Field-theoretical approach to open quantum systems and the Lindblad equation

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We develop a systematic field-theoretical approach to open quantum systems based on condensed-matter many-body methods. The time evolution of the reduced density matrix for the open quantum system is determined by a transmission matrix. Developing diagrammatic perturbation theory, invoking Wick's theorem in connection with a Caldeira-Leggett quantum oscillator environment in thermal equilibrium, the transmission matrix satisfies a Dyson equation characterized by an irreducible kernel. Unlike the Nakajima-Zwanzig and standard approaches, the Dyson equation is equivalent to a general non-Markovian master equation for the reduced density matrix, incorporating secular effects and independent of the initial preparation. The kernel is determined by a systematic diagrammatic expansion in powers of the interaction. We consider the Born approximation for the kernel. Applying a condensed-matter pole or, equivalently, a quasiparticle-type approximation, equivalent to the usual assumption of a timescale separation, we derive a master equation of the Markov type. Furthermore, imposing the rotating-wave approximation, we obtain a Markov master equation of the Lindblad form. To illustrate the method, we consider the standard example of a single qubit coupled to a thermal heat bath.

I. INTRODUCTION

There is a continuing strong interest in open quantum systems interacting with an environment both from a fundamental and an experimental point of view. The issue is important in many areas of science, spanning from chemistry over atomic, molecular, and optical physics to condensed-matter physics, quantum information, and quantum computers [1–8]. The theory of open systems has in particular been developed in the field of quantum optics [9–11], where atoms or cavity modes are coupled to the radiation field. In recent years, there has been a strong focus on decoherence, entanglement, and dissipation in the context of nano quantum systems and quantum computation [12–18].

There are a variety of theoretical approaches to open quantum systems [9–11,19–31]. Unlike the situation in condensed-matter physics where all degrees of freedom come into play [32,33], open quantum systems have a finite number of degrees of freedom, typically one or several qubits, atomic model systems, or cavity modes interacting with an environment or bath composed of many degrees of freedom. Like in classical statistical physics [34,35], the bath is usually assumed to be unperturbed by the open quantum system; note, however, recent work on system-bath correlations [36]. Defining the bath explicitly as a collection of independent quantum oscillators [37,38], the properties of the bath are characterised by a spectral density and a temperature; the vacuum state corresponds to zero temperature.

The combined system, consisting of the open quantum system and the bath, constitutes a closed quantum system evolving in time according to a unitary transformation [9,39]. On the other hand, the open quantum system evolves according to a nonunitary quantum map due to the interaction with the bath. This interaction gives rise to mixture, dissipation, decoherence, and entanglement [9].

Since a mixture of states is generated by the system-bath coupling, the starting point for an analysis is usually based on the von Neumann equation $d\rho(t)/dt = -i[H, \rho(t)]$ [9,39] for the density operator $\rho(t)$ for the combined closed system; here H is the total Hamiltonian. Subsequently, the density operator for the open quantum system (S), $\rho_S(t)$, is obtained by tracing out the bath (B) degrees of freedom. Consequently, $\rho_S(t) = \text{Tr}_B[\rho(t)]$ and formally $d\rho_S(t)/dt = -i\text{Tr}_B[H, \rho(t)]$; note that throughout the present paper, we set $\hbar = 1$.

In general, the time evolution of the reduced density operator $\rho_S(t)$ is governed by an inhomogeneous integral equation, or, correspondingly, a master equation incorporating memory effects. However, many studies of open quantum systems are based on the Markov assumption in combination with the Born approximation. Similar to the Langevin-Fokker-Planck scheme in classical statistical mechanics [2,35,40], the Markov approximation is based on an assumption of a separation of timescales. The intrinsic timescale of the bath, typically the fall-off time of bath correlations, is assumed to be short in comparison with the timescale associated with the small open quantum system, characteristically given by the inverse level spacing. In this limit, we can ignore memory effects and invoke the Markov approximation [9,41]; note, however, recent work on non-Markovian issues [42–47]. Finally, in the second-order Born approximation it follows that $\rho_S(t)$ is governed by the Lindblad equation [48,49],

$$\frac{d}{dt}\rho_S(t) = -i[H_S, \rho_S(t)]$$
$$+ \sum_{\alpha\beta, k} \gamma_k^{\alpha\beta}\left[S_k^\beta \rho_S(t)S_k^{\alpha\dagger} - \frac{1}{2}\{S_k^{\alpha\dagger}S_k^\beta, \rho_S(t)\}\right].$$

(1.1)

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Here H_S is the Hamiltonian for the open quantum system; note that here we ignore a renormalization of H_S due to a Lamb shift. The coupling to the bath is characterized by the jump operators S_k^α acting on the quantum system together with the positive dissipation coefficients $\gamma_k^{\alpha\beta}$. The commutator term characterizes the unitary time evolution in the absence of coupling to the bath according to the von Neumann equation $d\rho_S(t)/dt = -i[H_S, \rho_S(t)]$. The last term involving the jump operators S_k^α describes the coupling to the bath giving rise to dissipation, incoherence, and entanglement. The standard derivation of the Lindblad equation within the quantum optics approach is based on an iteration of the von Neumann equation to the second-order Born approximation in combination with a separation of timescales, the rotating-wave approximation, and a time coarse graining. The basic assumption is that bath correlations relax on a timescale much faster than the timescale associated with the evolution of the density operator; a standard source is the book by Breuer and Petruccione [9].

In the present paper, we approach the issue of open quantum systems and in particular the Lindblad equation from the point of view of condensed-matter physics using techniques from equilibrium and nonequilibrium many-body theory [32,33,50–53]. We believe that the present many-body approach to an open quantum system, the Markov approximation, and in particular the Lindblad equation sheds light on the assumptions underlying the standard approach [9].

Below we briefly sketch our procedure. The time evolution of the reduced density operator $\rho_S(t)$ describing the open quantum system interacting with the bath can be expressed in the form $\rho_S(t) = T(t, t_i)\rho_S(t_i)$, where $T(t, t_i)$ is a transmission (super)operator transporting the density operator forward in time from an initial time t_i to a final time t; as commonly assumed, the system is uncorrelated with the bath at the initial time t_i. Assuming that the bath is composed of noninteracting quantum oscillators and invoking Wick's theorem, it follows that the transmission operator $T(t, t')$ satisfies a Dyson equation of the form $T(t, t') = T^0(t, t') + \int dt'' dt''' T^0(t, t'') K(t'', t''') T(t''', t')$. Here $T^0(t, t')$ is the unperturbed transmission operator in the absence of coupling to the bath. The irreducible kernel or self-energy $K(t, t')$ can be determined perturbatively in terms of the coupling between the open quantum system and the bath. It then follows from the Dyson equation that the density operator satisfies the master equation $d\rho_S(t)/dt = -i[H_S, \rho_S(t)] + \int dt' K(t, t')\rho_S(t')$. Here H_S is the system Hamiltonian; we note that $T^0(t, t')$ solves the von Neumann equation $d\rho_S(t)/dt = -i[H_S, \rho_S(t)]$. In general, the memory kernel $K(t, t')$ depends on the free evolution of the system, $\exp(-iH_S t)$, and correlations characterizing the bath. In the Born approximation to second order in the system-bath coupling, the kernel $K(t, t')$ consists of four terms including a single bath correlation function. To implement the Markov approximation, we note that the behavior of $\rho_S(t)$ is determined by the transmission operator $T(t, t')$ or in Fourier space the pole structure of $\tilde{T}(\omega)$. From the Dyson equation for $\tilde{T}(\omega)$ we obtain formally $\tilde{T}(\omega)^{-1} = [\tilde{T}^0(\omega)^{-1} - \tilde{K}(\omega)]$, and the resonance structure is determined by $\det[\tilde{T}^0(\omega)^{-1} - \tilde{K}(\omega)] = 0$. Implementing from many-body theory the so-called quasiparticle approximation to leading order in the system-bath coupling by setting

$\det[\tilde{T}^0(\omega)^{-1}] = 0$, we obtain a set of frequencies ω_n. In this approximation, setting $\tilde{K}(\omega) = \tilde{K}(\omega_n)$, we obtain a Markov equation. Imposing furthermore the rotating-wave approximation, we arrive at the Lindblad equation.

Aiming at a self-contained exposition, the paper is organized as follows. In Sec. II we set up the general scheme deriving an expression for the reduced density matrix in terms of a transmission matrix; details are deferred to Appendix 1. In Sec. III we introduce the Caldeira-Leggett heat bath in terms of independent quantum oscillators, and we derive the bath correlation functions. Moreover, we introduce Wick's theorem with a derivation deferred to Appendix 2. Section IV is devoted to a discussion of the transmission matrix and a derivation of the Dyson equation. In Sec. V we present the general master equation for the reduced density matrix following from the Dyson equation. In Sec. VI we derive the general non-Markovian master equation to second order in the coupling between the system and the bath, i.e., the Born approximation. In Sec. VII the Lindblad equation is reviewed. In Sec. VIII we summarize the standard microscopic derivation of the Lindblad equation. In Sec. IX A we present a heuristic derivation of the Lindblad equation, in Sec. IX B a discussion of the pole structure of the transmission matrix, and in Sec. IX C a detailed discussion of the quasiparticle approximation, resulting in a Markovian master equation. In Sec. X A we present a discussion of the equivalence between the field-theoretical approach and the standard derivation. Moreover, we discuss aspects of the timescale separation: in Sec. X B we discuss the approach by Diosi and Ferialdi [42,43] and its relation to the present work, in Sec. X C the Nakajima-Zwanzig approach to a nonMarkovian master equation, and finally in Sec. X D a discussion of the quasiparticle approximation in the simple case of a qubit coupled to a bath. In Sec. XI we give a short summary of our results. In Appendix 1 we discuss the interaction representation, and in Appendix 2 we present a derivation of Wick's theorem.

II. GENERAL ANALYSIS

Here we set up the field-theoretical approach to open quantum systems. Details regarding the interaction representation are deferred to Appendix 1. The methods used are well-known in condensed-matter theory both for equilibrium [32,33,50] and nonequilibrium systems [51–53], but they have not been used much in the context of open quantum systems [9–11]; see, however, recent work in [54,55]. In condensed-matter theory, the essential building block is the Green's function or propagator in the context of diagrammatic or functional perturbative expansions. These schemes have been developed both for equilibrium and nonequilibrium systems.

For open quantum systems, the standard tool is the reduced density operator $\rho_S(t)$ obtained by tracing out the degrees of freedom of the environment. Expectation values of observables A_S relating to the open quantum system are thus given by $\langle A_S \rangle(t) = \text{Tr}_S[\rho_S(t)A_S]$, which together with the regression theorem provide the standard tools in quantum optics [9,11] and related fields. In discussing open quantum systems, the starting point is a small system (S) with a finite number of degrees of freedom, typically one or several qubits, an atomic

system or a cavity mode characterized by a quantum oscillator, interacting with a large quantum system acting as a bath (B) or an environment composed of many or infinitely many degrees of freedom. The total system composed of the open quantum system interacting with the quantum bath is thus described by the Hamiltonian

$$H = H_S + H_B + H_{SB}, \tag{2.1}$$

$$H_{SB} = \sum_\alpha S^\alpha B^\alpha = \boldsymbol{S} \cdot \boldsymbol{B}. \tag{2.2}$$

Here H_S is the Hamiltonian for the quantum system, H_B is the Hamiltonian for the bath, and H_{SB} is the interaction describing the coupling between the quantum system and the bath. The system and the bath live in separate Hilbert spaces, i.e., $[H_B, H_S] = 0$; however, due to the interaction we have $[H_S, H_{SB}] \neq 0$ and $[H_B, H_{SB}] \neq 0$. S^α are operators acting on the quantum system, and B^α are operators monitoring the bath; $\boldsymbol{S} \cdot \boldsymbol{B}$ denotes the usual scalar product. Since $H_{SB}^\dagger = H_{SB}$, we have $\sum_\alpha S^\alpha B^\alpha = \sum_\alpha S^{\alpha\dagger} B^{\alpha\dagger}$; note that this relation does not imply that S^α or B^α individually are Hermitian.

The coupling between the quantum system and the bath gives rise to mixed states, relaxation, decoherence, and entanglement between system and bath, and it is customary to use the density operator formalism. The density operator has the form $\rho(t) = \sum_n P_n |\Psi_n(t)\rangle \langle \Psi_n(t)|$, where the probabilities $P_n \geqslant 0$ and $\sum_n P_n = 1$ [56]. The density operator is Hermitian, positive, and has unit trace, i.e., $\rho(t)^\dagger = \rho(t)$, $\langle \Phi | \rho(t) | \Phi \rangle > 0$, and $\mathrm{Tr}[\rho(t)] = 1$. The purity of a quantum state is defined as $\gamma(t) = \mathrm{Tr}[\rho(t)^2]$; for a pure state, $\gamma = 1$; for a mixed state, $\gamma < 1$.

