Linear response theory for open systems: Quantum master equation approach

Masashi Ban,[1] Sachiko Kitajima,[1] Toshihico Arimitsu,[2] and Fumiaki Shibata[1]

[1]*Graduate School of Humanities and Sciences, Ochanomizu University, 2-1-1 Otsuka, Bunkyo-ku, Tokyo 112-8610, Japan*

[2]*Institute of Physics, University of Tsukuba, 1-1-1 Tennoudai, Tsukuba, Ibaraki 305-8577, Japan*

(Received 29 December 2016; published 27 February 2017)

A linear response theory for open quantum systems is formulated by means of the time-local and time-nonlocal quantum master equations, where a relevant quantum system interacts with a thermal reservoir as well as with an external classical field. A linear response function that characterizes how a relaxation process deviates from its intrinsic process by a weak external field is obtained by extracting the linear terms with respect to the external field from the quantum master equation. It consists of four parts. One represents the linear response of a quantum system when system-reservoir correlation at an initial time and correlation between reservoir states at different times are neglected. The others are correction terms due to these effects. The linear response function is compared with the Kubo formula in the usual linear response theory. To investigate the properties of the linear response of an open quantum system, an exactly solvable model for a stochastic dephasing of a two-level system is examined. Furthermore, the method for deriving the linear response function is applied for calculating two-time correlation functions of open quantum systems. It is shown that the quantum regression theorem is not valid for open quantum systems unless their reduced time evolution is Markovian.

DOI: 10.1103/PhysRevA.95.022126

I. INTRODUCTION

The linear response theory established by Kubo [1–3] in 1957 has long been utilized not only for analyzing experimental results but also for investigating theoretical models in various fields of physics and chemistry. A general expression of the linear response function is frequently referred to as the Kubo formula, which is very useful since it provides directly measurable quantities such as the magnetic susceptibility and the electrical conductivity. To calculate the Kubo formula, one usually employs the methods of the thermal Green function and the double-time Green function. The former was initiated by Matsubara [4] and the latter was elaborated by Zubarev [5]. Furthermore, the Kubo formula has more fundamental implications in nonequilibrium statistical mechanics of irreversible processes. In fact, the linear response theory establishes and generalizes the Einstein relations [6], the Nyquist theorem [7], and the Onsager reciprocal relations [8,9] from a unified viewpoint of the fluctuation-dissipation theorem [2].

In the linear response theory [1–3], usually a whole system is assumed to be in an equilibrium state at an initial time t_0, where the limit $t_0 \to -\infty$ is taken. So the density matrix at the initial time commutes with the Hamiltonian of the system, which consists of a relevant quantum system and a thermal reservoir if the relevant one is an open system under the influence of a thermal reservoir [3,10]. Then a weak external field is applied adiabatically to the quantum system. The density matrix evolves in time, the time evolution of which is governed by the Liouville–von Neumann equation. The system that we consider here is illustrated in Fig. 1(a).

An averaged value of an observable B under the influence of the external field is calculated up to the first order with respect to the external field, which is schematically shown in Fig. 1(b), where $\langle B\rangle$ stands for the equilibrium average of B and $\Delta\langle B(t)\rangle$ is a deviation from its equilibrium value. The usual linear response theory describes an irreversible process which is not far from equilibrium. In contrast, we develop in this paper a linear response theory for an open quantum system which is initially prepared in an arbitrary state. In this case, as shown in Fig. 1(c), the average value $\langle B(t)\rangle$ in the nonequilibrium state becomes time dependent even if an external field is not applied to the system. Using the extended linear response theory developed in this paper, we can systematically calculate the deviation $\Delta\langle B(t)\rangle$ from the time-dependent average value $\langle B(t)\rangle$ that is caused by the external field. For this purpose, we will use two kinds of quantum mechanical master equations which are derived by the projection operator method. One is the time-nonlocal equation (or the time-convolution equation) [10–12] and the other is the time-local equation (or the time-convolutionless equation) [10,13–16].

Recently, many authors have investigated the linear response theory for open quantum systems by means of various methods [17–27], where a relevant quantum system interacts not only with an external field but also with a thermal reservoir [see Fig. 1(a)]. In Refs. [17,18], applying the projection operator method, the authors have formulated a general theory of the linear response of open quantum systems to an external field. However, they have assumed that the whole system of the relevant quantum system and the thermal reservoir is in a stationary state before applying the external field to the system. So an average value of an observable remains its equilibrium value in the absence of the external field [see Fig. 1(b)]. In Refs. [19,20], using an exactly solvable dephasing model, the authors have investigated the transient linear response of a two-level system. In particular, the effect of initial correlation between the quantum system and the thermal reservoir on the linear response of the system has been discussed in detail. Furthermore, the adiabatic response of an open quantum system, which has a dephasing coupling with a thermal reservoir, has been considered in Refs. [21,22]. In Ref. [23], the nonlinear response of a topological insulator influenced by a thermal reservoir has been investigated. In Ref. [24], the linear response of an open quantum system has been studied by means of the Lindblad equation. In this work,

