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Hierarchical Equations for Open System Dynamics in Fermionic and Bosonic Environments

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Abstract We present two approaches to the dynamics of an open quantum system coupled linearly to a non-Markovian fermionic or bosonic environment. In the first approach, we obtain a hierarchy of stochastic evolution equations of the diffusion type. For the bosonic case such a hierarchy has been derived and proven suitable for efficient numerical simulations recently (Suess et al. in Phys. Rev. Lett. 113, 150403, 2014). The stochastic fermionic hierarchy derived here contains Grassmannian noise, which makes it difficult to simulate numerically due to its anti-commutative multiplication. Therefore, in our second approach we eliminate the noise by deriving a related hierarchy for density matrices. A similar reformulation of the bosonic hierarchy of pure states to a master equation hierarchy and its relation to the hierarchical equations of motion of Tanimura and Kubo is also presented.

Keywords Non-Markovian · Stochastic Schrödinger equation · Master equation · Quantum trajectories · Fermionic · Bosonic

1 Introduction

The theory of open quantum systems has become an important topic in modern physics and its applications, since many interesting phenomena of quantum systems emerge only when the influence of the system's environment is taken into account [1–3]. However, realistic system-bath models are often analytically and numerically intractable even if one is only interested in the relevant degrees of freedom, i.e. the reduced density operator. Nevertheless, a coarse-grained description is often sufficient for most practical purposes. One standard example

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is an environment of uncoupled harmonic oscillators, which is a good approximation for weakly coupled delocalized modes [4] or shifted harmonic Born–Oppenheimer surfaces in molecules [2,5]. Another standard model is an environment of non-interacting fermions.

These environmental models are often simplified further using the Born–Markov approximation for a "memory-less", weakly coupled environment [1–3]. Recently, there has been a growing interest in going beyond this Born–Markov approximation, which fails in particular if the coupling between system and bath is not weak or if one deals with a structured environment [1,2]. Because of its importance, e.g., for transport through molecules or quantum dots, various approaches have been developed to go beyond the Born–Markov regime in fermionic environments (see for example [6–10]).

One powerful approach to obtain the dynamics of the system for such a non-Markovian setting is non-Markovian quantum state diffusion (NMQSD). The NMQSD approach was originally derived for a harmonic oscillator environment [11–14]. A related NMQSD equation for fermionic environments has been derived recently [15,16]. Within NMQSD one obtains a stochastic Schrödinger equation of the diffusion type that lives in the Hilbert space of the system. Its solutions—so called quantum trajectories—yield the reduced density operator by an average over randomly chosen realizations. The main difference between the bosonic and the fermionic theory is the noise processes entering the equations of motion: While the influence of a bosonic environment can be described exactly by a complex-valued colored Gaussian process, fermionic environments require the use of Grassmannian colored noise.

An obstacle of the NMQSD equations is that it contains the noise not only as a multiplicative term, but also within a functional derivative under a memory integral. To tackle this problem, we have recently derived a hierarchy of pure states (HOPS) for the bosonic case [17]. This hierarchy consists of a set of coupled equations where the noise enters only linearly. The price one has to pay is that instead of an intractable memory integral one deals with an infinite hierarchy of coupled stochastic equations. Fortunately, it turns out that one can often truncate the hierarchy at quite low order resulting in a system of equations which can be solved numerically in an efficient way.

One main result of the present work is the derivation of such a hierarchy of pure states for the fermionic theory, which has a structure very similar to the bosonic one. In contrast to the bosonic case, where one can easily generate the complex Gaussian noise, a numerical simulation of the fermionic HOPS seems to be unfeasible due to the anti-commuting Grassmannian processes. Therefore, we will go one step further and eliminate the noise by deriving a hierarchy of master equations based on the fermionic HOPS, which can be solved numerically efficient. The corresponding master equation for a bosonic environment based on the established HOPS [17] is also presented. Although the hierarchical description of pure state dynamics is quite new, hierarchical equations of motion for density operators of open quantum systems coupled to bosonic [18–21] or fermionic [6] environments are well established tools. In certain special cases, the hierarchy obtained from HOPS is closely related to the established hierarchical equations of motion (HEOM) of Tanimura and Kubo.

The paper is organized as follows: In Sect. 2 we elaborate on the fermionic theory. First, we recapitulate the fermionic NMQSD approach in Sect. 2.1 in order to recall the established theory and introduce our notation. Then, Sect. 2.2 is devoted to the fermionic HOPS and the hierarchy of density matrices is derived in the following Sect. 2.3. Finally, we discuss finite-temperature environments in Sect. 2.4. The corresponding theory for bosonic environments is treated in Sect. 3: Sect. 3.1 summarizes the HOPS construction from [17]. The novel result for bosonic environments, namely the hierarchy of master equations, can be found in Sect. 3.2. Its relation to the established HEOM is discussed in Sect. 3.3. Finally, we provide numerical examples for both methods in Sect. 4. Throughout the paper, we use units where $k_B = \hbar = 1$.

2 Fermionic Environments

Let us consider the (total) Hamiltonian

$$H_{\text{tot}} = H + H_{\text{env}} + H_{\text{int}}, \tag{1}$$

where H captures the system's free dynamics and $H_{\text{env}} = \sum_{j,\lambda} \omega_{j,\lambda} b^\dagger_{j,\lambda} b_{j,\lambda}$ the dynamics of an environment consisting of indistinguishable spin-1/2 particles described by fermionic ladder operators obeying canonical anti-commutator relations

$$\left\{b_{j,\lambda}, b_{j',\lambda'}\right\} = 0 \quad \text{and} \quad \left\{b_{j,\lambda}, b^\dagger_{j',\lambda'}\right\} = \delta_{jj'}\delta_{\lambda\lambda'}. \tag{2}$$

The interaction of system and environment is modeled by a linear coupling Hamiltonian

$$H_{\text{int}} = \sum_{j,\lambda} \left(g^*_{j,\lambda} L_j b^\dagger_{j,\lambda} + g_{j,\lambda} L^\dagger_j b_{j,\lambda}\right). \tag{3}$$

Here, L_j are system operators and $g_{j,\lambda}$ are complex numbers quantifying the coupling strength of the respective fermion (j,λ) to the system. Throughout this paper, we use j to label independent environments coupling to the system and λ to label distinct physical modes (i.e. fermions) within each environment. We assume that all system operators commute with environment operators or, put differently, that the system is distinguishable from the environment. Such a model arises, for example, in the description of tunneling through a quantum dot or molecules [7]. It is convenient to encode the frequency dependence of the interaction strengths in the so called spectral densities

$$J_j(\omega) = \sum_\lambda |g_{j,\lambda}|^2 \delta(\omega - \omega_{j,\lambda}), \tag{4}$$

which are typically assumed to be continuous functions of frequency.

