Stochastic decoupling approach to the spin-boson dynamics: Perturbative and nonperturbative treatments

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We develop a hierarchical functional derivative (HFD) method to investigate the reduced dynamics of a spin-boson model within the framework of a stochastic decoupling description. Keeping only the lowest-order truncation of the hierarchical functional derivatives, one can recover the second-order Nakajima-Zwanzig quantum master equation. Taking into account the higher-order corrections, our method can be implemented as a highly efficient numerical scheme for a general spin-boson model beyond the usual Markovian, rotating-wave, and weak-coupling approximations.

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I. INTRODUCTION

A quantum dissipative system, which describes the interaction between a quantum subsystem and its surrounding bath, plays a very important role in many research fields, such as quantum information [1], condensed-matter physics [2], and biochemistry [3]. The spin-boson model is a common, but fundamental model in quantum dissipative systems, providing a universal model to simulate the relaxation and the decoherence of a quantum subsystem embedded in a bosonic bath [4]. Despite the long history and the wide interest of the spin-boson model, there are still relatively few methods which are capable of exactly describing its reduced dynamical behavior [4]. To obtaining an analytical result of the quantum dissipative dynamics, different approximations were used in many previous studies, say, the Markovian approximation, the rotating-wave approximation, and the weak-coupling approximation. Though these approximations are acceptable in certain parameter regimes, some important physical information may be ignored when using these approximations. For example, as shown in Refs. [5–12], the counter-rotating-wave terms [5–7] and the non-Markovianity [8–12] have significant effects on the short-time dynamical behavior as well as the steady-state characteristics of the spin-boson model. In this sense, it is highly desirable to develop a method which can accurately capture the reduced dynamics of the spin-boson model in a wide parameter regime. An alternative approach to realize a nonperturbative dynamics of the spin-boson model is presented in this paper which can be generalized to many other quantum dissipative systems as well.

The main difficulty in solving a quantum dissipative dynamical problem lies in the interaction between the quantum subsystem and its surrounding bath. The most simple and straightforward strategy is to decouple the system-bath interaction; by doing so, the evolution of the bath will be no longer involved in the dynamics of the quantum subsystem. Such a decoupling scheme can be achieved by making use of the Hubbard-Stratonovich transformation [13], or alternatively, the characteristics of the Itô calculus [13–15]. There are several decoupling techniques reported in previous articles [13–15]. In this paper, we adopt the stochastic decoupling (SD) scheme originally proposed by Shao *et al.* [13]. The main merit of the SD scheme is its conceptual simplicity: the sole effect of the bath is providing a stochastic field exerting on the evolution of the quantum subsystem. However, the stochastic field induced by the SD scheme randomizes the original equation of motion; one has to perform some stochastic simulations, which are rather time-consuming. On the other hand, a stochastic dynamical method is usually employed to simulate the short-time dynamical behavior of a quantum dissipative system, because it becomes unstable and uncontrollable in the long-time regime [16–18]. In this sense, comparing with a stochastic method, a deterministic approach is always preferred. To achieve this aim, one needs to eliminate all the noise terms and extract a deterministic quantum master equation in the SD scheme.

Unfortunately, the quantum master equation derived from the SD scheme contains some time-nonlocal functional derivatives [19,20], which are generally unknown and severely restrict the application range of the SD scheme. Only for a few special models, such as the quantum Brownian model [21] and the damped Jaynes-Cummings model [22], these time-nonlocal functional derivatives can be exactly worked out. For most realistic quantum dissipative dynamical problems, these functional derivatives cannot be explicitly determined. To overcome the difficulty, in this paper we develop a systematic approach, the HFD method, which can greatly extend the application range of the SD scheme. It is shown that these unknown functional derivatives can be constructed in a hierarchy equation in which each individual functional derivative couples to other functional derivatives with different orders. Although these functional derivatives have been explored in some previous articles [19,20], in this paper we reveal their physical meanings: they are not only ancillary matrices in the hierarchical equations, but also related to multitime bath correlation functions contributing to the reduced dynamical behavior. In a perturbative approximation, one

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can only consider the lowest-order contribution; in this case, the HFD result recovers the well-known Nakajima-Zwanzig quantum master equation [23,24] up to the second order. In a nonperturbative treatment, these hierarchical equations can be numerically solved with a suitable truncation by making use of the traditional Runge-Kutta method. In this situation, the HFD method is beyond the Markovian, rotating-wave, and weak-coupling approximations, which are usually adopted in many previous studies.

This paper is organized as follows: In Sec. II, we present our main theory and show how to handle the spin-boson dynamical problem in the framework of the SD scheme. In Sec. III, we make some comparisons between the numerical results from the HFD method and the results obtained by other approaches [the non-Markovian quantum state diffusion (QSD) approach [25–27] and the Born-Markov quantum master equation]. Some concerned discussions and the main conclusions of this paper are drawn in Sec. IV. In several Appendixes, we provide some additional details about the main text. Throughout the paper, we set $\hbar = k_{\rm B} = 1$, and all the other units are dimensionless as well.

II. THEORY

In this section, we present our main results, i.e., how to use the HFD method to handle the reduced dynamics of a general spin-boson model. In Sec. II A, we first briefly outline the SD scheme proposed by Shao, which is totally beyond the usual Markovian, rotating-wave, and weak-coupling approximations. Unfortunately, due to the difficulty in treating the time-nonlocal functional derivative terms, it is difficult to perform a deterministic simulation for most quantum dissipative models within the SD scheme. To eliminate this problem, in Secs. II C and II D we employ the HFD method, which can greatly generalize the application range of the SD scheme. In a perturbative treatment, the HFD method is equivalent to the Nakajima-Zwanzig quantum master equation in the second order. In a nonperturbative treatment, the higher-order HFD method provides a highly efficient numerical scheme for quantum dissipative dynamics as well.

A. General formulation

In this paper, we consider a generalized Hamiltonian of a quantum dissipative system, which can be described by

$$\hat{H} = \hat{H}_{\rm s} + \hat{H}_{\rm b} + f(\hat{s})g(\hat{b}), \tag{1}$$

where $\hat{H}_{\rm s}$ is the quantum subsystem of interest, and the operator $f(\hat{s})$ denotes the quantum subsystem's dissipative operator, which couples to the surrounding bath. The Hamiltonian of the bath is given by $\hat{H}_{\rm b} \equiv \sum_k \omega_k \hat{b}_k^\dagger \hat{b}_k$, where $\hat{b}_k$ and $\hat{b}_k^\dagger$ are the bosonic annihilation and creation operators of the kth environmental mode with frequency ω_k, respectively. The operator $g(\hat{b}) \equiv \sum_k g_k(\hat{b}_k^\dagger + \hat{b}_k)$ is the environmental dissipative operator, where the parameter g_k quantifies the coupling strength between the quantum subsystem and the kth environmental mode. It is necessary to point out that the SD scheme still works when the interaction term contains a finite number of factorized components, i.e., $\hat{H}_{\rm sb} = \sum_{\rm n} f_{\rm n}(\hat{s})g_{\rm n}(\hat{b})$ [13].

The time evolution of the whole system is governed by the quantum von Neumann equation,

$$i\frac{d}{dt}\hat{\rho}_{\rm sb}(t) = \hat{H}^\times \hat{\rho}_{\rm sb}(t), \tag{2}$$

where we have introduced the commutation superoperator $\hat{X}^\times \hat{Y} \equiv [\hat{X}, \hat{Y}] = \hat{X}\hat{Y} - \hat{Y}\hat{X}$. The straightforward computation of Eq. (2) can be very intractable due to the fact that the size of the whole system scales exponentially with respect to a large number of environmental modes. To avoid a direct calculation of Eq. (2), we adopt the SD method, which was originally proposed by Shao *et al.* [13]. In the SD scheme, the density operator of the whole system can be decoupled as two uncorrelated parts: $\hat{\rho}_{\rm sb}(t) = \mathcal{M}\{\hat{\rho}_{\rm s}(t)\hat{\rho}_{\rm b}(t)\}$, where we have assumed that the whole system is initially prepared in a product state $\hat{\rho}_{\rm sb}(0) = \hat{\rho}_{\rm s}(0)\hat{\rho}_{\rm b}(0)$. In our study, $\hat{\rho}_{\rm b}(0) = \hat{\rho}_{\rm th} \equiv \exp(-\beta\hat{H}_{\rm b})/{\rm tr}_{\rm b}[\exp(-\beta\hat{H}_{\rm b})]$, with β being the inverse temperature. The notation $\mathcal{M}$ denotes the statistical average over all the involved stochastic processes. The two stochastic operators $\hat{\rho}_{\rm s}(t)$ and $\hat{\rho}_{\rm b}(t)$ obey the following stochastic differential equations, respectively [13–15]:

$$\begin{aligned} i d\hat{\rho}_{\rm s}(t) = {} & \hat{H}_{\rm s}^\times \hat{\rho}_{\rm s}(t)dt + \frac{1}{2} f(\hat{s})^\times \hat{\rho}_{\rm s}(t) d\varpi_{1t} \\ & + \frac{i}{2} f(\hat{s})^\circ \hat{\rho}_{\rm s}(t) d\varpi_{2t}^*, \end{aligned} \tag{3}$$

$$\begin{aligned} i d\hat{\rho}_{\rm b}(t) = {} & \hat{H}_{\rm b}^\times \hat{\rho}_{\rm b}(t)dt + \frac{1}{2} g(\hat{b})^\times \hat{\rho}_{\rm b}(t) d\varpi_{2t} \\ & + \frac{i}{2} g(\hat{b})^\circ \hat{\rho}_{\rm b}(t) d\varpi_{1t}^*, \end{aligned} \tag{4}$$

where $\varpi_{1t} \equiv \int_0^t [w_1(t') + i w_4(t')]dt'$ and $\varpi_{2t} \equiv \int_0^t [w_2(t') + i w_3(t')]dt'$ are two complex-valued Wiener processes, and $w_{\rm i}(t)$ (with i = 1, 2, 3, 4) are four uncorrelated white noises, which obey $\mathcal{M}\{w_{\rm i}(t)\} = 0$ and $\mathcal{M}\{w_{\rm i}(t)w_{\rm i'}(t')\} = \delta_{\rm ii'}\delta(t - t')$. For the sake of simplicity, in Eqs. (3) and (4), we have also defined the anticommutation superoperator as $\hat{X}^\circ \hat{Y} \equiv \{\hat{X}, \hat{Y}\} = \hat{X}\hat{Y} + \hat{Y}\hat{X}$. It should be stressed that all the stochastic differential (and integral) equations in this paper are expressed in the Itô sense, which means $\mathcal{M}\{d\varpi_{1t} d\varpi_{1t}^*\} = \mathcal{M}\{d\varpi_{2t} d\varpi_{2t}^*\} = 2dt$ [13,15,28].

The dynamics of the reduced density operator of the quantum subsystem, i.e., $\hat{\varrho}_{\rm s}(t) \equiv {\rm tr}_{\rm b}[\hat{\rho}_{\rm sb}(t)] = \hat{\rho}_{\rm s}(t){\rm tr}_{\rm b}[\hat{\rho}_{\rm b}(t)]$, can be obtained from Eqs. (3) and (4) by employing a Girsanov transformation [21,29,30]. The result is given by

$$\begin{aligned} i d\hat{\varrho}_{\rm s}(t) = {} & [\hat{H}_{\rm s} + f(\hat{s})\bar{g}(t)]^\times \hat{\varrho}_{\rm s}(t)dt + \frac{1}{2} f(\hat{s})^\times \hat{\varrho}_{\rm s}(t) d\varpi_{1t} \\ & + \frac{i}{2} f(\hat{s})^\circ \hat{\varrho}_{\rm s}(t) d\varpi_{2t}^*, \end{aligned} \tag{5}$$

where $\bar{g}(t)$ denotes the bath-induced stochastic field and fully characterizes the influence of the bath on the reduced dynamics of the quantum subsystem. As demonstrated in Ref. [13], $\bar{g}(t)$ plays a similar role to that of the influence functional in the path-integral treatment. The expression of the bath-induced stochastic field $\bar{g}(t)$ can be derived from Eq. (4), as shown in Refs. [13,15,21]. The result is given by

$$\bar{g}(t) = \int_0^t \alpha(t - t_1) d\mu_{t_1}^* + \int_0^t \alpha^*(t - t_1) d\nu_{t_1}^*, \tag{6}$$

where $d\mu_t^* \equiv \frac{1}{2}d\varpi_{1t}^* - \frac{i}{2}d\varpi_{2t}$ and $d\nu_t^* \equiv \frac{1}{2}d\varpi_{1t}^* + \frac{i}{2}d\varpi_{2t}$ are combined complex-valued Wiener processes, and $\alpha(t) \equiv \mathrm{tr_b}[\exp(i\hat{H}_\mathrm{b}t)g(\hat{b})\exp(-i\hat{H}_\mathrm{b}t)g(\hat{b})\hat{\rho}_\mathrm{th}]$ is the two-time bath correlation function. For a bosonic bath consisting of numerous noninteracting harmonic oscillators, it is easy to find the explicit expression of $\alpha(t)$ as follows:

$$\alpha(t) = \int_0^\infty d\omega \mathcal{J}(\omega)\left[\coth\left(\frac{\beta\omega}{2}\right)\cos(\omega t) - i\sin(\omega t)\right], \tag{7}$$

where $\mathcal{J}(\omega) \equiv \sum_k g_k^2\delta(\omega-\omega_k)$ is the so-called bath density spectral function.

In Refs. [13,15], the authors proved that the stochastic equation of motion given by Eq. (5) is equivalent to the stochastic Liouville equation studied in Refs. [16,17] by a simple rearrangement of involved noise terms. Under this circumstance, one can directly perform stochastic simulations of Eq. (5) and obtain a convergent trajectory by a sufficiently large statistical sample. Nevertheless, a stochastic simulation is unstable in the long-time regime and is usually less efficient than solving a deterministic quantum master equation. Therefore, we prefer to extract a deterministic quantum master equation by taking the statistical averages on both sides of Eq. (5). Finally, we obtain the evolution equation of the reduced density operator $\tilde{\varrho}_\mathrm{s}(t) \equiv \mathcal{M}\{\hat{\varrho}_\mathrm{s}(t)\}$ as follows:

$$i\frac{d}{dt}\tilde{\varrho}_\mathrm{s}(t) = \hat{H}_\mathrm{s}^\times\tilde{\varrho}_\mathrm{s}(t) + f(\hat{s})^\times\left[\int_0^t dt_1\alpha(t-t_1)\mathcal{M}\left\{\frac{\delta\hat{\varrho}_\mathrm{s}(t)}{\delta\mu(t_1)}\right\} + \int_0^t d\tau_1\alpha^*(t-\tau_1)\mathcal{M}\left\{\frac{\delta\hat{\varrho}_\mathrm{s}(t)}{\delta\nu(\tau_1)}\right\}\right], \tag{8}$$

where we have used the Furutsu-Novikov theorem [29], which states

$$\mathcal{M}\{w_\varsigma^*\mathfrak{F}[w_t]\} = \mathcal{M}\left\{\frac{\delta\mathfrak{F}[w_t]}{\delta w_\varsigma}\right\},$$

for any white noise $w_t = w(t)$ and its arbitrary functional $\mathfrak{F}[w_t] = \mathfrak{F}[w(t)]$. The characteristics of the involved noise in the SD scheme are different from those of a non-Markovian QSD approach. However, as long as the final reduced density matrix is noiseless, one can demonstrate the equivalence of the non-Markovian QSD and SD approaches [20].

By far, no approximations are used during the derivation of Eq. (8), which means Eq. (8) can be regarded as a rigorous quantum master equation. However, Eq. (8) contains some time-nonlocal functional derivative terms, i.e., $\mathcal{M}\{\delta\hat{\varrho}_\mathrm{s}(t)/\delta\mu(t_1)\}$ and $\mathcal{M}\{\delta\hat{\varrho}_\mathrm{s}(t)/\delta\nu(\tau_1)\}$, which are generally unknown. Only for a few particular models [21,22], one can exactly evaluate their explicit expressions. This problem severely hinders the application range of the SD scheme. Except for the common Markovian approximation (see Sec. II B), in this paper, a perturbative (but non-Markovian) treatment (see Sec. II C) and a rigorous nonperturbative numerical approach (see Sec. II D) are used to solve the problem.