It follows from the Schrödinger equation, $id|\Psi(t)\rangle/dt = H|\Psi(t)\rangle$ [39], that the density operator satisfies the von Neumann equation [56,57]

$$\frac{d}{dt}\rho(t) = -i[H, \rho(t)], \tag{2.3}$$

with formal solution

$$\rho(t) = U(t, t_i)\rho(t_i)U(t, t_i)^\dagger, \tag{2.4}$$

$$U(t, t') = \exp\left(-iH(t - t')\right). \tag{2.5}$$

Here $\rho(t_i)$ is the density operator at an initial time t_i, and $U(t, t')$ is the usual unitary evolution operator. From (2.4) we infer the density operator for the system $\rho_S(t)$ by tracing over

the bath states, i.e.,

$$\rho_S(t) = \mathrm{Tr}_B[\rho(t)]. \tag{2.6}$$

In recent work [36], the issue of correlations between system and bath has been addressed. However, in the present context we assume that the bath and system are uncorrelated at an initial time t_i and that the bath composed of many degrees of freedom is maintained in a stationary thermodynamic state characterized by the density operator $\rho_B = \exp(-\beta H_B)/\mathrm{Tr}[\exp(-\beta H_B)]$ [35]; here β is the inverse temperature, and the vacuum state corresponds to $\beta = \infty$. The initial condition is thus given by the factorized density operator

$$\rho(t_i) = \rho_B \rho_S(t_i). \tag{2.7}$$

In terms of a superoperator or Liouville operator L, the time evolution in (2.4) can be written in the form $\rho(t) = L(t, t_i)\rho(t_i)$, where in a complete basis $|n\rangle$ we have $\rho(t)_{pp'} = \sum_{qq'} L_{pp',qq'}(t, t_i)\rho(t_i)_{qq'}$ and $L_{pp',qq'}(t, t_i) = U(t, t_i)_{pq}U(t, t_i)^\dagger_{q'p'}$. Likewise, due to the linearity, the time evolution of the reduced density operator $\rho_S(t) = \mathrm{Tr}_B[L(t, t_i)\rho_B \rho_S(t_i)]$ for the system can be expressed in a similar form,

$$\rho_S(t) = T(t, t_i)\rho_S(t_i), \tag{2.8}$$

$$\rho_S(t)_{pp'} = \sum_{qq'} T(t, t_i)_{pp',qq'}\rho_S(t_i)_{qq'}, \tag{2.9}$$

where $T(t, t_i)$ is a transmission (super) operator determining the time evolution of $\rho_S(t)$.

To proceed systematically, we turn to an expansion of the evolution operator $U(t, t_i)$ in powers of the system-bath interaction H_{SB}. Introducing the interaction representation [32,33] with respect to the unperturbed system and bath, we set

$$H_0 = H_S + H_B, \tag{2.10}$$

and we obtain in the interaction picture

$$H_{SB}(t) = \exp(iH_0 t)H_{SB}\exp(-iH_0 t), \tag{2.11}$$

$$\boldsymbol{B}(t) = \exp(iH_B t)\boldsymbol{B}\exp(-iH_B t), \tag{2.12}$$

$$\boldsymbol{S}(t) = \exp(iH_S t)\boldsymbol{S}\exp(-iH_S t). \tag{2.13}$$

Expanding $U(t, t')$ and the Hermitian conjugate $U(t, t')^\dagger$ in powers of $H_{SB}(t)$, we have

$$U(t, t') = +i\sum_{n=0}\int dt_n dt_{n-1}\cdots dt_1 G_R(t, t_n)\boldsymbol{S}_n G_R(t_n, t_{n-1})\boldsymbol{S}_{n-1}\cdots \boldsymbol{S}_2 G_R(t_2, t_1)\boldsymbol{S}_1 G_R(t_1, t')$$

$$\times e^{-iH_B t}\boldsymbol{B}_n(t_n)\boldsymbol{B}_{n-1}(t_{n-1})\cdots \boldsymbol{B}_2(t_2)\boldsymbol{B}_1(t_1)e^{iH_B t'}, \tag{2.14}$$

$$U(t, t')^\dagger = -i\sum_{n=0}\int du_n du_{n-1}\cdots du_1 G_A(t', u_1)\boldsymbol{S}_1 G_A(u_1, u_2)\boldsymbol{S}_2\cdots \boldsymbol{S}_{n-1}G_A(u_{n-1}, u_n)\boldsymbol{S}_n G_A(u_n, t)$$

$$\times e^{-iH_B t'}\boldsymbol{B}_1(u_1)\boldsymbol{B}_2(u_2)\cdots \boldsymbol{B}_{n-1}(u_{n-1})\boldsymbol{B}_n(u_n)e^{iH_B t}, \tag{2.15}$$

where we have used $S \cdot B = (S \cdot B)^\dagger$, introduced $H_{SB}(t_p) = \exp(iH_S t_p) S_p \exp(-iH_S t_p) B_p(t_p)$, and the retarded and advanced system Green's functions

$$G_R(t, t') = -i\eta(t - t') \exp(-iH_S(t - t')), \quad (2.16)$$

$$G_A(t, t') = +i\eta(t' - t) \exp(-iH_S(t - t')); \quad (2.17)$$

here the step function is given by $\eta(t) = 1$ for $t > 0$ and $\eta(t) = 0$ for $t < 0$, $\eta(0) = 1/2$. We also note that the step functions ensure that the integration range is from t' to t, thus ensuring causality, i.e., choosing the solution progressing forward in time. By insertion in (2.4) the global density operator assumes the form

$$\rho(t) = \sum_{n=0,m=0}^{\infty} \int dt_n \cdots dt_1 G_R(t, t_n) S_n G_R(t_n, t_{n-1}) \cdots G_R(t_2, t_1) S_1 G_R(t_1, t_i) \rho_S(t_i)$$

$$\times \int du_1 \cdots du_m G_A(t_i, u_1) S_1 G_A(u_1, u_2) \cdots G_A(u_{m-1}, u_m) S_m G_A(u_m, t)$$

$$\times e^{-iH_B t} B_n(t_n) \cdots B_1(t_1) \rho_B B_1(u_1) \cdots B_m(u_m) e^{iH_B t}; \quad (2.18)$$

note that this expression is completely equivalent to (2.4). For the reduced density operator $\rho_S(t)$ tracing over the bath and permuting the bath operators, we obtain

$$\rho_S(t) = \sum_{n=0,m=0}^{\infty} \int dt_n \cdots dt_1 G_R(t, t_n) S_n \cdots S_1 G_R(t_1, t_i) \rho_S(t_i)$$

$$\times \int du_1 \cdots du_m G_A(t_i, u_1) S_1 \cdots S_m G_A(u_m, t)$$

$$\times \text{Tr}_B[\rho_B B_1(u_1) \cdots B_m(u_m) B_n(t_n) \cdots B_1(t_1)]. \quad (2.19)$$

We note that the retarded and advanced Green's functions ensure that the operators S_p and B_p in (2.19) are chronologically ordered along the so-called Keldysh contour from time t to the initial time t_i and back to time t [52,53,58]. We also remark that the expansion (2.19) is completely equivalent to $\rho_S(t) = \text{Tr}_B[U(t, t_i)\rho(t_i)U(t_i, t)]$. Finally, inserting in (2.9) we obtain for the transmission matrix

$$T(t, t')_{pp',qq'}$$
$$= \sum_{n=0,m=0}^{\infty} \int dt_n \cdots dt_1 [G_R(t, t_n) S_n \cdots S_1 G_R(t_1, t')]_{pq}$$

$$\times \int du_1 \cdots du_m [G_A(t', u_1) S_1 \cdots S_m G_A(u_m, t)]_{q'p'}$$

$$\times \text{Tr}_B[\rho_B B_1(u_1) \cdots B_m(u_m) B_n(t_n) \cdots B_1(t_1)]. \quad (2.20)$$

In the unperturbed case, we have in particular

$$T^0(t, t')_{pp',qq'} = G_R(t, t')_{pq} G_A(t', t)_{q'p'}, \quad (2.21)$$

describing the unperturbed propagation of $\rho_S(t)_{pp'}$. Note that $B_p(t_p)$ forms a scalar product with S_p positioned between $G_R(t_{p+1}, t_p)$ and $G_R(t_p, t_{p-1})$; likewise, $B_q(u_q)$ is associated with S_q positioned between $G_A(u_{q-1}, u_q)$ and $G_A(u_q, u_{q+1})$.

The expression (2.20) provides a direct expansion of the transmission matrix $T(t, t')$ in powers of the interaction H_{SB} in terms of the Green's functions for the quantum system and the multitime bath correlations $\text{Tr}_B[\rho_B B_1(t_1) B_2(t_2) B_3(t_3) \cdots]$. We already note here that since $\rho_S(t) = T(t, t_i)\rho_S(t_i)$, we have $d\rho_S(t)/dt = (dT(t, t_i)/dt)\rho_S(t_i)$, and it follows that the expansion of T does not yield a proper master equation, independent of the initial condition $\rho_S(t_i)$. As a matter of fact, the expansion does not account for secular effects unless we make

further assumptions regarding the heat bath characterized by the density operator ρ_B and the operators B monitoring the bath.

III. BATH

At this stage, the expression (2.20) for the transmission matrix applies to a general bath characterized by the multibath correlations $\text{Tr}_B[\rho_B B_1(t_1) \cdots B_m(t_m)]$. To proceed and account for secular effects and in this connection invoke Wick's theorem, we specify the structure of the bath according to the Caldeira-Leggett prescription [37,38]. Details regarding Wick's theorem are deferred to Appendix 2.

A. Bath correlations

For simplicity, we consider a single bath with a bosonic structure, i.e., a collection of independent quantum oscillators, characterized by the bath Hamiltonian

$$H_B = \sum_k \Omega_k n_k, \quad (3.1)$$

where the occupation number $n_k = b_k^\dagger b_k$, and b_k is a Bose field with commutator $[b_k, b_p^\dagger] = \delta_{kp}$. The frequency associated with the wave number k is Ω_k. We assume that the bath is maintained at a temperature $1/\beta$ and characterized by the density operator [35]

$$\rho_B = \frac{\exp(-\beta H_B)}{\text{Tr}_B[-\exp(\beta H_B)]}. \quad (3.2)$$

From the Heisenberg equation of motion, $idb_k/dt = [b_b, H_B]$ [32,33], we infer

$$b_k(t) = b_k \exp(-i\Omega_k t), \quad (3.3)$$

$$b_k^\dagger(t) = b_k^\dagger \exp(+i\Omega_k t). \quad (3.4)$$

Moreover, the mean occupation number or Planck distribution [34,35] is given by

$$\langle n_k \rangle = \text{Tr}_B[\rho_B n_k] = \frac{1}{\exp(\beta\Omega_k) - 1}. \quad (3.5)$$

For the bath operators B^α entering in the coupling in (2.2) we choose for a single reservoir $\boldsymbol{B} = (B^1, B^2)$, where

$$B^1(t) = B(t) = \sum_k \lambda_k b_k(t), \quad (3.6)$$

$$B^2(t) = B^\dagger(t) = \sum_k \lambda_k b_k^\dagger(t); \quad (3.7)$$

note that the coupling constants λ_k can be chosen real by an appropriate choice of the phases of b_k. With this assignment, we have for the relevant bath correlations

$$D^{12}(t, t') = \text{Tr}_B[\rho_B B(t) B^\dagger(t')]$$
$$= \sum_k \lambda_k^2 (1 + \langle n_k \rangle) \exp(-i\Omega_k(t - t')), \quad (3.8)$$

$$D^{21}(t, t') = \text{Tr}_B[\rho_B B^\dagger(t) B(t')]$$
$$= \sum_k \lambda_k^2 \langle n_k \rangle \exp(+i\Omega_k(t - t')). \quad (3.9)$$

Introducing the spectral density of states $g(\Omega)$ and the Planck distribution $n(\Omega)$,

$$g(\Omega) = 2\pi \sum_k \lambda_k^2 \delta(\Omega - \Omega_k), \quad \Omega > 0, \quad (3.10)$$

$$n(\Omega) = \frac{1}{\exp(\beta\Omega) - 1}, \quad \Omega > 0, \quad (3.11)$$

we also have

$$D^{12}(t, t') = \int_0 \frac{d\Omega}{2\pi} g(\Omega)[1 + n(\Omega)] \exp(-i\Omega(t - t')), \quad (3.12)$$

$$D^{21}(t, t') = \int_0 \frac{d\Omega}{2\pi} g(\Omega) n(\Omega) \exp(+i\Omega(t - t')), \quad (3.13)$$

and introducing the Fourier transform, $D^{\alpha\beta}(t, t') = \int (d\omega/2\pi) \exp(-i\omega(t - t')) \tilde{D}^{\alpha\beta}(\omega)$,

$$\tilde{D}^{12}(\omega) = g(\omega)[1 + n(\omega)], \quad \omega > 0, \quad (3.14)$$

$$\tilde{D}^{21}(\omega) = g(-\omega) n(-\omega), \quad \omega < 0. \quad (3.15)$$

Note that the system-bath coupling λ_k is incorporated in the definition of the spectral density $g(\omega)$, which is of second order in λ_k. We also observe that in the classical limit for $\beta \to \infty$ we have $n(\omega) \to 1/\beta\Omega$, and the Ohmic

approximation requires $g(\Omega) \propto \Omega$ (with an appropriate high-frequency cutoff).

