Exact master equations for the non-Markovian decay of a qubit

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Exact master equations describing the decay of a two-state system into a structured reservoir are constructed. By employing the exact solution for the model, analytical expressions are determined for the memory kernel of the Nakajima-Zwanzig master equation and for the generator of the corresponding time-convolutionless master equation. This approach allows an explicit comparison of the convergence behavior of the corresponding perturbation expansions. Moreover, the structure of widely used phenomenological master equations with a memory kernel may be incompatible with a nonperturbative treatment of the underlying microscopic model. Several physical implications of the results on the microscopic analysis and the phenomenological modeling of non-Markovian quantum dynamics of open systems are discussed.

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I. INTRODUCTION

The field of open quantum systems [1–3] is of great interest because of its relevance in applications of quantum mechanics, as well as for a deeper understanding of the theory itself. Indeed the study of the interaction between a quantum system and its environment is an endeavor common to many fields, such as quantum measurement theory, quantum communication, quantum optics, condensed matter theory, and quantum chemistry to name a few. The field has been well assessed as far as Markovian dynamics is concerned. For this case the Gorini-Kossakowski-Sudarshan-Lindblad expression for the generator of a quantum dynamical semigroup [4,5] provides a benchmark result for both microscopic and phenomenological approaches. However, this situation is not satisfactory when one has to go beyond the Born-Markov approximation and it involves systems in which a separation of time scales between system and environment can no longer be assumed in a realistic description. Memory effects then become important, and a non-Markovian description is mandatory. For this case a general consistent theoretical framework has not yet been found, and partial results have been obtained as a result of intense efforts (see, e.g., Refs. [6–21]). An important step in the development of a general theory consists in the construction of a suitable measure that quantifies the degree of non-Markovianity for a given dynamical evolution [22,23].

In this article we obtain the exact Nakajima-Zwanzig kernel for a two-level system coupled to a Bosonic reservoir discussed in Ref. [2] and compare it to the exact time-convolutionless master equation, as well as to the Markovian approximation of the dynamics. This approach shows how involved the transition from the approximate Markovian level of description to the exact non-Markovian regime can be. Indeed, the non-Markovian memory kernel is found to have an operator structure which differs from the one that appears in the Born-Markov approximation. Often one tries to obtain dynamical equations of motion for non-Markovian systems by slight modifications with respect to the Markovian case, for example, by considering a master equation which involves a superoperator given by a convolution in time of the corresponding Markovian superoperator [24–29]. Our results show that such an approach, although justified as a phenomenological modeling, can be incompatible with a nonperturbative treatment of the underlying microscopic system-environment model. Moreover, different perturbation expansions such as time-convolutionless and Nakajima-Zwanzig projection operator technique turn out to have different ranges of validity. Indeed, the time-convolutionless expansion breaks down at finite time in the strong coupling limit. On the other hand, the Nakajima-Zwanzig approach does not preserve positivity if restricted to second order. Furthermore, the convergence to the exact solution is not uniform with respect to the expansion parameter: different matrix elements of the statistical operator such as coherences and populations are obtained with quite different accuracy at the same perturbative order.

The article is organized as follows. In Sec. II, the model and its exact solution are introduced and later exploited to obtain the exact equations of motion for the reduced statistical operator of the system. In Sec. III, we recall the structure of the time-convolutionless master equation, pointing out two different perturbation expansions for the generator. In Sec. IV, the Nakajima-Zwanzig integral kernel is derived, providing an alternative expansion with respect to the standard method. The two results are compared in Sec. V, which also builds on an exact analytic expression for all the quantities involved obtained considering a Lorentzian spectral density. Finally, conclusions are drawn in Sec. VI.

II. THE MODEL AND ITS EXACT SOLUTION

The total Hamiltonian of the model is given by

$$H = H_S + H_E + H_I = H_0 + H_I, \tag{1}$$

where

$$H_S = \omega_0 \sigma_+ \sigma_- \tag{2}$$

describes a two-state system (qubit) with ground state $|0\rangle$, excited state $|1\rangle$, and transition frequency ω_0. The operators $\sigma_+ = |1\rangle\langle 0|$ and $\sigma_- = |0\rangle\langle 1|$ are the raising and lowering operators of the qubit, respectively. The environmental

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Hamiltonian is taken to be

$$H_E = \sum_k \omega_k b_k^\dagger b_k, \quad (3)$$

describing a collection of harmonic oscillators with creation and annihilation operators $b_k^\dagger$ and b_k which satisfy Bosonic commutation relations $[b_k, b_{k'}^\dagger] = \delta_{kk'}$. The interaction Hamiltonian takes the form

$$H_I = \sum_k (g_k \sigma_+ \otimes b_k + g_k^* \sigma_- \otimes b_k^\dagger). \quad (4)$$

The model thus describes, for example, the coupling of the qubit to a reservoir of electromagnetic field modes labeled by the index k, with corresponding frequencies ω_k and coupling constants g_k, and has already been discussed in Ref. [2].