For now we will confine the discussion to the zero-temperature case with pure initial condition

$$|\Psi_0\rangle = |\psi_0\rangle \otimes |0\rangle \tag{5}$$

and treat the more general case of a thermal initial state in Sect. 2.4. Here, $|0\rangle$ denotes the vacuum with respect to all $b_{j,\lambda}$. Since the full dynamics governed by the Hamiltonian (1) is unitary, the full state of system and bath can be described by a pure state $|\Psi_t\rangle$ at all times. The reason for introducing a distinction between system and environment in the first place is that we are only interested in the reduced state of the former, namely

$$\rho_t = \text{Tr}_{\text{env}} |\Psi_t\rangle\langle\Psi_t|. \tag{6}$$

However, the NMQSD formalism recalled in the next section is formally equivalent to solving the full Schrödinger equation for $|\Psi_t\rangle$.

2.1 Fermionic NMQSD

The theory of non-Markovian quantum state diffusion for fermionic environments has been derived in [15,16]. Here, we will briefly recapitulate the crucial steps in order to establish the notation used throughout the paper.

Similar to the bosonic case, the fermionic NMQSD theory is based on a representation of the bath degrees of freedom in coherent states $|\mathbf{z}\rangle := \otimes_j \otimes_\lambda |z_{j\lambda}\rangle$, where a (non-normalized) fermionic coherent state is defined similarly to its bosonic counterpart by

$$|z_{j\lambda}\rangle = \mathrm{e}^{-z_{j\lambda} b^\dagger_{j\lambda}}|0\rangle = |0\rangle - z_{j\lambda} b^\dagger_{j,\lambda}|0\rangle \tag{7}$$

Here, the $z_{j\lambda}$ are anti-commuting Grassmann variables with $\{z_{j\lambda}, z_{j'\lambda'}\} = \{z^*_{j\lambda}, z_{j'\lambda'}\} = \delta_{j,j'}$ and $\{z_{j\lambda}, b_{j'\lambda'}\} = \{z_{j\lambda}, b^\dagger_{j'\lambda'}\} = \delta_{j,j'}$. For more details on these coherent states see e.g. [22,23].

We now expand the bath degrees of freedom of the full system-environment state with respect to the coherent states introduced above $\psi_t(\mathbf{z}^*) := \langle \mathbf{z}|\Psi_t\rangle$. We also absorb the free time evolution of the environment using the interaction picture with respect to H_{env}. The resulting Schrödinger equation for $\psi_t(\mathbf{z}^*)$ then reads

$$\begin{aligned}\partial_t \psi_t(\mathbf{z}^*) = &-\mathrm{i}H\psi_t(\mathbf{z}^*) - \mathrm{i}\sum_{j,\lambda} g^*_{j,\lambda} L_j \mathrm{e}^{\mathrm{i}\omega_{j,\lambda}t} z^*_{j,\lambda}\psi_t(\mathbf{z}^*)\\ &-\mathrm{i}\sum_{j,\lambda} g_{j,\lambda} L^\dagger_j \mathrm{e}^{-\mathrm{i}\omega_{j,\lambda}t} \overrightarrow{\partial}_{z^*_{j,\lambda}} \psi_t(\mathbf{z}^*),\end{aligned} \tag{8}$$

where $\overrightarrow{\partial}_{z^*_{j,\lambda}}$ denotes the left-derivative with respect to $z^*_{j,\lambda}$. In the following we will drop the arrow from left-derivatives (and only indicate right-derivatives explicitly).

The main result of [15,16] is that (8) can be recast into a *stochastic Schrödinger equation* of the quantum state diffusion type, namely

$$\begin{aligned}\partial_t \psi_t(Z^*) = &-\mathrm{i}H\psi_t(Z^*) + \sum_j L_j Z^*_j(t)\psi_t(Z^*)\\ &-\sum_j L^\dagger_j \int_0^t \alpha_j(t-s)\frac{\delta\psi_t(Z^*)}{\delta Z^*_j(s)}\,\mathrm{d}s\end{aligned} \tag{9}$$

where the stochastic process $Z^*_j(t) = -\mathrm{i}\sum_\lambda g^*_{j,\lambda}\mathrm{e}^{\mathrm{i}\omega_{j,\lambda}t} z^*_{j,\lambda}$ is characterized by its correlation function

$$\alpha_j(t) = \sum_\lambda |g_{j,\lambda}|^2 \mathrm{e}^{-\mathrm{i}\omega_{j,\lambda}t} \tag{10}$$

through

$$\mathbb{E}Z_j(t) = \mathbb{E}\left(Z_j(t)Z_{j'}(s)\right) = 0, \quad \mathbb{E}\left(Z_j(t)Z^*_{j'}(s)\right) = \delta_{jj'}\alpha_j(t-s). \tag{11}$$

Note that the Z^*_t are *Grassmannian processes*, i.e. values at different times anti-commute hindering an efficient numerical generation of these processes. Furthermore, Eq. (10) is the well-known relation of the zero-temperature bath correlation function (BCF) and the spectral density (4) through a (one-sided) Fourier transform [1].

Besides describing the correlation of the noise process, the BCF also weights the functional derivative at different times under the memory integral in (9). Since this term is non-local in time as well as in the realization of the processes, it is unclear how it should be evaluated

in general. We will abbreviate the full memory integral introducing the (left-)derivation operator[1]

$$D_{j,t}\psi_t(Z^*) = \int \alpha_j(t-s)\frac{\delta\psi_t(Z^*)}{\delta Z_j^*(s)}\,\mathrm{d}s. \tag{12}$$

In previous works [15,24], this functional derivative was replaced by an operator ansatz acting in the system's Hilbert space $\delta\psi_t(Z^*)/\delta Z_j^*(s) = Q(t,s,Z^*)\psi_t(Z^*)$. For certain simple models, this ansatz is exact and the Q-operator $Q(t,s,Z^*)$ is independent of the noise [15]. The latter property can be used to derive a master equation for the reduced density operator (6). However, no feasible scheme for calculating the Q-operator and, hence, solving the fermionic NMQSD equation (9) has been found so far. Therefore, we will present a different approach in the next section that does not rely on the Q-operator ansatz.