B. Wick's theorem

For a bath composed of independent quantum oscillators or bosons and assuming that the bath operator $\boldsymbol{B}$ is a a linear combination of Bose creation and annihilation operators and, moreover, assuming that the density operator ρ_B either corresponds to the Bose vacuum, i.e., $\rho_B = |0\rangle\langle0|$ or a bath described by the density operator $\rho_B = \exp(-\beta H_B)/\text{Tr}_B[\exp(-\beta H_B)]$, Wick's theorem holds for the multi-bath correlations in (2.20).

Wick's theorem [30,32,33,59,60] implies that the multibath correlations $\text{Tr}_B[\rho_B \boldsymbol{B}_1(t_1) \cdots \boldsymbol{B}_m(t_m)]$ can be broken up into all possible pairings or contractions; we note that $\text{Tr}_B[\rho_B \mathbf{B}(\mathbf{t})] = 0$. In the case of four bath operators we have for example

$$\text{Tr}_B[\rho_B B^\alpha(t_1) B^\beta(t_2) B^\gamma(t_3) B^\delta(t_4)]$$
$$= D^{\alpha\beta}(t_1, t_2) D^{\gamma\delta}(t_3, t_4) + D^{\alpha\gamma}(t_1, t_3) D^{\beta\delta}(t_2, t_4)$$
$$+ D^{\alpha\delta}(t_1, t_4) D^{\beta\gamma}(t_2, t_3), \quad (3.16)$$

where we note that the time ordering is preserved; we have introduced the bath correlation function

$$D^{\alpha\beta}(t, t') = \text{Tr}_B[\rho_B B^\alpha(t) B^\beta(t')]. \quad (3.17)$$

Wick's theorem is basically a generalisation of the contraction of Gaussian multi-correlation functions in terms of a single correlation function [60] to noncommuting operators. Usually the derivation of Wick's theorem is applied to the vacuum expectation value of time ordered products or in the finite temperature case imaginary time ordered products [32,33]. In the present context we summarize in Appendix 2 an elegant proof by Gaudin [59] directly applied to a thermal average of operator products relevant to the present analysis. We also state Wick's theorem in generator form [60].

IV. TRANSMISSION MATRIX

The transmission operator $T(t, t')$ is of central importance in our analysis; With an explicit bath prescription and Wick's theorem, we are in a position to discuss the transmission matrix (2.20) in more detail. Inserting the identity $\eta(t) + \eta(-t) = 1$, we can express (2.20) in the form

$$T(t, t')_{pp', qq'} = \sum_{n=0, m=0, ll', nn'} \int dt_n \cdots dt_1 G_R(t, t_n)_{pl} (\boldsymbol{S}_n \cdots \boldsymbol{S}_1)_{ll'} G_R(t_1, t')_{l'q}$$

$$\times \int du_1 \cdots du_m G_A(t', u_1)_{q'n} (\boldsymbol{S}_1 \cdots \boldsymbol{S}_m)_{nn'} G_A(u_m, t)_{n'p'} [\eta(t_n - u_m) + \eta(u_m - t_n)][\eta(t_1 - u_1) + \eta(u_1 - t_1)]$$

$$\times \text{Tr}_B[\rho_B \boldsymbol{B}_1(u_1) \cdots \boldsymbol{B}_m(u_m) \boldsymbol{B}_n(t_n) \cdots \boldsymbol{B}_1(t_1)], \quad (4.1)$$

yielding the sum of four individual contributions. Considering, for example, the term containing the product $\eta(t_n - u_m)\eta(t_1 - u_1)$ and using the identities

$$G_R(t, t')_{pq} = +i \sum_l G_R(t, t'')_{pl} G_R(t'', t')_{lq}, \quad (4.2)$$

$$G_A(t, t')_{pq} = -i \sum_l G_A(t, t'')_{pl} G_A(t'', t')_{lq}, \quad (4.3)$$

following from the definitions (2.16) and (2.17), we make the replacements

$$G_R(t_1, t') = iG_R(t_1, u_1)G_R(u_1, t'),$$ (4.4)

$$G_A(u_m, t) = -iG_A(u_m, t_n)G_A(t_n, t),$$ (4.5)

and we include the dummy arguments u_1 and t_n in the integrations over u_1 and t_n. Applying this procedure to all contributions, we can express the transmission matrix in the form

$$T(t, t')_{pp',qq'} = \sum_{ss',kk'} \int dt'' dt''' T^0(t, t'')_{pp',ss'} M(t'', t''')_{ss',kk'} T^0(t''', t')_{kk',qq'},$$ (4.6)

where $T^0(t, t')_{pp',qq'}$ is given by (2.21), and the reducible kernel $M(t, t')_{pp',qq'}$ takes the form

$$
\begin{aligned}
M(t, t')_{pp',qq'} = & -\sum_{n=0,m=0} \int dt_{n-1} \cdots dt_2 [S_n G_R(t, t_{n-1}) \cdots G_R(t_2, t')S_1]_{pq} \int du_1 \cdots du_m [G_A(t', u_1)S_1 \cdots S_m G_A(u_m, t)]_{q'p'} \\
& \times \mathrm{Tr}_B[\rho_B B_1(u_1) \cdots B_m(u_m)B_n(t) \cdots B_1(t')] \\
& -\sum_{n=0,m=0} \int dt_n \cdots dt_1 [G_R(t, t_n)S_n \cdots S_1 G_R(t_1, t')]_{pq} \int du_2 \cdots du_{m-1}[S_1 G_A(t', u_2) \cdots G_A(u_{m-1}, t)S_m]_{q'p'} \\
& \times \mathrm{Tr}_B[\rho_B B_1(t') \cdots B_m(t)B_n(t_n) \cdots B_1(t_1)] + \sum_{n=0,m=0} \int dt_{n-1} \cdots dt_1 [S_n G_R(t, t_{n-1}) \cdots S_1 G_R(t_1, t')]_{pq} \\
& \times \int du_2 \cdots du_m [S_1 G_A(t', u_2) \cdots S_m G_A(u_m, t)]_{q'p'} \mathrm{Tr}_B[\rho_B B_1(t') \cdots B_m(u_m)B_n(t) \cdots B_1(t_1)] \\
& + \sum_{n=0,m=0} \int dt_n \cdots dt_2 [G_R(t, t_n)S_n \cdots G_R(t_2, t')S_1]_{pq} \int du_1 \cdots du_{m-1}[G_A(t', u_1)S_1 \cdots G_A(u_{m-1}, t)S_m]_{q'p'} \\
& \times \mathrm{Tr}_B[\rho_B B_1(u_1) \cdots B_m(t)B_n(t_n) \cdots B_1(t')];
\end{aligned}
$$ (4.7)

the example above applies to the third term in (4.7). Inspecting Fig. 1 shows that the construction of the kernel $M(t, t')$ corresponds to removing the external legs $G_R(t, t_n)$, $G_R(t_1, t')$, $G_A(t', u_1)$, and $G_A(u_m, t)$ from the transmission matrix $T(t, t')$.

The next essential step accounting for secular effects is to identify a Dyson equation for the transmission matrix in (2.20). According to Wick's theorem, the vertices S_p in the reducible kernel $M(t, t')$ in (4.7) are connected pairwise to the bath operators B_p in the bath correlation function $D^{\alpha\beta}(t, t')$. As a result, $M(t, t')$ can be broken up in irreducible parts $K(t, t')$ connected by a pair of Green's functions G_R and G_A constituting the unperturbed transmission matrix $T^0(t, t')$ (2.21). Proceeding schematically term by term, we have
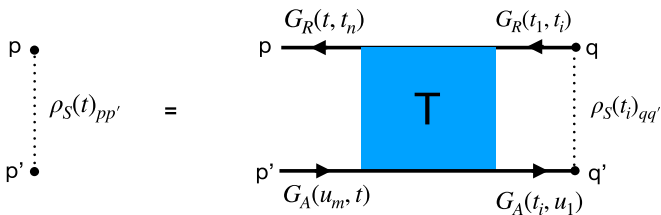

FIG. 1. Here we depict the transmission matrix $T(t, t_i)$ shown as a shaded box describing the evolution of the reduced density operator $\rho_S(t)$ from the initial time t_i to the final time t. The legs on the reducible kernel M, and the retarded and advanced Green's functions G_R and G_A, are denoted by directed arrows.

$M = I + K + KT^0K + \cdots$ and by insertion in $T = T^0 M T^0$ in (4.6) the expansion $T = T^0 + T^0 K T^0 + \cdots$, yielding the Dyson equation

$$
\begin{aligned}
T(t, t')_{pp',qq'} = & T^0(t, t')_{pp',qq'} + \sum_{ll',ss'} \int dt'' dt''' T^0(t, t'')_{pp',ll'} \\
& \times K(t'', t''')_{ll',ss'} T(t''', t')_{ss',qq'}.
\end{aligned}
$$ (4.8)

In the diagrammatic representation, the kernel $K(t, t')_{pp',qq'}$ is irreducible in the sense that is cannot be disconnected by the insertion of $T^0(t, t')_{pp',qq'}$. In Fig. 2 we have depicted the structure of the Dyson equation for $T(t, t')_{pp',qq'}$.

V. MASTER EQUATION

The Dyson equation (4.8) for $T(t, t')$ is a crucial result ensuring that secular effects are properly included. In a condensed-matter context, the irreducible kernel $K(t, t')$ serves as a self-energy or mass operator [32,33,50]. Schematically (4.8) has the form $T = T^0 + T^0 K T$. Inserted in (2.9), $\rho_S = T\rho_S(t_i)$, we have $\rho_S = T^0 \rho_S(t_i) + T^0 K T \rho_S(t_i)$ or $\rho_S = T^0 \rho_S(t_i) + T^0 K \rho_S$. We thus obtain the following general inhomogeneous integral equation for the reduced density operator $\rho_S(t)$:

$$
\begin{aligned}
\rho_S(t)_{pp'} = & \sum_{qq'} T^0(t, t_i)_{pp',qq'} \rho_S(t_i)_{qq'} + \sum_{ss',qq'} \int dt' dt'' T^0 \\
& \times (t, t')_{pp',ss'} K(t', t'')_{ss',qq'} \rho_S(t'')_{qq'}.
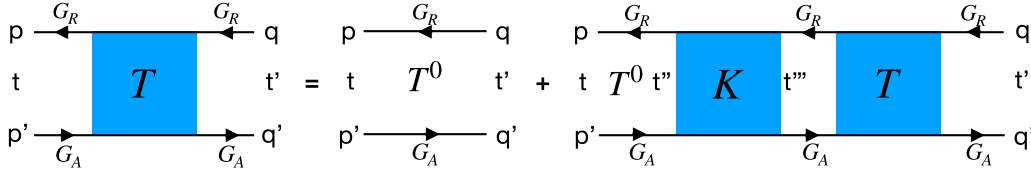\end{aligned}
$$ (5.1)

FIG. 2. Here we depict the Dyson equation for the transmission matrix. $T(t, t')$ and the irreducible kernel $K(t', t'')$ characterized by shaded boxes. $T^0(t, t')$ denotes the unperturbed transmission matrix, and the retarded and advanced Green's functions $G_R(t, t')$ and $G_A(t', t)$ are denoted by directed arrows.

The integral equation (5.1) represents an integrated form of the master equation and depends on the initial condition $\rho_S(t_i)$.