In the following we work in the interaction picture with respect to $H_0 = H_S + H_E$. In this picture the Schrödinger equation reads

$$\frac{d}{dt}|\Psi(t)\rangle = -iH_I(t)|\Psi(t)\rangle, \quad (5)$$

where the interaction Hamiltonian is given by

$$H_I(t) = \sigma_+(t) \otimes B(t) + \sigma_-(t) \otimes B^\dagger(t) \quad (6)$$

with

$$\sigma_\pm(t) = \sigma_\pm e^{\pm i\omega_0 t} \quad (7)$$

and

$$B(t) = \sum_k g_k b_k e^{-i\omega_k t}. \quad (8)$$

It is easy to verify that the operator

$$N = \sigma_+\sigma_- + \sum_k b_k^\dagger b_k \quad (9)$$

for the number of excitations in the system commutes both with the total Hamiltonian H and with the interaction Hamiltonian $H_I(t)$. This is a consequence of the fact that the rotating-wave approximation has been used in the interaction Hamiltonian (4). It follows that any initial state of the form

$$|\Psi(0)\rangle = c_0|0\rangle \otimes |0\rangle_E + c_1(0)|1\rangle \otimes |0\rangle_E + \sum_k c_k(0)|0\rangle \otimes |k\rangle_E \quad (10)$$

evolves after time t into the state

$$|\Psi(t)\rangle = c_0|0\rangle \otimes |0\rangle_E + c_1(t)|1\rangle \otimes |0\rangle_E + \sum_k c_k(t)|0\rangle \otimes |k\rangle_E. \quad (11)$$

The state $|0\rangle_E$ denotes the vacuum state of the reservoir, and $|k\rangle_E = b_k^\dagger|0\rangle_E$ is the state with one particle in mode k. Note that the amplitudes $c_1(t)$ and $c_k(t)$ depend on time, while the amplitude c_0 is constant in time because $H_I(t)|0\rangle \otimes |0\rangle_E = 0$. Substituting Eq. (11) into the Schrödinger equation (5), one finds

$$\frac{d}{dt}c_1(t) = -i\sum_k g_k e^{i(\omega_0-\omega_k)t} c_k(t), \quad (12)$$

$$\frac{d}{dt}c_k(t) = -ig_k^* e^{-i(\omega_0-\omega_k)t} c_1(t). \quad (13)$$

We assume in the following that $c_k(0) = 0$. This means that the environment is in the vacuum state initially, and that the total initial state is given by the product state

$$|\Psi(0)\rangle = [c_0|0\rangle + c_1(0)|1\rangle] \otimes |0\rangle_E \equiv |\psi(0)\rangle \otimes |0\rangle_E. \quad (14)$$

Expressing $c_k(t)$ in terms of $c_1(t)$ by means of Eq. (13), and substituting the result into Eq. (12), one obtains an integrodifferential equation for the amplitude $c_1(t)$:

$$\frac{d}{dt}c_1(t) = -\int_0^t dt_1 f(t-t_1)c_1(t_1). \quad (15)$$

Given the solution of this equation, which can be found through a Laplace transformation, the amplitudes $c_k(t)$ are determined by Eq. (13). The kernel $f(t-t_1)$ of Eq. (15) is given by a certain two-point correlation function of the reservoir,

$$f(t-t_1) = \langle 0|B(t)B^\dagger(t_1)|0\rangle e^{i\omega_0(t-t_1)} = \sum_k |g_k|^2 e^{i(\omega_0-\omega_k)(t-t_1)}, \quad (16)$$

on which no restrictive hypothesis is made, so that the present results are valid for an environment with a generic spectral density.

With the help of the procedure described above, and already used by Weisskopf and Wigner in their classical paper [30], one finds the solution of the Schrödinger equation of the total system with initial states of the form of Eq. (14) lying in the sector of the Hilbert space corresponding to zero or one excitation. By means of this solution, we can construct the exact dynamical map describing the time evolution of the reduced density matrix of the qubit. This is given by

$$\rho(t) = \mathrm{tr}_E\{|\Psi(t)\rangle\langle\Psi(t)|\} = \begin{pmatrix} \rho_{11}(t) & \rho_{10}(t) \\ \rho_{01}(t) & \rho_{00}(t) \end{pmatrix}, \quad (17)$$

where $\rho_{ij}(t) = \langle i|\rho(t)|j\rangle$ for $i,j = 0,1$. Using Eq. (11), we find

$$\rho_{11}(t) = 1 - \rho_{00}(t) = |c_1(t)|^2, \quad (18)$$

$$\rho_{10}(t) = \rho_{01}^*(t) = c_0^* c_1(t). \quad (19)$$

It is convenient to define the function $G(t)$ as the solution of the equation

$$\frac{d}{dt}G(t) = -\int_0^t dt_1 f(t-t_1)G(t_1) \quad (20)$$

corresponding to the initial condition $G(0) = 1$. We then have $c_1(t) = G(t)c_1(0)$ and, hence, the dynamics of the elements of the reduced density matrix can be represented as follows:

$$\rho_{11}(t) = |G(t)|^2\rho_{11}(0), \quad (21)$$

$$\rho_{00}(t) = \rho_{00}(0) + (1-|G(t)|^2)\rho_{11}(0), \quad (22)$$

$$\rho_{10}(t) = G(t)\rho_{10}(0), \quad (23)$$

$$\rho_{01}(t) = G^*(t)\rho_{01}(0). \quad (24)$$

These equations have been derived for the pure product initial state (14); that is, they describe the time evolution corresponding to the pure reduced system's initial state $\rho(0) = |\psi(0)\rangle\langle\psi(0)|$. However, since any mixed initial state can be represented as a convex-linear combination of pure initial states, and since the function $G(t)$ introduced above does not depend on the initial condition, Eqs. (21)–(24) hold true for any pure or mixed initial state. They thus represent the exact dynamical map $\Phi(t)$ which transforms the initial states into the states at time t:

$$\Phi(t): \ \rho(0) \mapsto \rho(t) = \Phi(t)\rho(0), \quad t \geqslant 0. \tag{25}$$

Since we have constructed this map from the exact solution of the model, it is clear from the general theory of open quantum systems that $\Phi(t)$ is completely positive and trace preserving.