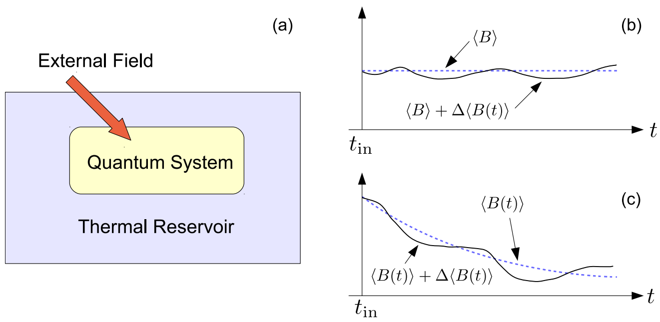

FIG. 1. (a) A schematic representation of the system, where an external field is applied to a relevant quantum system which is influenced by a thermal reservoir. (b) Time evolution of the average value of the system which is in a stationary state before applying an external field, and (c) time evolution when the system is in a nonstationary state.

an external field is phenomenologically introduced into the quantum master equation. So the effect of the external field on the damping operator of the quantum master equation cannot be taken into account, though the effect might be negligible if a reduced time evolution of the quantum system is Markovian. Furthermore, the linear response of a postselected system has been investigated within the framework of the Aharonov-Bergmann-Lebowitz formalism [26,27]. For a postselected system, the linear response is described by both advanced and retarded functions. In many works on the linear response theory of open quantum systems, it is assumed that a whole system is in a stationary state before applying an external field. Therefore, in this paper, using the projection operator method, we consider the linear response of an open quantum system which is prepared in an arbitrary initial state. The time evolution of an average value of a system observable is deviated from its intrinsic irreversible evolution by the external field [see Fig. 1(c)]. We also take into account the initial correlation between a relevant quantum system and a thermal reservoir. Therefore, we formulate the linear response theory in the most general setting. Furthermore, using the method developed for the linear response theory, we can discuss the violation of the quantum regression theorem for open quantum systems [28–37]. Although initial correlation between a quantum system and a thermal reservoir is usually ignored in the consideration of the quantum regression theorem [28–37], our approach makes it possible to take into account the system-reservoir initial correlation.

This paper is organized as follow. In Sec. II, we briefly review the time-local and time-nonlocal quantum master equations derived by the projection operator method [10–16]. A relevant quantum system interacts not only with a thermal reservoir, but also with an external classical field. Although the external field does not interact directly with the thermal reservoir, the damping operator of the quantum master equation depends on the external field in a complicated way. In Sec. III, we first linearize the quantum master equations in the Born approximation with respect to the external field. Solving the linearized equation, we derive the linear response function of an open quantum system. It is found that the linear response function consists of four parts, each of which has a clear physical meaning. Furthermore, the result is compared with the Kubo formula of the usual linear response theory [1–3]. As a simple example, we investigate the linear response of a two-level system, where a thermal reservoir is modeled by a fluctuating classical field which has a dephasing coupling with the relevant quantum system [3,38,39]. In Sec. IV, we generalize the linear response function beyond the Born approximation of the quantum master equation. The structure of the linear response function is the same as that obtained in the Born approximation. In Sec. V, using the method for deriving the linear response function, we obtain an explicit expression of a two-time correlation function for an open quantum system. The result clearly shows that a finite correlation time of a thermal reservoir violates the quantum regression theorem [35,36]. In Sec. VI, we provide a brief summary of this paper.

II. QUANTUM MASTER EQUATION WITH AN EXTERNAL FIELD

We consider reduced time evolution of an open quantum system, where the relevant quantum system interacts not only with a thermal reservoir, but also with a classical external field [see Fig. 1(a)]. Although the external field does not interact directly with the thermal reservoir, it significantly affects a damping operator of the quantum master equation. A Hamiltonian of the total system consisting of the relevant quantum system and the thermal reservoir can be written as

$$H(t) = H_S(t) + H_R + H_{SR}, \tag{1}$$

where H_R is a Hamiltonian of the thermal reservoir and H_{SR} is an interaction Hamiltonian between the quantum system and the thermal reservoir. The Hamiltonian $H_S(t)$ of the quantum system consists of an intrinsic part H_S and a system-field interaction part $H_{\text{ext}}(t)$, that is, $H_S(t) = H_S + H_{\text{ext}}(t)$. Since the external field is assumed not to interact directly with the thermal reservoir, the Hamiltonian $H_{\text{ext}}(t)$ can be written as

$$H_{\text{ext}}(t) = -F(t)A, \tag{2}$$

where $F(t)$ is a time-dependent classical field and A is a system observable coupled to the field. To formulate the linear response theory, it is convenient to use the Liouvillian superoperators. In the following, we denote $L(t) = -(i/\hbar)H^{\times}(t)$, $L_S(t) = -(i/\hbar)H_S^{\times}(t)$, $L_R = -(i/\hbar)H_R^{\times}$, $L_{SR} = -(i/\hbar)H_{SR}^{\times}$, and $L_{\text{ext}}(t) = (i/\hbar)F(t)A^{\times}$ with Kubo's notation $X^{\times}Y = [X,Y]$ [1]. Then, we have the Liouville–von Neumann equation,

$$\frac{\partial}{\partial t}W(t) = L(t)W(t), \tag{3}$$

for a density operator $W(t)$ of the whole system.