Choosing for simplicity an energy basis, $H_S|n\rangle = E_n|n\rangle$, and using the identity

$$\frac{d}{dt}T^0(t, t')_{pp', qq'} = -i(E_p - E_{p'})T^0(t, t')_{pp', qq'}$$
$$+ \delta(t - t')\delta_{pq}\delta_{q'p'}, \quad (5.2)$$

following from (2.21) we infer the general master equation for $\rho_S(t)$,

$$\frac{d}{dt}\rho_S(t)_{pp'} = -i[H_S, \rho_S(t)]_{pp'}$$
$$+ \sum_{qq'} \int dt' K(t, t')_{pp', qq'} \rho_S(t')_{qq'}. \quad (5.3)$$

This is a fundamental result showing that the Dyson equation for the transmission matrix implies a general non-Markovian master equation independent of the the initial condition. Moreover, we note that the only assumptions

underlying the structure of the master equation (5.3) are the Caldeira-Leggett heat bath in combination with Wick's theorem. In the absence of coupling to the heat bath, the kernel $K(t, t')$ vanishes and the system evolves in time according to the von Neumann equation $d\rho_S(t)/dt = -i[H_S, \rho_S(t)]$. Since $\text{Tr}_S[\rho_S(t)] = 1$, consistency requires that the trace of K vanishes, i.e., $\sum_p K(t, t')_{pp, qq'} = 0$, in the ensuing approximations.

VI. BORN APPROXIMATION

The master equation in (5.3) has a general non-Markovian form. To provide a concrete realization we can, in principle, expand the kernel $K(t, t')$ to any desired order in the interaction H_{SB} by identifying the relevant diagrams and applying Wick's theorem. However, in many applications it is customary to assume that the coupling to the bath is weak and that it is sufficient only to consider the Born approximation, i.e., an expansion to second order in H_{SB}. In the Born approximation, only a single bath correlation function enters. By inspection of (4.7) and inserting (3.17) we obtain the following expression for the irreducible kernel $K(t, t')$:

$$K(t, t')_{pp', qq'} = -\sum_{\alpha\beta}[S^\alpha G_R(t, t')S^\beta]_{pq}G_A(t', t)_{q'p'}D^{\alpha\beta}(t, t') - \sum_{\alpha\beta}G_R(t, t')_{pq}[S^\alpha G_A(t', t)S^\beta]_{q'p'}D^{\alpha\beta}(t', t)$$

$$+ \sum_{\alpha\beta}[S^\alpha G_R(t, t')]_{pq}[S^\beta G_A(t', t)]_{q'p'}D^{\beta\alpha}(t', t) + \sum_{\alpha\beta}[G_R(t, t')S^\alpha]_{pq}[G_A(t', t)S^\beta]_{q'p'}D^{\beta\alpha}(t, t'). \quad (6.1)$$

The four irreducible contributions to the kernel (6.1) in the Born approximation are depicted in Fig. 3; here diagrams (a) and (b) correspond to level populations, whereas diagrams (c) and (d) are associated with coherences.

It is straightforward to proceed to higher order in H_{SB} by identifying the corresponding irreducible contributions to the kernel K. Thus to fourth order in H_{SB} one identifies 20 individual contributions to the kernel K. The issue of a stronger system-bath coupling is important but will not be pursued in the present context.
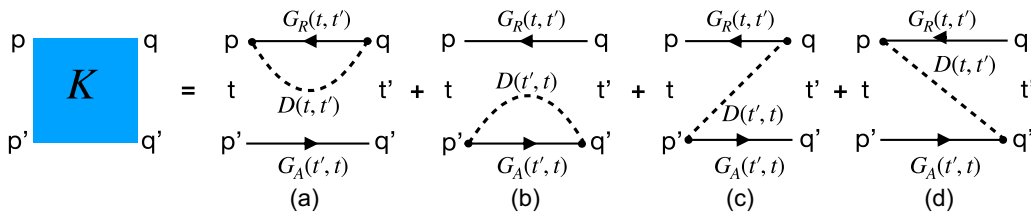

FIG. 3. Here we depict the irreducible kernel $K(t, t')$ in the Born approximation. The retarded and advanced Green's functions $G_R(t, t')$ and $G_A(t', t)$ are denoted by directed lines. The bath correlation function $D(t, t')$ is denoted by a dotted line. The vertices S are denoted by dots. Diagrams (a) and (b) correspond to populations, diagrams (c) and (d) to coherences.

Inserting (6.1) in (5.3), we obtain the master equation

$$\frac{d}{dt}\rho_S(t)_{pp'} = -i[H_S, \rho_S(t)]_{pp'} - \int dt' \sum_{\alpha\beta,qq'} [S^\alpha G_R(t,t')S^\beta]_{pq} G_A(t',t)_{q'p'}\rho_S(t')_{qq'}D^{\alpha\beta}(t,t')$$

$$- \int dt' \sum_{\alpha\beta,qq'} G_R(t,t')_{pq}[S^\alpha G_A(t',t)S^\beta]_{q'p'}\rho_S(t')_{qq'}D^{\alpha\beta}(t',t)$$

$$+ \int dt' \sum_{\alpha\beta,qq'} [S^\alpha G_R(t,t')]_{pq}[S^\beta G_A(t',t)]_{q'p'}\rho_S(t')_{qq'}D^{\beta\alpha}(t',t)$$

$$+ \int dt' \sum_{\alpha\beta,qq'} [G_R(t,t')S^\alpha]_{pq}[G_A(t',t)S^\beta]_{q'p'}\rho_S(t')_{qq'}D^{\beta\alpha}(t,t'), \tag{6.2}$$

or in operator form

$$\frac{d}{dt}\rho_S(t) = -i[H_S, \rho_S(t)] - \int dt' \sum_{\alpha\beta} S^\alpha G_R(t,t')S^\beta \rho_S(t')G_A(t',t)D^{\alpha\beta}(t,t')$$

$$- \int dt' \sum_{\alpha\beta} G_R(t,t')\rho_S(t')S^\alpha G_A(t',t)S^\beta D^{\alpha\beta}(t',t) + \int dt' \sum_{\alpha\beta} S^\alpha G_R(t,t')\rho_S(t')S^\beta G_A(t',t)D^{\beta\alpha}(t',t)$$

$$+ \int dt' \sum_{\alpha\beta} G_R(t,t')S^\alpha \rho_S(t')G_A(t',t)S^\beta D^{\beta\alpha}(t,t'). \tag{6.3}$$

We already discern here the structure of the Lindblad equation in (1.1). Applying the trace operation to (6.3) and cyclically permuting the operators under the trace, we readily infer $d\mathrm{Tr}_S[\rho_S(t)]/dt = 0$ and, consequently, $\mathrm{Tr}_S[\rho_S(t)] = \mathrm{Tr}_S[\rho_S(t_i)] = 1$, thus providing a consistency check of the Born approximation. The trace condition applied to the kernel K reads $\sum_p K_{pp,qq'} = 0$, which by inspection of (6.1) is easily verified. In terms of the diagrams in Fig. 3, the trace condition is obtained by setting $p = p'$ and noting that diagram (a) cancels with diagram (c) and diagram (b) with diagram (d).

Introducing the Fourier transform $\tilde{\rho}_S(\omega) = \int dt \exp(i\omega t)\rho_s(t)$, noting that the bath is in a stationary state, $D^{\alpha\beta}(t,t') = D^{\alpha\beta}(t - t')$, and from (2.16) and (2.17) the Green's function resolvents $\tilde{G}_R(\omega) = 1/(\omega - H_S + i\epsilon)$ and $\tilde{G}_A(\omega) = 1/(\omega - H_S - i\epsilon)$, we obtain the master equation in Fourier space,

$$-i\omega\tilde{\rho}_S(\omega)_{pp'} = -iE_{pp'}\tilde{\rho}_S(\omega)_{pp'} + \sum_{qq'}\tilde{K}(\omega)_{pp',qq'}\tilde{\rho}_S(\omega)_{qq'}, \tag{6.4}$$

with kernel

$$\tilde{K}(\omega)_{pp',qq'} = -i\delta_{p'q'}\sum_{\alpha\beta,l}\int\frac{d\omega'}{2\pi}\frac{S^\alpha_{pl}S^\beta_{lq}\delta(E_{pl}+E_{lq})\tilde{D}^{\alpha\beta}(\omega')}{\omega-\omega'+E_{p'l}+i\epsilon} - i\delta_{pq}\sum_{\alpha\beta,l}\int\frac{d\omega'}{2\pi}\frac{S^\alpha_{q'l}S^\beta_{lp'}\delta(E_{q'l}+E_{lp'})\tilde{D}^{\alpha\beta}(-\omega')}{\omega-\omega'+E_{lp}+i\epsilon}$$

$$+i\sum_{\alpha\beta}\int\frac{d\omega'}{2\pi}\frac{S^\alpha_{pq}S^\beta_{q'p'}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\beta\alpha}(-\omega')}{\omega-\omega'+E_{p'q}+i\epsilon} + i\sum_{\alpha\beta}\int\frac{d\omega'}{2\pi}\frac{S^\alpha_{pq}S^\beta_{q'p'}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\beta\alpha}(\omega')}{\omega-\omega'+E_{q'p}+i\epsilon}. \tag{6.5}$$

Here the matrix element of the system operator is given by $S^\alpha_{pq} = \langle p|S^\alpha|q\rangle$ and the associated energy level splitting is given by $E_{pq} = E_p - E_q$. We have, moreover, in order to implement the rotating-wave approximation introduced in a somewhat ad hoc manner, the δ-function constraints $\delta(E_{pq} + E_{q'p'})$ in order to ensure energy conservation in connection with the combined transitions $S^\alpha_{pq}S^\beta_{q'p'}$. We note that this implementation of selection rules does not follow automatically from the diagrammatic expansion; this is an issue that remains to be examined.

Applying the Plemejl formula $1/(\omega + i\epsilon) = \mathrm{P}(1/\omega) - i\pi\delta(\omega)$ [60], where P denotes the principal value, to the kernel (6.5), we obtain, setting $\tilde{K}(\omega)_{pp',qq'} = \tilde{L}(\omega)_{pp',qq'} + i\tilde{\Delta}(\omega)_{pp',qq'}$, the shift

$$\tilde{\Delta}(\omega)_{pp',qq'} = -\delta_{p'q'}\sum_{\alpha\beta,l}\mathrm{P}\int\frac{d\omega'}{2\pi}\frac{S^\alpha_{pl}S^\beta_{lq}\delta(E_{pl}+E_{lq})\tilde{D}^{\alpha\beta}(\omega')}{\omega-\omega'+E_{p'l}} - \delta_{pq}\sum_{\alpha\beta,l}\mathrm{P}\int\frac{d\omega'}{2\pi}\frac{S^\alpha_{q'l}S^\beta_{lp'}\delta(E_{q'l}+E_{lp'})\tilde{D}^{\alpha\beta}(-\omega')}{\omega-\omega'+E_{lp}}$$

$$+\sum_{\alpha\beta}\mathrm{P}\int\frac{d\omega'}{2\pi}\frac{S^\alpha_{pq}S^\beta_{q'p'}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\beta\alpha}(-\omega')}{\omega-\omega'+E_{p'q}} + \sum_{\alpha\beta}\mathrm{P}\int\frac{d\omega'}{2\pi}\frac{S^\alpha_{pq}S^\beta_{q'p'}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\beta\alpha}(\omega')}{\omega-\omega'+E_{q'p}}, \tag{6.6}$$

and the dissipative kernel

$$\tilde{L}(\omega)_{pp',qq'} = -\delta_{p'q'}\frac{1}{2}\sum_{\alpha\beta,l}S^\alpha_{pl}S^\beta_{lq}\delta(E_{pl}+E_{lq})\tilde{D}^{\alpha\beta}(\omega+E_{p'l}) - \delta_{pq}\frac{1}{2}\sum_{\alpha\beta,l}S^\alpha_{q'l}S^\beta_{lp'}\delta(E_{q'l}+E_{lp'})\tilde{D}^{\alpha\beta}(-\omega-E_{lp})$$

$$+\frac{1}{2}\sum_{\alpha\beta}S^\beta_{pq}S^\alpha_{q'p'}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\alpha\beta}(-\omega-E_{p'q}) + \frac{1}{2}\sum_{\alpha\beta}S^\beta_{pq}S^\alpha_{q'p'}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\alpha\beta}(\omega+E_{q'p}), \tag{6.7}$$

and by insertion in (6.4) the final Fourier form of the master equation,

$$-i\omega\tilde{\rho}_S(\omega)_{pp'} = -i\sum_{qq'}[E_{pp'}\delta_{pq}\delta_{p'q'} - \tilde{\Delta}(\omega)_{pp',qq'}]\tilde{\rho}_S(\omega)_{qq'} + \sum_{qq'}\tilde{L}(\omega)_{pp',qq'}\tilde{\rho}_S(\omega)_{qq'}. \tag{6.8}$$

By inspection we note that $\sum_p\tilde{\Delta}(\omega)_{pp,qq'}=0$ and $\sum_p\tilde{L}(\omega)_{pp,qq'}=0$, implying unity trace. Moreover, we observe the general symmetry inferred from the Dyson equation (4.8) together with (2.21) or in the Born case from (6.5),

$$K(t-t')^*_{pp',qq'} = K(t-t')_{p'p,q'q}, \tag{6.9}$$

$$\tilde{K}(\omega)^*_{pp',qq'} = \tilde{K}(-\omega)_{p'p,q'q}. \tag{6.10}$$

The master equation (6.8) is local in Fourier space and non-Markovian. The coupling of an energy level to a continuum of states, be it the vacuum or a heat reservoir, typically gives rise to both a damping given by $\tilde{L}$ and a shift given by $\tilde{\Delta}$. In the general case, both shift and damping are frequency-dependent characteristics of memory effects. In the case of a Markovian behavior, the shift is constant and appears as a Lamb shift that can be absorbed in a renormalization of the energy levels, corresponding to a counter term in the system Hamiltonian H_S [9].