III. THE TIME-CONVOLUTIONLESS MASTER EQUATION

A. Exact master equation in time-convolutionless form

The exact solution determined in Sec. II enables the construction of the exact generator $\mathcal{K}_{\rm TCL}$ of the time-convolutionless master equation

$$\frac{d}{dt}\rho(t) = \mathcal{K}_{\rm TCL}(t)\rho(t) \tag{26}$$

governing the dynamics of the reduced density matrix. The time-convolutionless generator is defined in terms of the dynamical map $\Phi(t)$ by means of

$$\mathcal{K}_{\rm TCL}(t) = \dot{\Phi}(t)\Phi^{-1}(t), \tag{27}$$

provided the inverse map $\Phi^{-1}(t)$ exists. Then, using Eqs. (21)–(24), one can show that the generator takes the following form [2]:

$$\mathcal{K}_{\rm TCL}(t)\rho = -\frac{i}{2}S(t)[\sigma_+\sigma_-,\rho] + \gamma(t)\left[\sigma_-\rho\sigma_+ - \frac{1}{2}\{\sigma_+\sigma_-,\rho\}\right], \tag{28}$$

where we have introduced the definitions

$$\gamma(t) = -2\,\mathrm{Re}\left[\frac{\dot{G}(t)}{G(t)}\right], \quad S(t) = -2\,\mathrm{Im}\left[\frac{\dot{G}(t)}{G(t)}\right]. \tag{29}$$

By construction, Eq. (26) with generator (28) represents an exact time-local master equation. Note that the generator is well defined as long as $G(t) \neq 0$. The quantity $S(t)$ plays the role of a time-dependent frequency shift, and $\gamma(t)$ can be interpreted as a time-dependent decay rate. We observe that the structure of $\mathcal{K}_{\rm TCL}$ is similar to that of a Lindblad generator. However, due to the time dependence of the coefficients $S(t)$ and $\gamma(t)$, Eq. (26) does generally not yield a quantum dynamical semigroup. Moreover, the time-dependent rate $\gamma(t)$ may become negative, signifying strong non-Markovian behavior of the reduced system dynamics.

B. Perturbation expansions of the generator

In most cases of interest, the time-convolutionless generator can only be determined through a perturbation expansion. Here we investigate two methods of expanding the exact master equation (26) with respect to the strength of the interaction Hamiltonian H_I. To this end, we introduce a small overall expansion parameter α, replacing the coupling constants g_k in the interaction Hamiltonian (4) by αg_k. The two-point correlation function $f(t)$, which is proportional to α^2, is then to be regarded as a quantity of second order.

The first method consists of using Eq. (20) to obtain a perturbative expression for $G(t)$, from which one directly finds an expansion for the coefficients $\gamma(t)$ and $S(t)$ appearing in the master equation. The expansion of $G(t)$ is obviously of the form

$$G(t) = \sum_{n=0}^{\infty} \alpha^{2n} G^{(2n)}(t), \tag{30}$$

where $G^{(0)}(t) \equiv 1$ because of the required initial condition $G(0) = 1$, and Eq. (20) leads to the following recursion relation:

$$G^{(2n)}(t) = -\int_0^t dt_1 \int_0^{t_1} dt_2 f(t_1 - t_2) G^{(2n-2)}(t_2). \tag{31}$$

To illustrate the procedure, we determine the frequency shift and the decay rate to fourth order in α:

$$-\frac{1}{2}[\gamma(t) + iS(t)] = \frac{\dot{G}(t)}{G(t)} = \alpha^2 \dot{G}^{(2)}(t) + \alpha^4[\dot{G}^{(4)}(t) - \dot{G}^{(2)}(t)G^{(2)}(t)] + O(\alpha^6). \tag{32}$$

With the help of these expressions, one obtains the second- and fourth-order contributions for the coefficients of the master equation:

$$\gamma^{(2)}(t) + iS^{(2)}(t) = 2\int_0^t dt_1 f(t - t_1),$$
$$\gamma^{(4)}(t) + iS^{(4)}(t) = 2\int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 [f(t - t_2)f(t_1 - t_3) + f(t - t_3)f(t_1 - t_2)]. \tag{33}$$

Another possibility for the construction of the perturbation expansion is to use the general method of expanding the time-convolutionless generator in terms of the ordered cumulants. This procedure allows a closed expression to be written for the coefficients of the master equation which takes the following form (for details, see Ref. [2] and references therein):

$$\gamma^{(2n)}(t) + iS^{(2n)}(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{2n-2}} dt_{2n-1} 2(-1)^{n+1} \times \langle f(t - t_1) f(t_2 - t_3) \cdots f(t_{2n-2} - t_{2n-1}) \rangle_{\rm oc}. \tag{34}$$

IV. THE NAKAJIMA-ZWANZIG MASTER EQUATION

A. The exact memory kernel

The Nakajima-Zwanzig master equation is given by

$$\frac{d}{dt}\rho(t) = \int_0^t dt_1 \mathcal{K}_{\rm NZ}(t - t_1)\rho(t_1), \tag{35}$$

where the superoperator $\mathcal{K}_{\rm NZ}(\tau)$ represents the memory kernel. We construct the form of this kernel from the exact solution of our model obtained in Sec. II. To this end, we employ the following ansatz,

$$\mathcal{K}_{\rm NZ}(\tau)\rho = -i\varepsilon(\tau)[\sigma_+\sigma_-,\rho] + k_1(\tau)\left[\sigma_-\rho\sigma_+ - \tfrac{1}{2}\{\sigma_+\sigma_-,\rho\}\right] + k_2(\tau)\left[\sigma_+\sigma_-\rho\sigma_+\sigma_- - \tfrac{1}{2}\{\sigma_+\sigma_-,\rho\}\right], \tag{36}$$

where the functions $\varepsilon(\tau)$, $k_1(\tau)$, and $k_2(\tau)$ are real, such that the master equation preserves Hermiticity and trace. The equations of motion for the population $\rho_{11}(t)$ and the coherence $\rho_{10}(t)$ obtained from this master equation read

$$\frac{d}{dt}\rho_{11}(t) = -\int_0^t dt_1 k_1(t-t_1)\rho_{11}(t_1), \tag{37}$$

and

$$\frac{d}{dt}\rho_{10}(t) = -\int_0^t dt_1 \left\{\frac{1}{2}[k_1(t-t_1)+k_2(t-t_1)] + i\varepsilon(t-t_1)\right\}\rho_{10}(t_1). \tag{38}$$

