Systematic perturbation theory for dynamical coarse-graining

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We demonstrate how the dynamical coarse-graining approach can be systematically extended to higher orders in the coupling between system and reservoir. Up to second order in the coupling constant, we explicitly show that dynamical coarse-graining unconditionally preserves positivity of the density matrix—even for bath density matrices that are not in equilibrium and also for time-dependent system Hamiltonians. By construction, the approach correctly captures the short-time dynamics; i.e., it is suitable for analyzing non-Markovian effects. We compare the dynamics with the exact solution for highly non-Markovian systems and find a remarkable quality of the coarse-graining approach. The extension to higher orders is straightforward but rather tedious. The approach is especially useful for bath correlation functions of simple structure and for small system dimensions.

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I. INTRODUCTION

The insight that quantum computers may solve certain problems such as number factoring [1] and database search [2] more efficiently than conventional computers has given rise to the field of quantum information (for an overview see, e.g., [3]). The conventional paradigm of quantum computation relies on unitary operations that act on low-dimensional subspaces of the 2^n-dimensional Hilbert space of n two-level systems—conventionally called qubits. Unfortunately, the dynamics of open quantum systems is not always unitary [4], such that the impact of decoherence has to be taken into account. This problem also affects alternative schemes such as one-way [5,6], holonomic [7], or adiabatic [8] quantum computation. Beyond this, the study of decoherence effects is of general interest in the control of quantum systems.

Often, the dynamics of open quantum systems is analyzed within the Born-Markov approximation scheme [4,9]. An important criticism raised against this scheme is that it does not generally preserve positivity of the reduced density matrix [10–13], which however is necessary for its probability interpretation [14]. In addition, the Born-Markov master equation cannot be expected to yield good results for short times, which may in the context of quantum computation, for example, lead to false error estimates on required gate operation times, etc. [15,16].

A possible resolution for the latter problem is to study non-Markovian master equations (that explicitly depend on the density matrix at all previous times via a memory kernel). However, except for some special cases [17], non-Markovian master equations are also not guaranteed to preserve positivity, and corresponding counterexamples can be easily constructed [18]. Technically, master equations with memory can, for example, be solved efficiently when the bath correlation functions can be approximated by a few decaying exponentials [19]. In the general case, they are however difficult if not impossible to solve analytically. This difficulty transfers to the numerical solution as well. In order to evolve the density matrix at time t, one would generally have to evaluate the solution at all previous times t' < t, which corresponds to significant computational and storage efforts.

It is therefore interesting to investigate alternatives such as the dynamical coarse-graining (DCG) approach. Recently, it has been analyzed up to second order in the system-reservoir coupling constant (Born approximation) [20]. Instead of solving a single quantum master equation, the coarse-graining approach defines a continuous set of master equations,

\[ \dot{\rho}_S = \mathcal{L}\rho_S(t), \]

parametrized by the coarse-graining time \( \tau \) and then interpolates through the set of solutions at \( t = \tau \),

\[ \overline{\rho}_S(t) = e^{\mathcal{L}\tau} \rho_S(0). \]

Since the Liouville superoperators \( \mathcal{L} \) are of Lindblad form [21] for all \( \tau > 0 \), the second-order dynamical coarse-graining (DCG2) approach preserves positivity of the density matrix at all times [20]. Note that in the general case, the above solution cannot be obtained by solving a single Lindblad form master equation merely equipped with time-dependent coefficients and should therefore be regarded as truly non-Markovian [22]. The conventional Born-Markov secular limit is obtained by the limit \( \tau \to \infty \), i.e.,

\[ \rho_S \to \mathcal{L} = \text{BMS}, \]

whereas in the short-time limit, the exact full solution is approximated. In addition, it was found for some simple examples considered in [20] that in the weak-coupling limit, the method approximated the results of the non-Markovian master equation for all times remarkably well.

The purpose of the present paper is twofold. By introducing coarse-graining in the interaction picture in Sec. II, we rigorously demonstrate that the method will approximate the exact solution for short times by construction; i.e., the method is suitable for studying non-Markovian effects. By including higher orders in the coupling constant, the agreement between coarse-graining and exact solution can be further improved. In addition, we show that up to second order

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the method unconditionally preserves positivity of the density matrix, i.e., even for bath density matrices that do not commute with the bath Hamiltonian and/or for time-dependent system Hamiltonians. We will give several examples for finite-size "baths" (Secs. III A and III B) and the spin-boson model in Sec. III C, and we also consider fermionic models with transport in Sec. III D.

II. DCG IN THE INTERACTION PICTURE

A. Preliminaries

We consider systems where the time-independent Hamiltonian can be divided into three parts,

\[ H = H_S + H_{SB} + H_B, \]

where \( H_S \) denotes the system Hamiltonian, \( H_B \) denotes the bath (reservoir) Hamiltonian (with \([H_S, H_B] = 0\)), and

\[ H_{SB} = \lambda \sum_a A_a \otimes B_a \]

(4)
couples the two by system \((A_a)\) and bath \((B_a)\) operators. Note that thereby one has by construction \([A_a, B_B] = 0\) (see Sec. III D for obtaining such a decomposition for fermionic systems with transport).

Note that Hermiticity of \( H_{SB} = H_{SB}^\dagger \) imposes some constraints on the coupling operators. For example, it is always possible to perform a suitable redefinition of operators by splitting into Hermitian and anti-Hermitian parts \((A_a = A_a^H + A_a^\dagger)\) and \((B_a = B_a^H + B_a^\dagger)\), for system and bath operators, respectively, to obtain \( H_{SB} = \frac{1}{2}(H_{SB} + H_{SB}^\dagger) = \sum_a [A_a^H B_a^\dagger - i A_a^\dagger B_a^H]\), such that one can always assume Hermitian coupling operators \(A_a = A_a^\dagger\) as well as \(B_a = B_a^\dagger\) [4]. For the sake of convenience however, we will not assume this form here unless stated otherwise. We will use \(\lambda < 1\) as a perturbation parameter (\(\alpha\)-dependent coupling constants can be absorbed in the operator definitions).

In the interaction picture (where we will denote all operators by bold symbols)

\[ \rho(t) = e^{iH_S t + H_B t} \rho(0) e^{-iH_S t + H_B t}, \]

\[ A_a(t) = e^{iH_S t} A_a e^{-iH_S t}, \]

\[ B_a(t) = e^{iH_S t} B_a e^{-iH_S t}, \]

(5)
the von Neumann equation reads

\[ \dot{\rho} = -i[H_S(t), \rho(t)], \]

(6)
which is formally solved by \(\rho(t) = U(t) \rho_0 U(t)\).

B. Perturbative expansion

The time-evolution operator in the interaction picture is governed by \(\dot{U} = -iH_{SB}(t) U(t)\), which can be solved iteratively. We can define the truncated time-evolution operator in the interaction picture via

\[ U_n(t) = \sum_{k=0}^{n} (-i)^k \int_0^t H_{SB}(t_1) \cdots H_{SB}(t_k) \times \Theta(t_1 - t) \cdots \Theta(t_{k-1} - t) dt_1 \cdots dt_k, \]

(7)
where the time ordering is expressed by Heaviside step functions. The above operator is unitary up to order of \(\lambda^n\) (assuming that \(H_{SB} = O(\lambda)\)), i.e., \(U_n(t) = I + O(\lambda^{n+1})\). Specifically, one has up to fourth order

\[ U_4(t) = 1 - i a \nu_1(t) - \lambda^2 \nu_2(t) + i \lambda^3 \nu_3(t) + \lambda^4 \nu_4(t), \]

(8)
where we can use Eq. (4) to find for the operators

\[ \nu_1(t) = \sum_a \int_0^t dt_1 A_a(t_1) B_a(t_1), \]

\[ \nu_2(t) = \sum_{a \beta} \int_0^t dt_1 dt_2 \Theta(t_1 - t_2) A_a(t_1) B_a(t_1) A_{\beta} B_{\beta}(t_2), \]

\[ \nu_3(t) = \sum_{a \beta \gamma} \int_0^t dt_1 dt_2 dt_3 \Theta(t_1 - t_2) \Theta(t_2 - t_3) \]

\[ \times A_a(t_1) B_a(t_1) A_{\beta} B_{\beta}(t_2) A_{\gamma} B_{\gamma}(t_3), \]

\[ \nu_4(t) = \sum_{a \beta \gamma \delta} \int_0^t \Theta(t_1 - t_2) \Theta(t_2 - t_3) \Theta(t_3 - t_4) \]

\[ \times A_a(t_1) B_a(t_1) A_{\beta} B_{\beta}(t_2) A_{\gamma} B_{\gamma}(t_3) A_{\delta} B_{\delta}(t_4), \]