2.2 Fermionic Hierarchy of Pure States

In analogy to the bosonic case of Ref. [17] we define auxiliary states[2] using the derivation operators (12)

$$\psi_t^{(\mathbf{k})} := D_{1,t}^{k_1} D_{2,t}^{k_2} \ldots \psi_t = \mathbf{D}_t^{\mathbf{k}}\psi_t. \tag{13}$$

In contrast to HOPS for a harmonic oscillator environment, the order of functional derivative operators is relevant here, since they anti-commute. For the same reason, all auxiliary states with some $k_j > 1$ vanish as expected from the fact that the $D_{i,t}$ are linear combinations of fermionic annihilation operators from the microscopic point of view (8). With this notation the memory integrals in (9) can be written as

$$\psi_t^{(1,0,\ldots)} := D_{1,t}\psi_t, \quad \psi_t^{(0,1,\ldots)} := D_{2,t}\psi_t, \quad \ldots \tag{14}$$

Identifying ψ_t with $\psi_t^{(\mathbf{0})}$ allows us to rewrite (9) as

$$\partial_t\psi_t^{(0)} = -\mathrm{i}H\psi_t^{(0)} + \sum_j L_j Z_j^*(t)\psi_t^{(0)} - \sum_j L_j^\dagger \psi_t^{(\mathbf{e}_j)}. \tag{15}$$

Here, we introduce a more compact notation of (14) using the jth unit vector in $\mathbb{R}^N$ $\mathbf{e}_j$ with N being the number of processes in (9).

In order to obtain the equations of motion for the auxiliary states (13) we assume that the BCFs have an exponential form

$$\alpha_j(t) = p_j \mathrm{e}^{-\gamma_j|t| - \mathrm{i}\Omega_j t}. \tag{16}$$

In the following we use the short hand $w_j = \gamma_j + \mathrm{i}\Omega_j$ for the exponent and refer to the tuple (p_j, w_j) as a "mode". It is easy to generalize to a sum of exponentials similar to the bosonic

[1] The integral boundaries in (9) arise from the specific vacuum initial conditions (5); see [17, Footnote 42] for details.

[2] To obtain dimensionless auxiliary states, one can absorb the dimension of the derivative operators $D_{j,t}^{k_j}$ into the system's coupling operators L, which then has the dimension of energy. To be consistent, one also has to rescale Z_t^* accordingly.

case [17]. Such BCFs arise naturally for many models in the finite-temperature case $T > 0$ as discussed in Sect. 2.4.

For such exponential BCFs we obtain (see Appendix 1 for the details) the hierarchy

$$\partial_t \psi_t^{(\mathbf{k})} = \left(-\mathrm{i}H - \mathbf{k}\cdot\mathbf{w} + (-1)^{|\mathbf{k}|} \sum_j Z_j^*(t) L_j \right) \psi_t^{(\mathbf{k})} + \sum_j (-1)^{|\mathbf{k}|_j} p_j L_j \psi_t^{(\mathbf{k}-\mathbf{e}_j)} - \sum_j (-1)^{|\mathbf{k}|_j} L_j^\dagger \psi_t^{(\mathbf{k}+\mathbf{e}_j)} \tag{17}$$

Here, we have introduced $\mathbf{k} = (k_1, \ldots, k_N)$, $\mathbf{w} = (w_1, \ldots, w_N)$, and $\mathbf{k}\cdot\mathbf{w} = k_1 w_1 + \cdots + k_N w_N$. Furthermore, we use the notation $|\mathbf{k}| = k_1 + \cdots + k_J$ for the sum over all k_j and $|\mathbf{k}|_j = k_{j+1} + \cdots + k_N$ denotes the sum *omitting* the first j components. In (17) all states with some $k_j \notin \{0, 1\}$ vanish as mentioned below Eq. (13). The initial conditions (5) translate to

$$\psi_0^{(0)} = \psi_0 \quad \text{and} \quad \psi_0^{(\mathbf{k})} = 0 \text{ for } \mathbf{k} \neq 0. \tag{18}$$

Equation (17) is our first important result of this paper. It will serve as a starting point for the derivation of a density matrix hierarchy. Although, an exponential BCF (16) corresponds to an environment of infinitely many fermions, the hierarchy (17) is completely equivalent to the unitary time evolution of the system and its environment.

Note that (17) is a *finite* system of 2^N coupled equations (as before, N denotes the number of modes). This is in marked contrast to the bosonic case of Ref. [17], where one has an *infinite* system of equations, which has to be truncated for all practical purposes. Although now the system (17) is finite, for a large number of modes N it becomes intractable large for numerical simulations quickly. Therefore, as in the bosonic case, it is useful to truncate the system in an appropriate way. One possibility would be to exclude all states with $|\mathbf{k}| > \mathcal{K}$, where $\mathcal{K}$ is called the truncating order. Comparing calculations with increasing order allows for a systematic check of convergence. Another possible truncation criterion is $|\mathbf{w}\cdot\mathbf{k}| > \mathcal{W}$, where $\mathcal{W}$ is a "maximal energy". However, even with a suitable truncation, direct simulation as for the bosonic HOPS is problematic since Grassmannian stochastic processes are hard to simulate.

2.3 Fermionic Master Equation Hierarchy

From the stochastic trajectories we obtain the reduced density operator by (see Ref. [15], Eq. (17))

$$\rho_t = \mathbb{E}\big(|\psi_t(Z^*)\rangle\langle\psi_t(-Z^*)|\big) = \mathbb{E}\big(|\psi_t(Z^*)\rangle\langle\tilde{\psi}_t(Z^*)|\big), \tag{19}$$

where we have defined $\tilde{\psi}_t(Z^*) = \psi_t(-Z^*)$. Put into words: $\tilde{\psi}_t(Z^*)$ is the solution of (9) evaluated at $-Z^*$. Therefore, for each realization $\psi_t(Z^*)$ one would have to compute $\psi_t(-Z^*)$ as well by propagating (17) with Z^* replaced by $-Z^*$.