Applying the projection operator method [10–16], we eliminate the reservoir variables from the Liouville–von Neumann equation (3). Then we can derive the two different quantum master equations. One is the time-nonlocal or time-convolution equation [10–12,15,16] and the other is the time-local or time-convolutionless equation [10,13–16]. Usually a projection operator is defined by $P\bullet = \rho_R \text{Tr}_R \bullet$, with ρ_R being a reservoir density operator, where Tr_R stands for the trace operation over a Hilbert space of the thermal reservoir. To derive the quantum master equations, we first move the Liouville–von Neumann equation (3) from the Schrödinger picture into the interaction picture. Then we have the Liouville–von Neumann equation in the interaction picture,

$$\frac{\partial}{\partial t}\hat{W}(t) = \hat{L}_{SR}(t)\hat{W}(t), \tag{4}$$

with $\hat{W}(t) = U_{SR}^{-1}(t,t_{\rm in})W(t)$ and $\hat{L}_{SR}(t) = U_{SR}^{-1}(t,t_{\rm in})L_{SR}U_{SR}(t,t_{\rm in})$. Here, $t_{\rm in}$ stands for an initial time and the superoperator $U_{SR}(t,t_{\rm in})$ is given by

$$U_{SR}(t,t_{\rm in}) = \exp_{\leftarrow}\left\{\int_{t_{\rm in}}^{t} dt'\,[L_S(t') + L_R]\right\}, \tag{5}$$

where $\exp_{\leftarrow}$ stands for the time-ordered exponential in which operators are placed from the right to the left in chronological order. In this paper, we denote operators and superoperators with a hat in the interaction picture. The time-nonlocal quantum master equation for the reduced density operator $\hat{W}_S(t) = {\rm Tr}_R\hat{W}(t)$ of the relevant quantum system [10–12,15,16] is given by

$$\frac{\partial}{\partial t}\hat{W}_S(t) = \langle\hat{L}_{SR}(t)\rangle_R\hat{W}_S(t) + \int_{t_{\rm in}}^{t} ds\,\hat{\Phi}(t,s)\hat{W}_S(s) + \hat{J}(t), \tag{6}$$

where $\langle\bullet\rangle_R$ means the partial average with the reservoir density operator ρ_R, namely, $\langle\bullet\rangle_R = {\rm Tr}_R[\bullet\rho_R]$. In this equation, the memory kernel superoperator $\hat{\Phi}(t,s)$ and the inhomogeneous term $\hat{J}(t)$ are given, respectively, by

$$\hat{\Phi}(t,s) = \langle\hat{L}_{SR}(t)\hat{G}_P(t,s)(1-P)\hat{L}_{SR}(s)\rangle_R, \tag{7}$$

$$\hat{J}(t) = {\rm Tr}_R[\hat{L}_{SR}(t)\hat{G}_P(t,t_{\rm in})(1-P)\hat{W}(t_{\rm in})], \tag{8}$$

with

$$\hat{G}_P(t,s) = \exp_{\leftarrow}\left[\int_s^t dt'\,(1-P)\hat{L}_{SR}(t')\right]. \tag{9}$$

The memory kernel superoperator $\hat{\Phi}(t,s)$ can be expanded in terms of the partial cumulants of the Liouvillian superoperators $\hat{L}_{SR}(t)$ [15,16]. On the other hand, we can derive from Eq. (4) the time-local quantum master equation for the reduced density operator $\hat{W}_S(t)$ [10,13–16],

$$\frac{\partial}{\partial t}\hat{W}_S(t) = \hat{K}(t)\hat{W}_S(t) + \hat{I}(t), \tag{10}$$

with

$$\hat{K}(t) = \langle\hat{L}_{SR}(t)[1-\hat{\Sigma}(t)]^{-1}\rangle_R, \tag{11}$$

$$\hat{I}(t) = {\rm Tr}_R[\hat{L}_{SR}(t)[1-\hat{\Sigma}(t)]^{-1}\hat{G}_P(t,t_{\rm in})(1-P)\hat{W}(t_{\rm in})]. \tag{12}$$

In these equations, the superoperator $\hat{\Sigma}(t)$ is given by

$$\hat{\Sigma}(t) = \int_{t_{\rm in}}^{t} ds\,\hat{G}_P(t,s)(1-P)\hat{L}_{SR}(s)P\hat{G}^{-1}(t,s), \tag{13}$$

with

$$\hat{G}(t,s) = \exp_{\leftarrow}\left[\int_s^t dt'\,\hat{L}_{SR}(t')\right]. \tag{14}$$

In deriving Eq. (10), we have assumed that the superoperator $1-\hat{\Sigma}(t)$ is invertible. The superoperator $\hat{K}(t)$ can be expanded in terms of the time-ordered cumulants of the Liouvillian superoperators $\hat{L}_{SR}(t)$ [10,14–16].