(9)
Using these expressions in the formal solution of the density matrix and collecting all terms of the same order, we obtain

\[ \rho(t) = \rho_0 - i \lambda [ - \rho_0 \nu_1(t) + \nu_1(t) \rho_0 + \lambda^2 \nu_2(t) \]

\[ + \nu_1(t) \rho_0 \nu_1(t) - \nu_2(t) \rho_0 - i \lambda [ \nu_2(t) \rho_0 \nu_1(t) - \nu_1(t) \rho_0 \nu_2(t) \]

\[ + \nu_2(t) \rho_0 \nu_2(t) - \nu_3(t) \rho_0 - \lambda^2 \nu_3(t) \]

\[ + \nu_2(t) \rho_0 \nu_3(t) - \nu_3(t) \rho_0 \nu_2(t) + \nu_3(t) \rho_0 + O(\lambda^3) \]

(10)
In order to obtain the reduced density matrix, we have to perform the trace over the bath degrees of freedom. We assume that at \(t_0 = 0\) the density matrix factorizes such that we have \(\rho_{SB} = \text{Tr}_B[\rho_0]\). Then we can define \(\rho_{SB}(t) = \text{Tr}_B[\rho(t)]\) and calculate the reduced density matrix at time \(t\),

\[ \rho_{SB}(t) = \rho_0 - i \lambda \text{Tr}_B[\rho_0 \nu_1(t) + \nu_1(t) \rho_0]\]

\[ + \lambda^2 \text{Tr}_B[\rho_0 \nu_2(t) + \nu_2(t) \rho_0 - \nu_3(t) \rho_0 - \nu_3(t) \rho_0 \nu_1(t) + \nu_3(t) \rho_0 \nu_2(t)\]

\[ - i \lambda^3 \text{Tr}_B[\rho_0 \nu_3(t) + \nu_3(t) \rho_0 \nu_1(t) + \nu_3(t) \rho_0 \nu_3(t) + \nu_3(t) \rho_0 \nu_3(t)\]

\[ + \lambda^4 \text{Tr}_B[\rho_0 \nu_4(t) + \nu_4(t) \rho_0 - \nu_4(t) \rho_0 \nu_1(t) + \nu_4(t) \rho_0 \nu_2(t)\]

\[ - \nu_4(t) \rho_0 + \nu_4(t) \rho_0 \nu_3(t) + \nu_4(t) \rho_0 \nu_3(t) + \nu_4(t) \rho_0 \nu_4(t) + O(\lambda^5) \]

(11)
where we can evaluate the right-hand side by using the reservoir correlation functions; see below.

\[ T'_{\alpha} \rho^S_\alpha = -i \sum_{\alpha} \int_{t_0}^{t} dt_1 \left\{ -C_\alpha(t_1) \rho^S_\alpha(t_1) + C_\alpha(t_1) \mathcal{A}_\alpha(t_1) \rho^S_\alpha(t_1) \right\}. \]

\[ T'_\alpha \rho^S_\beta = \sum_{\alpha \beta} \int_{t_0}^{t} dt_1 dt_2 dt_3 \left\{ -C_{\alpha \beta}(t_1, t_2) \Theta(t_2 - t_1) \rho^S_\beta(t_1) \mathcal{A}_\alpha(t_1) \mathcal{A}_\beta(t_2) + C_{\alpha \beta}(t_1, t_2) \mathcal{A}_\beta(t_2) \rho^S_\alpha(t_1) - C_{\alpha \beta}(t_1, t_2) \Theta(t_1 - t_2) \mathcal{A}_\alpha(t_1) \mathcal{A}_\beta(t_2) \rho^S_\beta(t_2) \right\}. \]

\[ \rho^S_\alpha(t) = \left\{ 1 + \lambda \tau \mathcal{L}_\alpha + \lambda^2 \left[ \tau \mathcal{L}_\alpha^2 + \frac{\tau^2}{2} \mathcal{L}_\alpha \right] \right\} \rho^S_\alpha + O(\lambda^5) \]

\[ \rho^S_\alpha(t) = \left\{ 1 + \lambda \tau \mathcal{L}_\alpha + \lambda^2 \left[ \tau \mathcal{L}_\alpha^2 + \frac{\tau^2}{2} \mathcal{L}_\alpha \right] + \frac{\lambda^3}{6} \left[ \tau \mathcal{L}_\alpha^3 + \frac{\tau^2}{2} (\mathcal{L}_\alpha \mathcal{L}_\alpha^2 + \mathcal{L}_\alpha^2 \mathcal{L}_\alpha) + \frac{\tau^3}{6} \mathcal{L}_\alpha \mathcal{L}_\alpha \mathcal{L}_\alpha \right] \right\} \rho^S_\alpha + O(\lambda^5) \]

We can clearly match this with Eq. (11) evaluated at \( t = \tau \) order by order to solve for
\[ \mathcal{L}^r \rho_S^0 = \frac{1}{\tau} \mathcal{T}^r \rho_S^0, \]

\[ \mathcal{L}^\ast_0 \rho_S^0 = \frac{1}{\tau} \left[ \frac{3}{2} \mathcal{L}^\ast_0 \mathcal{L}^\ast_0 - \frac{1}{2} \mathcal{L}^\ast_0 \mathcal{L}^\ast_0 \right] \rho_S^0, \]

\[ \mathcal{L}^\ast_0 \rho_S^0 = \frac{1}{\tau} \left[ \frac{3}{2} \mathcal{L}^\ast_0 \mathcal{L}^\ast_0 + \frac{3}{6} \mathcal{L}^\ast_0 \mathcal{L}^\ast_0 \right] \rho_S^0, \]

where \( \mathcal{T}_1^\ast, \mathcal{T}_1^\ast, \mathcal{T}_1^\ast, \mathcal{T}_1^\ast \) can be extracted from Eq. (13). Since these equations have to hold for all initial conditions \( \rho_S^0 \), we can infer the matrix elements of each Liouvillian by comparing coefficients of the matrix elements of \( \rho_S^0 \). Equation (15) defines in combination with Eq. (13) our coarse-graining Liouvillian. Evidently, we automatically approximate the short-time dynamics of the true solution very well by construction with this scheme.

### E. Unconditional positivity of DCG2

Here we will show that DCG2 always preserves positivity—regardless of whether the first-order correlation functions vanish or not. We do not even require that \( [H_B, \rho_S^0] = 0 \). For simplicity we assume Hermitian coupling operators \( A_a = A^\dagger_a \) and \( B_a = B^\dagger_a \). Then, we obtain from Eq. (13)

\[ \mathcal{T}_1^\dagger \rho_S = -i \sum_a \int_0^\tau dt_1 C_a(t_1) [A_a(t_1) \rho_S - \rho_S A_a(t_1)], \]

\[ \mathcal{T}_1^\ast \rho_S = \sum_{ab} \int_0^\tau dt_1 dt_2 C_{ab}(t_1, t_2) \left[ A_b(t_2) \rho_S A_a(t_1) \right. \]

\[ \left. - \frac{1}{2} \rho_S A_a(t_1) A_b(t_2) - \frac{1}{2} A_a(t_1) A_b(t_2) \rho_S \right] \]

\[ - i \sum_{ab} \frac{1}{2} \int_0^\tau C_{ab}(t_1, t_2) \text{sgn}(t_1 - t_2) \]

\[ \times [A_a(t_1) A_b(t_2)] \rho_S [dt_1 dt_2], \]

where we have used \( \Theta(x) = \frac{1}{2} [1 + \text{sgn}(x)] \) and \( \text{sgn}(-x) = -\text{sgn}(x) \) in the last equation. From the first of the above equations, we obtain that the first-order Liouvillian just generates a unitary evolution

\[ \mathcal{L}^\ast_1 \rho_S = \frac{1}{\tau} \int_0^\tau C_a(t_1) A_a(t_1) dt_1 \rho_S = -\frac{i}{\tau} [H_{eff}^\ast \rho_S], \]

where Hermiticity of the Lamb-shift Hamiltonian follows directly from Hermiticity of the coupling operators (which also implies real-valued first-order correlation functions). In addition, we obtain from consecutive application

\[ \frac{1}{\tau} \mathcal{T}^\ast_1 \mathcal{T}^\dagger_1 \rho_S = \sum_{ab} \int_0^\tau dt_1 dt_2 C_{ab}(t_1) C_{ab}(t_2) \]

\[ \times [A_b(t_2) \rho_S A_a(t_1) - \frac{1}{2} (\rho_S A_a(t_1) \rho_S A_b(t_2) + \rho_S A_b(t_2) \rho_S A_a(t_1))]. \]