B. Markovian approximation

In the Markovian approximation, the bath correlation function can be approximately viewed as a Dirac-δ function, namely, $\alpha(t-t_1) \simeq \frac{1}{2}\Gamma\delta(t-t_1)$ and $\alpha^*(t-\tau_1) \simeq \frac{1}{2}\Gamma\delta(t-\tau_1)$, where Γ is assumed to be a positive value and can be explained as the decay rate [see Eq. (10)]. Then the quantum master equation given by Eq. (8) reduces to

$$i\frac{d}{dt}\tilde{\varrho}_\mathrm{s}(t) \simeq \hat{H}_\mathrm{s}^\times\tilde{\varrho}_\mathrm{s}(t) + \frac{1}{2}\Gamma f(\hat{s})^\times\left[\mathcal{M}\left\{\frac{\delta\hat{\varrho}_\mathrm{s}(t)}{\delta\mu(t)}\right\} + \mathcal{M}\left\{\frac{\delta\hat{\varrho}_\mathrm{s}(t)}{\delta\nu(t)}\right\}\right]. \tag{9}$$

The explicit expressions of the time-local functional derivatives in Eq. (9) can be easily obtained with the help of Eq. (3). The results are given by

$$\mathcal{M}\left\{\frac{\delta\hat{\varrho}_\mathrm{s}(t)}{\delta\mu(t)}\right\} = -if(\hat{s})\mathcal{M}\{\hat{\varrho}_\mathrm{s}(t)\} = -if(\hat{s})\tilde{\varrho}_\mathrm{s}(t),$$
$$\mathcal{M}\left\{\frac{\delta\hat{\varrho}_\mathrm{s}(t)}{\delta\nu(t)}\right\} = i\mathcal{M}\{\hat{\varrho}_\mathrm{s}(t)\}f(\hat{s}) = i\tilde{\varrho}_\mathrm{s}(t)f(\hat{s}).$$

Substituting the above expressions into Eq. (9), we can obtain the following Markovian quantum master equation:

$$\frac{d}{dt}\tilde{\varrho}_\mathrm{s}(t) = -i\hat{H}_\mathrm{s}^\times\tilde{\varrho}_\mathrm{s}(t) - \frac{1}{2}\Gamma f(\hat{s})^\times f(\hat{s})^\times\tilde{\varrho}_\mathrm{s}(t). \tag{10}$$

One can find Eq. (10) has the same form with the famous Gorini-Kossakowski-Sudarshan-Lindblad quantum master equation [31], which has wide applications in quantum information [1] and quantum optics [32].

C. Perturbative treatment

To solve the quantum master equation [Eq. (8)] beyond the Markovian approximation, one needs to treat the time-nonlocal functional derivative terms in a more rigorous way. For this purpose, we define a series of functional derivative operators [19,20]:

$$\hat{O}(t;t_1,t_2,\ldots,t_m;\tau_1,\tau_2,\ldots,\tau_n) = \mathcal{M}\left\{\frac{\delta^{m+n}\hat{\varrho}_\mathrm{s}(t)}{\delta\mu(t_1)\delta\mu(t_2)\ldots\delta\mu(t_m)\delta\nu(\tau_1)\delta\nu(\tau_2)\ldots\delta\nu(\tau_n)}\right\}. \tag{11}$$

Then the quantum master equation given by Eq. (8) can be rewritten as

$$i\frac{d}{dt}\tilde{\varrho}_\mathrm{s}(t) = \hat{H}_\mathrm{s}^\times\tilde{\varrho}_\mathrm{s}(t) + f(\hat{s})^\times\left[\int_0^t dt_1\alpha(t-t_1)\hat{O}(t;t_1;) + \int_0^t d\tau_1\alpha^*(t-\tau_1)\hat{O}(t;;\tau_1)\right]. \tag{12}$$

It is found that the dynamics of $\tilde{\varrho}_\mathrm{s}(t)$ can be fully determined if one can obtain the expression or the evolution equation of $\hat{O}(t;t_1,t_2,\ldots,t_m;\tau_1,\tau_2,\ldots,\tau_n)$. To this aim, we take the time derivative to $\hat{O}(t;t_1,t_2,\ldots,t_m;\tau_1,\tau_2,\ldots,\tau_n)$ and use the following consistency condition [26,27]:

$$\frac{d}{dt}\mathcal{M}\left\{\frac{\delta\mathfrak{F}[w_t]}{\delta w_\varsigma}\right\} = \mathcal{M}\left\{\frac{\delta}{\delta w_\varsigma}\frac{d\mathfrak{F}[w_t]}{dt}\right\}. \tag{13}$$

Then, the time evolution equation of $\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n)$ can be derived as

$$\begin{aligned} i\frac{d}{dt}\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n) &= \hat{H}_s^{\times}\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n) \\ &+ f(\hat{s})^{\times}\int_0^t dt_{m+1}\alpha(t-t_{m+1}) \\ &\times \hat{O}(t;t_1,t_2,...,t_m,t_{m+1};\tau_1,\tau_2,...,\tau_n) \\ &+ f(\hat{s})^{\times}\int_0^t d\tau_{n+1}\alpha^*(t-\tau_{n+1}) \\ &\times \hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n,\tau_{n+1}), \end{aligned} \tag{14}$$

with the initial conditions

$$\begin{aligned} &\hat{O}(t_m;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n) \\ &= -if(\hat{s})\hat{O}(t_m;t_1,t_2,...,t_{m-1};\tau_1,\tau_2,...,\tau_n), \end{aligned} \tag{15}$$

$$\begin{aligned} &\hat{O}(\tau_n;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n) \\ &= i\hat{O}(\tau_n;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_{n-1})f(\hat{s}). \end{aligned} \tag{16}$$

It is clear to see that the evolution of each individual functional derivative depends on not only itself, but also its two neighboring functional derivatives with different orders. There are infinite coupled equations in Eq. (14). In a practical calculation, one needs to employ a truncation technique for obtaining a finite and closed set of equations. For the simplest truncation it is assumed that

$$\begin{aligned} i\frac{d}{dt}\hat{O}(t;t_1,t_2,...,t_M;\tau_1,\tau_2,...,\tau_N) &\simeq \hat{H}_s^{\times}\hat{O}(t;t_1,t_2,...,t_M;\tau_1,\tau_2,...,\tau_N) \end{aligned} \tag{17}$$

in the $(M+N)$th order, and all the higher-order corrections are set to be zero.

In the lowest-order truncation, we find that

$$\hat{O}(t;t_1;) \simeq -ie^{-i\hat{H}_s(t-t_1)}f(\hat{s})\hat{\varrho}_s(t_1)e^{i\hat{H}_s(t-t_1)} \tag{18}$$

and

$$\hat{O}(t;;\tau_1) \simeq ie^{-i\hat{H}_s(t-\tau_1)}\hat{\varrho}_s(\tau_1)f(\hat{s})e^{i\hat{H}_s(t-\tau_1)}. \tag{19}$$

Inserting Eqs. (18) and (19) into Eq. (12), an approximated but non-Markovian quantum master equation is obtained as follows:

$$\begin{aligned} i\frac{d}{dt}\tilde{\varrho}_s(t) &= \hat{H}_s^{\times}\tilde{\varrho}_s(t) - if(\hat{s})^{\times}\int_0^t dt_1\alpha(t-t_1) \\ &\times e^{-i\hat{H}_s(t-t_1)}f(\hat{s})\hat{\varrho}_s(t_1)e^{i\hat{H}_s(t-t_1)} \\ &+ if(\hat{s})^{\times}\int_0^t d\tau_1\alpha^*(t-\tau_1) \\ &\times e^{-i\hat{H}_s(t-\tau_1)}\hat{\varrho}_s(\tau_1)f(\hat{s})e^{i\hat{H}_s(t-\tau_1)}. \end{aligned} \tag{20}$$

As shown in Appendix A, Eq. (20) is equivalent to the well-known Nakajima-Zwanzig quantum master equation [23,24] up to the second order. The truncation technique given by Eq. (17) is the simplest, but it may not produce good numerical results for a small value of $M+N$. An alternative choice is to assume that [33]

$$\begin{aligned} &\int_0^t dt_{m+1}\alpha(t-t_{m+1})\hat{O}(t;t_1,t_2,...,t_m,t_{m+1};\tau_1,\tau_2,...,\tau_n) \\ &\simeq \xi(t)\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n), \\ &\int_0^t d\tau_{n+1}\alpha^*(t-\tau_{n+1})\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n,\tau_{n+1}) \\ &\simeq \zeta(t)\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n), \end{aligned}$$

where $\xi(t)$ and $\zeta(t)$ are undeterminate terminators with several possible expressions [33], such as $\xi(t)=\int_0^t\alpha(t')dt'$ and $\zeta(t)=\int_0^t\alpha^*(t')dt'$. We want to stress that different truncation techniques may have different numerical performances; they should provide the same physical result when the truncation order is sufficiently large.