VII. LINDBLAD EQUATION

The time evolution of the density operator $\rho(t)$ for the closed system composed of the open quantum system under investigation and the bath is unitary and governed by the von Neumann master equation (2.3) with solution $\rho(t) = U(t,t_i)\rho(t_i)U(t,t_i)^\dagger$. By construction, the density operator is Hermitian, positive, and has unit trace. However, due to entanglement with the bath, the reduced density operator $\rho_S(t) = \text{Tr}_B[\rho(t)]$ for the open quantum system does not develop in time according to a unitary transformation, and consequently it does not conserve probability or entropy. On the other hand, a proper physical interpretation requires that $\rho_S(t)$ is Hermitian, positive, and has unit trace. These requirements imply that $\rho_S(t)$ conforms to the Kraus representation $\rho_S(t) = \Lambda(t,t_i)\rho_S(t_i) = \sum_\alpha K_\alpha(t,t_i)\rho_S(t_i)K_\alpha^\dagger(t,t_i)$, where $\sum_\alpha K_\alpha^\dagger K_\alpha = I$, defining a so-called quantum channel [9,49,61].

In the Markov approximation, assuming a separation of the fast timescale of the bath and the slower time scale of the open system, yielding a memoryless kernel, we have $d\rho_S(t)/dt = G\rho_S(t)$, with solution $\rho_S(t) = \exp(G(t-t_i))\rho_S(t_i)$; here G is the generator of a quantum dynamical semigroup. The issue of the most general form of the generator G has been addressed by Gorini, Kossakowski, Sudarshan, and Lindblad (GKSL) [48,49,62,63]; for a review, see also [64]. The GKSL or

Lindblad master equation has the form given in (1.1), where we note that the trace of the right-hand side of the equation vanishes, yielding a constant trace $\text{Tr}[\rho_S(t)] = 1$. Hermiticity, moreover, implies $\gamma_k^{\alpha\beta} = (\gamma_k^{\beta\alpha})^*$, i.e., the dissipation coefficients form a Hermitian matrix.

VIII. STANDARD DERIVATION OF THE LINDBLAD EQUATION

Referring for details to the standard text by Breuer and Petruccione [9] (see also [49]), the customary microscopic derivation of the Lindblad equation takes as its starting point the the von Neumann equation in the interaction representation and its integrated form,

$$\frac{d}{dt}\rho^I(t) = -i[H_{SB}(t), \rho^I(t)], \tag{8.1}$$

$$\rho^I(t) = \rho(0) - i\int_0^t dt'[H_{SB}(t'), \rho^I(t')], \tag{8.2}$$

with initial value $\rho(0)$; here $\rho^I(t) = \exp(iH_0t)\rho(t)\exp(-iH_0t)$. Inserting (8.2) in (8.1), tracing over the bath, and assuming $\text{Tr}_B[H_{SB}(t), \rho(t_i)] = 0$, we obtain to second-order Born for the reduced density operator $\rho_S^I(t) = \exp(iH_St)\rho_S(t)\exp(-iH_St)$

$$\frac{d}{dt}\rho_S^I(t) = -\int_0^t dt'\text{Tr}_B[H_{SB}(t), [H_{SB}(t'), \rho^I(t')]]. \tag{8.3}$$

Assuming a weak coupling to the reservoir and introducing the physical approximation

$$\rho^I(t) \approx \rho_S^I(t)\rho_B, \tag{8.4}$$

we obtain closure with respect to $\rho_S^I(t)$ yielding

$$\frac{d}{dt}\rho_S^I(t) = -\int_0^t dt'\text{Tr}_B[H_{SB}(t), [H_{SB}(t'), \rho_S^I(t')\rho_B]]. \tag{8.5}$$

We note that (8.5) is not a proper master equation since it depends on the initial value at $t = 0$; this issue will be addressed in more detail in Sec. X.

Next implementing the Markov approximation by locking $\rho_S^I(t')$ onto $\rho_S^I(t)$, we obtain at this stage the Redfield equation [21]

$$\frac{d}{dt}\rho_S^I(t) = -\int_0^t dt'\text{Tr}_B[H_{SB}(t), [H_{SB}(t'), \rho_S^I(t)\rho_B]]. \tag{8.6}$$

The Redfield equation, although local in time and often used in quantum optics, is [for the same reason as (8.5)] not a full

Markov equation since it depends on the initial preparation. However, assuming a timescale separation between the fast bath relaxation time τ_B and the slower system timescale τ_S, i.e., $\tau_S \gg \tau_B$, one obtains a Markovian master equation. For the resulting master equation to correspond to the generator of a dynamical semigroup, one finally makes a further secular approximation averaging over oscillating terms, the so-called rotating-wave approximation (RWA),

To implement the RWA, one projects the system operator S^α onto the energy eigenspace of the system Hamiltonian H_S and defines (note that $E_{k'k} = E_{k'} - E_k$)

$$S^\alpha(\omega) = \sum_{kk'} |k\rangle\langle k|S^\alpha|k'\rangle\langle k'|\delta(\omega - E_{k'k}). \quad (8.7)$$

Upon further manipulations, see [9] for details, averaging over oscillating terms (the RWA), and returning to the Schrödinger picture, we obtain the Lindblad equation (1.1) in the form

$$\frac{d}{dt}\rho_S(t) = -i[H_{LS}, \rho_S(t)] + L^{ST}\rho_S(t),$$

$$L^{ST}\rho_S(t) = \sum_{\alpha\beta,\omega} \gamma_{\alpha\beta}(\omega)\left(S^\beta(\omega)\rho_S(t)S^{\alpha\dagger}(\omega) \right.$$
$$\left. - \frac{1}{2}\{S^{\alpha\dagger}(\omega)S^\beta(\omega), \rho_S(t)\}\right). \quad (8.8)$$

Here L^{ST} denotes the dissipator in the standard derivation, H_{LS} is the system Hamiltonian including a Lamb shift, and bath correlations $\gamma_{\alpha\beta}(\omega) = \text{Tr}_B[\rho_B B^{\alpha\dagger}B^\beta](\omega)$.

Summarizing, the customary approach in the microscopic derivation of the Lindblad equation found in the literature on open quantum systems is based on a series of physical approximations: (i) weak coupling to the bath, i.e., the Born approximation, (ii) timescale separation, (iii) the Markov approximation, and (iv) the RWA.

IX. QUASIPARTICLE APPROXIMATION

In the master equation in the Born approximation (6.3) or in the general form (5.3), the time-dependent kernel $K(t, t')$ describes the coupling to the bath. However, the presence of memory effects makes an analysis difficult, and it is customary to apply the Markov approximation [35,40]. This approximation is based on the assumption of a timescale separation between the fast timescale or decay time of correlations in the bath and the slower timescale associated with the time evolution of the reduced density operator. This approach corresponds to the Langevin or equivalent Fokker-Planck scheme in classical statistical mechanics [35,40].

A. Heuristic derivation of the Lindblad equation

By inspection of (6.3) we note that assuming that the bath correlations $D^{\alpha\beta}(t, t')$ fall off on a short timescale, τ_B, compared to the timescale of the evolution of the system, τ_S, i.e., $\tau_B \ll \tau_S$, and setting $D^{\alpha\beta}(t, t') \to \delta(t - t')D^{\alpha\beta}$ together with the limits $G_R(t, t) = -i(1/2)$ and $G_A(t, t) = +i(1/2)$, we recover the Lindblad equation in (1.1). Note, however, that this heuristic argument does not provide the actual form of $D^{\alpha\beta}$ and its dependence on the bath parameters.

B. Pole structure of the transmission matrix

From (2.9) we have in Fourier space

$$\tilde{\rho}_S(\omega)_{pp'} = \sum_{qq'} \tilde{T}(\omega)_{pp',qq'} \exp(i\omega t_i)\rho_S(t_i)_{qq'}, \quad (9.1)$$

and the time behavior of $\rho_S(t)_{pp'}$ is determined by the pole structure of the transmission matrix $\tilde{T}(\omega)_{pp',qq'}$. From the Dyson equation (4.8) in Fourier space,

$$\tilde{T}(\omega)_{pp',qq'} = \tilde{T}^0(\omega)_{pp',qq'}$$
$$+ \sum_{ss',ll'} \tilde{T}^0(\omega)_{pp',ss'}\tilde{K}(\omega)_{ss',ll'}\tilde{T}(\omega)_{ll',qq'}, \quad (9.2)$$

and defining the inverse transmission matrix according to $\sum_{ll'} \tilde{T}^{-1}_{pp',ll'}\tilde{T}_{ll',qq'} = \delta_{pq}\delta_{p'q'}$ we infer

$$\tilde{T}(\omega)^{-1}_{pp',qq'} = \tilde{T}^0(\omega)^{-1}_{pp',qq'} - \tilde{K}(\omega)_{pp',qq'}, \quad (9.3)$$

yielding the resonance condition. From (2.16) and (2.17) inserted in (2.21) we have in the energy basis $\tilde{T}^0(\omega)^{-1}_{pp',qq'} = -i\delta_{pq}\delta_{p'q'}(\omega - E_{pp'})$ and we obtain, splitting off the diagonal part of $\tilde{K}(\omega)_{pp',qq'}$, the resonance condition given by

$$\det[D_{pp',qq'} - \tilde{K}^{OD}(\omega)_{pp',qq'}] = 0, \quad (9.4)$$

$$D_{pp',qq'} = [-i(\omega - E_{pp'}) - \tilde{K}(\omega)_{pp',pp'}]\delta_{pq}\delta_{p'q'}, \quad (9.5)$$

where $\tilde{K}^{OD}(\omega)_{pp',qq'} = (1 - \delta_{pq}\delta_{p'q'})\tilde{K}(\omega)_{pp',qq'}$ is the off-diagonal part (OD).

C. Quasiparticle approximation

To proceed in deriving a Markov master equation, we invoke the so-called quasiparticle approximation employed in condensed-matter many-body theory [32,33,50,51]. Here the basic building block is the single-particle Green's function $\tilde{G}(\omega, k)$ describing the propagation of a quantum particle with energy ω and momentum k in a many-body environment. Suppressing the k dependence, the Green's function for a noninteracting system has the form $\tilde{G}_0(\omega) = 1/(\omega - E)$, where E is an energy level. In the simplest case, diagrammatic perturbation theory gives rise to a Dyson equation of the form $\tilde{G}(\omega) = \tilde{G}_0(\omega) + \tilde{G}_0(\omega)\tilde{\Sigma}(\omega)\tilde{G}(\omega)$, whose solution is the generic form $\tilde{G}(\omega) = 1/[\omega - E - \tilde{\Sigma}(\omega)]$; here the self-energy or mass operator $\tilde{\Sigma}(\omega)$ is determined perturbatively. The time dependence of the propagation of the quasiparticle is thus given by the resonance condition $\tilde{G}(\omega)^{-1} = \omega - E - \tilde{\Sigma}(\omega) = 0$. To leading order, the quasiparticle approximation corresponds to $\tilde{\Sigma}(\omega) \to \tilde{\Sigma}(E)$. Separating $\tilde{\Sigma}(E)$ in a real and imaginary part, i.e., $\tilde{\Sigma}(E) = \tilde{\Delta} + i\tilde{\Gamma}$, we have $G(t) \propto e^{-i(E+\tilde{\Delta})t}e^{-\tilde{\Gamma}t}$; the real part $\tilde{\Delta}$ gives rise to a quasiparticle energy shift, while the imaginary part $\tilde{\Gamma}$ yields a damping of the quasiparticle. Both energy shift and damping are caused by interaction with the many-body environment. For the quasiparticle to preserve its identity, we must assume that the damping is small. It is important to note that the Dyson equation automatically incorporates secular effects in producing an energy shift and a damping.

