2})$$

Here we already incorporate that $\psi_t(Z^*)$ needs to be independent of Z_s^* for $s < 0$ and $s > t$ by using a bounded integral domain.¹ We use a basis for $\psi_t(Z^*)$ which renders σ_z diagonal and denote the corresponding components by “+” and “-”. The “+”-component of our NMSSE with the ansatz (A.2) reads

$$\begin{aligned} \dot{\psi}^+(t) + \psi_t^+(t) Z_t^* + \int_0^t \dot{\psi}_s^+ Z_s^* ds \\ = -i \frac{\omega}{2} \left(\psi^+(t) + \int_0^t \psi_s^+(t) Z_s^* ds \right) - c \int_0^t \alpha(t-s) \psi_s^-(t) ds. \end{aligned} \quad (\text{A.3})$$

¹A consistent treatment of these independence-conditions is more tricky than it seems at first glance, since we are mixing a distributional object Z_s^* with a discontinuous function $s \mapsto \psi_s(t)$. As $\delta \psi_t(Z^*) / \delta Z_s^* = 0$ translates to $\psi_s(t) = 0$ the correct integral boundary reads $\int_{0-\varepsilon}^{t+\varepsilon}$ with $\varepsilon > 0$ but arbitrary otherwise.

A. Analytic Solution of Dissipative Two-level System

In order to separate contributions of different order in Z_s^* , we apply the functional derivative $\delta/\delta Z_s^*$ with arbitrary s to the last equation

$$\psi_t^+(t)\delta(t-s) + \int_0^t \dot{\psi}_{s'}^+(t)\delta(s-s')ds' = -i\frac{\omega}{2} \int_0^t \psi_{s'}^+(t)\delta(s-s')ds'. \quad (\text{A.4})$$

Choosing $s \in (0, t)$ leaves us with a simple ordinary differential equation with solution $\psi_s^+(t) = C_s e^{-i\omega t/2}$. By further investigating the singular term in Eq. (A.4) we find that $\psi_s^+(s)$ and therefore the constant C_s must vanish. This amounts formally to integrating (A.4) over a small interval $(t-\varepsilon, t+\varepsilon)$ with respect to s . In the limit $\varepsilon \rightarrow 0$ all terms except the first go to zero. On the other hand we can isolate all terms independent of Z_s^* in Eq. (A.3) simply by taking the expectation value. The resulting equation

$$\dot{\psi}^+(t) = -i\frac{\omega}{2}\psi^+(t) - c \int_0^t \alpha(t-s)\psi_s^-(t)ds \quad (\text{A.5})$$

is treated later, once we have an explicit expression for $\psi_s^-(t)$.

The “-” component of Eq. (A.1) is quite similar to (A.3)

$$\begin{aligned} \dot{\psi}^-(t) + \psi_t^-(t)Z_t^* + \int_0^t \dot{\psi}_s^-(t)Z_s^*ds \\ = i\frac{\omega}{2} \left(\psi^-(t) + \int_0^t \psi_s^-(t)Z_s^* \right) + c\psi^+(t)Z_t^*, \end{aligned} \quad (\text{A.6})$$

where we already used that $\psi_s^+(t) = 0$. The solution

$$\psi^-(t) = \psi^-(0) e^{i\frac{\omega}{2}t} \quad (\text{A.7})$$

can be read off Eq. (A.6) directly by comparing all terms independent of Z_t^* . In the same manner as we derived Eq. (A.4) we can treat all terms proportional to Z_t^* . Again we find a solution of the form $\psi_s^-(t) = C_s e^{i\omega t/2}$; the only difference is an additional singular term due to the driving process in Eq. (A.1). It gives rise to the boundary condition $\psi_t^-(t) = c\psi^+(t)$. Together with ?? we obtain a closed equation for $\psi^+(t)$

$$\dot{\psi}^+(t) = -i\frac{\omega}{2}\psi^+(t) - c^2 \int_0^t \alpha(t-s)e^{i\frac{\omega}{2}(t-s)}\psi^+(s)ds. \quad (\text{A.8})$$

Hence we may replace the original NMSSE (A.1) by a \mathbb{C} -valued equation similar in structure.