When we investigate the effects of an external field on the reduced time evolution of the relevant quantum system, it is convenient to move the quantum master equations back into the Schrödinger picture. Then the time-nonlocal quantum master equation given by Eq. (6) becomes

$$\frac{\partial}{\partial t}W_S(t) = [L_S + L_{\rm ext}(t)]W_S(t) + \Phi_1(t)W_S(t) + \int_{t_{\rm in}}^{t} ds\,\Phi(t,s)W_S(s) + J(t), \tag{15}$$

with

$$\Phi_1(t) = U_S(t,t_{\rm in})\langle\hat{L}_{SR}(t)\rangle_R U_S^{-1}(t,t_{\rm in}), \tag{16}$$

$$\Phi(t,s) = U_S(t,t_{\rm in})\hat{\Phi}(t,s)U_S^{-1}(s,t_{\rm in}), \tag{17}$$

$$J(t) = U_S(t,t_{\rm in})\hat{J}(t), \tag{18}$$

and the time-local equation (10) is rewritten into

$$\frac{\partial}{\partial t}W_S(t) = [L_S + L_{\rm ext}(t)]W_S(t) + K(t)W_S(t) + I(t), \tag{19}$$

with

$$K(t) = U_S(t,t_{\rm in})\hat{K}(t)U_S^{-1}(t,t_{\rm in}), \tag{20}$$

$$I(t) = U_S(t,t_{\rm in})\hat{I}(t). \tag{21}$$

In these equations, the superoperator $U_S(t,t_{\rm in})$ of the quantum system is given by

$$U_S(t,t_{\rm in}) = \exp_{\leftarrow}\left\{\int_{t_{\rm in}}^{t} dt'[L_S + L_{\rm ext}(t')]\right\}, \tag{22}$$

where the equality $U_{SR}(t,t_{\rm in}) = U_S(t,t_{\rm in})e^{L_R(t-t_{\rm in})}$ holds. It is important to note that $\Phi_1(t)$, $\Phi(t,s)$, and $J(t)$ in Eq. (15) and $K(t)$ and $I(t)$ in Eq. (19) include the effect of the external field. The external field that is not directly coupled to the thermal reservoir can affect the irreversible part of the quantum master equation via the system-reservoir interaction. As will be shown in the later sections, this effect is closely related to the non-Markovianity of the reduced time evolution of the quantum system. In order to investigate the linear response of the quantum system to the external field, we need to extract the first-order terms with respect to the external field $F(t)$ from Eqs. (15) and (19).

III. LINEAR RESPONSE IN THE BORN APPROXIMATION

In this section, we derive the linear response function of the open quantum system when the coupling strength between the quantum system and the thermal reservoir is sufficiently weak. In this case, the Born approximation (the second-order approximation) [10] can be applied. First we suppose that the reduced time evolution is described by the time-local quantum master equation. Later we develop the linear response theory by means of the time-nonlocal quantum master equation. In our consideration, we take into account the initial correlation between the quantum system and the thermal reservoir. In this section, we assume that the partial average of $\hat{L}_{SR}(t)$ with the reservoir density operator ρ_R is zero without a loss of generality.

A. First-order solution of the time-local master equation

We derive the solution of the time-local quantum master equation up to the first order with respect to the external field. In the Born approximation of the time-local quantum master equation, we obtain, from Eq. (19),

$$\frac{\partial}{\partial t}W_S(t) = [L_S + L_{\rm ext}(t)]W_S(t) + K^{(2)}(t)W_S(t) + I^{(1)}(t) + I^{(2)}(t), \tag{23}$$

with

$$K^{(2)}(t) = \int_{t_{\rm in}}^{t} dt_1\, U_S(t,t_{\rm in})\langle \hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\rangle_R U_S^{-1}(t,t_{\rm in}), \tag{24}$$

$$I^{(1)}(t) = U_S(t,t_{\rm in}){\rm Tr}_R[\hat{L}_{SR}(t)\delta W], \tag{25}$$

$$I^{(2)}(t) = \int_{t_{\rm in}}^{t} dt_1\, U_S(t,t_{\rm in}){\rm Tr}_R[\hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\delta W], \tag{26}$$

where we set $\delta W = W - W_S\rho_R$ with $W_S = {\rm Tr}_R W$ and $W = W(t_{\rm in})$, and ρ_R is the density operator of the thermal reservoir which is used in the definition of the projection operator P. It is important to note that $I^{(1)}(t) \neq 0$ in general, even if $\langle \hat{L}_{SR}\rangle_R = 0$. If there is no initial correlation between the quantum system and the thermal reservoir, we can write the initial state as $W = W_S W_R$. In this case, if we set $\rho_R = W_R$ in the definition of the projection operator P, the inhomogeneous terms vanish, that is, $I^{(1)}(t) = I^{(2)}(t) = 0$. In the following, we extract the terms from Eq. (23) up to the first order with respect to the external field.