On the other hand, Eq. (23) together with Eq. (20) yields

$$\frac{d}{dt}\rho_{10}(t) = -\int_0^t dt_1 f(t-t_1)\rho_{10}(t_1), \tag{39}$$

where we have set the expansion parameter α equal to 1, and we only have to remember that $f(t)$ is a quantity of second order. Comparing Eq. (39) with Eq. (38), we see that the expression within the curly braces of Eq. (38) must be equal to $f(t-t_1)$; that is, we get the conditions

$$\varepsilon(\tau) = f_2(\tau), \tag{40}$$

$$k_1(\tau) + k_2(\tau) = 2f_1(\tau), \tag{41}$$

where $f_1(\tau)$ and $f_2(\tau)$ denote the real and the imaginary part of the correlation function

$$f(\tau) = f_1(\tau) + i f_2(\tau). \tag{42}$$

In order for Eq. (37) to reproduce the correct solution (21), we have to choose $k_1(\tau)$ in such a way that the solution of the equation

$$\frac{d}{dt}z(t) = -\int_0^t dt_1 k_1(t-t_1)z(t_1), \quad z(0)=1, \tag{43}$$

is given by

$$z(t) = |G(t)|^2. \tag{44}$$

Formulated in Laplace space, this means that

$$\hat{k}_1(u) = \frac{1-u\hat{z}(u)}{\hat{z}(u)}. \tag{45}$$

Since superoperator (36) preserves the Hermiticity and the trace of the density matrix, Eqs. (21)–(24) follow directly from Eqs. (37) and (38). Thus, we find that Eq. (36) represents the exact memory kernel of the model for any given two-point correlation function. In fact, given $f(\tau)$, the functions $\varepsilon(\tau)$, $k_1(\tau)$, and $k_2(\tau)$ are uniquely determined by Eqs. (40), (41), and (45). In view of this result, the memory kernel (36) can now be written in the form

$$\mathcal{K}_{\rm NZ}(\tau)\rho = -if_2(\tau)[\sigma_+\sigma_-,\rho] - f_1(\tau)\{\sigma_+\sigma_-,\rho\} + k_1(\tau)\sigma_-\rho\sigma_+ + [2f_1(\tau)-k_1(\tau)]\sigma_+\sigma_-\rho\sigma_+\sigma_-. \tag{46}$$

This expression only involves the real and the imaginary parts of the correlation function $f(\tau)$ and the function $k_1(\tau)$, to be determined from Eq. (45).

We note that the various coefficients in the memory kernel exhibit a very different convergence behavior. In fact, we see that the commutator and the anticommutator term in Eq. (46) come out exactly in second order in α. It follows that the equation of motion for the coherence ρ_{10} [see Eq. (39)] is already reproduced exactly within second order, while the exact representation of the equation for the population ρ_{11} generally requires the inclusion of all orders of the expansion. This nonuniform convergence behavior of the elements of the density matrix has also been observed in other, more complicated models [31] and seems to be a typical feature of the perturbation expansion of the memory kernel.

As shown below, relations (20) together with Eqs. (43)–(45) provide a direct perturbation approach to the determination of the functions appearing in the memory kernel. This provides an alternative approach with respect to the standard Nakajima-Zwanzig perturbation expansion. Moreover, this set of equations in some cases allows a closed analytical expression for the memory kernel to be derived.

B. Perturbation expansions of the memory kernel

Here we discuss two methods of expanding the exact memory kernel with respect to the strength of the interaction Hamiltonian H_I.

The first expansion method relies on the expansion for $G(t)$ given by Eq. (30). Indeed, as shown in Sec. IV A, to obtain the memory kernel we only need to know the function $k_1(t)$. A perturbative expression for the latter can be easily obtained by relying on the expansion for $G(t)$, noting that, thanks to Eq. (43), the Laplace transform of $k_1(t)$ can be directly expressed by means of the Laplace transform of the function $z(t) = |G(t)|^2$, according to Eq. (45). This procedure leads to the following expansion:

$$k_1(t) = \sum_{n=0}^{\infty} k_1^{(2n)}(t), \tag{47}$$

as described in detail in Appendix A, where the zero-order contribution is immediately seen to be zero.

Here we consider for the sake of simplicity only the second-order contribution, which is readily obtained. According to Eq. (31) together with the initial condition $G(0)=1$, the expression for $G(t)$ up to second order is given by

$$G(t) \approx 1 - \int_0^t dt_1 \int_0^{t_1} dt_2 f(t_2), \tag{48}$$

so that in the same approximation, recalling that the two-point correlation function $f(t)$ is a quantity of second order, one has

$$z(t) \approx 1 - 2\int_0^t dt_1 \int_0^{t_1} dt_2 f_1(t_2), \tag{49}$$

where $f_1(t)$ denotes the real part of the correlation function. The Laplace transform of this quantity is now easily expressed in terms of the Laplace transform of the correlation function according to

$$\hat{z}(u) \approx \frac{u - 2\hat{f}_1(u)}{u^2}, \tag{50}$$

and, by further exploiting Eq. (45), we find

$$\hat{k}_1(u) \approx 2\hat{f}_1(u). \tag{51}$$

This immediately implies, for the second-order contributions to the kernel (36),

$$k_1^{(2)}(t) = 2f_1(\tau), \tag{52}$$

and therefore, due to Eq. (41),

$$k_2^{(2)}(\tau) = 0. \tag{53}$$

As shown in Appendix A, the fourth-order contribution reads

$$k_1^{(4)}(t-t_1) = -2\,\mathrm{Re}\int_{t_1}^{t} dt_2 \int_{t_1}^{t_2} dt_3 [f(t-t_3)f(t_1-t_2) + f(t-t_1)f(t_3-t_2)], \tag{54}$$

so that $k_2^{(4)} = -k_1^{(4)}$. Indeed Eq. (41) generally implies $k_2^{(2n)} = -k_1^{(2n)}$ for $n \geqslant 2$; therefore, Eq. (47) also provides an expansion for $k_2(t)$.