Our aim is to get rid of the problematic noise processes in (17) by constructing a hierarchy of density operators similar to that for the pure states (13). To this end, we introduce auxiliary density operators by

$$\rho_t^{(\mathbf{m},\mathbf{n})} = \mathbb{E}\left(|\psi_t^{(\mathbf{m})}\rangle\langle\tilde{\psi}_t^{(\mathbf{n})}|\right) \tag{20}$$

with $\tilde{\psi}_t^{(\mathbf{n})}(Z^*) = \psi_t^{(\mathbf{n})}(-Z^*)$. Using the equations of motion (17) we find

$$\begin{aligned}
\partial_t \rho_t^{(\mathbf{m},\mathbf{n})} = &-\mathrm{i}\left[H, \rho_t^{(\mathbf{m},\mathbf{n})}\right] - (\mathbf{m}\cdot\mathbf{w} + \mathbf{n}\cdot\mathbf{w}^*)\rho_t^{(\mathbf{m},\mathbf{n})} \\
&+ (-1)^{|\mathbf{m}|}\sum_j L_j \mathbb{E}\left(Z_j^*(t)|\psi_t^{(\mathbf{m})}\rangle\langle\tilde{\psi}_t^{(\mathbf{n})}|\right) \\
&+ (-1)^{|\mathbf{n}|}\sum_j \mathbb{E}\left(|\psi_t^{(\mathbf{m})}\rangle\langle\tilde{\psi}_t^{(\mathbf{n})}|(-Z_j(t))\right) L_j^\dagger \\
&+ \sum_j \left((-1)^{|\mathbf{m}|_j} p_j L_j \rho_t^{(\mathbf{m}-\mathbf{e}_j,\mathbf{n})} + (-1)^{|\mathbf{n}|_j} p_j^* \rho_t^{(\mathbf{m},\mathbf{n}-\mathbf{e}_j)} L_j^\dagger\right) \\
&- \sum_j \left((-1)^{|\mathbf{m}|_j} L_j^\dagger \rho_t^{(\mathbf{m}+\mathbf{e}_j,\mathbf{n})} + (-1)^{|\mathbf{n}|_j} \rho_t^{(\mathbf{m},\mathbf{n}+\mathbf{e}_j)} L_j\right)
\end{aligned} \tag{21}$$

This equation still contains the Grassmann processes in the averages. However, these can be eliminated using the Grassmannian Novikov theorem (see Ref. [15] and Appendix 2). Finally, we obtain the following hierarchy of density operators

$$\begin{aligned}
\partial_t \rho_t^{(\mathbf{m},\mathbf{n})} = &-\mathrm{i}\left[H, \rho_t^{(\mathbf{m},\mathbf{n})}\right] - \left(\mathbf{m}\cdot\mathbf{w} + \mathbf{n}\cdot\mathbf{w}^*\right)\rho_t^{(\mathbf{m},\mathbf{n})} \\
&+ \sum_j (-1)^{|\mathbf{m}|_j} p_j L_j \rho_t^{(\mathbf{m}-\mathbf{e}_j,\mathbf{n})} + \sum_j (-1)^{|\mathbf{n}|_j} p_j^* \rho_t^{(\mathbf{m},\mathbf{n}-\mathbf{e}_j)} L_j^\dagger \\
&- \sum_j \left((-1)^{|\mathbf{m}|_j} L_j^\dagger \rho_t^{(\mathbf{m}+\mathbf{e}_j,\mathbf{n})} - (-1)^{|\mathbf{n}|} \rho_t^{(\mathbf{m}+\mathbf{e}_j,\mathbf{n})} L_j^\dagger\right) \\
&+ \sum_j \left((-1)^{|\mathbf{m}|} L_j \rho_t^{(\mathbf{m},\mathbf{n}+\mathbf{e}_j)} - (-1)^{|\mathbf{n}|_j} \rho_t^{(\mathbf{m},\mathbf{n}+\mathbf{e}_j)} L_j\right)
\end{aligned} \tag{22}$$

This constitutes the second main result of the present work. As for the stochastic fermionic hierarchy (17), this is a finite set of 2^{2N} equations. The initial conditions follow trivially from the initial conditions for the pure states hierarchy (18) and the definition of the auxiliary density operators (20):

$$\rho_0^{(\mathbf{0},\mathbf{0})} = |\psi_0\rangle\langle\psi_0| \quad \text{and} \quad \rho_0^{(\mathbf{m},\mathbf{n})} = 0 \text{ for } \mathbf{m},\mathbf{n} \neq \mathbf{0}. \tag{23}$$

For numerical purposes it might again be advantageous to use a suitable truncation. Also, the accuracy of a truncated hierarchy can be increased by approximating truncated auxiliary operators using a so called terminator [17].

2.4 Thermal Initial State of the Bath

Clearly, the pure state hierarchy (17)—and therefore also the density operator hierarchy (22)—depends crucially on the bounded domains of the memory integrals. These only arise for an initial vacuum state of the bath (5) as indicated in the footnote on page 6. Remarkably, it is possible to map the equations of motion corresponding to an initial thermal state of the bath

$$\rho_{\mathrm{tot}}(t=0) = |\psi_0\rangle\langle\psi_0| \otimes \rho_{\mathrm{th}} \tag{24}$$

to the established zero-temperature NMQSD equation (9). Here, the thermal bath state for temperature T is given by $\rho_{\mathrm{th}} = \mathrm{e}^{-\frac{H_{\mathrm{env}}-\mu N_{\mathrm{env}}}{T}}/\mathcal{Z}$, with the chemical potential μ and the partition function $\mathcal{Z} = \mathrm{Tr}_{\mathrm{env}}\,\mathrm{e}^{-\frac{H_{\mathrm{env}}-\mu N_{\mathrm{env}}}{T}}$. Note that ρ_{tot} in (24) describes an uncorrelated state of the system and the bath and, therefore, is not the genuine thermal equilibrium state of the total system, i.e. with respect to H_{tot} in Eq. (1). By doubling the degrees of freedom using the well-known Bogoliubov transformation [14,24,25] the resulting NMQSD equation reads [16]

$$\begin{aligned}\partial_t\psi_t = &-\mathrm{i}H\psi_t + \sum_j L_j Z_j^*(t)\psi_t + \sum_j L_j^\dagger W_j^*(t)\psi_t \\ &- \sum_j L_j^\dagger \int_0^t \alpha_j(t-s)\frac{\delta\psi_t}{\delta Z_j^*(s)}\,\mathrm{d}s \\ &- \sum_j L_j \int_0^t \beta_j(t-s)\frac{\delta\psi_t}{\delta W_j^*(s)}\,\mathrm{d}s,\end{aligned} \tag{25}$$

where we now have another auxiliary process $W_j^*(t)$ for each original process $Z_j^*(t)$. The correlation functions $\alpha_j(t)$ and $\beta_j(t)$ in (25) also characterize these noise processes:

$$\mathbb{E}\left(Z_j(t)Z_j^*(s)\right) = \alpha_j(t-s) = \int_0^\infty J_j(\omega)(1-\bar{n}_j(\omega))\mathrm{e}^{-\mathrm{i}\omega(t-s)}\,\mathrm{d}\omega \tag{26}$$

$$\mathbb{E}\left(W_j(t)W_j^*(s)\right) = \beta_j(t-s) = \int_0^\infty J_j(\omega)\bar{n}_j(\omega)\mathrm{e}^{\mathrm{i}\omega(t-s)}\,\mathrm{d}\omega, \tag{27}$$

with all other relations being zero similar to (11). Here, $\bar{n}_j(\omega)$ denotes the Fermi-Dirac distribution function (now with a possibly different chemical potential for each independent bath) $\bar{n}_j(\omega) = (\mathrm{e}^{(\omega-\mu_j)/T}+1)^{-1}$.