Before moving on to the next section, we would like to make some remarks on the functional derivative operators $\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n)$ defined by Eq. (11). It is necessary to point out that these functional derivative operators have their corresponding physical interpretations. They are not mere ancillary matrices or certain mathematical tricks, when solving Eq. (8). Though the SD scheme was originally proposed to study the reduced dynamics of a quantum subsystem coupled to a bosonic bath, this stochastic theory can be also generalized to fermionic bath and spin bath situations [15]. For a more generalized bath, beyond the bosonic bath considered in this paper, the expression of the bath-induced stochastic field $\bar{g}(t)$ is given by [15]

$$\begin{aligned} \bar{g}(t) &= \Phi_1(t) + \int_0^t \Phi_2(t,t_1)d\mu_{t_1}^* + \int_0^t \Phi_2^*(t,t_1)d\nu_{t_1}^* + \int_0^t\int_0^{t_1}\Phi_3(t,t_1,t_2)d\mu_{t_1}^*d\mu_{t_2}^* \\ &+ \int_0^t\int_0^{t_1}\Phi_3^*(t,t_1,t_2)d\nu_{t_1}^*d\nu_{t_2}^* + \cdots + \int_0^t\int_0^{t_1}\int_0^{t_2}...\int_0^{t_{n-1}}\Phi_{n+1}(t,t_1,t_2,..,t_n)d\mu_{t_1}^*d\mu_{t_2}^*...d\mu_{t_n}^* \\ &+ \int_0^t\int_0^{t_1}\int_0^{t_2}...\int_0^{t_{n-1}}\Phi_{n+1}^*(t,t_1,t_2,..,t_n)d\nu_{t_1}^*d\nu_{t_2}^*...d\nu_{t_n}^* + \cdots, \end{aligned} \tag{21}$$

where $\Phi_n(t,t_1,t_2,...,t_{n-1})$ denotes the n-time bath correlation function. The odd-time correlation functions can be neglected if one chooses a suitable thermal equilibrium state as the environmental initial state. Note that Eq. (21) is quite similar to the so-called functional expansion technique used in Ref. [34]. However, in contrast to that of Ref. [34], the expansion coefficients in our scheme have specific physical meanings: they are multitime bath correlation functions. For

the bosonic bath and the fermionic bath, one can only consider that the contributions arise from the two-time bath correlation functions due to the Gaussian characteristic; in this situation, Eq. (21) recovers Eq. (6). However, for a spin bath, one should take into account the effects of the higher-order multitime bath correlation functions.

By substituting Eq. (21) into Eq. (5), one can obtain a more generalized quantum master equation by averaging out the noises as follows:

$$i\frac{d}{dt}\tilde{\varrho}_{\rm s}(t)=\hat{H}_{\rm s}^{\times}\tilde{\varrho}_{\rm s}(t)+f(\hat{s})^{\times}\Bigg[\sum_{m=0}^{\infty}\int_0^t dt_1\int_0^{t_1}dt_2...\int_0^{t_{m-1}}dt_m\,\Phi_{m+1}(t,t_1,t_2,...,t_m)\hat{O}(t;t_1,t_2,...t_m;)$$
$$+\sum_{n=0}^{\infty}\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2...\int_0^{\tau_{n-1}}d\tau_n\,\Phi^*_{n+1}(t,\tau_1,\tau_2,...,\tau_n)\hat{O}(t;;\tau_1,\tau_2,...\tau_n)\Bigg]. \tag{22}$$

From the above quantum master equation, the functional derivative operators $\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n)$ can be regarded as the weights of the multitime bath correlation functions contributing to the final reduced dynamics. In other words, $\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n)$ is closed associated with the influence of its corresponding multitime bath correlation function on a quantum dissipative dynamics. Considering the fact that $\Phi_n\propto\langle g_k\rangle^n$, where $\langle g_k\rangle$ stands for the mean coupling strength, the contribution from higher-order multitime bath correlation functions can be neglected when the system-bath coupling function is not too strong. This result suggests that the reduced dynamical behavior in a spin bath is similar to that of a bosonic (or a fermionic) bath in the weak-coupling regime, where their corresponding two-time correlation functions may have similar structures under certain conditions, owing to the fact that the two-time bath correlation is the leading-order correction in this situation. A similar conclusion has also been reported in Ref. [35].

D. Nonperturbative treatment

As shown in many previous literatures [36], the Nakajima-Zwanzig quantum master equation may give an incorrect dynamical description as a result of neglecting higher-order corrections in the strong-coupling regime. Thus, it is necessary to develop a nonperturbative approach, which can include all the contributions arising from higher-order functional derivative terms.

Due to the non-Markovian characteristics, the evolution equation of $\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n)$, i.e., Eq. (14), contains many integro-differential terms, which are difficult to handle in a numerical simulation. Motivated by the hierarchical equations of motion (HEOM) formalism [37–39], we define a set of auxiliary operators which fold these integro-differential terms in their auxiliary constructions and convert Eq. (14) into a set of ordinary differential equations. To this aim, first, we need to introduce a complete set of orthonormal functions $\{\phi_j(t)\}$ with $j=1,2,3,...,\epsilon$ [15,40,41], and then the bath correlation function can be decomposed in the form of $\alpha(t)=\sum_j c_j\phi_j(t)$, where c_j denotes the expansion coefficients in the basis $\{\phi_j(t)\}$. Owing to the completeness of $\{\phi_j(t)\}$, $\frac{d}{dt}\alpha(t)$ can be fully expanded over the same basis as

$$\frac{d}{dt}\alpha(t)=\sum_{j,j'}c_j\eta_{jj'}\phi_{j'}(t). \tag{23}$$

Then, let us define the auxiliary operators $\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(t)$ as follows:

$$\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(t)\equiv\sum_{\mathbf{j}}c_{\mathbf{j}}\int_0^t dt_1\phi_{j_1}(t-t_1)\int_0^t dt_2\phi_{j_2}(t-t_2)...$$
$$\times\int_0^t dt_m\phi_{j_m}(t-t_m)$$
$$\times\int_0^t d\tau_1\phi^*_{j_1}(t-\tau_1)\int_0^t d\tau_2\phi^*_{j_2}(t-\tau_2)...$$
$$\times\int_0^t d\tau_n\phi^*_{j_n}(t-\tau_n)\hat{O}_{mn}, \tag{24}$$

where $\hat{O}_{mn}=\hat{O}(t;t_1,t_2,...,t_m;\tau_1,\tau_2,...,\tau_n)$, $c_{\mathbf{j}}$ is the expansion coefficient of $\prod_{l=1}^m\alpha_l(t)\prod_{\ell=1}^n\alpha^*_\ell(t)$ in the basis $\{\phi_j(t)\}$, and $\sum_{\mathbf{j}}$ means the summation runs over all the j_l and j_ℓ. The subscript $m+n$ indicates the order of the hierarchy expansion, and the superscript $\mathbf{J}_n^m$, which is a matrix with indefinite size, is employed to identify different auxiliary operators with the same hierarchy order. The expression of $\mathbf{J}_n^m$ is

$$\mathbf{J}_n^m\equiv\begin{pmatrix}j_1, & j_2, & ..., & j_l, & ..., & j_m, & 0, & ...\\ j_1, & j_2, & ..., & j_\ell, & ..., & j_n, & 0, & ...\end{pmatrix}.$$

By doing so, the quantum master equation given by Eq. (12) becomes

$$i\frac{d}{dt}\tilde{\varrho}_{\rm s}(t)=\hat{H}_{\rm s}^{\times}\tilde{\varrho}_{\rm s}(t)+f(\hat{s})^{\times}\hat{\mathcal{O}}^{\mathbf{J}_0^1}_{1+0}(t)+f(\hat{s})^{\times}\hat{\mathcal{O}}^{\mathbf{J}_1^0}_{0+1}(t). \tag{25}$$