This defines the second-order Liouvillian as

\[ \mathcal{L}^\ast_2 \rho_S = -i \int_0^\tau \sum_{ab} \int_0^\tau C_{ab}(t_1, t_2) \text{sgn}(t_1 - t_2) \]

\[ \times [A_a(t_1) A_b(t_2) dt_1 dt_2 \rho_S] \]

\[ + \frac{1}{\tau} \sum_{ab} \int_0^\tau dt_1 dt_2 [C_{ab}(t_1, t_2) - C_{ab}(t_1) C_{ab}(t_2)] \]

\[ \times [A_b(t_2) \rho_S A_a(t_1) - \frac{1}{2} (A_a(t_1) A_b(t_2) \rho_S) + (A_b(t_2) A_a(t_1) \rho_S)]. \]

The first commutator term induces a unitary evolution where Hermiticity of the corresponding effective Hamiltonian follows directly from \( C_{ab}(t_1, t_2) = C_{ab}(t_1, t_2) \). However, in contrast to the standard Born-Markov secular approximation \([4]\), here we have in general \( [H_{eff}^\ast, H_b] \neq 0 \). In order to see that the last expression corresponds to a Lindblad dissipator, we insert identities at suitable places \( 1 = \sum_{a} |a\rangle \langle a| \) to obtain

\[ \mathcal{L}^\ast_\Theta \rho_S = \frac{1}{\tau} \int_0^\tau \sum_{ab} \int_0^\tau C_{ab}(t_1, t_2) \text{sgn}(t_1 - t_2) \]

\[ \times [A_{ab}(t_1, t_2) - C_{ab}(t_1) C_{ab}(t_2)] |a\rangle A_b(t_2) \langle b| \]

\[ \langle c| A_a(t_1) |d\rangle \langle d| A_b(t_2) dt_1 dt_2, \]

where we have abbreviated the operators \( L_{ab} = |a\rangle \langle b| \). The dampening matrix elements can be most conveniently evaluated in the energy eigenbasis \( H_b(a) = E_a \). However, independent of the basis choice it remains to be shown that the dampening matrix is positive semidefinite to get a Lindblad form. In order to see this, we calculate with Eq. (12).
 Whereas the first term in the last line appears positive, one might fear that positivity can be spoiled by the existence of the additional second term. However, we can bound the second term via the Cauchy-Schwarz trace inequality \[ |Tr(AB)|^2 \leq Tr(A^2)Tr(B^2), \] with \[ A = K(\tau)\sqrt{\rho_B^0} \] and \[ B = \sqrt{\rho_B^0} \] (which exists as \( \rho_B^0 \) is positive semidefinite),

\[
|Tr_B[K(\tau)\rho_B^0]|^2 = |Tr_B[K(\tau)\sqrt{\rho_B^0}\sqrt{\rho_B^0}]|^2 \\
\leq Tr_B[\sqrt{\rho_B^0}K^*(\tau)K(\tau)\sqrt{\rho_B^0}]Tr_B[\sqrt{\rho_B^0}\sqrt{\rho_B^0}] \\
= Tr_B[K^*(\tau)K(\tau)\rho_B^0].
\]

Remembering that \( Tr_B[K^*(\tau)K(\tau)\rho_B^0] \geq 0 \) for any operator \( K(\tau) \), we therefore obtain for \( \tau \geq 0 \)

\[
\sum_{abcd} x_{ab}^* y_{abcd}^2 x_{cd} \geq 0,
\]

i.e., we have generated a Lindblad form master equation. This result goes beyond Ref. [20] in several aspects. Not only is the case \( C_{ab}(t_1) \neq 0 \) considered but, in addition, we do not constrain ourselves to bath density matrices in thermal equilibrium; i.e., one also has positivity for \( [\rho_B^0, H_B] \neq 0 \). It is an interesting avenue of further research to compare DCG with other methods within the context of nonequilibrium environments [24]. Beyond that, all of the above arguments go through if the system Hamiltonian is time dependent. In this case, the coupling operators in the interaction picture have to obey \( \dot{A}_{ab} = +i[H_S(t), A_{ab}(t)] \), such that the challenge is then to calculate the matrix elements \( \langle c|A_{ab}(t)|b \rangle \).

Under the assumptions \( C_{ab}(t_1) = 0 \) (no first-order correlation functions) and \( C_{ab}(t_1, t_2) = C_{ab}(t_1 - t_2) = Tr_B[B_{ab}(t_1 - t_2)B_{ab}^0] \) (reservoir in thermal equilibrium), we can insert the Fourier transforms of \( C_{ab}(t_1 - t_2) \) and \( C_{ab}(t_1 - t_2)sgn(t_1 - t_2) \). In addition, the system Hamiltonian is time independent, we may calculate the time integrals analytically. Then, we can make use of the identity for discrete \( a \) and \( b \) (see, e.g., Appendix F of Ref. [20]),

\[
\lim_{\tau \to \infty} \tau \operatorname{sinc} \left[ \frac{(\omega + a) \tau}{2} \right] \operatorname{sinc} \left[ \frac{(\omega + b) \tau}{2} \right] \sim 2 \pi \delta_{ab} \operatorname{sinc}(\omega + a),
\]

to calculate the large-time limit of the DCG2 approach. In complete analogy to Ref. [20], we obtain the Born-Markov secular approximation [4] in this limit.

Unfortunately, the unconditional preservation of positivity is not preserved by higher orders within DCG (although of course, in the weak-coupling limit the nice properties of DCG2 will dominate).

### III. EXAMPLES

In the following, we will test the DCG approach with simple examples for which at least in special cases an analytical solution exists. For finite-size reservoirs the correlation functions are nondecaying and these systems are inherently non-Markovian (exhibiting, for example, recurrences); cf. the examples in Secs. III A and III B. For quasicontinuous reservoirs we will compare the performance of the DCG approach with the Born-Markov approximation; see Secs. III C and III D.

#### A. DCG2 for two spins

We consider a highly non-Markovian system (S) by using a very small reservoir (B), namely, just a single further spin,

\[ H_S = \omega \sigma_3^0, \quad H_B = \Omega \sigma_B^0, \]

\[ H_{SB} = \lambda \tilde{\sigma}_S \cdot \tilde{\sigma}_B = \lambda [\sigma^x_3 \otimes \sigma^x_B + \sigma^y_3 \otimes \sigma^y_B + \sigma^z_3 \otimes \sigma^z_B], \]

(25)

i.e., the index of the coupling operators runs from one to three. Note that all coupling operators are Hermitian, such that we may omit overbars and daggers in Eq. (13). We as-
sume that the initial bath density matrix is diagonal in order to simplify all expressions,
\[ \rho_0^B = \begin{pmatrix} \rho_{00}^B & 0 \\ 0 & 1 - \rho_{00}^B \end{pmatrix}. \]

The exact solution can be obtained by exponentiating the Hamiltonian and tracing out the bath spin (not shown). As in Sec. II E we decompose the Liouville operator into unitary and nonunitary contributions, where we have first- and second-order contributions in the unitary action of decoherence, \( H_{\text{eff}}^1 = H_{\text{eff}}^1 + H_{\text{eff}}^2 \), and second-order contributions for the dissipative action \( \gamma_{ab,cd}^2 = \gamma_{ab,cd}^2 \).

Transforming the coupling operators into the interaction picture, we obtain \( B_1(t) = \cos(2\Omega t)\sigma_B^z - \sin(2\Omega t)\gamma_{B}^z \), \( A_1(t) = \cos(2\Omega t)\sigma_B^x - \sin(2\Omega t)\gamma_{B}^x \), \( B_3(t) = \cos(2\Omega t)\sigma_B^y + \sin(2\Omega t)\gamma_{B}^y \), \( A_3(t) = \sigma_B^0 \), and \( A_3(t) = \sigma_B^0 \). From this, we obtain the time-independent first-order correlation functions
\[ C_1(t) = 0, \quad C_2(t) = 0, \quad C_3(t) = 2\rho_{00}^B - 1, \]
which yield for the first-order Lamb shift Hamiltonian from Eq. (17) the following:
\[ H_{\text{eff}}^1 = \lambda(2\rho_{00}^B - 1)\sigma_S^z. \]

The nonvanishing second-order correlation functions equate to
\[ C_{11} = \cos[2(t_1 - t_2)\Omega] - i(1 - 2\rho_{00}^B)\sin[2(t_1 - t_2)\Omega], \]
\[ C_{12} = -i(1 - 2\rho_{00}^B)\cos[2(t_1 - t_2)\Omega] - \sin[2(t_1 - t_2)\Omega], \]
\[ C_{21} = i(1 - 2\rho_{00}^B)\cos[2(t_1 - t_2)\Omega] + \sin[2(t_1 - t_2)\Omega], \]
\[ C_{22} = \cos[2(t_1 - t_2)\Omega] - i(1 - 2\rho_{00}^B)\sin[2(t_1 - t_2)\Omega], \]
\[ C_{33} = 1, \]
where we have omitted the time dependencies for brevity. This can be inserted in the expression for the second-order Lamb-shift Hamiltonian in Eq. (19) to yield
\[ H_{\text{eff}}^2 = \frac{2\lambda^2}{\Omega - \omega}(1 - \sin[2(\Omega - \omega)]) \left[ \left( \rho_{00}^B - \frac{1}{2} \right) s_1 - \frac{1}{2} \sigma_S^z \right], \]

which commutes with the system Hamiltonian.