The second expansion method is to employ the general Nakajima-Zwanzig projection operator technique [32,33], in which the memory kernel is expressed in terms of the full propagator of the total system. The details of this method for our model are presented in Appendix B, where it is shown that the projection operator technique reproduces, as expected, the preceding results obtained by the direct expansion of the coefficients in the memory kernel.

V. DISCUSSION

A. Comparison of the time-convolutionless and the Nakajima-Zwanzig master equations

It is interesting to compare the exact time-convolutionless and Nakajima-Zwanzig master equations. For the considered model, the functions appearing in Eqs. (28) and (36) are given by Eqs. (29) and Eqs. (40) and (41), respectively.

Comparing Eqs. (28) and (36), we see that the superoperator structure of the memory kernel differs from that of the time-convolutionless generator. In fact, the memory kernel [Eq. (36)] contains the term proportional to $k_2(\tau)$, which involves the projection $\sigma_+\sigma_- = |1\rangle\langle 1|$ onto the excited state. Without such a term, Eqs. (37) and (38) for the population and the coherence would be incompatible with the exact expressions (21) and (23). However, a term with this structure is missing in the time-convolutionless generator (28). A further remarkable point is the fact that in second order $k_2(\tau) = 0$, according to Eqs. (41) and (52). This shows that the difference in the superoperator structure of the memory kernel and the time-convolutionless generator is visible only in higher orders of the perturbation expansion.

The preceding discussion leads to some important conclusions for the modeling of non-Markovian dynamics through phenomenological master equations. In the Markovian limit our model yields the following Lindblad generator $\mathcal{L}$ describing a quantum dynamical semigroup:

$$\mathcal{L}\rho = -\frac{i}{2}S_M[\sigma_+\sigma_-,\rho] + \gamma_M\left[\sigma_-\rho\sigma_+ - \frac{1}{2}\{\sigma_+\sigma_-,\rho\}\right], \tag{55}$$

with constant frequency shift S_M and decay rate $\gamma_M \geqslant 0$. Usually master equations of this form are derived by applying the Markov approximation and second-order perturbation theory (Born-Markov approximation). A natural and widely used non-Markovian generalization is the following. One keeps the structure of the Lindblad generator $\mathcal{L}$ and introduces a kernel function $h(\tau)$, thus obtaining a master equation of the form [24–28]

$$\frac{d}{dt}\rho(t) = \int_0^t dt_1 h(t-t_1)\mathcal{L}\rho(t_1). \tag{56}$$

Although this equation is perfectly justified as a phenomenological ansatz, it does not generally represent the correct structure of the memory kernel of the underlying microscopic model. In fact, we see that even for the simple model studied here the true memory kernel (36) is not of the form of Eq. (56). It is rather given by a linear combination of terms of the form of Eq. (56), where, going beyond the Born approximation, other operator structures appear besides the Markovian Lindblad generator: the latter are still in Lindblad form but with different Lindblad operators. This observation seems to be of particular relevance for the analysis of the positivity and the complete positivity of the dynamical maps obtained from phenomenological equations of motion.

B. Example

These considerations can be nicely illustrated considering the example of an exponential correlation function, corresponding to a Lorentzian spectral density [2]

$$f(\tau) = \tfrac{1}{2}\gamma_0\lambda e^{-\lambda|\tau|}, \tag{57}$$

where the parameters γ_0 and λ are real and positive. For this case both the time-convolutionless generator and the Nakajima-Zwanzig kernel can be exactly calculated. Indeed by means of Eq. (20) one obtains for the function $G(t)$ the expression

$$G(t) = e^{-\lambda t/2}\left[\cosh\left(\frac{\lambda t}{2}\delta\right) + \frac{1}{\delta}\sinh\left(\frac{\lambda t}{2}\delta\right)\right], \tag{58}$$

where $\delta = \sqrt{1-2\gamma_0/\lambda}$. Note that this function is always real. Furthermore, it stays positive for any time t in the weak coupling regime $\gamma_0 < \lambda/2$, while for strong coupling ($\gamma_0 > \lambda/2$), the parameter δ becomes purely imaginary and the function $G(t)$ starts to oscillate. In particular it goes through zero for the first time when t is equal to the smallest positive solution of

$$t = \frac{2}{\lambda\hat{\delta}}(n\pi - \arctan\hat{\delta}), \tag{59}$$

where $\hat{\delta} = \sqrt{2\gamma_0/\lambda - 1}$ and $n \in \mathbb{N}$. Building on Eq. (58), one can obtain the exact expressions for the functions $\gamma(t)$ and $S(t)$ appearing in the time-convolutionless generator, given by $S(t) = 0$ and

$$\gamma(t) = 2\gamma_0\frac{\sinh\left(\frac{\lambda t}{2}\delta\right)}{\delta\cosh\left(\frac{\lambda t}{2}\delta\right) + \sinh\left(\frac{\lambda t}{2}\delta\right)}. \tag{60}$$

In order to obtain the Nakajima-Zwanzig kernel, one considers the Laplace transform of the function $z(t) = |G(t)|^2$, which is found to be

$$\hat{z}(u) = \frac{(u+\lambda)(u+2\lambda)+\gamma_0\lambda}{(u+\lambda)[(u+\lambda)^2-\lambda^2+2\gamma_0\lambda]}, \quad (61)$$

so that according to Eq. (45) one has

$$\hat{k_1}(u) = \gamma_0\lambda\frac{u+2\lambda}{(u+\lambda)(u+2\lambda)+\gamma_0\lambda}. \quad (62)$$

Transforming back to the time domain we finally get

$$k_1(t) = \gamma_0\lambda e^{-3\lambda t/2}\left[\cosh\left(\frac{\lambda t}{2}\delta'\right)+\frac{1}{\delta'}\sinh\left(\frac{\lambda t}{2}\delta'\right)\right], \quad (63)$$

where $\delta' = \sqrt{1-4\gamma_0/\lambda}$. Substituting this result into Eq. (46), we find the exact memory kernel for the case of an exponential correlation function.