First we consider the third term on the right-hand side of Eq. (23). Since the equalities ${\rm Tr}_R[e^{L_R(t-t_{\rm in})}\bullet] = {\rm Tr}_R[\bullet]$ and $U_{SR}(t,t_{\rm in}) = U_S(t,t_{\rm in})e^{L_R(t-t_{\rm in})}$ hold, we obtain

$$U_S(t,t_{\rm in})\big\langle U_{SR}^{-1}(t,t_{\rm in})\bullet\big\rangle_R = U_S(t,t_{\rm in})\big\langle e^{L_R(t-t_{\rm in})}U_{SR}^{-1}(t,t_{\rm in})\bullet\big\rangle_R = \langle\bullet\rangle_R. \tag{27}$$

Then the second-order superoperator $K^{(2)}(t)$ becomes

$$K^{(2)}(t) = \int_{t_{\rm in}}^{t} dt_1\, \langle L_{SR}U_{SR}(t,t_1)L_{SR}U_{SR}(t_1,t_{\rm in})\rangle_R U_S^{-1}(t,t_{\rm in}). \tag{28}$$

Here, up to the first order with respect to the external field, the superoperators $U_{SR}(t,s)$ and $U_S^{-1}(t,s)$ can be approximated by

$$U_{SR}(t,s) = e^{L_0(t-s)} + \int_s^t d\tau\, e^{L_0(t-\tau)}L_{\rm ext}(\tau)e^{L_0(\tau-s)}, \tag{29}$$

$$U_S^{-1}(t,s) = e^{-L_S(t-s)} - \int_s^t d\tau\, e^{-L_S(\tau-s)}L_{\rm ext}(\tau)e^{-L_S(t-\tau)}, \tag{30}$$

where the Liouvillian superoperator L_0 is given by

$$L_0 = L_S + L_R = -\frac{i}{\hbar}(H_S + H_R)^{\times}. \tag{31}$$

Substituting Eqs. (29) and (30) into the right-hand side of Eq. (28) and discarding the second-order terms, we obtain

$$K^{(2)}(t) = \Pi_0(t) + \Pi_1(t), \tag{32}$$

where $\Pi_0(t)$ is the damping operator of the time-local quantum master equation in the absence of the external field,

$$\Pi_0(t) = \int_{t_{\rm in}}^{t} dt_1\, \langle L_{SR}e^{L_0(t-t_1)}L_{SR}e^{L_0(t_1-t_{\rm in})}\rangle_R e^{-L_S(t-t_{\rm in})}, \tag{33}$$

and $\Pi_1(t)$ is the first-order correction to the damping operator, which is given by

$$\Pi_1(t) = \Pi_1^{(a)}(t) + \Pi_1^{(b)}(t) + \Pi_1^{(c)}(t), \tag{34}$$

with

$$\Pi_1^{(a)}(t) = \int_{t_{\rm in}}^{t} dt_1 \int_{t_1}^{t} d\tau\, \langle L_{SR}e^{L_0(t-\tau)}L_{\rm ext}(\tau)e^{L_0(\tau-t_1)} \times L_{SR}e^{L_0(t_1-t_{\rm in})}\rangle_R e^{-L_S(t-t_{\rm in})}, \tag{35}$$

$$\Pi_1^{(b)}(t) = \int_{t_{\rm in}}^{t} dt_1 \int_{t_{\rm in}}^{t_1} d\tau\, \langle L_{SR}e^{L_0(t-t_1)}L_{SR}e^{L_0(t_1-\tau)} \times L_{\rm ext}(\tau)e^{L_0(\tau-t_{\rm in})}\rangle_R e^{-L_S(t-t_{\rm in})}, \tag{36}$$

$$\Pi_1^{(c)}(t) = -\int_{t_{\rm in}}^{t} dt_1 \int_{t_{\rm in}}^{t} d\tau\, \langle L_{SR}e^{L_0(t-t_1)}L_{SR}e^{L_0(t_1-t_{\rm in})}\rangle_R \times e^{-L_S(\tau-t_{\rm in})}L_{\rm ext}(\tau)e^{-L_S(t-\tau)}. \tag{37}$$

We rewrite the damping operators $\Pi_0(t)$ and $\Pi_1(t)$ in convenient forms for deriving the linear response function of the quantum system. Defining the Liouvillian superoperator,

$$L_{SR}(t) = e^{-L_0(t-t_{\rm in})}L_{SR}e^{L_0(t-t_{\rm in})}, \tag{38}$$

with Eq. (31) and using the relation,

$$\langle e^{L_0(t-t_{\rm in})}\bullet\rangle_R = e^{L_S(t-t_{\rm in})}\langle e^{L_R(t-t_{\rm in})}\bullet\rangle_R = e^{L_S(t-t_{\rm in})}\langle\bullet\rangle_R, \tag{39}$$

we obtain, from Eq. (33),

$$\Pi_0(t) = \int_{t_{\rm in}}^{t} dt_1\, e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)\rangle_R e^{-L_S(t-t_{\rm in})}. \tag{40}$$