And the auxiliary operators $\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(t)$ obey the following hierarchical equations (see Appendix B for more details):

$$i\frac{d}{dt}\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(t)=\hat{H}_s^{\times}\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(t)+f(s)^{\times}\sum_{j_{m+1}}c_{j_{m+1}}\hat{\mathcal{O}}^{\mathbf{J}_n^{m+1}}_{m+1+n}(t)$$
$$+f(s)^{\times}\sum_{j_{n+1}}c^*_{j_{n+1}}\hat{\mathcal{O}}^{\mathbf{J}_{n+1}^m}_{m+n+1}(t)$$
$$+i\sum_{l,l'}\eta_{j_l j_{l'}}\hat{\mathcal{O}}^{\mathbf{J}_n^{m(j_l\to j_{l'})}}_{m+n}(t)$$
$$+i\sum_{\ell,\ell'}\eta^*_{j_\ell j_{\ell'}}\hat{\mathcal{O}}^{\mathbf{J}^m_{n(j_\ell\to j_{\ell'})}}_{m+n}(t)$$

$$+\sum_{l}\phi_{j_l}(0)f(\hat{s})\hat{\mathcal{O}}^{\mathbf{J}_n^{m(m-j_l)}}_{m-1+n}(t)$$

$$-\sum_{\ell}\phi^{*}_{j_\ell}(0)\hat{\mathcal{O}}^{\mathbf{J}^{m}_{n(n-j_\ell)}}_{m+n-1}(t)f(\hat{s}), \tag{26}$$

where $m(j_l \to j_{l'})$ and $n(j_\ell \to j_{\ell'})$ denote an element replacement in the first and the second row of matrix $\mathbf{J}_n^m$, respectively. We use $m(m-j_l)$ and $n(n-j_\ell)$ to mean removing an element in the first and the second row of matrix $\mathbf{J}_n^m$, respectively. Together with Eq. (25), the reduced dynamics of the quantum subsystem is fully determined. This result [Eq. (26)] is beyond the result in Ref. [39] and already resembles that in Ref. [40]. However, we want to emphasize that the choice of the auxiliary operators in our scheme is completely different from those of Refs. [37,38,42,43]. In this sense, we propose an alternative way to establish the HEOM. The HEOM can be viewed as a bridge linking the original quantum Liouville equation and a computable numerical scheme, which should be elaborately designed without losing any important dynamical information. How to build such a bridge is the crucial step of the HEOM treatment. In most of the previous studies [37,38], the construction of HEOM was achieved by an influence functional approach instead of the stochastic process adopted in this paper. We believe that a different viewpoint of building HEOM would be helpful to obtain more physical insights into this research field.

If the bath correlation function $\alpha(t)$ is an Ornstein-Uhlenbeck–type correlation function, i.e., $\alpha(t)=\Omega e^{-\gamma t}$ or $\alpha(t)=\sum_q \Omega_q e^{-\gamma_q t}$, which can be viewed as a special situation of the above derivation, the hierarchical equations given by Eq. (26) can be greatly simplified (see Appendix C). Similar to the hierarchy equation of $\hat{O}_{mn}$, there are infinite auxiliary operators in Eq. (26), thus we need to truncate the number of coupled equations for a sufficiently large integer $M+N$ and let all the terms with $m>M$ and $n>N$ equal zero. In a numerical simulation, the initial conditions for the auxiliary operators are $\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(0)=0$, and we keep on increasing the truncation order until the final result converges.

In Ref. [27], Luo *et al.* introduced a set of auxiliary operators which are very similar to the $\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(t)$ defined in our paper. However, these auxiliary operators employed by Luo *et al.* still contain some complex-valued colored noises, and they obey a series of *stochastic* differential equations. In our treatment, the auxiliary operators $\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(t)$ are noise-free, and evolution equations of $\hat{\mathcal{O}}^{\mathbf{J}_n^m}_{m+n}(t)$ are *ordinary* differential equations. Thus, the key advantage of our formulation is its highly efficient; it is easier to numerically solve a set of (deterministic and noiseless) ordinary differential equations, comparing with computing stochastic differential equations, because it is usually difficult to have a convergent stochastic averaging. In Sec. III, a specific example is provided to verify this algorithm is superior to the non-Markovian QSD approach in numerical efficiency.

III. RESULTS

In this section, we show some numerical results obtained by the HFD method and make some comparisons between our results with those of some other approaches (the non-Markovian QSD approach [27] and the Born-Markov quantum master equation).

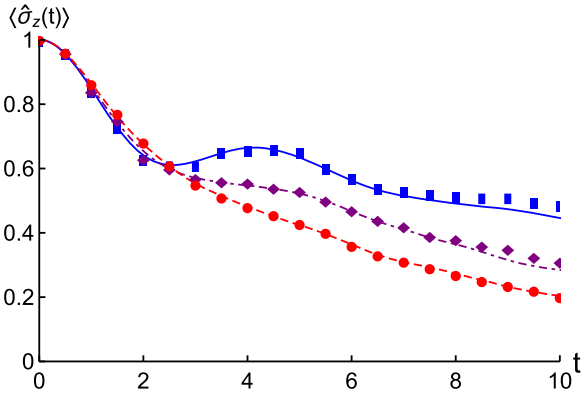

FIG. 1. The dynamics of the population difference $\langle\hat{\sigma}_z(t)\rangle$ of the spin-boson model at zero temperature with different spectrum widths: $\lambda=0.2$ (non-Markovian QSD approach: solid blue line, our numerical results: blue rectangles), $\lambda=0.4$ (non-Markovian QSD approach: dot-dashed purple line, our numerical results: purple diamonds), and $\lambda=0.8$ (non-Markovian QSD approach: dashed red line, our numerical results: red circles). Other parameters are chosen as $\omega_0=1$ and $\Lambda\gamma=0.2$.

We first consider the standard spin-boson model without the rotating-wave approximation, i.e., $\hat{H}_{\rm s}=\frac{1}{2}\omega_0\hat{\sigma}_z$ and $f(\hat{s})=\hat{\sigma}_x$. We assume the initial state of the quantum subsystem is given by $\tilde{\varrho}_s(0)=|e\rangle\langle e|$, where $|e\rangle$ is the excited state of the Pauli z operator, and the environment is initially prepared in its Fock vacuum state. In this numerical simulation, the bath density spectral function is chosen as the Lorentzian spectrum, i.e., $2\pi\mathcal{J}_{\rm L}(\omega)=\Lambda\lambda^2/(\omega^2+\lambda^2)$, where Λ denotes the coupling constant, and λ can be explained as the broadening width of the bath mode, which is connected to the bath correlation time λ^{-1}. In this case, the bath correlation function is given by $\alpha_{\rm L}(t)=\frac{1}{2}\lambda\Lambda e^{-\lambda t}$ at zero temperature, which is the simplest situation in this numerical scheme.

As shown in Fig. 1, our numerical results are in excellent agreement with results from the non-Markovian QSD approach in the short-time regime. A small deviation is found in the long-time regime, probably because of the instability induced by the stochastic simulation in the non-Markovian QSD approach. More importantly, the simulation of the curve $\lambda=0.2$ needs about 2 h for the non-Markovian QSD approach on an Intel core-i7 CPU PC cluster (the truncation order is 50th with 1000 noise realizations), but in our numerical scheme, it only needs a few seconds with the same computational resource.