Here we apply a corresponding "quasiparticle approximation" to the transmission operator $\tilde{T}(\omega)_{pp',qq'}$ for open quantum systems in order to incorporate secular effects. In a

slightly compressed form, expressing $\det[D(\omega) - \tilde{K}^{\text{OD}}(\omega)]$ in the form $\det[D(\omega)]\det[I - D(\omega)^{-1}\tilde{K}(\omega)^{\text{OD}}]$, using the expansion $\det[I - D^{-1}\tilde{K}^{\text{OD}}] = 1 - \text{Tr}[D^{-1}\tilde{K}^{\text{OD}}]$, and noting that $\text{Tr}[D^{-1}\tilde{K}^{\text{OD}}] = 0$ (by construction), we obtain to leading order the resonance condition $\det[D] = 0$, i.e.,

$$\det[[-i(\omega - E_{pp'}) - \tilde{K}(\omega)_{pp',pp'}]\delta_{pq}\delta_{p'q'}] = 0. \quad (9.6)$$

For vanishing coupling for $\tilde{K}(\omega)_{pp',qq'} = 0$ the resonance condition is given by

$$\det[(\omega - E_{pp'})\delta_{pq}\delta_{p'q'}] = 0, \quad (9.7)$$

yielding the roots $\omega = E_{pp'}$ or equivalently $\omega = E_{qq'}$. Consequently, the quasiparticle approximation corresponds to replacing the frequency ω in diagonal kernel $\tilde{K}(\omega)_{pp',pp'}$ by the unperturbed value $\omega = E_{pp'}, E_{qq'}$. As a result, inserting in (6.6) and (6.7) we obtain for the shift and dissipative kernel in the quasiparticle approximation the constant shift

$$
\begin{aligned}
\tilde{\Delta}_{pp',qq'} = &-\delta_{p'q'}\sum_{\alpha\beta,l}\text{P}\int\frac{d\omega}{2\pi}\frac{S_{pl}^{\alpha}S_{lq}^{\beta}\delta(E_{pl}+E_{lq})\tilde{D}^{\alpha\beta}(\omega)}{E_{pl}-\omega}\\
&-\delta_{pq}\sum_{\alpha\beta,l}\text{P}\int\frac{d\omega}{2\pi}\frac{S_{q'l}^{\alpha}S_{lp'}^{\beta}\delta(E_{q'l}+E_{lp'})\tilde{D}^{\alpha\beta}(-\omega)}{E_{lp'}-\omega}\\
&+\sum_{\alpha\beta}\text{P}\int\frac{d\omega}{2\pi}\frac{S_{pq}^{\beta}S_{q'p'}^{\alpha}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\alpha\beta}(\omega)}{E_{q'p'}-\omega}\\
&+\sum_{\alpha\beta}\text{P}\int\frac{d\omega}{2\pi}\frac{S_{pq}^{\beta}S_{q'p'}^{\alpha}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\alpha\beta}(-\omega)}{E_{pq}-\omega},
\end{aligned}
$$
$$(9.8)$$

and the constant dissipative kernel

$$
\begin{aligned}
\tilde{L}_{pp',qq'} = &-\delta_{p'q'}\frac{1}{2}\sum_{\alpha\beta,l}S_{pl}^{\alpha}S_{lq}^{\beta}\delta(E_{pl}+E_{lq})\tilde{D}^{\alpha\beta}(E_{pl})\\
&-\delta_{pq}\frac{1}{2}\sum_{\alpha\beta,l}(S^{\alpha})_{q'l}(S^{\beta})_{lp'}\delta(E_{q'l}+E_{lp'})\tilde{D}^{\alpha\beta}(E_{p'l})\\
&+\frac{1}{2}\sum_{\alpha\beta}S_{pq}^{\beta}S_{q'p'}^{\alpha}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\alpha\beta}(E_{q'p'})\\
&+\frac{1}{2}\sum_{\alpha\beta}S_{pq}^{\beta}S_{q'p'}^{\alpha}\delta(E_{pq}+E_{q'p'})\tilde{D}^{\alpha\beta}(E_{qp}). \quad (9.9)
\end{aligned}
$$

Finally, inserting (9.8) and (9.9) in (6.4), we obtain the Fourier form of the master equation in the quasiparticle approximation

$$
\begin{aligned}
-i\omega\tilde{\rho}_S(\omega)_{pp'} = &-i\sum_{qq'}(E_{pp'}\delta_{pq}\delta_{p'q'} - \tilde{\Delta}_{pp',qq'})\tilde{\rho}_S(\omega)_{qq'}\\
&+\sum_{qq'}\tilde{L}_{pp',qq'}\tilde{\rho}_S(\omega)_{qq'}. \quad (9.10)
\end{aligned}
$$

Correspondingly, the master equation takes the form

$$
\begin{aligned}
\frac{d}{dt}\rho_S(t)_{pp'} = &-i\sum_{qq'}(E_{pp'}\delta_{pq}\delta_{p'q'} - \tilde{\Delta}_{pp',qq'})\rho_S(t)_{qq'}\\
&+\sum_{qq'}\tilde{L}_{pp',qq'}\rho_S(t)_{qq'}. \quad (9.11)
\end{aligned}
$$

This is our main result, which comes from a standard field-theoretical analysis in combination with a quasiparticle approximation and an imposed rotating-wave approximation. The master equation is memoryless, i.e., Markovian. By inspection we note that $\sum_p \tilde{K}_{pp,qq'} = 0$, $\sum_p \tilde{\Delta}_{pp,qq'} = 0$, and $\sum_p E_{pp} = 0$ yielding a constant trace of ρ_S.

There is an important issue that we have not addressed, namely the positivity of the reduced density matrix ρ_S required from general principles; see, e.g., [9]. We have shown that $\text{Tr}\rho_S = 1$ both in the non-Markovian case and in the Lindblad case. The present diagrammatic approach, however, does not ensure positivity of ρ_S. This issue has been discussed in [65].

X. DISCUSSION

Here we discuss the Markovian Lindblad equation and non-Markovian approaches by Diosi-Feriadi and Nakajima-Zwanzig.

A. Lindblad equation

To establish the equivalence between the present field-theoretical approach and the standard derivation of the Lindblad equation, we express (8.8) in matrix form. Using

$$\langle k|S^{\alpha}(\omega)|k'\rangle = S_{kk'}^{\alpha}\delta(\omega - E_{k'k}), \quad (10.1)$$

$$\langle k|S^{\alpha}(\omega)^{\dagger}|k'\rangle = S_{kk'}^{\alpha\dagger}\delta(\omega + E_{k'k}), \quad (10.2)$$

summing over ω, and symmetrizing the first term, we obtain for the dissipative kernel

$$
\begin{aligned}
L_{pp',qq'}^{\text{ST}} = &+\frac{1}{2}\sum_{\alpha\beta}\gamma^{\alpha\beta}(E_{qp})S_{pq}^{\beta}\rho_S(t)_{qq'}S_{q'p'}^{\alpha\dagger}\delta(E_{pq}+E_{q'p'})\\
&+\frac{1}{2}\sum_{\alpha\beta}\gamma^{\alpha\beta}(E_{q'p'})S_{pq}^{\beta}\rho_S(t)_{qq'}S_{q'p'}^{\alpha\dagger}\delta(E_{pq}+E_{q'p'})\\
&-\frac{1}{2}\delta_{p'q'}\sum_{\alpha\beta,l}\gamma^{\alpha\beta}(E_{pl})S_{pl}^{\alpha\dagger}S_{lq}^{\beta}\rho_S(t)_{qq'}\delta(E_{pl}+E_{lq})\\
&-\frac{1}{2}\delta_{pq}\sum_{\alpha\beta,l}\gamma^{\alpha\beta}(E_{p'l})\rho_S(t)_{qq'}S_{q'l}^{\alpha\dagger}S_{lp'}^{\beta}\delta(E_{q'l}+E_{lp'}).
\end{aligned}
$$
$$(10.3)$$

Using $\sum_\alpha S^\alpha B^\alpha = \sum_\alpha S^{\alpha\dagger}B^{\alpha\dagger}$ we obtain complete agreement with the field-theoretical expression in (9.9). This equivalence demonstrates that the field-theoretical approach in combination with a RWA approximation yields the same expression for the Lindblad equation as the standard approach. With the exception of the added RWA, the quasiparticle approximation replaces the physical approximation in the standard approach.

The assumption of separation of timescales is essential in obtaining a Markov master equation and is used throughout in the standard derivation of the Lindblad equation. In the present field-theoretical approach, the timescale separation is implicit in the quasiparticle approximation locking the frequency in the kernel $\tilde{K}(\omega)$ onto the level energy separation ΔE. For this approximation to be valid, we must assume that the kernel varies slowly over a frequency range of order

$\tau_S \approx 1/\Delta E$. To illustrate this point, we assume that $K(t)$ due to the fast decay of the bath correlation behaves approximately like $K(t) \approx \exp(-t/\tau_B)$, where τ_B is the bath correlation time. In Fourier space we then have $\tilde{K}(\omega) \approx 1/[\omega^2 + (1/\tau_B)^2]$, and the slow variation of $\tilde{K}$ implies the timescale separation $\tau_B \ll \tau_S$. We note that a simple version of the many-body quasi-particle approximation is also encountered in the standard Wigner-Weisskopf analysis of spontaneous emission [66], where the timescale $1/\omega_0$ associated with the level splitting ω_0 is assumed to be slow compared to the fast timescale associated with the radiation field. The resulting pole approximation corresponds to the Markov approximation.

B. Diosi-Ferialdi approach

There is currently a strong interest in non-Markovian features of open quantum systems [43–47]. In this subsection we address recent work by Diosi and Ferialdi [42,43], who present an exact analytical expression for the transmission operator.

Choosing the initial time $t_i = 0$ and inserting from Appendix 1 Eqs. (A16) and (A17), the transmission operator is given by a formal expression in terms of time-ordered and anti-time-ordered products according to

$$T(t, 0) = \exp(-iH_S t) \text{Tr}_B \left\{ \left[\exp\left(-i \int_0^t dt' \boldsymbol{S}(t') \boldsymbol{B}(t') \right) \right]_+ \rho_B \left[\exp\left(+i \int_0^t dt' \boldsymbol{S}(t') \boldsymbol{B}(t') \right) \right]_- \right\} \exp(+iH_S t). \quad (10.4)$$

Here the time-ordered term $[\cdots]_+$ refers to the upper branch of the so-called Keldysh contour from $t = 0$ to t, and the anti-time-ordered term $[\cdots]_-$ refers to the lower branch of the Keldysh contour from t to $t = 0$ [52,53]. In the context of nonequilibrium many-body theory, the two branches are adjacent and Wick's theorem in its generator form from Appendix 2 (A28) can be applied to the path-ordered operators along the complete Keldysh contour from $t = 0$ to t and back to $t = 0$. This is the basis for diagrammatic nonequilibrium many-body theory [52,53].

In the case of an open quantum system, as exemplified in (10.4), the density operator ρ_B entering in the bath average separates the two Keldysh branches, and Wick's theorem cannot be directly applied. In a series of intriguing papers, Diosi and Ferialdi [42,43], see also [67,68], have remedied this feature by introducing "left" and "right" operators according to the prescription $\boldsymbol{S}_L \boldsymbol{B}_L \rho_B \rho_S(0) = \boldsymbol{S} \boldsymbol{B} \rho_B \rho_S(0)$ and $\boldsymbol{S}_R \boldsymbol{B}_R \rho_B \rho_S(0) = \rho_B \rho_S(0) \boldsymbol{S} \boldsymbol{B}$. In this case, the equation of motion for the density operator in the interaction representation, $\rho^I(t) = \exp(iH_0 t) \rho(t) \exp(-iH_0 t)$, $i d\rho^I(t)/dt = [\boldsymbol{S}(t)\boldsymbol{B}(t), \rho^I(t)]$, takes the form $i d\rho^I(t)/dt = [\boldsymbol{S}_L(t)\boldsymbol{B}_L(t) - \boldsymbol{S}_R(t)\boldsymbol{B}_R(t)]\rho^I(t)$ with time-ordered solution $\rho^I(t) = [\exp(-i \int_0^t dt' [\boldsymbol{S}_L(t)\boldsymbol{B}_L(t) - \boldsymbol{S}_R(t)\boldsymbol{B}_R(t)])]_+ \rho(0)$, corresponding to the transmission operator

$$T(t, 0) = \exp(-iH_0 t) \text{Tr}_B \left(\left[\exp\left(-i \int_0^t dt' (\boldsymbol{S}_L(t')\boldsymbol{B}_L(t') - \boldsymbol{S}_R(t')\boldsymbol{B}_R(t')) \right) \right]_+ \rho_B \right) \exp(+iH_0 t). \quad (10.5)$$

By means of this procedure the two Keldysh branches become adjacent, and Wick's theorem can be applied to the complete Keldysh contour in order to explicitly average over the bath, yielding a closed formal expression for the transmission matrix. Referring to [42,43] for details one arrives at

$$T(t, 0) = \exp(-iH_0 t) \left[\exp\left(\int_0^t dt' \int_0^t dt'' D^{\alpha\beta}(t', t'') Q_{RL}^{\alpha\beta}(t', t'') \right) \right]_+ \exp(+iH_0 t)$$

$$Q_{RL}^{\alpha\beta}(t', t'') = S_L^\beta(t'') S_R^\alpha(t') - \theta(t' - t'') S_R^\alpha(t') S_L^\beta(t'') - \theta(t'' - t') S_R^\beta(t'') S_R^\alpha(t'), \quad (10.6)$$

where the time-ordering $[\cdots]_+$ still applies to the system operators $S_{R,L}^{\alpha,\beta}(t)$ in $Q_{RL}^{\alpha\beta}(t', t'')$. The result (10.6) provides an exact formal non-Markovian expression for the transmission operator. In further developments in [43] Ferialdi also discusses the Hu-Paz-Zhang model for quantum Brownian motion [69].