Substituting $L_{\rm ext}(t) = (i/\hbar)F(t)A^{\times}$ into Eqs. (35)–(37) and using Eq. (39), we can derive

$$\Pi_1^{(a)}(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{t_{\rm in}}^{\tau} dt_1 e^{L_S(t-t_{\rm in})} \times \langle L_{SR}(t)A^{\times}(\tau)L_{SR}(t_1)\rangle_R e^{-L_S(t-t_{\rm in})}, \tag{41}$$

$$\Pi_1^{(b)}(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{\tau}^{t} dt_1 e^{L_S(t-t_{\rm in})} \times \langle L_{SR}(t)L_{SR}(t_1)A^{\times}(\tau)\rangle_R e^{-L_S(t-t_{\rm in})}, \tag{42}$$

$$\Pi_1^{(c)}(t) = -\frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{t_{\rm in}}^{t} dt_1 e^{L_S(t-t_{\rm in})} \times \langle L_{SR}(t)L_{SR}(t_1)\rangle_R A^{\times}(\tau)e^{-L_S(t-t_{\rm in})}, \tag{43}$$

with

$$A^{\times}(t) = e^{-L_S(t-t_{\rm in})}A^{\times}e^{L_S(t-t_{\rm in})} = (e^{(i/\hbar)H_S(t-t_{\rm in})}Ae^{-(i/\hbar)H_S(t-t_{\rm in})})^{\times}. \quad (44)$$

In the derivation of Eqs. (41)–(43), we have rearranged the order of integrations so that the integration with respect to time which the external field $F(\tau)$ depends on is performed last. Furthermore, using $\langle \bullet A^{\times}(\tau)\rangle_R = \langle\bullet\rangle_R A^{\times}(\tau)$ and $\int_{t_{\rm in}}^t dt_1 = \int_\tau^t dt_1 + \int_{t_{\rm in}}^\tau dt_1$, we obtain the correction to the damping operator,

$$\Pi_1(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\, F(\tau)\int_{t_{\rm in}}^\tau dt_1\, e^{L_S(t-t_{\rm in})} \times \langle L_{SR}(t)[A^{\times}(\tau),L_{SR}(t_1)]\rangle_R e^{-L_S(t-t_{\rm in})}$$
$$= \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\, F(\tau)\int_{t_{\rm in}}^\tau dt_1\, e^{L_S(t-t_{\rm in})} \times \langle L_{SR}(t)[A^{\times}(\tau)]^{\times L_{SR}}L_{SR}(t_1)\rangle_R e^{-L_S(t-t_{\rm in})}. \quad (45)$$

In this equation, the symbol $X^{\times L_{SR}}$ represents the commutation relation between X and $L_{SR}(t)$'s which are placed on the right of X, regardless of the reservoir averages with the density operator ρ_R. For instance, we have

$$YX^{\times L_{SR}}L_{SR}(t_1)\cdots L_{SR}(t_n)Z = Y[X,L_{SR}(t_1)\cdots L_{SR}(t_n)]Z, \quad (46)$$

and

$$\langle YX^{\times L_{SR}}L_{SR}(t_1)\cdots L_{SR}(t_\ell)\rangle_R\langle L_{SR}(t_{\ell+1})\cdots L_{SR}(t_n)\rangle_R Z$$
$$= \langle YXL_{SR}(t_1)\cdots L_{SR}(t_\ell)\rangle_R\langle L_{SR}(t_{\ell+1})\cdots L_{SR}(t_n)\rangle_R Z - \langle YL_{SR}(t_1)\cdots L_{SR}(t_\ell)\rangle_R\langle L_{SR}(t_{\ell+1})\cdots L_{SR}(t_n)X\rangle_R Z. \quad (47)$$

This notation is used throughout this paper. Hence, up to the first order with respect to the external field $F(\tau)$, we have obtained the damping operator of the time-local quantum master equation.

Next we calculate the inhomogeneous terms $I^{(1)}(t)$ and $I^{(2)}(t)$ of the quantum master equation (23). Up to the first order with respect to the external field, we obtain, from Eq. (25),

$$I^{(1)}(t) = I_0^{(1)}(t) + I_1^{(1)}(t), \quad (48)$$

with

$$I_0^{(1)}(t) = e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)\delta W], \quad (49)$$

$$I_1^{(1)}(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\, F(\tau)\mathcal{I}_1(t,\tau), \quad (50)$$

where $\mathcal{I}_1(t,\tau)$ is given by

$$\mathcal{I}_1(t,\tau) = e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)A^{\times}(\tau)\delta W]. \quad (51)$$