The second-order dissipative terms must be calculated from the dissipative parts of Eq. (19), where we obtain for the nonvanishing matrix elements of the dampering matrix \( \gamma_{00,00}^2 = 4\lambda^2(1 - \rho_{00}^B)\rho_{00}^B \), \( \gamma_{01,10}^2 = -4\lambda^2(1 - \rho_{00}^B)\rho_{00}^B \), \( \gamma_{10,01}^2 = -4\lambda^2(1 - \rho_{00}^B)\rho_{00}^B \), \( \gamma_{11,11}^2 = 4\lambda^2(1 - \rho_{00}^B)\rho_{00}^B \), \( \gamma_{10,10}^2 = 4\lambda^2\rho_{00}^B \), \( \gamma_{11,01}^2 = 4\lambda^2\rho_{00}^B \), \( \gamma_{11,00}^2 = 4\lambda^2\rho_{00}^B \), \( \gamma_{10,00}^2 = 4\lambda^2\rho_{00}^B \), which shows (e.g., by the Gershgorin circle theorem [25]) that \( \gamma_{ab,cd}^2 \) is positive semidefinite. The solution of the coarse-graining master equation \( \rho_B^2(t) = \mathcal{L} \rho_B^2(t) \) can be conveniently obtained by exploiting that diagonal and off-diagonal matrix elements decouple. From the diagonal equations
\[ \rho_0^B(t) = \gamma_{00,00}^2 \rho_0^B(t) - \frac{4\lambda^2}{(\Omega - \omega)^2} \sin^2[\pi(\Omega - \omega)] \rho_0^B(t) \]
\[ + \left[ 1 - \exp \left( -\frac{4\lambda^2}{(\Omega - \omega)^2} \sin^2[\pi(\Omega - \omega)] \right) \right] \rho_0^B(t), \]
which does admit for complete recurrences of the populations; see Fig. 1(a). For the off-diagonal equation
\[ \rho_{10}^B(t) = \gamma_{00,00}^2 \rho_{10}^B(t) - \frac{4\lambda^2}{(\Omega - \omega)^2} \sin^2[\pi(\Omega - \omega)] \rho_{10}^B(t), \]
we obtain the solution at \( \tau = t \),
\[ \rho_{10}^B(t) = \gamma_{10,00}^2 \rho_{10}^B(t) - \frac{4\lambda^2}{(\Omega - \omega)^2} \sin^2[\pi(\Omega - \omega)] \rho_{10}^B(t) \]
\[ + \left[ 1 - \exp \left( -\frac{4\lambda^2}{(\Omega - \omega)^2} \sin^2[\pi(\Omega - \omega)] \right) \right] \rho_{10}^B(t), \]
which corresponds to the exact solution for \( \rho_{10}^B(t) \), as shown in Fig. 1(b).
the density matrix as in Sec.III A.1 not the complete Hamiltonian and tracing out the second spin in the interaction picture.

We consider a single system spin coupled to a bath of bosonic modes

\[ \hat{H}_S = \omega_0 \sigma_z, \quad \hat{H}_B = \Omega \sigma_z, \quad \hat{H}_{SB} = \lambda \sigma_z \otimes \sigma_z. \]

where we also here the coupling operators are Hermitian. In this case, the bath correlation functions are all time independent, which enables a convenient calculation of the Liouvillian matrix elements. The example is of course a bit trivial, since the exact solution for the reduced density matrix does not depend on \( \Omega \). Note, however, that unlike the pure dephasing limit considered in [26] this case still holds some time dependence that can be found in the system operator in the interaction picture.

The exact solution can be calculated by exponentiating the complete Hamiltonian and tracing out the second spin in the solution for the density matrix as in Sec. III A. In a similar manner we determine the DCG1, DCG2, DCG3, and DCG4 solutions by directly determining the 4 \( \times 4 \) Liouvillian matrix as described in Sec. II D (not shown). The solution is then obtained by exponentiating the Liouvillian. The resulting solution for the diagonals is displayed in Fig. 2(a) and that for the off-diagonals in Fig. 2(b).
nal. By doing so it becomes obvious that the number of creation and annihilation operators in each term of the bath-bath correlation functions must be balanced for all modes to obtain a nonvanishing result. Therefore, we conclude (since only one operator is involved, we may omit the indices) that \( C(t_1) = 0 = C(t_1, t_2, t_3) \). In the interaction picture, the annihilation and creation operators transform according to \( b_k(t) = e^{-i\omega t} b_k, e^{i\omega t} b_k \) and the Hermitian conjugate, respectively.

The second-order correlation function then evaluates to

\[
C(t_1, t_2) = \frac{1}{2\pi} \int_0^\infty d\omega G(\omega) \{ n(\omega) e^{i\omega(t_1 - t_2)} \\
+ [1 + n(\omega)] e^{-i\omega(t_1 - t_2)} \} = \frac{1}{2\pi} \int_0^\infty d\omega \left[ G(\omega) e^{i\omega(t_1 - t_2)} \right. \\
\left. + \frac{1}{\beta \omega_c} \frac{1}{\beta - i\omega} \right] e^{i\omega(t_1 - t_2)} d\omega, \tag{36}
\]

where the bosonic occupation number is given by \( n(\omega) = \frac{1}{e^{\beta \omega_c} - 1} \). In the above equation, we have assumed a quasicontinuous spectral density \( G(\omega) = 2\pi \sum_i |b_i|^2 \delta(\omega - \omega_i) \) to convert the sum into an integral. When we parametrize the spectral density as

\[
G(\omega) = G_0 e^{\omega \omega_c}, \tag{37}
\]

where \( \omega_c \) denotes a cutoff frequency and the parameter \( S \) governs the slope at \( \omega = 0 \), we can obtain an analytic solution for the correlation function [27,28]

\[
C(t_1, t_2) = \frac{G_0 \Gamma(1+S)}{2\pi \beta^{1+S}} \left( \frac{1}{\beta \omega_c} - \frac{(t_1 - t_2)}{\beta} \right) \\
+ \left( \frac{1}{\beta \omega_c} + \frac{(t_1 - t_2)}{\beta} \right) \right], \tag{38}
\]

in terms of generalized Riemann zeta functions \( \zeta(x, y) \).

The next nonvanishing correlation function is fourth order, where we obtain

\[
C(t_1, t_2, t_3, t_4) = C(t_1, t_2) C(t_1, t_3) + C(t_1, t_3) C(t_1, t_4) + C(t_1, t_2) C(t_1, t_4), \tag{39}
\]

which can, for example, be obtained using Wick’s theorem (for a special case see also Eq. (61) of [16]).

2. Pure dephasing

The case of pure dephasing \( A = \alpha \) is exactly solvable [29,30] and it is known that DCG2 already yields the exact result [20]. The exact solution predicts time-independent diagonal matrix elements and a decay of the off-diagonal matrix element according to (in the interaction picture, cf. Eq. (82) of [30]) in the limit of a continuous bath spectrum

\[
\rho_{0i}(t) = e^{-\Gamma(t)} \rho_{0i}(0),
\]

\[
\Gamma(t) = \frac{8N^2}{2\pi} \int_0^\infty G(\omega) \frac{\sin^2(\omega t/2)}{\omega^2} \coth \left( \frac{\beta \omega}{2} \right) d\omega, \tag{40}
\]

where the additional factor of \( \frac{1}{2\pi} \) in comparison to [20] results from a different definition of the spectral density \( G(\omega) \).

By taking the time derivative of the exact solution density matrix, we obtain a closed master equation that is not of Lindblad form (not even with time-dependent coefficients) but nevertheless must—as it is exact—preserve positivity. Hence, one would regard this case as truly non-Markovian [22]. The corresponding steady state is also derived by the equation of motion method in Appendix A. For pure dephasing we have \( A(t) = \alpha \). We observe a decoupled evolution of diagonal and off-diagonal matrix elements of the density matrix.