In the case of self-adjoint coupling operators $L_j = L_j^\dagger$ one can go on step further and combine each $Z_j^*(t)$ and $W_j^*(t)$ into the sum processes $\tilde{Z}_j^*(t) = Z_j^*(t) + W_j^*(t)$. Remarkably, the corresponding correlation function

$$\tilde{\alpha}_j(t) = \alpha_j(t) + \beta_j(t) = \int_0^\infty J_j(\omega)\left\{\cos(\omega t) - \mathrm{i}\tanh\left(\frac{\omega}{2T}\right)\sin(\omega t)\right\}\mathrm{d}\omega \tag{28}$$

is also the well known thermal correlation function for a spin bath [1]. In conclusion, the resulting finite temperature NMQSD equation in this case is identical to the zero-temperature version (9) except for the thermal BCF.

Besides incorporating the effects of finite temperature on the system's dynamics, the thermal BCF (28) also provides a natural way to obtain the crucial decomposition of the BCF as a sum of exponentials. Here, we will only sketch the idea following the detailed exposition for a bosonic environment in Ref. [26]: Due to the symmetric behavior under reflection at the origin of the term in braces in (28), a symmetric continuation

$$\tilde{J}(\omega) = \begin{cases} J(\omega) & : \omega \geq 0 \\ J(-\omega) & : \omega < 0 \end{cases} \tag{29}$$

lets us expand the integral boundaries in (28) to the whole real axis

$$\alpha(t) = \frac{1}{2}\int_{-\infty}^{\infty} \tilde{J}(\omega)\left\{\cos(\omega t) - \mathrm{i}\tanh\left(\frac{\omega}{2T}\right)\sin(\omega t)\right\}\mathrm{d}\omega. \tag{30}$$

Closing the integral contour in the upper/lower complex half plane and employing the residual theorem yields exactly the sought after sum of exponentials provided we can express the integrand as a sum of poles. A BCF given as a sum of exponentials like (16) can then be treated completely analogous to the zero-temperature case. Many realistic spectral densities can be well approximated as a finite sum of poles [25,27–29]. Combined with a suitable sum-over-poles scheme for the hyperbolic tangent, i.e. the Matsubara decomposition [30], continued fraction expansion [31], or the Padé decomposition [32], one obtains the BCF as a sum of exponentials.[3] We also note that systems coupled to non-Markovian environments with exponential BCFs are sometimes treated within the framework of "pseudo modes" [5,33–35], where the system is amended by suitably chosen oscillators, that are coupled to Markovian environments.

3 Bosonic Environments

The bosonic microscopical Hamiltonian is identical to (1)—the Hamiltonian used in the previous section—except that the environment's creation and annihilation operators $b^\dagger_{j,\lambda}$ and $b_{j,\lambda}$ are replaced by their bosonic counterparts. In other words, the anti-commutators in Eq. (2) are replaced by commutators

$$\left[b_{j,\lambda}, b_{j',\lambda'}\right] = 0 \quad \text{and} \quad \left[b_{j,\lambda}, b^\dagger_{j',\lambda'}\right] = \delta_{jj'}\delta_{\lambda\lambda'}. \tag{31}$$

Without any approximation, such a model leads to the well-known NMQSD equation [11–14], which agrees with Eq. (9) except for the noise Z^*: In contrast to the fermionic case, the noise process in the bosonic equation is complex valued and, therefore, efficiently implementable for large classes of BCFs [36].

3.1 Bosonic Hierarchy of Pure States

Due to the similarities of the underlying NMQSD equations, the fermionic (Eq. (17)) and bosonic hierarchy of pure states [17, Eq. (13)]

$$\begin{aligned}\partial_t \psi_t^{(\mathbf{k})} = &\left(-\mathrm{i}H - \mathbf{k}\cdot\mathbf{w} + \sum_j Z_j^*(t) L_j\right)\psi_t^{(\mathbf{k})} \\ &+ \sum_j k_j p_j L_j \psi_t^{(\mathbf{k}-\mathbf{e}_j)} - \sum_j L_j^\dagger \psi_t^{(\mathbf{k}+\mathbf{e}_j)}\end{aligned} \tag{32}$$

are remarkably similar. Besides the obvious sign prefactors in (17) and the different noise processes, the crucial difference is number of auxiliary states: Whereas the fermionic hierarchy is always finite due to the condition $k_j \in \{0, 1\}$, its bosonic counterpart is originally infinite with $k_j \in \mathbb{N}_0$. Only a suitable truncation turns the latter into a practical scheme [17].

3.2 Bosonic Master Equation Hierarchy

Clearly, the similarities of the two pure state hierarchies carries over to the hierarchy of density matrices: The construction of the *bosonic master equation hierarchy*

[3] Of course, one can also try to approximate the BCF directly by a sum of exponentials.

$$\begin{aligned}\partial_t \rho_t^{(\mathbf{m},\mathbf{n})} &= -\mathrm{i}\left[H, \rho_t^{(\mathbf{m},\mathbf{n})}\right] - \left(\mathbf{m}\cdot\mathbf{w} + \mathbf{n}\cdot\mathbf{w}^*\right)\rho_t^{(\mathbf{m},\mathbf{n})} \\ &+ \sum_j m_j p_j L_j \rho_t^{(\mathbf{m}-\mathbf{e}_j,\mathbf{n})} + \sum_j n_j p_j^* \rho_t^{(\mathbf{m},\mathbf{n}-\mathbf{e}_j)} L_j^\dagger \\ &- \sum_j \left[L_j^\dagger, \rho_t^{(\mathbf{m}+\mathbf{e}_j,\mathbf{n})}\right] + \sum_j \left[L_j, \rho_t^{(\mathbf{m},\mathbf{n}+\mathbf{e}_j)}\right]\end{aligned} \tag{33}$$

runs along the same lines as the derivation in Sect. 2.2. In contrast to Eq. (20), the density operators arise without a reflection of the noise for the $\langle\psi_t^{(\mathbf{n})}(Z^*)|$, i.e.