In the same way, we can derive, from Eq. (26),

$$I^{(2)}(t) = I_0^{(2)}(t) + I_1^{(2)}(t), \quad (52)$$

where $I_0^{(2)}(t)$ and $I_1^{(2)}(t)$ are given by

$$I_0^{(2)}(t) = \int_{t_{\rm in}}^t dt_1\, e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)L_{SR}(t_1)\delta W], \quad (53)$$

$$I_1^{(2)}(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\, F(\tau)\mathcal{I}_2(t,\tau), \quad (54)$$

with

$$\mathcal{I}_2(t,\tau) = \int_{t_{\rm in}}^\tau dt_1 e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)A^{\times}(\tau)L_{SR}(t_1)\delta W] + \int_\tau^t dt_1 e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)L_{SR}(t_1)A^{\times}(\tau)\delta W]. \quad (55)$$

Here we define $I_0(t)$ and $I_1(t)$ by

$$I_0(t) = I_0^{(1)}(t) + I_0^{(2)}(t) = e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)\delta W] + \int_{t_{\rm in}}^t dt_1\, e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)L_{SR}(t_1)\delta W], \quad (56)$$

$$I_1(t) = I_1^{(1)}(t) + I_1^{(2)}(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\, F(\tau)\mathcal{I}(t,\tau), \quad (57)$$

with

$$\mathcal{I}(t,\tau) = \mathcal{I}_1(t,\tau) + \mathcal{I}_2(t,\tau) = e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)A^{\times}(\tau)\delta W] + \int_{t_{\rm in}}^\tau dt_1 e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)A^{\times}(\tau)L_{SR}(t_1)\delta W] + \int_\tau^t dt_1 e^{L_S(t-t_{\rm in})}{\rm Tr}_R[L_{SR}(t)L_{SR}(t_1)A^{\times}(\tau)\delta W]. \quad (58)$$

The operator $I_0(t)$ represents the inhomogeneous term of the time-local quantum master equation in the absence of the external field and the operator $I_1(t)$ is the first-order correction to the inhomogeneous term.

Finally we obtain the time-local quantum master equation up to the first order with respect to the external field $F(t)$,

$$\frac{\partial}{\partial t}W_S(t) = \left[L_S + \frac{i}{\hbar}F(t)A^{\times}\right]W_S(t) + [\Pi_0(t)+\Pi_1(t)]W_S(t) + I_0(t) + I_1(t). \quad (59)$$

To solve this equation, we define the superoperator $V(t,t_{\rm in})$ of the quantum system by

$$V(t,t_{\rm in}) = \exp_{\leftarrow}\left\{\int_{t_{\rm in}}^t d\tau\,[L_S + \Pi_0(\tau)]\right\}. \quad (60)$$

Then we can obtain the solution of Eq. (59),

$$W_S(t) = V(t,t_{\rm in})U(t,t_{\rm in})W_S + \int_{t_{\rm in}}^t dt_1\, V(t,t_{\rm in})U(t,t_1)V^{-1}(t_1,t_{\rm in})I_0(t_1) + \int_{t_{\rm in}}^t dt_1\, V(t,t_{\rm in})U(t,t_1)V^{-1}(t_1,t_{\rm in})I_1(t_1), \quad (61)$$

where $U(t,t_1)$ is given by

$$\begin{aligned} U(t,t_1) &= \exp_{\leftarrow}\left\{\int_{t_1}^{t} d\tau\, V^{-1}(\tau,t_{\rm in})\left[\frac{i}{\hbar}F(\tau)A^{\times}+\Pi_1(\tau)\right]V(\tau,t_{\rm in})\right\} \\ &= 1+\int_{t_1}^{t} d\tau\, V^{-1}(\tau,t_{\rm in})\left[\frac{i}{\hbar}F(\tau)A^{\times}+\Pi_1(\tau)\right]V(\tau,t_{\rm in}) \\ &\equiv 1+\Delta U(t,t_1). \end{aligned} \tag{62}$$

In the second equality, we have neglected all the terms higher than the first order with respect to the external field. Then substituting Eq. (62) into Eq. (61) and discarding the second-order terms, we obtain the solution of Eq. (59),

$$W_S(t)=W_{S,0}(t)+W_{S,1}(t), \tag{63}$$

with

$$W_{S,0}(t)=V(t,t_{\rm in})W_S+\int_{t_{\rm in}}^{t} dt_1\, V(t,t_1)I_0(t_1), \tag{64}$$

$$\begin{aligned} W_{S,1}(t) &= V(t,t_{\rm in})\Delta U(t,t_{\rm in})W_S \\ &\quad+\int_{t_{\rm in}}^{t} dt_1\, V(t,t_{\rm in})\Delta U(t,t_1)V^{-1}(t_1,t_{\rm in})I_0(t_1) \\ &\quad+\int_{t_{\rm in}}^{t} dt_1\, V(t,t_1)I_1(t_1). \end{aligned} \tag{65}$$

The operator $W_{S,0}(t)$ represents the reduced density matrix of the quantum system in the absence of the external field and the operator $W_{S,1}(t)$ is the first-order correction which yields the linear response function of the quantum system.