For the second-order contribution we have \([\text{using } \Theta(t_1 - t_2) + \Theta(t_2 - t_3) = 1]\) from Eq. (13)

\[
\mathcal{T}_{\tilde{\rho}_S}^0 \rho^0_0 = \int_0^t C(t_1, t_2) [\alpha^0 \rho^0_0 - \rho^0_0 \alpha^0] dt_1 dt_2, \tag{41}
\]

from which we obtain

\[
\langle 0 \mid \mathcal{T}_{\tilde{\rho}_S}^0 \rho^0_0 \rangle = \langle 1 \mid \mathcal{T}_{\tilde{\rho}_S}^0 \rho^0_1 \rangle = 0, \tag{42}
\]

\[
\langle 0 \mid \mathcal{T}_{\tilde{\rho}_S}^0 \rho^0_1 \rangle = -2 \int_0^t C(t_1, t_2) dt_1 dt_2 \rho^0_1 \tag{43}
\]

which leads to the same exponential decay as with exact solution (40); i.e., as noted earlier [20], DCG2 yields the exact solution in this case. Note that this example demonstrates explicitly that the DCG2 solution cannot be generally written as the solution of a single Lindblad-type master equation with time-dependent coefficients, such that it should be classified as non-Markovian [22].

Using the relation

\[
0 = \Theta(t_4 - t_3) \Theta(t_3 - t_2) \Theta(t_2 - t_1) + \Theta(t_3 - t_4) \Theta(t_2 - t_1) + \Theta(t_1 - t_2) \Theta(t_2 - t_1)
\]

we obtain for the fourth-order contribution

\[
\langle 0 \mid \mathcal{T}_{\tilde{\rho}_S}^0 \rho^0_0 \rangle = 1 \mid \mathcal{T}_{\tilde{\rho}_S}^0 \rho^0_1 \rangle = 0, \tag{44}
\]

\[
\langle 0 \mid \mathcal{T}_{\tilde{\rho}_S}^0 \rho^0_1 \rangle = 2 \rho^0_1 \int_0^t dt_1 dt_2 dt_3 dt_4 C(t_1, t_2, t_3, t_4)
\]

\[
\times [\Theta(t_3 - t_2) \Theta(t_2 - t_1) + \Theta(t_3 - t_4) \Theta(t_4 - t_3)]
\]

\[
= 2 \int_0^t dt_1 dt_2 dt_3 dt_4 C(t_1, t_2, t_3, t_4) \rho^0_1, \tag{45}
\]

where we have exploited the symmetries of the fourth-order correlation functions under exchange of the arguments and the relation

\[
2 = \Theta(t_2 - t_3) \Theta(t_3 - t_4) + \Theta(t_3 - t_4) \Theta(t_4 - t_3)
\]

\[
+ \Theta(t_1 - t_2) \Theta(t_2 - t_3) + \Theta(t_2 - t_3) \Theta(t_3 - t_4)
\]

\[
+ \Theta(t_4 - t_1) \Theta(t_1 - t_2) + \Theta(t_1 - t_2) \Theta(t_2 - t_3)
\]
The nonvanishing off-diagonal contribution has to be compared with the counterterm arising from the second order,

\[
\frac{1}{2} \left( 2 \left[ \int_0^\tau C(t_1, t_2) dt_1 dt_2 \right] \rho_{S}^0 \right)^2
\]

\[
= 2 \left[ \int_0^\tau C(t_1, t_2) dt_1 dt_2 \right] \rho_{S}^0 \]

\[
\times \left[ \int_0^\tau C(t_1, t_2) dt_1 dt_2 \right] \rho_{S}^0.
\]

Since the diagonal elements of the density matrix are neither affected by \( T \bar{T} \) nor by \( T \), we conclude that we have for pure dephasing

\[
T_4 = \frac{1}{2} T_2 \bar{T} T_2,
\]

such that DCG4 will yield the same result as DCG2. Since DCG2 is already exact for this case, this cancellation is a strong indicator of the correctness of our fourth-order correlation function (39).

\[
T_2 = \int_0^\tau C(t_1, t_2) \begin{pmatrix}
- e^{-i \epsilon_{d}(t_1-t_2)} & 0 & 0 & + e^{i \epsilon_{d}(t_1-t_2)} \\
0 & - e^{-i \epsilon_{d}(t_1-t_2)} + e^{i \epsilon_{d}(t_1+t_2)} & 0 & 0 \\
0 & e^{i \epsilon_{d}(t_1+t_2)} - e^{-i \epsilon_{d}(t_1-t_2)} & 0 & 0 \\
+ e^{-i \epsilon_{d}(t_1-t_2)} & 0 & 0 & - e^{i \epsilon_{d}(t_1-t_2)} \\
\end{pmatrix} dt_1 dt_2,
\]

such that we observe a decoupled evolution of diagonal and off-diagonal matrix elements. Defining

\[
m_{11}(\tau) = (T_2)_11 = - \frac{\tau^2}{2\pi} \int_{-\infty}^{\infty} \frac{G(|\omega|)}{|e^{i \omega \tau} - 1|} \sin^2 \left( \omega - \epsilon_d \frac{\tau}{2} \right) d\omega,
\]

\[
m_{14}(\tau) = (T_2)_14 = \frac{\tau^2}{2\pi} \int_{-\infty}^{\infty} \frac{G(|\omega|)}{|e^{i \omega \tau} - 1|} \sin^2 \left( \omega + \epsilon_d \frac{\tau}{2} \right) d\omega,
\]

\[
m_{41}(\tau) = (T_2)_41 = - m_{11}(\tau),
\]

\[
m_{44}(\tau) = (T_2)_44 = - m_{14}(\tau),
\]

we obtain the second-order solution for the diagonals (using trace conservation),

\[
\rho^\tau_{00} = \rho^{00}_{0} \exp\left\{ \lambda^2 [m_{11}(\tau) - m_{14}(\tau)] \right\} + \frac{1 - \exp\left\{ \lambda^2 [m_{11}(\tau) - m_{14}(\tau)] \right\}}{m_{14}(\tau)}.
\]

In Eq. (49) it is already obvious that for finite times \( \tau \) all frequencies will contribute to the matrix elements \( m_{ij}(\tau) \), in contrast to the Markov approximation, where only \( G(\epsilon_d) \) is relevant. Using the fact that the bandfilter sinc functions transform into Dirac delta functions in Eq. (24), we can perform the limit \( \tau \rightarrow \infty \) to obtain the steady state

\[
\rho^{\infty}_{00} = \frac{1}{1 + e^{-\beta \epsilon_d}},
\]

which corresponds to the thermalized system density matrix that is consistent with our expectations; compare also Appendix A. The same stationary state can also be obtained by using the method of equation of motion and truncating correlations at second order between system and reservoir.

The Markovian limit is obtained by using
where we have again used identity (24) in Eq. (49). Evidently, the above equation leads to the same steady state as Born-Markov approximation (51).

By virtue of Eq. (42) and using \( C(t_1, t_2, t_3, t_4) = [C(t_4, t_3, t_2, t_1)]^* \), we obtain for the diagonal part

\[
\langle 0 | \mathcal{T} \hat{p}^0_S(0) | 0 \rangle = 2 \int_0^\infty \text{Re} \{ C(t_1, t_2, t_3, t_4) e^{-i\omega_d t_1 + i\omega_d t_3} \} \times \Theta(t_2 - t_3) \Theta(t_3 - t_4) dt_1 dt_2 dt_3 dt_4 \rho^{00}_S
\]

\[
- 2 \int_0^\infty \text{Re} \{ C(t_1, t_2, t_3, t_4) e^{i\omega_d t_1 - i\omega_d t_3 + i\omega_d t_4} \} \times \Theta(t_2 - t_3) \Theta(t_3 - t_4) dt_1 dt_2 dt_3 dt_4 \rho^{11}_S
\]

\[
= p_{11}(\tau) \rho^{00}_S + p_{14}(\tau) \rho^{11}_S. 
\]

This result has to be combined with the counterterm arising from the squared second-order contribution. Defining

\[
\tilde{p}_{11}(\tau) = \lambda^2 m_{11}(\tau) - \frac{\lambda^4}{2} \left[ m_{11}(\tau) m_{11}(\tau) + m_{14}(\tau) m_{41}(\tau) \right]
\]

\[
+ \lambda^4 p_{11}(\tau),
\]

\[
\tilde{p}_{14}(\tau) = \lambda^2 m_{14}(\tau) - \frac{\lambda^4}{2} \left[ m_{11}(\tau) m_{14}(\tau) + m_{14}(\tau) m_{44}(\tau) \right]
\]

\[
+ \lambda^4 p_{14}(\tau),
\]

we therefore obtain the fourth-order solution

\[
\rho^{00}_{11}(\tau) = \rho^{00}_0 \exp\left[ -\tilde{p}_{11}(\tau) - \tilde{p}_{14}(\tau) \right] + \frac{1 - \exp\left[ -\tilde{p}_{11}(\tau) - \tilde{p}_{14}(\tau) \right]}{1 - \frac{\tilde{p}_{11}(\tau)}{\tilde{p}_{14}(\tau)}}.
\]

A general exact solution is unfortunately not available for this case. However, it is interesting to note that when considering the Markov limit \( \beta=0 \), \( \omega_c \to \infty \), and \( S=0 \) (where the Markov approximation becomes exact), the correlation function (36) becomes a \( \delta \) function and we see that in this limit, the fourth-order term is canceled by the squared second-order counterterm. For comparison, we plot Born-Markov solution, DCG2, and DCG4 solutions in Fig. 3.