In the above illustrative example, the quantum subsystem is a spin-$\frac{1}{2}$ system. It should be emphasized that our method can be accommodated to some more complex situations. For instance, our method can also be used to describe the reduced dynamics of an arbitrary size spin interacting with a bosonic reservoir. For this generalized model, $\hat{H}_{\rm s}=\varepsilon\hat{J}_z+\Delta\hat{J}_x$ and $f(\hat{s})=\hat{J}_z$, where $\hat{J}_{x,z}$ are components of a spin vector $\vec{J}$ of an arbitrary spin-J size. Such a large-spin model has been extensively employed to investigate the dynamical property of two-component Bose-Einstein condensates [44]. Unfortunately, no exact solution is found for the reduced dynamics of

the large-spin model. In order to verify the feasibility of the HFD method, we also employ a second-order Born-Markov quantum master equation to compute the reduced dynamics of the large-spin model as a benchmark. The quantum master equation is given by [42]

$$\frac{d}{dt}\tilde{\varrho}_{\rm s}(t) = [-i\hat{H}_{\rm s}^{\times} - f(\hat{s})^{\times}\Upsilon(\hat{s})^{\times} + f(\hat{s})^{\times}\Xi(\hat{s})^{\circ}]\tilde{\varrho}_{\rm s}(t), \tag{27}$$

where

$$\Upsilon(\hat{s}) \equiv \int_0^{\infty} dt\alpha_{\rm R}(t)\hat{f}_{\rm s}(-t),$$
$$\Xi(\hat{s}) \equiv -i\int_0^{\infty} dt\alpha_{\rm I}(t)\hat{f}_{\rm s}(-t),$$

with $\hat{f}_{\rm s}(t) \equiv e^{i\hat{H}_{\rm s}t}f(\hat{s})e^{-i\hat{H}_{\rm s}t}$, and $\alpha_{\rm R(I)}(t)$ being the real (imaginary) part of the bath correlation function $\alpha(t)$. One can further simplify the expressions of $\Upsilon(\hat{s})$ and $\Xi(\hat{s})$ by making use of the trick proposed in Ref. [42], which would be very helpful to a numerical calculation.

We now consider a spin-1 model as the second illustrative example. In this spin-1 model, the matrix expressions of $\hat{J}_{z,x}$ are given by

$$\hat{J}_z = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \quad \hat{J}_x = \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}.$$

These matrices are written in the basis $\{|J=1, J_{\rm m}=1\rangle, |J=1, J_{\rm m}=0\rangle, |J=1, J_{\rm m}=-1\rangle\}$, where $|J, J_{\rm m}\rangle$ are the eigenstates of $\hat{J}_z$ with $\hat{J}_z|J, J_{\rm m}\rangle = J_{\rm m}|J, J_{\rm m}\rangle$. In the simulation, the bath density spectral function is an Ohmic spectrum with Debye-type cutoff

$$\mathcal{J}_{\rm D}(\omega) = \frac{1}{\pi}\frac{2\chi\omega_c\omega}{\omega^2+\omega_c^2},$$

where χ denotes the system-bath coupling strength and ω_c is the cutoff frequency. At finite temperature, the corresponding two-time bath correlation function can be expressed as a series of exponential functions [37–39],

$$\alpha_{\rm D}(t) = \left[\chi\omega_{\rm c}\cot\left(\frac{\beta\omega_{\rm c}}{2}\right) - i\chi\omega_{\rm c}\right]e^{-\omega_{\rm c}t} + \frac{4\chi\omega_{\rm c}}{\beta}\sum_{\rm j=1}^{\infty}\frac{\vartheta_{\rm j}}{\vartheta_{\rm j}^2-\omega_{\rm c}^2}e^{-\vartheta_{\rm j}t},$$

where $\vartheta_{\rm j} \equiv 2{\rm j}\pi/\beta$ denotes the jth Matsubara frequency. In a practical numerical simulation, we can only consider the first few terms in the series, which is reliable when the bath temperature is not very low.

In Fig. 2, we make a comparison between the result obtained by the HFD method and that of the Born-Markov quantum master equation given by Eq. (27). It is clear to see that our numerical result is in agreement with the result from the Born-Markov quantum master equation in short-time regime. In the long-time limit, whether our numerical result or that of the Born-Markov quantum master equation, it is found that $\langle\hat{J}_z(t)\rangle$ finally decays to a same steady value $\langle\hat{J}_z(t\to\infty)\rangle$. In the limit of zero coupling $\chi\to 0$, the steady value

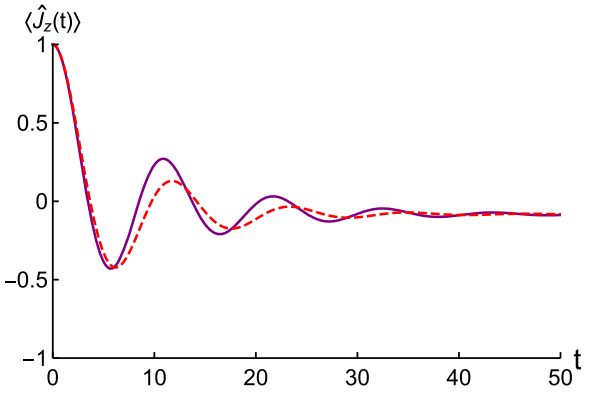

FIG. 2. The dynamics of the spin z component $\langle\hat{J}_z(t)\rangle$ of the spin-1 model at finite temperature $\beta\omega_c = 0.5$. The solid purple line is the numerical result according to our HFD method, and the red dashed line denotes the approximate solution given by the Born-Markov quantum master equation. The initial state is $|J=1, J_{\rm m}=1\rangle$. Other parameters are chosen as $\varepsilon = 0.25\omega_c$, $\Delta = 0.5\omega_c$, $\chi = 0.05$, and $\omega_c = 1$.

$\langle\hat{J}_z(t\to\infty)\rangle$ coincides the thermodynamic expression of $\hat{J}_z$, i.e.,

$$\langle\hat{J}_z(t\to\infty)\rangle = \langle\hat{J}_z\rangle_{\rm th} = -\frac{\varepsilon}{\sqrt{\varepsilon^2+\Delta^2}}\frac{2\sinh(\beta\sqrt{\varepsilon^2+\Delta^2})}{1+2\cosh(\beta\sqrt{\varepsilon^2+\Delta^2})},$$

where $\langle\hat{J}_z\rangle_{\rm th} \equiv {\rm tr_s}[\hat{J}_z\exp(-\beta\hat{H}_{\rm s})]/{\rm tr_s}[\exp(-\beta\hat{H}_{\rm s})]$. This result is physically reasonable and convinces us that the HFD method truly captures the dynamical behavior of a generalized spin-boson model.

IV. CONCLUSIONS

Compared to that of the standard bosonic bath case, where one can easily generate complex Gaussian noises in a practical numerical simulation [43], a straightforward stochastic simulation for a fermionic bath case can be trackable due to the difficulty in generating generic anticommuting Grassmannian noises [15,45]. A natural way to eliminate this problem is to average out the Grassmannian variables and derive a corresponding noise-free quantum master equation, which suggests that the HFD method presented in this paper is a promising candidate for a fermionic bath system. On the other hand, with a suitable truncation, Eq. (22) also provides a perturbative (but non-Markovian) quantum master equation for a spin bath system. In this sense, the SD scheme is a unified methodology for the reduced dynamics of a quantum dissipative system.

In summary, we develop an alternative HFD method to investigate the reduced dynamics of a spin-boson model and greatly extend the application range of the SD scheme. The main idea is to convert the time-nonlocal functional derivative terms in the original SD scheme into a set of hierarchical equations. We also uncover the physical meaning of these functional derivative terms. The sacrifice of the HFD method is that, instead of intractable functional derivatives, we need to deal with infinite integro-differential equations. In a perturbative approximation, one can only consider the lowest-order

truncation of these hierarchical equations and rederive the second-order Nakajima-Zwanzig quantum master equation. In a nonperturbative treatment, we can further eliminate these integro-differential terms by deriving a corresponding hierarchy equation of the auxiliary operators, which can be easily solved by employing the traditional Runge-Kutta algorithm. In this case, our method also provides a highly efficient numerical formalism for a general quantum dissipative system without the usual Markovian, rotating-wave, and weak-coupling approximations. Finally, due to the generality of the quantum dissipative system, we expect our approach to be of interest for a wide range of applications in quantum optics and quantum information processing.