A.2. Exponential Correlation Function

As Eq. (A.8) still contains a memory integral there is no analytic solution in general. The situation is noticeably simpler for a bath correlation of the form

$$\alpha(t-s) = \sum_{j=1}^N g_j e^{-\gamma_j |t-s| - i\Omega_j(t-s)}. \quad (\text{A.9})$$

Since ψ^+ only depends on values of $\alpha(t)$ for $t \geq 0$ we can assume $\gamma = 0$ without loss of generality. Similar to our hierarchical equations of motion elaborated in Sect. 3.2 we absorb the problematic terms into auxiliary functions

$$\phi_j(t) := \int_0^t \alpha_j(t-s) e^{i\frac{\omega}{2}(t-s)} \psi^+(s) ds. \quad (\text{A.10})$$

This allows us to rewrite Eq. (A.8) as a system of $(N+1)$ ordinary differential equations with constant coefficients

$$\begin{aligned} \dot{\psi}^+(t) &= -i\frac{\omega}{2}\psi^+(t) - c^2 \sum_{j=1}^N \phi_j(t) \\ \dot{\phi}_j(t) &= g_j \psi^+(t) + i\left(\frac{\omega}{2} - \Omega\right) \phi_j(t). \end{aligned}$$

and initial conditions $\phi_j(0) = 0$. In the special case $N = 1$ the diagonalization can be carried out analytically. With the shorthand notation $\tilde{\Omega} = \sqrt{(\omega - \Omega)^2 + 4c^2g}$ the solution to Eq. (A.8) reads

$$\psi^+(t) = \frac{\psi^+(0)}{2\tilde{\Omega}} \left((\omega - \Omega + \tilde{\Omega}) e^{-i\frac{\Omega+\tilde{\Omega}}{2}t} - (\omega - \Omega - \tilde{\Omega}) e^{-i\frac{\Omega-\tilde{\Omega}}{2}t} \right). \quad (\text{A.11})$$

B. Expansions of Hyperbolic Cotangens

Before presenting the two expansion schemes for the hyperbolic cotangens function mentioned in Sect. 3.3 we need to introduce a few ideas from complex analysis. For more details and proofs we refer to the book of Rudin [Rud87].

First recall that a function f with an isolated singularity $a \in \mathbb{C}$ can always be expanded in a Laurent series around a

$$f(z) = \sum_{j=0}^{\infty} a_j (z-a)^j + \sum_{j=1}^{\infty} \frac{a_{-j}}{(z-a)^j} \quad (0 < |z-a| < \varepsilon). \quad (\text{B.1})$$

The first sum is called *analytical part* while the second one is called *principal part*. Clearly if a is a removable singularity, that is the principal part vanishes, we can continue f to a function analytical for all z with $|z-a| < \varepsilon$. A complex function f is said to be *meromorphic* in an open set $\Omega \subset \mathbb{C}$ if it is analytical on Ω except a set $A \subset \Omega$ of isolated points, which are poles for f .

B.1. Matsubara Spectrum Decomposition

In Sect. 3.3 we use the expansion of the hyperbolic cotangens in terms of Matsubara frequencies [Mah00]

$$\omega_n = \frac{2\pi n}{\beta} \quad (n \in \mathbb{Z}). \quad (\text{B.2})$$

It provides a sum over poles representation, namely

$$\coth\left(\frac{\beta z}{2}\right) = \frac{2}{\beta} \sum_{n=-\infty}^{\infty} \frac{1}{i\omega_n - z}, \quad (\text{B.3})$$

with singularities evenly distributed along the imaginary axis.

In order to proof Eq. (B.3) notice that $i\omega_n$ are exactly the isolated singularities of

$z \mapsto \coth(\beta z/2)$. Indeed, using the representation

$$f(z) := \coth\left(\frac{\beta z}{2}\right) = \frac{1 + e^{-\beta z}}{1 - e^{-\beta z}} \quad (\text{B.4})$$

we see that the numerator as well as the denominator are analytical for all $z \in \mathbb{C}$, but the latter vanishes for $z = i\omega_n$. A short calculation reveals that these z are simple poles with $\text{Res } f(i\omega_n) = 2/\beta$.

Let us now define g by the right hand side of Eq. (B.3); in order to enforce convergence of the infinite series, we need to take the symmetric limit

$$g(z) = \frac{2}{\beta} \lim_{N \rightarrow \infty} \sum_{n=-N}^N \frac{1}{i\omega_n - z} = -\frac{2}{\beta} \left(\sum_{n=0}^{\infty} \frac{2z}{\omega_n^2 + z^2} + \frac{1}{z} \right) \quad (\text{B.5})$$

where we used $\omega_{-n} = -\omega_n$ in the second step. Since $\omega \sim n$ the infinite series for g in Eq. (B.5) converges absolutely unless $z = i\omega_n$ and defines a meromorphic function with the same poles and principal parts as f . This leads to the conclusion that $h := f - g$ can be continued to an analytical function on the entire complex plane. We now show that h is bounded on \mathbb{C} and therefore a constant function by Liouville's Theorem. Expanding both f and g in a Laurent series about 0 allows us to calculate $f() = g()$ = as In conclusion we find $h = 0$, or put differently $f = g$ —the Matsubara expansion (B.3) is valid on the entire complex plane.

B.2. Padé Spectrum Decomposition

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