For the dissipative spin-boson model and Ohmic dissipation \( (S=1) \), one obtains an exponential decay of the expectation value \( \langle \sigma^z(\tau) \rangle \) in the long-time limit \( [31] \) (note the rotations \( \sigma^z \to \sigma^z \) and \( \sigma^z \to -\sigma^z \)). This corresponds to a decay of the off-diagonal matrix elements and is of course also reproduced by the DCG approach.

### D. Fano-Anderson model

We consider the Fano-Anderson model [32,33]: two leads that are connected by a single quantum dot, through which electrons may tunnel from one lead to the other. The Hamiltonian is given by

\[
H = H_S + H_B + H_{SB},
\]

\[
H_S = \varepsilon_d c^\dagger c + \sum_{ka} \omega_k c^\dagger_{ka} c_{ka} + \text{H.c.},
\]

\[
H_{SB} = \hbar \sum_{ka} \left[ t_k d^\dagger_{ka} c^\dagger_{ka} - t_k^* d^\dagger_{ka} c_{ka} \right] + \text{H.c.}
\]

with fermionic operators creating an electron with momentum \( k \) in the left or right lead \( a \in \{L,R\} \) or in the quantum dot for \( d^\dagger \). Due to the fermionic anticommutation relations, the model (56) can be solved exactly, for example, with Green’s functions [34,35]. We provide a simplified derivation based on the equation of motion method in Appendix B.
In contrast to our assumptions in Sec. II, the operators $d$ and $c_{k\sigma}$ do not act on separate Hilbert spaces, which is evident from their anticommutation relations $\{d, c_{k\sigma}\} = 0$. This becomes even more explicit via the decomposition

$$
d = |0\rangle\langle 1| \otimes 1,
$$

$$
c_{k\sigma} = |0\rangle\langle 0| - |1\rangle\langle 1| \otimes \bar{c}_{k\sigma}, \quad (57)
$$

where $|0\rangle$ and $|1\rangle$ denote the empty and filled dot states, respectively, and the fermionic operators $\bar{c}_{k\sigma}$ act only on the (distinct) Fock space of the remaining sites in the leads with $\{\bar{c}_{k\sigma}, \bar{c}_{k'\sigma'}\} = \delta_{k\sigma_k\sigma'}$. Naturally, the above decomposition obeys the original anticommutation relations such that $\{d, c_{k\sigma}\} = 0$ and we conclude for the operators in the interaction Hamiltonian $dc_{k\sigma} = -|0\rangle\langle 1| \otimes \bar{c}_{k\sigma}$, and $d^k c_{k\sigma} = +|1\rangle\langle 0| \otimes \bar{c}_{k\sigma}$, such that now the new operators commute by construction. Similar decompositions are also possible for systems containing more than one site. We identify

$$
A_1 = -|0\rangle\langle 1|, \quad A_2 = -|1\rangle\langle 0|,

B_1 = \sum_{k\sigma} t_{k\sigma} \bar{c}_{k\sigma}, \quad B_2 = \sum_{k\sigma} t_{k\sigma}^* \bar{c}_{k\sigma}^\dagger. \quad (58)
$$

We assume that there are no correlations between left and right leads. Here we will just consider the infinite-bias limit (although this is not crucial, it enables an analytic calculation of all integrals). Taking the chemical potentials to plus or minus infinity for the left and right leads, respectively ($\mu_L \to +\infty$ and $\mu_R \to -\infty$), we obtain for the fermionic occupation number

$$
\langle \bar{c}_{k\sigma}^\dagger \bar{c}_{k\sigma} \rangle = \frac{1}{e^{\beta \omega_{k\sigma}} + 1} \rightarrow 1,
$$

$$
\langle \bar{c}_{k\sigma}^\dagger \bar{c}_{k\sigma} \rangle = \frac{1}{e^{\beta \omega_{k\sigma}} + 1} \rightarrow 0. \quad (59)
$$

We observe that the coupling operators are non-Hermitian in this case. The correlation functions relevant for the evolution of the diagonal matrix elements can be calculated by using continuum tunneling rates via $\Gamma_\sigma(\omega) = 2\pi \sum_{k\sigma} t_{k\sigma}^2 \delta(\omega - \omega_{k\sigma})$,

$$
C_L(t_1 - t_2) = C_{12}(t_1, t_2) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \Gamma_L(\omega) e^{i\omega(t_1 - t_2)} d\omega,
$$

$$
C_R(t_2 - t_1) = C_{21}(t_1, t_2) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \Gamma_R(\omega) e^{-i\omega(t_1 - t_2)} d\omega,
$$

$$
C_{1212}(t_1, t_2, t_3, t_4) = C_{L}(t_1 - t_2) C_{L}(t_3 - t_4)
$$

$$
+ C_{L}(t_1 - t_3) C_{R}(t_2 - t_4),
$$

$$
C_{2121}(t_1, t_2, t_3, t_4) = C_{R}(t_1 - t_2) C_{R}(t_3 - t_4)
$$

$$
+ C_{R}(t_1 - t_3) C_{L}(t_2 - t_4). \quad (60)
$$

From Eq. (13), we can derive the second-order approximation

$$
\mathcal{T}_2^\sigma \rho_0^\sigma = \int_0^\tau dt_1 dt_2 [C_{21}(t_1, t_2) \Theta(t_2 - t_1) \rho_0^\sigma A_1^\dagger(t_1) A_1^\dagger(t_2) + C_{12}(t_1, t_2) \Theta(t_2 - t_1) \rho_0^\sigma A_2^\dagger(t_1) A_2^\dagger(t_2) + C_{21}(t_1, t_2) \rho_0^\sigma A_1^\dagger(t_1) A_2^\dagger(t_2) + C_{12}(t_1, t_2) \rho_0^\sigma A_2^\dagger(t_1) A_1^\dagger(t_2)]
$$

$$
+ C_{1212}(t_1, t_2, t_3, t_4) \rho_0^\sigma A_1^\dagger(t_1) A_2^\dagger(t_2) A_1^\dagger(t_3) A_2^\dagger(t_4) + C_{2121}(t_1, t_2, t_3, t_4) \rho_0^\sigma A_2^\dagger(t_1) A_1^\dagger(t_2) A_2^\dagger(t_3) A_1^\dagger(t_4),
$$

$$
= \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \Gamma_\sigma(\omega) \int_0^\tau dt_1 dt_2 e^{-i(\omega - \epsilon_{\sigma}) (t_1 - t_2)} \left[ -\rho_0^\sigma |1\rangle\langle 1| \Theta(t_2 - t_1) + |0\rangle\langle 1| \rho_0^\sigma |1\rangle\langle 1| - |1\rangle\langle 0| \rho_0^\sigma |1\rangle\langle 1| \right]
$$

$$
+ \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \Gamma_\sigma(\omega) \int_0^\tau dt_1 dt_2 e^{i(\omega - \epsilon_{\sigma}) (t_1 - t_2)} \left[ -\rho_0^\sigma |0\rangle\langle 0| \Theta(t_2 - t_1) + |1\rangle\langle 0| \rho_0^\sigma |0\rangle\langle 0| - |0\rangle\langle 0| \rho_0^\sigma |0\rangle\langle 0| \right], \quad (61)
$$

from which we can infer the matrix elements of the second-order Liouvillian via $\mathcal{L}_2^\sigma \rho_0^\sigma = \frac{d}{d\tau} \mathcal{T}_2^\sigma \rho_0^\sigma$. The Born-Markov secular approximation is obtained by the Liouvillian $\mathcal{L}_2^\sigma$ with the help of Eq. (24).