ACKNOWLEDGMENTS

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APPENDIX A: COMPARISON WITH NAKAJIMA-ZWANZIG EQUATION

The general Nakajima-Zwanzig quantum master equation is given by [23]

$$i\frac{d}{dt}\tilde{\varrho}_{\rm s}(t)=\hat{H}_{\rm s}^{\times}\tilde{\varrho}_{\rm s}(t)+\int_0^t d\tau\,\hat{\Sigma}(t-\tau)\tilde{\varrho}_{\rm s}(\tau),\tag{A1}$$

where $\hat{\Sigma}(t)$ is the self-energy superoperator, which gives rise to memory effects, i.e., the time evolution of $\tilde{\varrho}_{\rm s}(t)$ depends on the state $\tilde{\varrho}_{\rm s}(t)$ at all earlier times $\tau\leqslant t$. The rigorous expression of $\hat{\Sigma}(t)$ is given by

$$\hat{\Sigma}(t)=-i{\rm tr}_{\rm b}(\hat{H}_{\rm sb}^{\times}e^{-it\hat{\mathcal{Q}}\hat{H}^{\times}}\hat{H}_{\rm sb}^{\times}\hat{\rho}_{\rm th}),\tag{A2}$$

where $\hat{H}_{\rm sb}=f(\hat{s})g(\hat{b})$ and $\hat{\mathcal{Q}}\equiv 1-\hat{\rho}_{\rm th}{\rm tr}_{\rm b}$ is the projection superoperator. In the second-order (and Born) approximation, one can find the expression of $\hat{\Sigma}^{(2)}(t)$ is given by [24]

$$\begin{aligned}\hat{\Sigma}^{(2)}(t)\tilde{\varrho}_{\rm s}(t)&=-i{\rm tr}_{\rm b}\{[\hat{H}_{\rm sb},e^{-i\hat{H}_0t}[\hat{H}_{\rm sb},\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}]e^{i\hat{H}_0t}]\}\\&=-i{\rm tr}_{\rm b}\{\hat{H}_{\rm sb}e^{-i\hat{H}_0t}\hat{H}_{\rm sb}\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}e^{i\hat{H}_0t}\}+i{\rm tr}_{\rm b}\{e^{-i\hat{H}_0t}\hat{H}_{\rm sb}\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}e^{i\hat{H}_0t}\hat{H}_{\rm sb}\}\\&\quad+i{\rm tr}_{\rm b}\{\hat{H}_{\rm sb}e^{-i\hat{H}_0t}\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}\hat{H}_{\rm sb}e^{i\hat{H}_0t}\}-i{\rm tr}_{\rm b}\{e^{-i\hat{H}_0t}\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}\hat{H}_{\rm sb}e^{i\hat{H}_0t}\hat{H}_{\rm sb}\},\end{aligned}\tag{A3}$$

where $\hat{H}_0=\hat{H}_{\rm s}+\hat{H}_{\rm b}$. The first term in the second row of Eq. (A3) can be calculated as follows:

$$\begin{aligned}{\rm tr}_{\rm b}\{\hat{H}_{\rm sb}e^{-i\hat{H}_0t}\hat{H}_{\rm sb}\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}e^{i\hat{H}_0t}\}&={\rm tr}_{\rm b}[g(\hat{b})e^{-i\hat{H}_{\rm b}t}g(\hat{b})\hat{\rho}_{\rm th}e^{i\hat{H}_{\rm b}t}]f(\hat{s})e^{-i\hat{H}_{\rm s}t}f(s)\tilde{\varrho}_{\rm s}(t)e^{i\hat{H}_{\rm s}t}\\&={\rm tr}_{\rm b}[e^{i\hat{H}_{\rm b}t}g(\hat{b})e^{-i\hat{H}_{\rm b}t}g(\hat{b})\hat{\rho}_{\rm th}]f(\hat{s})e^{-i\hat{H}_{\rm s}t}f(s)\tilde{\varrho}_{\rm s}(t)e^{i\hat{H}_{\rm s}t}\\&=\alpha(t)f(\hat{s})e^{-i\hat{H}_{\rm s}t}f(s)\tilde{\varrho}_{\rm s}(t)e^{i\hat{H}_{\rm s}t}.\end{aligned}$$

By making use of the same method, we can finally obtain

$$\begin{aligned}{\rm tr}_{\rm b}\{e^{-i\hat{H}_0t}\hat{H}_{\rm sb}\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}e^{i\hat{H}_0t}\hat{H}_{\rm sb}\}&=\alpha(t)e^{-i\hat{H}_{\rm s}t}f(\hat{s})\tilde{\varrho}_{\rm s}(t)e^{i\hat{H}_{\rm s}t}f(\hat{s}),\\{\rm tr}_{\rm b}\{\hat{H}_{\rm sb}e^{-i\hat{H}_0t}\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}\hat{H}_{\rm sb}e^{i\hat{H}_0t}\}&=\alpha^*(t)f(\hat{s})e^{-i\hat{H}_{\rm s}t}f(\hat{s})\tilde{\varrho}_{\rm s}(t)e^{i\hat{H}_{\rm s}t},\\{\rm tr}_{\rm b}\{e^{-i\hat{H}_0t}\tilde{\varrho}_{\rm s}(t)\otimes\hat{\rho}_{\rm th}\hat{H}_{\rm sb}e^{i\hat{H}_0t}\hat{H}_{\rm sb}\}&=\alpha^*(t)e^{-i\hat{H}_{\rm s}t}f(\hat{s})\tilde{\varrho}_{\rm s}(t)e^{i\hat{H}_{\rm s}t}f(\hat{s}).\end{aligned}$$

Inserting these expressions into Eq. (A1), one can obtain the quantum master equation given by Eq. (20) in the main text.

APPENDIX B: THE DERIVATION OF EQ. (26)

In this Appendix, we shall show how to derive the HEOM given by Eq. (26). Taking the time derivative of $\hat{\mathcal{O}}_{m+n}^{\mathbf{J}_n^m}(t)$, we find that

$$i\frac{d}{dt}\hat{\mathcal{O}}_{m+n}^{\mathbf{J}_n^m}(t)=\mathbf{I}+\mathbf{II}+\mathbf{III},$$

where the first term $\mathbf{I}$ can be worked out with the help of Eq. (14):

$$\begin{aligned}\mathbf{I}&=\sum_{\mathbf{j}}c_{\mathbf{j}}\int_0^t dt_1\phi_{j_1}(t-t_1)...\int_0^t dt_m\phi_{j_m}(t-t_m)\int_0^t d\tau_1\phi_{j_1}^*(t-\tau_1)...\int_0^t d\tau_n\phi_{j_n}^*(t-\tau_n)\partial_t(i\hat{O}_{mn})\\&=\hat{H}_s^{\times}\hat{\mathcal{O}}_{m+n}^{\mathbf{J}_n^m}(t)+f(s)^{\times}\sum_{j_{m+1}}c_{j_{m+1}}\hat{\mathcal{O}}_{m+1+n}^{\mathbf{J}_n^{m+1}}(t)+f(s)^{\times}\sum_{j_{n+1}}c_{j_{n+1}}^*\hat{\mathcal{O}}_{m+n+1}^{\mathbf{J}_{n+1}^m}(t).\end{aligned}$$

The second term **II** can be evaluated as

$$\begin{aligned}
\mathbf{II} &= i\sum_{\mathbf{j}} c_{\mathbf{j}} \int_0^t dt_1 ... \int_0^t dt_m \int_0^t d\tau_1 ... \int_0^t d\tau_n \hat{O}_{mn} \partial_t \left[\phi_{j_1}(t-t_1)...\phi_{j_m}(t-t_m)\phi^*_{j_1}(t-\tau_1)...\phi^*_{j_n}(t-\tau_n)\right] \\
&= i\sum_{\mathbf{j}} c_{\mathbf{j}} \int_0^t dt_1 ... \int_0^t dt_m \int_0^t d\tau_1 ... \int_0^t d\tau_n \hat{O}_{mn} \partial_t \left[\phi_{j_1}(t-t_1)...\phi_{j_m}(t-t_m)\right]\phi^*_{j_1}(t-\tau_1)...\phi^*_{j_n}(t-\tau_n) \\
&\quad + i\sum_{\mathbf{j}} c_{\mathbf{j}} \int_0^t dt_1 ... \int_0^t dt_m \int_0^t d\tau_1 ... \int_0^t d\tau_n \hat{O}_{mn} \phi_{j_1}(t-t_1)...\phi_{j_m}(t-t_m) \partial_t \left[\phi^*_{j_1}(t-\tau_1)...\phi^*_{j_n}(t-\tau_n)\right] \\
&= i\sum_{l,l'} \eta_{l,l'} \sum_{\mathbf{j}} c_{\mathbf{j}} \int_0^t dt_1 ... \int_0^t dt_{l'} ... \int_0^t dt_m \int_0^t d\tau_1 ... \int_0^t d\tau_n \hat{O}_{mn} \phi_{j_1}(t-t_1)...\phi_{j_{l'}}(t-t_{l'})...\phi_{j_m}(t-t_m) \\
&\quad \times \phi^*_{j_1}(t-\tau_1)...\phi^*_{j_n}(t-\tau_n) + i\sum_{\ell,\ell'} \eta^*_{\ell,\ell'} \sum_{\mathbf{j}} c_{\mathbf{j}} \int_0^t dt_1 ... \int_0^t dt_m \int_0^t d\tau_1 ... \int_0^t d\tau_{\ell'} ... \int_0^t d\tau_n \hat{O}_{mn} \\
&\quad \times \phi_{j_1}(t-t_1)...\phi_{j_m}(t-t_m)\phi^*_{j_1}(t-\tau_1)...\phi_{j_{\ell'}}(t-\tau_{\ell'})...\phi^*_{j_n}(t-\tau_n) \\
&= i\sum_{l,l'} \eta_{j_l j_{l'}} \hat{\mathcal{O}}^{\mathbf{J}^{m(j_l\to j_{l'})}_n}_{m+n}(t) + i\sum_{\ell,\ell'}^{n} \eta^*_{j_\ell j_{\ell'}} \hat{\mathcal{O}}^{\mathbf{J}^{m}_{n(j_\ell\to j_{\ell'})}}_{m+n}(t).
\end{aligned}$$