$$\rho_t^{(\mathbf{m},\mathbf{n})} = \mathbb{E}\left(|\psi_t^{(\mathbf{m})}\rangle\langle\psi_t^{(\mathbf{n})}|\right). \tag{34}$$

Here, the indices $m_j, n_j \in \mathbb{N}_0$ are not bounded from above and, therefore, the hierarchy is infinite in principle and has to be truncated for numerical simulations. Note the close relation between the fermionic and bosonic equations: besides the prefactors m_j and n_j in (33), which are irrelevant for $m_j, n_j \in \{0, 1\}$, the two hierarchies only differ in the additional sign factors $(-1)^{|\mathbf{m}|}$ and $(-1)^{|\mathbf{n}|}$ in (22). Furthermore, Eq. (33) is solved with the same initial conditions (23) as the fermionic hierarchy. For particular systems, our hierarchy Eq. (33) is similar to the ones derived in [37,38].

3.3 Relation to the Hierarchical Equation of Motion (HEOM)

For *Hermitian* system coupling operators and *real* ω_j, one can simplify Eq. (33) considerably by introducing new auxiliary operators that combine operators $\rho_t^{(\mathbf{m},\mathbf{n})}$ with the same order $|\mathbf{m}| + |\mathbf{n}|$:

$$\rho_t^{(\mathbf{n})} = (-\mathrm{i})^{|\mathbf{n}|} \sum_{l_1=0}^{n_1} \cdots \sum_{l_N=0}^{n_N} (-1)^{|\mathbf{l}|} \binom{n_1}{l_1} \cdots \binom{n_N}{l_N} \rho_t^{(\mathbf{n}-\mathbf{l},\mathbf{l})} \tag{35}$$

Under the stated assumptions, these distinct combinations of auxiliary operators satisfy closed equations of motion obtained from (33), namely

$$\begin{aligned}\dot{\rho}_t^{(\mathbf{n})} &= -\mathrm{i}[H, \rho_t^{(\mathbf{n})}] - \mathbf{n}\cdot\mathbf{w}\rho_t^{(\mathbf{n})} - \mathrm{i}\sum_j n_j \left(p_j L_j \rho_t^{(\mathbf{n}-\mathbf{e}_j)} - \rho_t^{(\mathbf{n}-\mathbf{e}_j)} L_j p_j^*\right) \\ &-\mathrm{i}\sum_j \left[L_j, \rho_t^{(\mathbf{n}+\mathbf{e}_j)}\right]\end{aligned} \tag{36}$$

This already resembles the hierarchical equations of motion [19, Eq. (4.4)] very closely. The two equations agree completely if we employ the exponential decomposition of the BCF used in the derivation of HEOM, namely a Matsubara expansion for the coth and a Drude-Lorentz spectral density. However, we typically prefer to use a Padé expansion of the coth as described in e.g. Refs. [25,26].

4 Numerical Examples

In order to demonstrate the feasibility and strengths of the master equation hierarchy and also highlight some of its shortcomings compared to the HOPS, we will provide some numerical examples in this section. For that matter, we choose the relatively simple spin-boson model [39], which nevertheless can displays highly non-Markovian behavior and is therefore well suited for benchmarking. This model consists of a two-level system with a free time

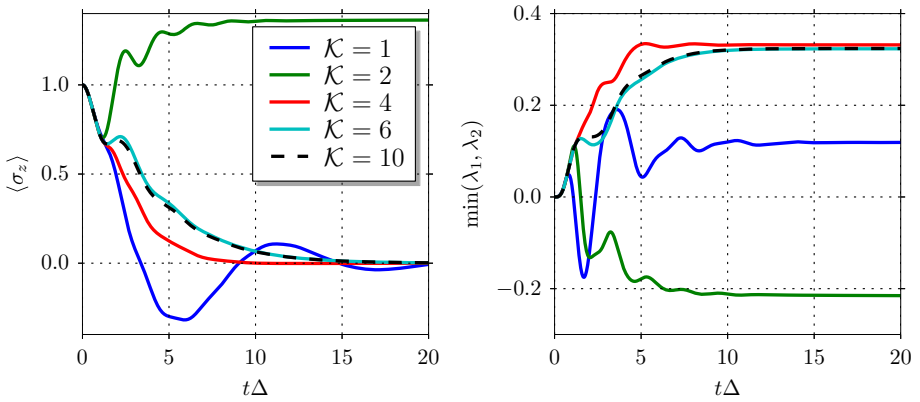

Fig. 1 Time evolution of the spin-boson model in the non-Markovian regime with $\epsilon = 0$, $p = 2\Delta$, $\gamma = \Delta/2$, and $\Omega = 2\Delta$ calculated using the master equation hierarchy (33) for different orders of the truncation order $\mathcal{K}$. We show the expectation value of the system's σ_z (*left*) and the smaller eigenvalue of the reduced density operator (*right*). Going to orders larger than $\mathcal{K} = 10$ (*black dashed line*) did not lead to an appreciable change in the results

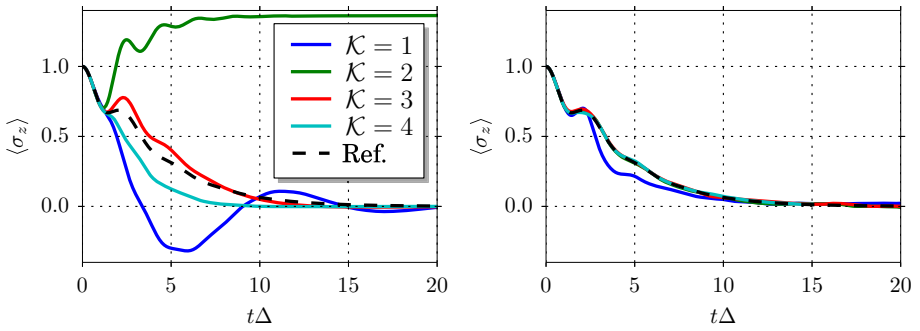

Fig. 2 Comparison of the master equation hierarchy (*left*) and the HOPS (*right*). The same parameters as in Fig. 1 are chosen. The HOPS results is averaged over 10,000 realizations. As a reference, the master equation hierarchy result of Fig. 1 with $\mathcal{K} = 10$ is shown

evolution governed by $H = -\frac{\Delta}{2}\sigma_x + \frac{\epsilon}{2}\sigma_z$ coupled to an environment of harmonic oscillators. We employ a single-mode bath correlation function $\alpha(t) = p \exp(-\gamma|t| - \mathrm{i}\Omega t)$ and a coupling operator $L = \sigma_z$. In order to truncate the (generally infinite) hierarchy (33), we set all auxiliary states $\rho_t^{(\mathbf{m},\mathbf{n})}$ with $|\mathbf{m}| + |\mathbf{n}| > \mathcal{K}$ to zero.