The exact expressions for Eqs. (60) and (63) already allow for an important comparison. While the function on the right-hand side of Eq. (63) represents an analytic function of γ_0 (remember that γ_0 is a quantity of second order in the expansion parameter α), so that the Nakajima-Zwanzig memory kernel has an infinite radius of convergence, the same does not hold true for the time-convolutionless generator. Indeed the time-convolutionless expansion breaks down in the strong coupling regime $\gamma_0 > \lambda/2$, when the function $G(t)$ given in Eq. (58) goes through zero. This behavior corresponds to the divergence of the decay rate $\gamma(t)$ given in Eq. (60).

Considering an expansion in γ_0 of the function $k_1(t)$ due to the fact that the correlation function Eq. (57) is real, one obtains

$$\mathcal{K}_{\rm NZ}^{(2)}(\tau)\rho = 2f(\tau)\big[\sigma_-\rho\sigma_+ - \tfrac{1}{2}\{\sigma_+\sigma_-,\rho\}\big], \quad (64)$$

so that up to second order the corresponding master equation is indeed of the form of Eq. (56) with the exponential kernel function $h(t) = 2f(t)$. However, in fourth order, further terms appear which are not present in Eq. (56):

$$\mathcal{K}_{\rm NZ}^{(4)}(\tau)\rho = k_1^{(4)}(\tau)(\sigma_-\rho\sigma_+ - \sigma_+\sigma_-\rho\sigma_+\sigma_-), \quad (65)$$

where

$$k_1^{(4)}(\tau) = \gamma_0^2[e^{-\lambda\tau}(1-\lambda\tau)-e^{-2\lambda\tau}]. \quad (66)$$

As shown in Ref. [9], this implies in particular that if one truncates the expansion to first order in γ_0 the complete positivity (and even the positivity) of the resulting dynamical map is violated for strong couplings in the Nakajima-Zwanzig case. On the contrary, the second-order time-convolutionless master equation always guarantees complete positivity, as can be seen considering the second-order approximation for Eq. (60) given by

$$\gamma^{(2)}(t) = \gamma_0(1-e^{-\lambda t}). \quad (67)$$

VI. CONCLUSIONS

We have constructed the exact Nakajima-Zwanzig memory kernel for a specific model describing the decay of a two-level system into a reservoir of field modes which is initially in a vacuum state. The construction of the memory kernel is based on the analytical solution of the Schrödinger equation within the Hilbert-space sector describing states with zero or one excitation and is valid for a generic spectral density. Since the dynamical map giving the reduced system dynamics of the two-state system is known exactly, there is, of course, no reason in principle to resort to any kind of master equation in order to determine the dynamical behavior of the system. However, the present results lead to several important implications, which are relevant for more realistic physical systems and their microscopic or phenomenological modeling and for which analytical results cannot be obtained. Indeed for this model both the time-convolutionless generator and the Nakajima-Zwanzig kernel can be exactly expressed in terms of functions for which perturbative expansions are given. Furthermore, the analytical expression of these functions has been obtained for a reservoir with an exponential correlation function, corresponding to a Lorentzian spectral density. This allows for a direct comparison of the two approaches expressing the dynamics in terms of a time-local and integrodifferential master equation, respectively. It turns out that, contrary to what is often expected, the Nakajima-Zwanzig master equation is not simply obtained by convolution of the Lindblad operator appearing in the non-Markovian case with a suitable kernel. It actually has a different operator structure, emerging when considering higher perturbative orders. Furthermore, this exact analytical result shows the different convergence behavior of the two approaches. While the Nakajima-Zwanzig kernel is an analytic function of the coupling strength, providing a well-defined master equation at any time, the time-convolutionless generator breaks down at finite time in the strong coupling regime, thus failing to reproduce the asymptotic behavior.

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APPENDIX A

In this appendix we consider how to obtain a perturbative expansion for the function $k_1(\tau)$, which according to Eq. (36) and Eqs. (40) and (41) determines the memory kernel in the Nakajima-Zwanzig master equation. To this end, one considers the solution of Eq. (20), which is of the form Eq. (30) with $G(0) = 1$ and $G^{(2n)}(t)$ explicitly given by

$$G^{(2n)}(t) = (-1)^n\int_0^t dt_1\int_0^{t_1} dt_2\cdots\int_0^{t_{2n-1}} dt_{2n} \times\prod_{i=1}^n f(t_{2i-1}-t_{2i}), \quad (A1)$$

where $f(t)$ is the two-point correlation function of the reservoir, so that

$$\begin{aligned} z(t) &= |G(t)|^2 \\ &= 1 + 2\,{\rm Re}\sum_{n=1}^\infty(-1)^n\int_0^t dt_1\int_0^{t_1} dt_2\cdots\int_0^{t_{2n-1}} dt_{2n} \\ &\quad\times\prod_{i=1}^n f(t_{2i-1}-t_{2i}) + \left|\sum_{n=1}^\infty(-1)^n\int_0^t dt_1\right. \\ &\quad\left.\times\int_0^{t_1} dt_2\cdots\int_0^{t_{2n-1}} dt_{2n}\prod_{i=1}^n f(t_{2i-1}-t_{2i})\right|^2. \end{aligned} \quad (A2)$$