There is here a parallel to the Feynman path integral representation in quantum field theory or condensed-matter many-body theory [60]. In both cases, the closed form permits a concise way of checking symmetries, etc. However, for practical purposes one must often resort to actual expansions in terms of the interaction, typically diagrammatic expansion organized according to appropriate Feynman rules.

In the present context of open quantum systems, the expansion of $T(t, 0)$ in (10.6) in powers of the interaction H_{SB}, applying the time-order prescription and rearranging the system operators $S_{R,L}^{\alpha,\beta}$, should reproduce the diagrammatic expansion discussed here. However, we should like to em-

phasize that the present approach based on diagrammatic perturbations theory allows for a derivation of the master equation, an identification of the irreducible kernel, and diagrammatic rules for the determination of K to any desired order in the interaction; we note that the use of Wick's theorem in expanding time-ordered products is a standard tool in condensed matter and field theory going back to the development of quantum electrodynamics.

C. Nakajima-Zwanzig approach

An approach to open quantum systems has also been formulated using the method by Nakajima and Zwanzig (NZ) [2,9,19,70–74]. This approach relies on a projection techniques yielding a formal expression for the master equation with a memory kernel. In the case of a memoryless kernel, the time-convolutionless projection operator formalism, yielding a perturbative expansion, has also been

developed [75–78]. For details, we refer to [9]; see also [74,79–88].

Below we briefly summarize the NZ approach. Referring to the exposition in [9], the starting point is the von Neumann equation in the interaction representation in the form

$$\frac{d\rho^I(t)}{dt} = L(t)\rho^I(t), \tag{10.7}$$

where $L(t)$ is the Liouville superoperator acting according to $L(t)\bullet = -i[H_{SB}(t), \bullet]$; note that $\rho^I(t) = \exp(iH_0 t)\rho(t)\exp(-iH_0 t)$. The formal solution of (10.7) is thus given by

$$\rho^I(t) = \left[\exp\left(\int_0^t dt' L(t')\right)\right]_+ \rho(0). \tag{10.8}$$

By expanding, it is easily verified that this expression is completely equivalent to the expression

$$\rho^I(t) = \left[\exp\left(-i\int_0^t dt H_{SB}(t')\right)\right]_+ \rho(0)$$
$$\times \left[\exp\left(+i\int_0^t dt' H_{SB}(t')\right)\right]_-, \tag{10.9}$$

forming the basis for diagrammatic perturbation theory; here $[\cdots]_\pm$ denotes the time-ordered and anti-time-ordered products, respectively.

In the NZ approach, one introduces a projection operator P according to the definition

$$P\rho^I(t) = \rho_S^I(t)\rho_B, \tag{10.10}$$

where $\rho_S^I(t)$ is the reduced density operator for the system, and ρ_B is the density operator for the bath. Correspondingly, defining $Q = 1 - P$, we have the relations $P + Q = 1$, $P^2 = P$, $Q^2 = Q$, and $PQ = QP = 0$. Applying this scheme to (10.7), we obtain coupled equations of motion for $P\rho^I$ and $Q\rho^I$. Solving the equation for $Q\rho^I$ with initial condition $Q\rho(0)$ and inserting in the equation for $P\rho^I$, we obtain the Nakajima-Zwanzig equation

$$\frac{dP\rho^I(t)}{dt} = PL(t)G(t,0)Q\rho(0) + PL(t)P\rho^I(t)$$
$$+ \int_0^t dt' K(t,t')P\rho^I(t'), \tag{10.11}$$

$$G(t,t') = \left[\exp\left(\int_{t'}^t dt' QL(t')\right)\right]_+, \tag{10.12}$$

$$K(t,t') = PL(t)G(t,t')QL(t')P. \tag{10.13}$$

Assuming that odd moments of H_{SB} vanish, and choosing a factorized initial condition $\rho(0) = \rho_S(0)\rho_B$, we obtain for $P\rho(t)$

$$\frac{dP\rho(t)}{dt} = \int_0^t dt' K(t,t')P\rho(t'). \tag{10.14}$$

To leading order in H_{SB} we have $G = I$, and we obtain $K(t,t') = PL(t)QL(t')P$, yielding the second-order NZ master equation

$$\frac{dP\rho(t)}{dt} = \int_0^t dt' PL(t)L(t')P\rho(t'), \tag{10.15}$$

which, implementing the definitions of L, P, and Q, agrees with the expression (8.5) in Sec. VIII.

Since the projection $P\rho^I(t) = \rho_S^I(t)\rho_B$ treats ρ_B as an inert background (bath) and to leading order yields the expression (8.5), it appears that the projection basically corresponds to the physical assumption $\rho^I(t) \approx \rho_S^I(t)\rho_B$ in the derivation of (8.5) in Sec. VIII.

Another issue regarding the NZ approach and the standard derivation in Sec. VIII is the dependence of the master equations (10.14), (10.15), and (8.5) on the initial preparation at $t = 0$, i.e., the lower integration limit. In the NZ approach, this feature is associated with inserting the solution of the equation for $Q\rho^I$ with initial condition $Q\rho(0)$ in the equation of motion for $P\rho^I$. Clearly, a proper evolution equation like the Schrödinger equation or the von Neumann equation cannot depend on the initial preparation. Likewise, this must hold for a proper non-Markovian evolution equation for the reduced density operator.

It seems that this dependence on the initial condition indicates that secular effects are not properly included in the NZ approach. In the standard derivation of the Lindblad equation in Sec. VIII, secular effects are included by applying the rotating-wave approximation.

In condensed-matter many-body theory, the issue of secular effects was discussed briefly in Sec. IX. Secular effects are properly included by the construction of the Dyson equation for the single-particle Green's function. Likewise, in the present diagrammatic approach to open quantum systems, a Dyson equation is constructed for the transmission matrix in Sec. IV. Schematically, the Dyson equation for T has the form $T = T^0 + T^0 KT$ given by (4.8), yielding in Sec. V the general non-Markovian master equation $\dot\rho_S = -i[H_S, \rho_S] + K\rho_S$ in (5.3); note that if we *incorrectly* make a direct expansion of the Dyson equation to leading order, i.e., $T \approx T^0 + T^0 KT^0$, the definition $\rho_S = T\rho_S(0) = (T^0 + T^0 KT^0)\rho_S(0)$ yields a master equation $\dot\rho_S = (\dot{T}^0 + \dot{T}^0 KT^0)\rho_S(0)$, depending on the initial condition at $t = 0$.

The fact that the NZ approach does not include secular effects implies that we cannot compare the NZ approach to the systematic diagrammatic method presented here. As discussed above, even to leading order we encounter a discrepancy. Also regarding the time-convolutionless projection operator formalism yielding a time-local or memoryless master equation, we are prevented from a direct comparison since the diagrammatic approach by construction accounts for memory effects.

D. Qubit coupled to a heat bath

To illustrate the field-theoretical scheme developed in the previous sections to a particular open quantum system interacting with a bath, we must specify the system Hamiltonian H_S, the system operators S^α, the bath Hamiltonian H_B, the bath operators B^α, and the corresponding bath correlations $D^{\alpha\beta}$. Here we consider the well-known and much studied case of a two-level system or qubit coupled to a single heat bath [9]. The isolated qubit is characterized by the two-state Hamiltonian

$$H_S = \frac{\omega_0}{2}\sigma^z, \tag{10.16}$$

where the two energy levels are denoted $|-\rangle$ and $|+\rangle$ with splitting ω_0. In the Pauli matrix basis [60] $\sigma^z, \sigma^\pm$ we have $\sigma^+|-\rangle = |+\rangle$, $\sigma^-|+\rangle = |-\rangle$, and $\sigma^z|\pm\rangle = \pm|\pm\rangle$. For the coupling to the bath we choose $H_{SB} = \sigma^+ \sum_k \lambda_k b_k + \sigma^- \sum_k \lambda_k b_k^\dagger$. With the assignment $S^1 = \sigma^+$, $S^2 = \sigma^-$, $B^1 = B$, and $B^2 = B^\dagger$, the coupling is

$$H_{SB} = S^1 B^1 + S^2 B^2. \tag{10.17}$$

By inspection of the shift (9.8) and the kernel (9.9), using $(S^1)_{+-} = 1$, $(S^2)_{-+} = 1$, and $E_{+-} = \omega_0$, and inserting $\tilde{D}^{12}$ and $\tilde{D}^{21}$ from (3.14) and (3.15), noting that $\tilde{D}^{11} = \tilde{D}^{22} = 0$, we obtain the nonvanishing shift elements

$$\tilde{\Delta}_{+-,+-} = -\tilde{\Delta}_{-+,-+} = -\mathrm{P} \int_0 \frac{d\omega}{2\pi} \frac{g(\omega)[1 + 2n(\omega)]}{\omega_0 - \omega} \tag{10.18}$$

and the nonvanishing kernel elements

$$\tilde{K}_{++,++} = -g(\omega_0)[1 + n(\omega_0)], \tag{10.19}$$

$$\tilde{K}_{--,++} = +g(\omega_0)[1 + n(\omega_0)], \tag{10.20}$$

$$\tilde{K}_{++,--} = +g(\omega_0)n(\omega_0), \tag{10.21}$$

$$\tilde{K}_{--,--} = -g(\omega_0)n(\omega_0), \tag{10.22}$$

$$\tilde{K}_{+-,+-} = -\tfrac{1}{2}g(\omega_0)[1 + 2n(\omega_0)], \tag{10.23}$$

$$\tilde{K}_{-+,-+} = -\tfrac{1}{2}g(\omega_0)[1 + 2n(\omega_0)]. \tag{10.24}$$

We note that the δ-function conditions originating from the RWA are automatically satisfied in the present case. With the notation $g(\omega_0) = g_0$, $n(\omega_0) = n_0$, and $\tilde{\Delta}_{+-,+-} = \Delta$, we

subsequently obtain the master equation

$$\frac{d}{dt}\rho_S(t)_{++} = -g_0(1 + n_0)\rho_S(t)_{++} + g_0 n_0 \rho_S(t)_{--}, \tag{10.25}$$

$$\frac{d}{dt}\rho_S(t)_{--} = +g_0(1 + n_0)\rho_S(t)_{++} - g_0 n_0 \rho_S(t)_{--}, \tag{10.26}$$

$$\frac{d}{dt}\rho_S(t)_{--} = -i(\omega_0 - \Delta)\rho_S(t)_{+-} - \frac{1}{2}g_0(1 + 2n_0)\rho_S(t)_{+-}, \tag{10.27}$$

$$\frac{d}{dt}\rho_S(t)_{-+} = +i(\omega_0 - \Delta)\rho_S(t)_{-+} - \frac{1}{2}g_0(1 + 2n_0)\rho_S(t)_{-+}, \tag{10.28}$$

where we note that the shift Δ can be absorbed in a renormalization of the level shift ω_0, i.e., a Lamb shift. Finally, introducing the operators σ^+, σ_-, and σ^z, the corresponding Lindblad master equation has the form

$$\frac{d}{dt}\rho_S = -i[(\omega_0 - \Delta)\sigma^z/2, \rho_S]$$
$$+ D^{-+}\left[\sigma^- \rho_S \sigma^+ - \frac{1}{2}\{\sigma^+\sigma^-, \rho_S\}\right]$$
$$+ D^{+-}\left[\sigma^+ \rho_S \sigma^- - \frac{1}{2}\{\sigma^-\sigma^+, \rho_S\}\right], \tag{10.29}$$

where we have set $D^{+-} = g_0 n_0$ and $D^{-+} = g_0(1 + n_0)$ in compliance with (1.1).