The last term can be easily formulated out with the help of the initial conditions given by Eqs. (15) and (16) as follows:

$$\begin{aligned}
\mathbf{III} &= i\sum_{l}\sum_{\mathbf{j}} c_{\mathbf{j}}\phi_{j_l}(0) \int_0^t dt_1 \phi_{j_1}(t-t_1)... \int_0^t dt_{l-1}\phi_{j_{l-1}}(t-t_{l-1}) \int_0^t dt_{l+1}\phi_{j_{l+1}}(t-t_{l+1})... \int_0^t dt_m \phi_{j_m}(t-t_m) \\
&\quad \times \int_0^t d\tau_1 \phi^*_{j_1}(t-\tau_1)... \int_0^t d\tau_n \phi^*_{j_n}(t-\tau_n) \hat{O}_{mn}(t;t_1,...,t_l=t,...,t_m;\tau_1,...\tau_n) \\
&\quad + i\sum_{\ell}\sum_{\mathbf{j}} c_{\mathbf{j}}\phi^*_{j_\ell}(0) \int_0^t dt_1 \phi_{j_1}(t-t_1)... \int_0^t dt_m \phi_{j_m}(t-t_m) \int_0^t d\tau_1 \phi^*_{j_1}(t-\tau_1)... \int_0^t d\tau_{\ell-1}\phi^*_{j_{\ell-1}}(t-\tau_{\ell-1}) \\
&\quad \times \int_0^t d\tau_{\ell+1}\phi^*_{j_{\ell+1}}(t-\tau_{\ell+1})... \int_0^t d\tau_n \phi^*_{j_n}(t-\tau_n) \hat{O}_{mn}(t;t_1,...,t_m;\tau_1,...,\tau_\ell=t,...,\tau_n) \\
&= \sum_{l} \phi_{j_l}(0) f(\hat{s}) \hat{\mathcal{O}}^{\mathbf{J}^{m(m-j_l)}_n}_{m-1+n}(t) - \sum_{\ell} \phi^*_{j_\ell}(0) \hat{\mathcal{O}}^{\mathbf{J}^{m}_{n(n-j_\ell)}}_{m+n-1}(t) f(\hat{s}).
\end{aligned}$$

Finally, we can recover Eq. (26) in the main text.

APPENDIX C: ORNSTEIN-UHLENBECK–TYPE BATH CORRELATION FUNCTION

If $\alpha(t)$ is an Ornstein-Uhlenbeck–type correlation function, namely, $\alpha(t) = \Omega e^{-\gamma t}$, one can redefine the auxiliary operators as

$$\begin{aligned}
\hat{\mathcal{O}}_{mn}(t) &= \int_0^t dt_1 \alpha(t-t_1) \int_0^t dt_2 \alpha(t-t_2)... \int_0^t dt_m \alpha(t-t_m) \\
&\quad \times \int_0^t d\tau_1 \alpha^*(t-\tau_1) \int_0^t d\tau_2 \alpha^*(t-\tau_2)... \\
&\quad \times \int_0^t d\tau_n \alpha^*(t-\tau_n) \hat{O}_{mn},
\end{aligned} \tag{C1}$$

where we have already omitted the hierarchy order index $m+n$ for the sake of simplicity. The subscript (m,n) is a two-dimensional index identifying different auxiliary operators. Making use of the similar procedure shown in Appendix B, one can also derive the hierarchical equations of $\hat{\mathcal{O}}_{mn}(t)$. The result is given by

$$\begin{aligned}
i\frac{d}{dt}\hat{\mathcal{O}}_{mn}(t) &= [\hat{H}^\times_s - i(m\gamma + n\gamma^*)]\hat{\mathcal{O}}_{mn}(t) + f(\hat{s})^\times \\
&\quad \times [\hat{\mathcal{O}}_{m+1,n}(t) + \hat{\mathcal{O}}_{m,n+1}(t)] \\
&\quad + m\Omega f(\hat{s})\hat{\mathcal{O}}_{m-1,n}(t) - n\Omega^* \hat{\mathcal{O}}_{m,n-1}(t) f(\hat{s}).
\end{aligned} \tag{C2}$$

Noticing that $\tilde{\varrho}_s(t) = \hat{\mathcal{O}}_{00}(t)$, then Eqs. (25) and (C2) can be combined and rewritten in a more compact form,

$$\begin{aligned}
\frac{d}{dt}\hat{\mathcal{O}}_{\vec{p}}(t) &= (-i\hat{H}^\times_s - \vec{p}\cdot\vec{\gamma})\hat{\mathcal{O}}_{\vec{p}}(t) - if(\hat{s})^\times \sum_{\mathrm{k}=1}^{2} \hat{\mathcal{O}}_{\vec{p}+\vec{e}_{\mathrm{k}}}(t) \\
&\quad + \sum_{\mathrm{k}=1}^{2} p_{\mathrm{k}} \hat{\Theta}_{\mathrm{k}} \hat{\mathcal{O}}_{\vec{p}-\vec{e}_{\mathrm{k}}}(t),
\end{aligned} \tag{C3}$$

where $\vec{p} = (m,n)$, $\vec{\gamma} = (\gamma,\gamma^*)$, $\vec{\Omega} = (\Omega,\Omega^*)$, $\vec{e}_1 = (1,0)$, and $\vec{e}_2 = (0,1)$ are two-dimensional indexes, and the super-

operator $\hat{\Theta}_{\rm k}$ is defined by

$$\hat{\Theta}_{\rm k} \equiv \frac{i}{2}\Omega_{\rm k}[(-1)^{\rm k} f(\hat{s})^{\circ} - f(\hat{s})^{\times}].$$

It is found that Eq. (C3) recovers the HEOM given by Ref. [39].

For the more general case, where the bath correlation function $\alpha(t)$ can be (or at least approximately) expressed as a sum of exponential functions,

$$\alpha(t) = \sum_{q=1}^{Q} \Omega_q e^{-\gamma_q t},$$

where Q is the cutoff integer. A similar hierarchy equation of $\hat{\mathcal{O}}_{\vec{p}}(t)$ can be derived,

$$\frac{d}{dt}\hat{\mathcal{O}}_{\vec{p}}(t) = (-i\hat{H}_{\rm s}^{\times} - \vec{p}\cdot\vec{\gamma})\hat{\mathcal{O}}_{\vec{p}}(t) - if(\hat{s})^{\times}\sum_{\rm k=1}^{2Q}\hat{\mathcal{O}}_{\vec{p}+\vec{e}_{\rm k}}(t) + \sum_{\rm k=1}^{2Q} p_{\rm k}\hat{\Theta}_{\rm k}\hat{\mathcal{O}}_{\vec{p}-\vec{e}_{\rm k}}(t), \tag{C4}$$

and in this case, $\vec{p} = (m_1, n_1, m_2, n_2, ..., m_Q, n_Q)$, $\vec{\gamma} = (\gamma_1, \gamma_1^*, \gamma_2, \gamma_2^*, ...\gamma_Q, \gamma_Q^*)$, $\vec{\Omega} = (\Omega_1, \Omega_1^*, \Omega_2, \Omega_2^*, ..., \Omega_Q, \Omega_Q^*)$, and $\vec{e}_{\rm k} = (0, 0, ..., 1_{\rm k}, ...0)$ become $2Q$-dimensional indexes.

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