In Fig. 1 we show the results of the master equation hierarchy for different values of the truncation order $\mathcal{K}$. Clearly, the lowest order calculations give unphysical results: The expectation value of σ_z for $\mathcal{K} = 2$ even converges to a limit greater than one. This is due to the reduced density operator ρ_t not being positive semidefinite, as one can see on the right hand side. For $\mathcal{K} = 1, 2$ the smallest eigenvalue of ρ_t becomes negative for some times. Only for higher orders does the reduced density operator become admissible, and the result for truncation order 6 is a good approximation to the exact result.

In contrast, the HOPS converges fast with respect to the truncation order as can be seen in Fig. 2. The results obtained from HOPS can be considered converged already at truncation order $\mathcal{K} = 2$. From the construction of the auxiliary density matrices in Eq. (34) it is clear that the pure state hierarchy contains higher order effects compared to the master equation hierar-

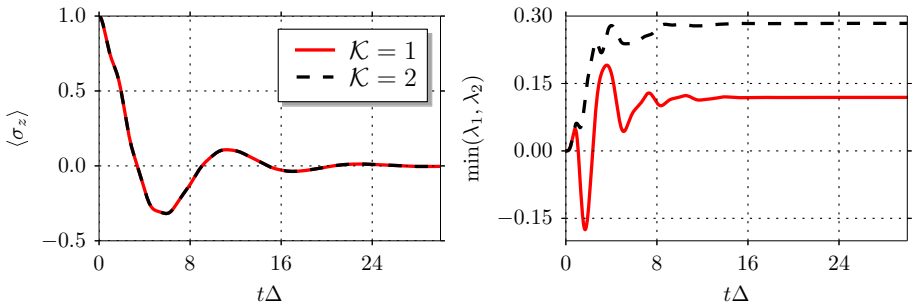

Fig. 3 Same as Fig. 1, but for a fermionic environment using the master equation hierarchy (22). Note that $\mathcal{K} = 2$ is exact up to numerical errors

chy at same truncation order. Finally, the reduced density operator obtained as an ensemble average over quantum trajectories $\mathbb{E}|\psi_t(Z^*)\rangle\langle\psi_t(Z^*)|$ will always be positive semidefinite.

The fermionic case, where the stochastic pure state formulation is not feasible for numeric implementation, is shown in Fig. 3. Since we only treat the case of a single exponential mode, there are only two nontrivial values for the truncation order, namely $\mathcal{K} = 1, 2$. Although the smallest eigenvalue in the truncated first-order case also becomes negative and deviates from the exact case ($\mathcal{K} = 2$), the expectation values of σ_z agree for both.

5 Conclusions

In the present work we have considered an open system model with a fermionic environment which is coupled linearly to the system. The two main results are the derivation of a hierarchy of pure states (17) and the corresponding hierarchy for density matrices (22). The starting point was the general stochastic NMQSD equation that contains Grassmannian noise as well as a functional derivative with respect to this noise under a memory integral. The hierarchy of pure states (17) no longer contains this functional derivative, however, it still contains the Grassmann noise, which hinders efficient numerical simulations. In contrast, the hierarchy for density matrices (22) also gets rid of the noise making it suitable for numerical simulations.

Both, the hierarchy of pure states (17) and the hierarchy for density matrices (22), are actually finite and allow to compute the reduced density operator of the system exactly. However, the number of coupled differential equations scales as 2^N for the pure state hierarchy and as 2^{2N} for the hierarchy for density matrices. Note that the objects entering these equations have the dimension D of the system Hilbert space in the case of the pure state hierarchy and D^2 for the matrix hierarchy. Since for a large number of modes N the size of the problem becomes numerically intractable, it is necessary to truncate the hierarchy in a suitable way. This can be done along the lines discussed at the end of Sect. 2.2.

In the second part of the paper we showed that the construction of a density operator hierarchy from a hierarchy of pure states is also feasible for bosonic environments. Since in this bosonic case, both, Eq. (32) and (33) are suitable for numerical simulations, a detailed comparison of their performance is needed to asses their respective strengths and weaknesses. However, the brief comparison in Sect. 4 indicates that HOPS converges faster with respect to the truncation order. Furthermore, it always yields physical (i.e., positive semidefinite) reduced density operators. The main disadvantage of HOPS, namely the necessity to calculate

many realizations, is easily coped with in practice since the computation of independent realizations is embarrassingly parallel.

Within the approaches presented in this work it is also possible to treat systems coupled to both types of environments. Generalizations to multiple distinguishable kinds of fermions or to a spin bath (e.g. arising as low-temperature limit of localized modes [40]) are possible as well. This makes the hierarchical approach presented in this a paper a highly flexible tool in the field of open quantum system dynamics.

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Appendix 1: Derivation of the Fermionic Hierarchy of Pure States

The derivation for the fermionic hierarchy of pure states is very similar to that of the bosonic one discussed in [17]. To have a compact notation, we ignore the condition $k_j \in \{0, 1\}$ for our derivation. In the end, it will turn out that these conditions are trivially incorporated due to the structure of the hierarchy.