When we parametrize the tunneling rates by Lorentzians [36]

$$
\Gamma_\sigma(\omega) = \frac{\Gamma_0^{\sigma}}{\omega - \epsilon_\sigma)^2 + \delta_\sigma^2}, \quad \Gamma_L(\omega) = \frac{\Gamma_0^L}{(\omega - \epsilon_L)^2 + \delta_L^2}, \quad (62)
$$

we obtain an analytic expression for the bath correlation functions in terms of a single decaying exponential,

$$
C_R(t) = \frac{\Gamma_0^{\sigma}}{2} e^{-|\delta_\sigma + ig\tau|^2}, \quad C_L(t) = \frac{\Gamma_0^L}{2} e^{-|\delta_L + ig\tau|^2}, \quad (63)
$$

such that we can analytically calculate the matrix elements of $\mathcal{T}_2^\sigma$ that govern the evolution of the diagonals.
\[
\begin{pmatrix}
m_{11}(\tau) & m_{14}(\tau) \\
m_{41}(\tau) & m_{44}(\tau)
\end{pmatrix} = \int_0^\tau dt_1 dt_2 e^{-i\bar{\lambda}(t_1-t_2)} \\
\times \left(-C_L(t_1-t_2) + C_R(t_1-t_2) \right) \\
+ C_L(t_1-t_2) - C_R(t_1-t_2) \right);
\]
\[\tag{64}
\]

Then, we obtain from \(\langle 0 | \mathcal{T} \hat{T}^\tau \hat{\rho}_0^0 | 0 \rangle\) for the evolution of the diagonal matrix element
\[
\hat{\rho}_0^0 = \rho_0^0(t) = \lambda^2 \left[ \frac{m_{11}(\tau)}{\tau} \rho_{\bar{\lambda}}(t) + \frac{m_{14}(\tau)}{\tau} \rho_{\bar{\lambda}}(t) \right],
\]
\[\tag{65}
\]
where we can exploit trace conservation \(\rho_{\bar{\lambda}}(t) = 1 - \rho_{\bar{\lambda}}(t)\) (this feature is trivially fulfilled by the coarse-graining approach). Afterward, the above equation can explicitly be solved for
\[
\rho_{\bar{\lambda}}(t) = \rho_0^0(0) \exp \left\{ \lambda^2 \left[ m_{11}(\tau) - m_{14}(\tau) \right] \frac{t}{\tau} \right\} \\
+ \frac{1 - \exp \left\{ \lambda^2 \left[ m_{11}(\tau) - m_{14}(\tau) \right] \frac{t}{\tau} \right\}}{- \lambda^2}. \tag{66}
\]

For the fourth-order contribution we obtain [extensively using \(C_{1112}(t_1,t_2,t_3,t_4) = C_{2121}(t_1,t_2,t_3,t_4), C_{1112}(t_1,t_2,t_3,t_4) = 0,\) etc.]

\[
\mathcal{T}^\tau \hat{\rho}_0^0 = \int_0^\tau dt_1 dt_2 dt_3 dt_4 \times \left[ C_{2121}(t_1,t_2,t_3,t_4) \frac{\Theta(t_1-t_2) \Theta(t_1-t_3) \Theta(t_1-t_2) \Theta(t_1-t_4) \rho_0^0(0) \rho_{\bar{\lambda}}(t_1) \rho_{\bar{\lambda}}(t_2) \rho_{\bar{\lambda}}(t_3) \rho_{\bar{\lambda}}(t_4)}{\rho_{\bar{\lambda}}(t_1) \rho_{\bar{\lambda}}(t_2) \rho_{\bar{\lambda}}(t_3) \rho_{\bar{\lambda}}(t_4)} \right]
\]
\[\tag{67}
\]

The relevant part in above equation for the evolution of the diagonals evaluates by using Eq. (42) to

\[
(\mathcal{T}^\tau \hat{\rho}_0^0)_{11} = m_{11}(\tau) \rho_0^0(t) + m_{14}(\tau) \rho_{\bar{\lambda}}(t),
\]

\[
p_{11}(\tau) = \int_0^\tau dt_1 dt_2 dt_3 dt_4 e^{-i\bar{\lambda} \left[ t_1-t_2 + t_2-t_3 + t_3-t_4 \right]} \\
\times \left[ C_L(t_1-t_2) C_L(t_3-t_4) + C_L(t_1-t_4) C_L(t_3-t_2) \right] \\
\times \left[ \Theta(t_1-t_2) \Theta(t_1-t_3) + \Theta(t_1-t_3) \Theta(t_1-t_2) \right],
\]

\[
\rho_0^0(0) \exp \left\{ \lambda^2 \left[ m_{11}(\tau) - m_{14}(\tau) \right] \frac{t}{\tau} \right\} \\
+ \frac{1 - \exp \left\{ \lambda^2 \left[ m_{11}(\tau) - m_{14}(\tau) \right] \frac{t}{\tau} \right\}}{- \lambda^2},
\]

where again all integrals can be solved analytically, yielding even lengthier expressions than before.

Together with

\[
\langle 0 | \mathcal{T}^\tau \mathcal{T}_3^\tau \hat{\rho}_0^0 | 0 \rangle = \left[ m_{11}(\tau) m_{14}(\tau) + m_{14}(\tau) m_{14}(\tau) \right] \rho_0^0 \\
+ \left[ m_{11}(\tau) m_{14}(\tau) + m_{14}(\tau) m_{14}(\tau) \right] \rho_{\bar{\lambda}}^0,
\]

we obtain from

\[
p_{14}(\tau) = \int_0^\tau dt_1 dt_2 dt_3 dt_4 e^{-i\bar{\lambda} \left[ t_1-t_2 + t_2-t_3 + t_3-t_4 \right]} \\
\times \left[ C_L(t_1-t_2) C_L(t_4-t_3) + C_L(t_4-t_3) C_L(t_2-t_3) \right] \\
\times \left[ \Theta(t_1-t_2) \Theta(t_1-t_3) + \Theta(t_1-t_3) \Theta(t_1-t_2) \right],
\]

\[
\rho_0^0(0) \exp \left\{ \lambda^2 \left[ m_{11}(\tau) - m_{14}(\tau) \right] \frac{t}{\tau} \right\} \\
+ \frac{1 - \exp \left\{ \lambda^2 \left[ m_{11}(\tau) - m_{14}(\tau) \right] \frac{t}{\tau} \right\}}{- \lambda^2},
\]

\[
0.032110-12
\]
the following differential equation for the diagonals:

\[
\dot{\rho}_{00}^\tau = \frac{1}{\tau} \left[ \lambda^2 m_{11}(\tau) - \frac{\lambda^4}{2} [m_{11}(\tau)m_{11}(\tau) + m_{14}(\tau)m_{41}(\tau)] 
+ \lambda^4 p_{11}(\tau) \right] \rho_{00}^\tau(t) 
+ \lambda^4 p_{11}(\tau) \rho_{00}^\tau(t) 
+ m_{14}(\tau)m_{41}(\tau) \right] \dot{\rho}_{11}^\tau(t).
\]

The above equation can be explicitly solved for \( \rho_{00}^\tau(\tau) \) by imposing trace conservation as in Sec. III C. It can be shown analytically that in the flatband limit \( \delta_R \to \infty, \delta_L \to \infty \) (where for infinite bias the Markovian approximation becomes exact), the fourth-order correction in the above equation is canceled by the counterterm from the second order for all graining times \( \tau \). Moreover, one can also show analytically that the apparent divergence for large graining times \( p_{11} \propto \tau^2 \) and \( p_{14} \propto \tau^2 \) is precisely canceled by the fourth-order counter terms arising from the second order.

In contrast, one obtains under the Born-Markov (the secular approximation has no effect for this particular example) approximation the solution

\[
\lambda^2 \mathcal{L}_{2}^\tau + \lambda^4 \mathcal{L}_{4}^\tau = \frac{\lambda^2}{\tau} \mathcal{T}_{2}^\tau + \frac{\lambda^4}{\tau} \left( \mathcal{T}_{4}^\tau - \frac{\tau}{2} \mathcal{L}_{2}^\tau \mathcal{L}_{2}^\tau \right)
= \frac{1}{\tau} \left( \lambda^2 \mathcal{T}_{2}^\tau + \lambda^4 \mathcal{T}_{4}^\tau - \frac{\lambda^4}{2} \mathcal{T}_{2}^\tau \mathcal{T}_{2}^\tau \right)
\]

FIG. 5. (Color online) Comparison of exact (solid black line), DCG2 (dashed red line), DCG4 (dot-dashed blue line), and Born-Markov (dotted green line) solutions for the Fano-Anderson model. In figure (a), we have considered symmetric maximum tunneling rates \( \Gamma^0_R = \Gamma^0_L = 1 \) for the weak-coupling limit \( \lambda^2 = 0.1 \) (bold lines) and the strong-coupling limit \( \lambda^2 = 1.0 \) (thin lines). It is visible that the steady state of BMS and DCG2 solutions does not depend on the coupling strength and in the strong-coupling limit, the steady state of DCG4 might actually be worse than that of DCG2. In figure (b), we have used \( \lambda^2 = 0.1 \) and \( \Gamma^0_R = 1.0 \) and varied the left maximum tunneling rate as \( \Gamma^0_L = 2.0 \) (bold line), \( \Gamma^0_L = 5.0 \) (medium-thickness line), and \( \Gamma^0_L = 10.0 \) (thin line). For small times, DCG4 always yields a better result than DCG2 and both DCG solutions are better than the BMS approximation. The latter performs particularly bad for small times as expected. The other parameters have (in both figures) been chosen as \( \delta_R = 1, \delta_L = 2, \epsilon_R = \epsilon_L = 0, \) and \( \epsilon_R = 1. \)

\[
\rho_{00}(\tau) = \frac{\Gamma_R(\epsilon_d)}{\Gamma_L(\epsilon_d) + \Gamma_R(\epsilon_d)} [1 - e^{-\lambda^2 [\Gamma_L(\epsilon_d) + \Gamma_R(\epsilon_d)] \tau}] 
+ \rho_{00}(0) e^{-\lambda^2 [\Gamma_L(\epsilon_d) + \Gamma_R(\epsilon_d)] \tau},
\]

which has been derived using Eq. (52) and identity (24). From Lorentzian tunneling rates (62) we see that the Markovian solution is completely independent of the width of the tunneling rates \( \delta_R \) and \( \delta_L \); see also Fig. 4. Especially for small widths \( \delta_R \) and \( \delta_L \) correlation functions (63) decay very slowly and we also observe a large difference between Born-
Markov and exact solutions, whereas the DCG solutions perform comparably well. With the exact solution from Appendix B we plot the Born–Markov solution, and DCG2 as well as DCG4 solutions in Fig. 5 for varying coupling strengths as well as for different model symmetries.