Considering terms up to fourth order in the expansion parameter, one has

$$z(t) = 1 - 2\,\mathrm{Re}\int_0^t dt_1 \int_0^{t_1} dt_2 f(t_2) + 2\,\mathrm{Re}\int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_4} dt_4 f(t_1-t_2) f(t_3-t_4) + \left|\int_0^t dt_1 \int_0^{t_1} dt_2 f(t_2)\right|^2 + \cdots, \tag{A3}$$

and, denoting real and imaginary parts of $f(t)$ as in Eq. (42), also

$$z(t) = 1 - 2\int_0^t dt_1 \int_0^{t_1} dt_2 f_1(t_2) + 2\int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \times \int_0^{t_3} dt_4 [f_1(t_1-t_2) f_1(t_3-t_4) - f_2(t_1-t_2) f_2(t_3-t_4)] + \left|\int_0^t dt_1 \int_0^{t_1} dt_2 f_1(t_2)\right|^2 + \left|\int_0^t dt_1 \int_0^{t_1} dt_2 f_2(t_2)\right|^2 + \cdots. \tag{A4}$$

Introducing the functions

$$h_i(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 f_i(t_2), \quad i = 1,2, \tag{A5}$$

one obtains, for the Laplace transform of $z(t)$,

$$\hat{z}(u) = \frac{u - 2u\widehat{f_1}(u)}{u^2} + \frac{2}{u^3}\left[\widehat{f_1}^2(u) - \widehat{f_2}^2(u)\right] + \widehat{h_1^2}(u) + \widehat{h_2^2}(u) + \cdots, \tag{A6}$$

and thanks to Eq. (45),

$$\hat{k}_1(u) = 2\widehat{f_1}(u) + \frac{2}{u}\left[\widehat{f_1}^2(u) + \widehat{f_2}^2(u)\right] - u^2\left[\widehat{h_1^2}(u) + \widehat{h_2^2}(u)\right] + \cdots. \tag{A7}$$

Now using the fact that the functions h_i are equal to zero together with their derivatives at $t = 0$ so that

$$\widehat{\frac{d^2}{dt^2} h_i^2}(u) = u^2 \widehat{h_i^2}(u), \tag{A8}$$

one has

$$k_1(\tau) = 2f_1(\tau) + 2\int_0^\tau dt_1 \int_0^{t_1} dt_2 f_1(t_1-t_2) f_1(t_2) + 2\int_0^\tau dt_1 \int_0^{t_1} dt_2 f_2(t_1-t_2) f_2(t_2) - 2\left|\int_0^\tau dt_1 f_1(t_1)\right|^2 - 2\left|\int_0^\tau dt_1 f_2(t_1)\right|^2 - 2f_1(\tau)\int_0^\tau dt_1 \int_0^{t_1} dt_2 f_1(t_1-t_2) - 2f_2(\tau)\int_0^\tau dt_1 \int_0^{t_1} dt_2 f_2(t_1-t_2) + \cdots. \tag{A9}$$

We now exploit the identity

$$\int_0^\tau dt_2 \int_0^{t_2} dt_3 f(t_2-t_3) f(t_3) - \left|\int_0^\tau dt_2 f_1(t_2)\right|^2 + \int_0^\tau dt_2 \int_0^{t_2} dt_3 f(\tau-t_3) f(t_2) = 0, \tag{A10}$$

which can be checked by noting that the function of t defined by the left-hand side of Eq. (A10) has a vanishing derivative and is equal to zero for $t = 0$. We are thus left with

$$k_1(\tau) = 2f_1(\tau) - 2\int_0^\tau dt_2 \int_0^{t_2} dt_3 [f_1(\tau-t_3) f_1(t_2) + f_1(\tau) f_1(t_2-t_3)] - 2\int_0^\tau dt_2 \int_0^{t_2} dt_3 [f_2(\tau-t_3) \times f_2(t_2) + f_2(\tau) f_2(t_2-t_3)] + \cdots \tag{A11}$$

and, thanks to the fact that real and imaginary parts of $f(t)$ are even and odd, respectively,

$$\begin{aligned} k_1(\tau) &= 2f_1(\tau) - 2\int_0^\tau dt_2 \int_0^{t_2} dt_3 [f_1(\tau-t_3) f_1(-t_2) - f_2(\tau-t_3) f_2(-t_2) + f_1(\tau) f_1(t_3-t_2) - f_2(\tau) f_2(t_3-t_2)] + \cdots \\ &= 2f_1(\tau) - 2\,\mathrm{Re}\int_0^\tau dt_2 \int_0^{t_2} dt_3 [f(\tau-t_3) f(-t_2) + f(\tau) f(t_3-t_2)] + \cdots. \end{aligned} \tag{A12}$$

Upon the change of variables $t_2 \to t_2 - t_1$, $t_3 \to t_3 - t_1$, one has the following for the second- and fourth-order contribution to $k_1(\tau)$:

$$k_1^{(2)}(t-t_1) = 2f_1(t-t_1) \tag{A13}$$

$$k_1^{(4)}(t-t_1) = -2\,\mathrm{Re}\int_{t_1}^t dt_2 \int_{t_1}^{t_2} dt_3 [f(t-t_3) f(t_1-t_2) + f(t-t_1) f(t_3-t_2)]. \tag{A14}$$