Here we discuss the quasiparticle approximation for the qubit-bath case in more detail. According to (9.3), the inverse transmission matrix has the form

$$\tilde{T}(\omega)^{-1} = \begin{pmatrix} -i\omega_{++} - \tilde{K}_{++,++}(\omega) & -\tilde{K}_{++,--}(\omega) & 0 & 0 \\ -\tilde{K}_{--,++}(\omega) & -i\omega_{--} - \tilde{K}_{--,--}(\omega) & 0 & 0 \\ 0 & 0 & -i\omega_{+-} - \tilde{K}_{+-,+-}(\omega) & 0 \\ 0 & 0 & 0 & -i\omega_{-+} - \tilde{K}_{-+,-+}(\omega) \end{pmatrix}, \tag{10.30}$$

with the notation $\omega_{pp'} = \omega - E_{pp'}$. The resonance condition is given by $\det[\tilde{T}(\omega)^{-1}] = 0$ and we obtain

$$\omega - \omega_0 + i\tilde{K}_{+-,+-}(\omega) = 0, \tag{10.31}$$

$$\omega + \omega_0 + i\tilde{K}_{-+,-+}(\omega) = 0, \tag{10.32}$$

$$[\omega - i\tilde{K}_{++,++}(\omega)][\omega - i\tilde{K}_{--,--}(\omega)] + \tilde{K}_{++,--}$$
$$\times(\omega)\tilde{K}_{--,++}(\omega) = 0. \tag{10.33}$$

Applying the quasiparticle approximation, we note that to leading order $\omega = \omega_0$ in (10.31), $\omega = -\omega_0$ in (10.32), and $\omega = 0$ in (10.33), yielding $\tilde{K}_{+-,+-}(\omega_0)$, $\tilde{K}_{-+,-+}(-\omega_0)$, $\tilde{K}_{++,++}(0)$, $\tilde{K}_{--,--}(0)$, $\tilde{K}_{++,--}(0)$, and $\tilde{K}_{--,++}(0)$. Note that $K_{--,++}$ and $K_{--,--}$ also follows from the trace condition $K_{++,++} + K_{--,++} = 0$ and $K_{++,--} + K_{--,--} = 0$.

XI. SUMMARY

In the present paper, we have applied condensed-matter many-body methods to open quantum systems. We have derived a general non-Markovian master equation for the reduced density operator characterized by an irreducible kernel allowing for a systematic diagrammatic expansion. We have in particular considered the Born approximation. We have shown that the quasiparticle approximation, common in condensed-matter many-body theory, in the context of open quantum systems is equivalent to the standard Markov approximation implying a separation of timescales. Implementing the rotating-wave approximation, we have demonstrated that the Lindblad equation follows from the Markov approximation. We have, moreover, discussed the Nakajima-Zwanzig method and its relation to the present diagrammatic approach. As an application of the many-body approach, we have discussed the coupling of a qubit to a thermal heat bath. Technical aspects

of the analysis are supported by an Appendix. We believe that the present approach correctly including secular effects allows for a more systematic microscopic analysis of open quantum systems. Finally, we list here further references relevant to the present approach [88–93].

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APPENDIX

1. Interaction representation

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The interaction representation in the context of perturbation theory is textbook material [32,33]. However, in order to render our presentation self-contained, we present the procedure below applied to open quantum systems.

The open quantum system (S) coupled to the bath (B) is described by the Hamiltonian

$$H = H_S + H_B + H_{SB}, \tag{A1}$$

$$H_{SB} = \sum_\alpha S^\alpha B^\alpha = \boldsymbol{S} \cdot \boldsymbol{B}, \tag{A2}$$

where H_S is the system Hamiltonian, H_B is the bath Hamiltonian, and H_{SB} is the system-bath interaction. Here $\boldsymbol{S}$ and $\boldsymbol{B}$ are the system and bath operators, respectively. The time evolution of the total system is governed by the unitary operator $U(t, t')$ satisfying an evolution equation, i.e.,

$$U(t, t') = \exp(-iH(t - t')), \tag{A3}$$

$$\frac{d}{dt}U(t, t') = -iHU(t, t'). \tag{A4}$$

To treat the system-bath coupling H_{SB} perturbatively, we apply the interaction representation. Introducing the Hamiltonian H_0 for the uncoupled bath and system,

$$H_0 = H_S + H_B, \tag{A5}$$

we have in the interaction representation

$$H_{SB}(t) = \exp(iH_0 t)H_{SB}\exp(-iH_0 t), \tag{A6}$$

$$\boldsymbol{B}(t) = \exp(iH_B t)\boldsymbol{B}\exp(-iH_B t), \tag{A7}$$

$$\boldsymbol{S}(t) = \exp(iH_S t)\boldsymbol{S}\exp(-iH_S t), \tag{A8}$$

$$\tilde{U}(t, t') = \exp(iH_0 t)U(t, t')\exp(-iH_0 t'), \tag{A9}$$

where $\tilde{U}(t, t')$ satisfies the evolution equation together with its integrated form,

$$\frac{d}{dt}\tilde{U}(t, t') = -iH_{SB}(t)\tilde{U}(t, t'), \tag{A10}$$

$$\tilde{U}(t, t') = I - i\int_{t'}^t dt'' H_{SB}(t'')\tilde{U}(t'', t'). \tag{A11}$$

Iterating (A11), we obtain expansions for $\tilde{U}(t, t')$ and its conjugate $\tilde{U}(t, t')^\dagger$ according to

$$\tilde{U}(t, t') = \sum_{n=0} (-i)^n \int_{t'}^t dt_n \int_{t'}^{t_n} dt_{n-1} \cdots \int_{t'}^{t_2} dt_1 H_{SB}(t_n) \cdots H_{SB}(t_1), \tag{A12}$$

$$\tilde{U}(t, t')^\dagger = \sum_{n=0} (+i)^n \int_{t'}^t dt_n \int_{t'}^{t_n} dt_{n-1} \cdots \int_{t'}^{t_2} dt_1 H_{SB}(t_1) \cdots H_{SB}(t_n). \tag{A13}$$

Introducing the time-order and anti-time-order according to the prescriptions

$$[H_{SB}(t)H_{SB}(t')]_+ = H_{SB}(t)H_{SB}(t')\eta(t - t') + H_{SB}(t')H_{SB}(t)\eta(t' - t), \tag{A14}$$

$$[H_{SB}(t)H_{SB}(t')]_- = H_{SB}(t)H_{SB}(t')\eta(t' - t) + H_{SB}(t')H_{SB}(t)\eta(t - t'), \tag{A15}$$

and using (A9), we have compactly for the evolution operators $U(t, t')$ and $U(t, t')^\dagger$ the time-ordered products [50,52]

$$U(t, t') = \exp(-iH_0 t)\left[\exp\left(-i\int_{t'}^t dt'' H_{SB}(t'')\right)\right]_+ \exp(+iH_0 t'), \tag{A16}$$

$$U(t, t')^\dagger = \exp(-iH_0 t')\left[\exp\left(+i\int_{t'}^t dt'' H_{SB}(t'')\right)\right]_- \exp(+iH_0 t). \tag{A17}$$

Expanding Eqs. (A14) and (A15) or Eqs. (A16) and (A17), inserting Eqs. (A6)–(A8), and the retarded and advanced Green's functions

$$G_R(t, t') = -i\eta(t - t')\exp(-iH_S(t - t')), \tag{A18}$$

$$G_A(t, t') = +i\eta(t' - t)\exp(-iH_S(t - t')), \tag{A19}$$

we obtain the expansions

$$U(t, t') = +i\sum_{n=0} \int dt_n dt_{n-1} \cdots dt_1 G_R(t, t_n)\boldsymbol{S}_n G_R(t_n, t_{n-1})\boldsymbol{S}_{n-1} \cdots \boldsymbol{S}_2 G_R(t_2, t_1)\boldsymbol{S}_1 G_R(t_1, t')$$

$$\times e^{-iH_B t}\boldsymbol{B}_n(t_n)\boldsymbol{B}_{n-1}(t_{n-1}) \cdots \boldsymbol{B}_2(t_2)\boldsymbol{B}_1(t_1)e^{iH_B t'}, \tag{A20}$$

$$U(t, t')^{\dagger} = -i \sum_{n=0} \int du_n du_{n-1} \cdots du_1 G_A(t', u_1) \boldsymbol{S}_1 G_A(u_1, u_2) \boldsymbol{S}_2 \cdots \boldsymbol{S}_{n-1} G_A(u_{n-1}, u_n) \boldsymbol{S}_n G_A(u_n, t)$$

$$\times e^{-iH_B t'} \boldsymbol{B}_1(u_1) \boldsymbol{B}_2(u_2) \cdots \boldsymbol{B}_{n-1}(u_{n-1}) \boldsymbol{B}_n(u_n) e^{iH_B t}, \tag{A21}$$

required for the analysis in Sec. II.

2. Wick's theorem

Within the Caldeira-Leggett prescription of the bath in terms of independent bosons (quantum oscillators) and assuming that the bath operators $B^{\alpha}(t)$ are linear combinations of creation and annihilation Bose operators, Wick's theorem implies that the average of a product of ordered bath operators $\mathrm{Tr}\rho_B B_1(t_1) \cdots B_n(t_n)$ can be broken up into all possible pairings or contractions with the time-order preserved.

a. Proof by Gaudin

Here we summarize a proof by Gaudin [59] directly applied to a thermal average of operator products relevant to the present analysis. Since the bath operators $B^{\alpha}(t)$ are linear combinations of the creation and annihilation operators pertaining to a specific wave number, and the Hamiltonian H_B is a sum of contributions from each node, it is sufficient to consider the thermal average of the ordered product $d_1 d_2 \cdots d_n$,

$$\langle d_1 d_2 \cdots d_n \rangle = \mathrm{Tr}[\rho d_1 d_2 \cdots d_n], \tag{A22}$$

with the abbreviation $d_1 \equiv d_1(t_1)$. Here d_n is either an annihilation operator $b_k(t)$ or a creation operator $b_k^{\dagger}(t)$ with time evolution given by (3.3) and (3.4); from (3.2) the density operator for the kth mode is $\rho = \exp(-\beta\Omega_k n_k)/\mathrm{Tr}[\exp(-\beta\Omega_k n_k)]$.

First cyclically moving ρ to the end of the trace and subsequently permuting d_1 to the end of the operator product, we obtain, moving the c-number commutator outside the trace, the intermediate expansion

$$\mathrm{Tr}[d_1 d_2 \cdots d_n \rho] = [d_1, d_2]\mathrm{Tr}[d_3 d_4 \cdots d_n \rho]$$
$$+ \cdots [d_1, d_n]\mathrm{Tr}[d_2 \cdots d_{n-1}\rho]$$
$$+ \mathrm{Tr}[d_2 \cdots d_n d_1 \rho], \tag{A23}$$

Next using the identity $d_1 \rho = \rho d_1 z$, where $z = \exp(\beta\Omega_k)$ for $d_1 = b_k^{\dagger}$ and $z = \exp(-\beta\Omega_k)$ for $d_1 = b_k$, we can exchange

ρ and d_1, and by permuting the operators under the trace we obtain the expansion with the commutators $[d_1, d_p]$ replaced by $[d_1, d_p]/(1 - z)$. Applying the scheme to the case $n = 2$, we infer

$$[d_1, d_2]/(1 - z) = \mathrm{Tr}[\rho d_1 d_2] = \langle d_1 d_2 \rangle. \tag{A24}$$

Finally, we have

$$\langle d_1 d_2 \cdots d_n \rangle = \langle d_1 d_2 \rangle \langle d_3 d_4 \cdots d_n \rangle + \cdots \langle d_1 d_n \rangle \langle d_2 \cdots d_{n-1} \rangle, \tag{A25}$$

and by induction Wick's theorem, i.e., the ordered average, is reduced to all possible pairings of two operators, where we note that the order is preserved as shown in (3.16).

b. Wick's theorem in generator form

In the expression (A22) we have assumed a specific order of the operators $\{d_n(t)\}$; however, introducing the time-order and anti-time-order prescriptions

$$[d(t_n)d(t_m)]_+ = d(t_n)d(t_m)\theta(t_n - t_m) + d_m d_n \theta(t_m - t_n), \tag{A26}$$

$$[d(t_n)d(t_m)]_- = d(t_n)d(t_m)\theta(t_m - t_n) + d(t_m)d(t_n)\theta(t_n - t_m), \tag{A27}$$

and noting that the operators commute under time-ordering or anti-time-ordering, we infer the functional Wick theorem [60,94]

$$\left\langle \left[\exp\left(-i \int dt\, \Omega(t)d(t)\right) \right]_{\pm} \right\rangle$$
$$= \exp\left(-\frac{1}{2} \int dt dt'\, \Omega(t)\langle [d(t)d(t')]_{\pm} \rangle \Omega(t')\right), \tag{A28}$$

where $\Omega(t)$ is a generator field.

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