IV. SUMMARY

The dynamical coarse-graining approach has been extended to higher orders. By performing the derivation in the interaction picture, we have directly demonstrated that the DCG solution must approximate the exact solution by construction for small times. This has been confirmed by several examples. Interestingly, the DCG method even reproduced the complete recurrences of the diagonal density matrix elements in case of small reservoirs. For continuous reservoirs, the short-time dynamics of DCG4 has always been superior to the short-time performance of DCG2. This however needs not be the case for the large-time limit. However, the performance of DCG2 is always better (short time) or equal (large time) to the performance of the Born-Markov secular approximation.

We have shown that DCG2 unconditionally preserves positivity. Going beyond [20], this also includes reservoirs that are not in equilibrium. In addition, the presentation in the interaction picture leads to a much simpler form of the interaction picture, which is unfortunately not closed. However, we can achieve closure by assuming factorization of the expectation values and stationarity of the reservoir, with the approximate steady state of the system.

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APPENDIX A: APPROXIMATE STEADY STATE OF THE SPIN-BOSON MODEL

Starting from Hamiltonian (34) we obtain with the abbreviations (we use bold symbols for operators in the Heisenberg picture)

\[
\dot{s}_k = h_s \hat{b}_k + h_s^* \hat{b}_k, \quad \dot{a}_k = \hat{a}_k (h_s \hat{b}_k - h_s^* \hat{b}_k^*) \quad (A1)
\]

a set of equations for the time evolutions of \(\langle \sigma^x \rangle, \langle \sigma^y \rangle, \langle \sigma^z \rangle, \langle \sigma^x \sigma_y \rangle, \langle \sigma^y \sigma_z \rangle, \langle \sigma^z \sigma_x \rangle, \langle \sigma^x \sigma_z \rangle, \langle \sigma^y \sigma_x \rangle\), and \(\langle \sigma^2 \rangle\) in the Heisenberg picture, which is unfortunately not closed. However, we can achieve closure by assuming factorization of the expectation values and stationarity of the reservoir,

\[
\left\langle \sigma^{x/y/z} \sum_k \left( s_k s_k + s_k s_k^* \right) \right\rangle = 2 \left\langle \sigma^{x/y/z} \sigma^2 \right\rangle,
\]

\[
\left\langle s_k^2 \right\rangle = |h_k|^2 [1 + 2n(\omega_k)],
\]

\[
\left\langle \sigma^{x/y/z} \sum_k \left( s_k a_k + a_k s_k^* \right) \right\rangle = 0,
\]

\[
\left\langle \sum_k \left( s_k a_k - a_k s_k^* \right) \right\rangle = -2i |h_k|^2,
\]

APPENDIX B: EXACT SOLUTION OF THE FANO-ANDERSON MODEL FOR LORENTZIAN TUNNELING RATES

From Hamiltonian (56) we can calculate the time evolution of the fermionic operators in the Heisenberg picture (we use bold operator symbols to denote the Heisenberg picture and exploit the anticommutation relations throughout,)

\[
d = -i e_d \hat{d} + i \lambda \sum_k \left[ t_{k\ell}^* \hat{c}_{k\ell} + t_{k\ell} \hat{c}_{k\ell}^* \right],
\]

\[
\hat{c}_{k\ell} = -i \omega_{k\ell} \hat{c}_{k\ell} + i \lambda t_{k\ell} \hat{d},
\]

\[
\hat{c}_{k\ell}^* = -i \omega_{k\ell} \hat{c}_{k\ell}^* + i \lambda t_{k\ell} \hat{d},
\]

which already form a closed set of equations. These equations can be Laplace transformed [where \(d(t) \rightarrow \tilde{d}(z)\) and \(\dot{d}(t) \rightarrow -d + \tilde{d}(z)\) and similarly for the other operators],

In Laplace space, we can eliminate \(\tilde{c}_{k\ell}(z)\) and \(\tilde{c}_{k\ell}^*(z)\) to solve the resulting equations for
In the large-time limit, this considerably simplifies

\[ \lim_{t \to \infty} \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} d\omega \frac{\hat{d}(\omega)}{z + i\omega} \]

where in the last line we have already assumed Lorentzian tunneling rates (62). The inverse Laplace transform (Bromwich integral [38]) can be performed by the theorem of residues \( d(t) = \sum \text{Res} \hat{d}(\omega) e^{\gamma t} \), where \( z_j \) denote the poles of \( \hat{d}(\omega) \). Denoting the roots of

\[ (z + i\epsilon)(z + \delta_L + i\epsilon_L)(z + \delta_R + i\epsilon_R) + \frac{\lambda^2}{2} \left( \Gamma_L^0 \delta_L(z + \delta_R + i\epsilon_R) + \Gamma_R^0 \delta_R(z + \delta_L + i\epsilon_L) \right) = (z - z_1)(z - z_2)(z - z_3) \]

by \( z_1, z_2, \) and \( z_3 \), respectively, we can easily calculate the residues. (In case of degenerate roots, one may either use residue formulas for higher-order poles or simply perform analytic continuation of the solution for first-order poles.) For \( z_1 \neq z_2, z_1 \neq z_3, \) and \( z_2 \neq z_3 \) we obtain the solution

\[ d(t) = \left[ \frac{(z_1 + \delta_L + i\epsilon_L)(z_1 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_1 - z_2)(z_1 - z_3)} + \frac{(z_2 + \delta_L + i\epsilon_L)(z_2 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_2 - z_1)(z_2 - z_3)} + \frac{(z_3 + \delta_L + i\epsilon_L)(z_3 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_3 - z_1)(z_3 - z_2)} \right] d + i\lambda \sum_k \left[ \frac{(z_1 + \delta_L + i\epsilon_L)(z_1 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_1 - z_2)(z_1 - z_3)} + \frac{(z_2 + \delta_L + i\epsilon_L)(z_2 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_2 - z_1)(z_2 - z_3)} + \frac{(z_3 + \delta_L + i\epsilon_L)(z_3 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_3 - z_1)(z_3 - z_2)} \right] \]

With taking the initial conditions as \( \langle c_{1L}^+ c_{1L} \rangle = \delta_{L1} f_L(\omega_L) \) and \( \langle c_{1R}^+ c_{1R} \rangle = \delta_{L1} f_R(\omega_R) \) and \( \langle c_{1L}^+ c_{1L} \rangle = 0 \), we obtain for \( n(t) \)

\[ n(t) = \left| \frac{(z_1 + \delta_L + i\epsilon_L)(z_1 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_1 - z_2)(z_1 - z_3)} + \frac{(z_2 + \delta_L + i\epsilon_L)(z_2 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_2 - z_1)(z_2 - z_3)} + \frac{(z_3 + \delta_L + i\epsilon_L)(z_3 + \delta_R + i\epsilon_R) e^{\gamma t}}{(z_3 - z_1)(z_3 - z_2)} \right|^2 \]

In the large-time limit, this considerably simplifies (using \( \text{Re} \, z_j < 0 \)). Conventionally, \( \lambda^2 \) is absorbed in \( \Gamma_L(\omega) \) and \( \Gamma_R(\omega) \) and by setting \( \lambda \to 1 \) we explicitly recover the well-known steady-state results in the literature. [Compare, e.g., Eq. (12.27) of Ref. [34] with using Lorentzian tunneling rates of form (62) and Eqs. (12.30) and (12.31) of Ref. [34] with \( n_0 = \frac{\lambda^2}{2\pi i} \int_{c-i\infty}^{c+i\infty} G^c(\omega) d\omega \).]