APPENDIX B

Here we derive the contributions to the memory kernel Eq. (36) up to fourth order by employing the standard Nakajima-Zwanzig projection operator technique. Since the initial state of the system and bath is of the factorized form of Eq. (14), we can employ the standard projection operator

$$\mathcal{P}w = \mathrm{Tr}_E(w) \otimes \rho_E, \tag{B1}$$

where w is a state of the system plus environment and ρ_E denotes the vacuum state of the reservoir. This projection operator is the same used to obtain Eq. (34), and for it the initial state Eq. (14) is indeed an eigenoperator. Introducing further the superoperators

$$\mathcal{L}(t)\rho(t) = -i[H_I(t), \rho(t)] \tag{B2}$$

with $H_I(t)$ as in Eq. (6), and

$$\mathcal{G}(t,t_1) = \mathcal{T}\exp\left[\int_{t_1}^t ds\, \mathcal{Q}\mathcal{L}(s)\right], \tag{B3}$$

where $\mathcal{T}$ denotes time ordering and $\mathcal{Q} = 1 - \mathcal{P}$, the Nakajima-Zwanzig memory kernel appearing in Eq. (35) is given by

$$\mathcal{K}_{\mathrm{NZ}}(t-t_1)\rho(t_1) = \mathrm{Tr}_E\,[\mathcal{L}(t)\mathcal{G}(t,t_1)\mathcal{Q}\mathcal{L}(t_1)\rho(t_1)\otimes\rho_E]\,. \tag{B4}$$

Noting that for this model $\mathcal{P}\mathcal{L}(t_1)\cdots\mathcal{L}(t_{2n+1})\mathcal{P}=0$, one has

$$\begin{aligned}\mathcal{K}_{\mathrm{NZ}}&(t-t_1)\rho(t_1)\\ &= \mathrm{Tr}_E[\mathcal{L}(t)\mathcal{L}(t_1)\rho(t_1)\otimes\rho_E]\\ &\quad+\int_{t_1}^{t}dt_2\int_{t_1}^{t_2}dt_3\,\{\mathrm{Tr}_E\,[\mathcal{L}(t)\mathcal{L}(t_2)\mathcal{L}(t_3)\mathcal{L}(t_1)\rho(t_1)\otimes\rho_E]\\ &\quad-\mathrm{Tr}_E\,[\mathcal{L}(t)\mathcal{L}(t_2)\mathcal{P}\mathcal{L}(t_3)\mathcal{L}(t_1)\rho(t_1)\otimes\rho_E]\}+\cdots.\end{aligned} \tag{B5}$$

Using Eqs. (6) and (16), one readily obtains

$$\begin{aligned}\mathrm{Tr}_E&[\mathcal{L}(t)\mathcal{L}(t_1)\rho(t_1)\otimes\rho_E]\\ &= (-i)^2[f(t-t_1)\sigma_+\sigma_-\rho(t_1)-f(t_1-t)\sigma_-\rho(t_1)\sigma_+\\ &\quad-f(t-t_1)\sigma_-\rho(t_1)\sigma_+ + f(t_1-t)\rho(t_1)\sigma_+\sigma_-],\end{aligned} \tag{B6}$$

so that the second-order contribution is given by

$$\begin{aligned}\mathcal{K}^{(2)}_{\mathrm{NZ}}(\tau)\rho &= -if_2(\tau)[\sigma_+\sigma_-,\rho]\\ &\quad+2f_1(\tau)\big[\sigma_-\rho\sigma_+-\tfrac{1}{2}\{\sigma_+\sigma_-,\rho\}\big],\end{aligned} \tag{B7}$$

which due to Eq. (46) confirms the result of Eq. (52). Setting

$$I_1(t,t_2,t_3,t_1)\rho(t_1) = \mathrm{Tr}_B[\mathcal{L}(t)\mathcal{L}(t_2)\mathcal{L}(t_3)\mathcal{L}(t_1)\rho(t_1)\otimes\rho_E] \tag{B8}$$

and

$$I_2(t,t_2,t_3,t_1)\rho(t_1) = \mathrm{Tr}_B[\mathcal{L}(t)\mathcal{L}(t_2)\mathcal{P}\mathcal{L}(t_3)\mathcal{L}(t_1)\rho(t_1)\otimes\rho_E], \tag{B9}$$

a lengthy but straightforward calculation leads to the results

$$\begin{aligned}I_2&(t,t_2,t_3,t_1)\rho(t_1)\\ &= f(t-t_2)f(t_3-t_1)\sigma_+\sigma_-\rho(t_1)+f(t_2-t)f(t_1-t_3)\\ &\quad\times\rho(t_1)\sigma_+\sigma_- + 2\,\mathrm{Re}[f(t-t_2)f(t_1-t_3)]\sigma_+\sigma_-\\ &\quad\times\rho(t_1)\sigma_+\sigma_- - 4f_1(t-t_2)f_1(t_1-t_3)\sigma_-\rho(t_1)\sigma_+\end{aligned} \tag{B10}$$

and

$$\begin{aligned}I_1&(t,t_2,t_3,t_1)\rho(t_1)\\ &= I_2(t,t_2,t_3,t_1)\rho(t_1) - 2\,\mathrm{Re}[f(t-t_3)f(t_1-t_2)+f(t-t_1)\\ &\quad\times f(t_3-t_2)]\sigma_-\rho(t_1)\sigma_+ + 2\,\mathrm{Re}[f(t-t_3)f(t_1-t_2)\\ &\quad+f(t-t_1)f(t_3-t_2)]\sigma_+\sigma_-\rho(t_1)\sigma_+\sigma_-.\end{aligned} \tag{B11}$$

Thus, for the fourth-order expression, one has

$$\begin{aligned}\mathcal{K}^{(4)}_{\mathrm{NZ}}&(t-t_1)\rho\\ &= -2\,\mathrm{Re}\int_{t_1}^{t}dt_2\int_{t_1}^{t_2}dt_3[f(t-t_3)f(t_1-t_2)+f(t-t_1)\\ &\quad\times f(t_3-t_2)][\sigma_-\rho\sigma_+-\sigma_+\sigma_-\rho\sigma_+\sigma_-],\end{aligned} \tag{B12}$$

which according to Eq. (46) confirms Eq. (54).

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