We start by taking the time derivative of $\psi_t^{(\mathbf{k})}$. Using its definition (13) we find

$$\partial_t \psi_t^{(\mathbf{k})} = (\partial_t \mathbf{D}_t^{\mathbf{k}})\psi_t + \mathbf{D}_t^{\mathbf{k}}(\partial_t \psi_t) \tag{37}$$

For the first term on the right hand side we use[4] $(\partial_t D_{j,t})\psi_t = -w_j D_{j,t}\psi_t$ due to the exponential BCF (16). For the second term on the right hand side we use that all system operators commute with all $D_{j,t}$ and obtain

$$\begin{aligned}\partial_t \psi_t^{(\mathbf{k})} &= -\mathbf{k}\cdot\mathbf{w}\,\psi_t^{(\mathbf{k})} \\ &\quad -\mathrm{i}H\psi_t^{(\mathbf{k})} + \underbrace{\sum_j L_j \mathbf{D}_t^{\mathbf{k}} Z_j^*(t)\psi_t}_{(*)} - \underbrace{\sum_j L_j^\dagger \mathbf{D}_t^{\mathbf{k}} D_{j,t}\psi_t}_{(**)}\end{aligned} \tag{38}$$

To obtain a closed equation for the auxiliary states, we want the $D_{j,t}$ ordered as in the definition (13). In $(**)$ we have to move $D_{j,t}$ to the correct position (note the ordering in (13)):

$$\begin{aligned}\mathbf{D}_t^{\mathbf{k}} D_{j,t} &= (-1)^{k_N} D_{1,t}^{k_1} \ldots D_{j,t} D_{N,t}^{k_N} \\ &= (-1)^{k_{j+1}+\cdots+k_N} D_{1,t}^{k_1} \ldots D_{j,t}^{k_j+1} \cdots D_{N,t}^{k_N}.\end{aligned} \tag{39}$$

In $(*)$ we have to bring $Z_j^*(t)$ in front of $\mathbf{D}_t^{\mathbf{k}}$. This can be achieved by noting that $\{D_{j,t}, Z_{j'}^*(s)\} = \delta_{jj'}\alpha_j(t_j - s)$. We then find

[4] The bounded integral domain in the memory integral that appears in the final equation is due to vacuum initial conditions (5).

$$\begin{aligned}
\mathbf{D}_t^{\mathbf{k}} Z_j^*(t) &= (-1)^{k_N} D_{1,t}^{k_1} \dots Z_j^*(t) D_{N,t}^{k_N} \\
&= (-1)^{k_{j+1}+\dots+k_N} D_{1,t} \dots D_{j,t}^{k_j} Z_j^*(t) \dots \\
&= (-1)^{|\mathbf{k}|_j} D_{1,t} \dots \left(-D_{j,t}^{k_j-1} Z_j^*(t) D_{j,t} + D_{j,t}^{k_j-1} p_j \right) \dots \\
&= \dots \left(D_{j,t}^{k_j-2} Z_j^*(t) D_{j,t}^2 - D_{j,t}^{k_j-1} p_j + D_{j,t}^{k_j-1} p_j \right) \dots \\
&= \dots \left((-1)^{k_j} Z_j^*(t) D_{j,t}^{k_j} + (k_j \bmod 2) p_j D_{j,t}^{k_j-1} \right) \dots \\
&= (-1)^{|\mathbf{k}|} Z_j^*(t) \mathbf{D}_t^{\mathbf{k}} + (-1)^{|\mathbf{k}|_j} (k_j \bmod 2) p_j \mathbf{D}_t^{\mathbf{k}-\mathbf{e}_j},
\end{aligned} \tag{40}$$

where $|\mathbf{k}|$ and $|\mathbf{k}|_j$ have been defined below Eq. (17).

Combining (39) and (40) leads to the (apparently) infinite hierarchy of pure states for fermionic environment

$$\begin{aligned}
\partial_t \psi_t^{(\mathbf{k})} = &\left(-\mathrm{i}H - \mathbf{k} \cdot \mathbf{w} + (-1)^{|\mathbf{k}|} \sum_j Z_j^*(t) L_j \right) \psi_t^{(\mathbf{k})} \\
&+ \sum_j (-1)^{|\mathbf{k}|_j} (k_j \bmod 2) p_j L_j \psi_t^{(\mathbf{k}-\mathbf{e}_j)} \\
&- \sum_j (-1)^{|\mathbf{k}|_j} (-1)^{|\mathbf{k}|_j} L_j^\dagger \psi_t^{(\mathbf{k}+\mathbf{e}_j)}.
\end{aligned} \tag{41}$$

Note that all states with some $k_j \notin \{0, 1\}$—which should be zero actually—only couple to other states also satisfying this condition: Due to the modulo function in the term coupling to states "below" in the hierarchy, states with some $k_j \notin \{0, 1\}$ that are initially zero always remain zero. Therefore, the closed and finite hierarchy with all $k_j \in \{0, 1\}$ and equation (41) can be written as (17).

Appendix 2: Derivation of Master Equation Hierarchy

In this appendix we provide the Novikov theorem, which is essential to get from Eqs. (21) to (22). The Novikov theorem allows us to get rid of the explicit dependence of the Grassmann processes in the second and third line of (21) by a "partial integration". For the fermionic case the Novikov theorem has been discussed in [15] (see Eqs. (22) and (23) therein). We need two variants of the Novikov theorem:

$$\begin{aligned}
\mathbb{E}\left(|\psi_t^{(\mathbf{m})}\rangle\langle\tilde{\psi}_t^{(\mathbf{n})}| Z_j(t) \right) &= -\mathbb{E}\left(\int \mathrm{d}s\, \alpha_j(t-s) \frac{\overrightarrow{\delta}}{\delta Z_j^*(s)} |\psi_t^{(\mathbf{m})}\rangle\langle\tilde{\psi}_t^{(\mathbf{n})}| \right) \\
&= -\rho_t^{(\mathbf{m}+\mathbf{e}_j,\mathbf{n})}
\end{aligned} \tag{42}$$

and

$$\begin{aligned}
\mathbb{E}\left(Z_j^*(t) |\psi_t^{(\mathbf{m})}\rangle\langle\tilde{\psi}_t^{(\mathbf{n})}| \right) &= -\mathbb{E}\left(\int \mathrm{d}s\, \alpha_j(t-s)^* \left| \psi_t^{(\mathbf{m})}\rangle\langle\tilde{\psi}_t^{(\mathbf{n})} \right| \frac{\overleftarrow{\delta}}{\delta Z_j(s)} \right) \\
&= \rho_t^{(\mathbf{m},\mathbf{n}+\mathbf{e}_j)},
\end{aligned} \tag{43}$$

where in the second line of each equation we have used the definition of the auxiliary matrices (20) and the definitions (12) and (13). In the second equation the right-functional derivative appears and we have used

$$\frac{\overrightarrow{\delta}\,\tilde{\psi}_t(Z^*)}{\delta Z_j^*(s)} = \frac{\overrightarrow{\delta}\,\psi_t(-Z^*)}{\delta Z_j^*(s)} = -\left.\frac{\overrightarrow{\delta}\,\psi_t(Z'^*)}{\delta Z_j'^*(s)}\right|_{Z'^*=-Z^*}. \tag{44}$$

These two equations (42) and (43) show that it is possible to express the averages in the second and third line of (21) containing the noise process explicitly by the auxiliary density operators.

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