B. Linear response function of open quantum systems

To obtain the linear response function of the quantum system, we calculate the change of the average value of an observable B caused by the external field, which is calculated by $\Delta\langle B(t)\rangle=\mathrm{Tr}_S[BW_{S,1}(t)]$. First we obtain the contribution $\Delta B_1(t)$ from the first term on the right-hand side of Eq. (65). It is easy to obtain from Eq. (62),

$$\begin{aligned} \Delta B_1(t) &= \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, \mathrm{Tr}_S[BV(t,\tau)A^{\times}V(\tau,t_{\rm in})W_S]F(\tau) \\ &\quad+\int_{t_{\rm in}}^{t} dt_1\, \mathrm{Tr}_S[BV(t,t_1)\Pi_1(t_1)V(t_1,t_{\rm in})W_S]. \end{aligned} \tag{66}$$

Substituting Eq. (45) into the second term on the right-hand side of this equation and rearranging the order of integrations, we can express the contribution $\Delta B_1(t)$ in the following form:

$$\Delta B_1(t)=\int_{t_{\rm in}}^{t} d\tau\big[\phi_{BA}^{(0)}(t,\tau)+\phi_{BA}^{(\rm c)}(t,\tau)\big]F(\tau), \tag{67}$$

with

$$\phi_{BA}^{(0)}(t,\tau)=\frac{i}{\hbar}\mathrm{Tr}_S[BV(t,\tau)A^{\times}V(\tau,t_{\rm in})W_S], \tag{68}$$

$$\begin{aligned} \phi_{BA}^{(\rm c)}(t,\tau) &= \frac{i}{\hbar}\int_{\tau}^{t} dt_1\int_{t_{\rm in}}^{\tau} dt_2\, \mathrm{Tr}_S\{BV(t,t_1)e^{L_S(t_1-t_{\rm in})} \\ &\quad\times\langle L_{SR}(t_1)[A^{\times}(\tau)]^{\times_{L_{SR}}}L_{SR}(t_2)\rangle_R \\ &\quad\times e^{-L_S(t_1-t_{\rm in})}V(t_1,t_{\rm in})W_S\}, \end{aligned} \tag{69}$$

which are parts of the linear response function of the open quantum system.

Next we obtain the contributions $\Delta B_2(t)$ and $\Delta B_3(t)$ from the second and third terms on the right-hand side of Eq. (65), which stem from the inhomogeneous terms of the quantum master equation. The contribution $\Delta B_2(t)$ is calculated to be

$$\begin{aligned} \Delta B_2(t) &= \int_{t_{\rm in}}^{t} d\tau\, F(\tau)\frac{i}{\hbar}\int_{t_{\rm in}}^{\tau} dt_1\, \mathrm{Tr}_S[BV(t,\tau)A^{\times}V(\tau,t_1)I_0(t_1)] \\ &\quad+\int_{t_{\rm in}}^{t} d\tau\int_{t_{\rm in}}^{\tau} dt_1\, \mathrm{Tr}_S[BV(t,\tau)\Pi_1(\tau)V(\tau,t_1)I_0(t_1)] \\ &= \int_{t_{\rm in}}^{t} d\tau\,\big[\phi_{BA}^{(i,1)}(t,\tau)+\phi_{BA}^{(i,2)}(t,\tau)\big]F(\tau), \end{aligned} \tag{70}$$

with

$$\phi_{BA}^{(i,1)}(t,\tau)=\frac{i}{\hbar}\int_{t_{\rm in}}^{\tau} dt_1\, \mathrm{Tr}_S[BV(t,\tau)A^{\times}V(\tau,t_1)I_0(t_1)], \tag{71}$$

$$\begin{aligned} \phi_{BA}^{(i,2)}(t,\tau) &= \frac{i}{\hbar}\int_{\tau}^{t} dt_1\int_{t_{\rm in}}^{t_1} dt_2\int_{t_{\rm in}}^{\tau} dt_3\, \mathrm{Tr}_S\{BV(t,t_1)e^{L_S(t_1-t_{\rm in})} \\ &\quad\times\langle L_{SR}(t_1)[A^{\times}(\tau)]^{\times_{L_{SR}}}L_{SR}(t_3)\rangle_R \\ &\quad\times e^{-L_S(t_1-t_{\rm in})}V(t_1,t_2)I_0(t_2)\}. \end{aligned} \tag{72}$$

In deriving Eq. (72), we have used Eq. (45). The contribution $\Delta B_3(t)$ from the third term is given by

$$\Delta B_3(t)=\int_{t_{\rm in}}^{t} d\tau\,\phi_{BA}^{(i,3)}(t,\tau)F(\tau), \tag{73}$$

with

$$\phi_{BA}^{(i,3)}(t,\tau)=\frac{i}{\hbar}\int_{\tau}^{t} dt_1\, \mathrm{Tr}_S[BV(t,t_1)\mathcal{I}(t_1,\tau)], \tag{74}$$

where $\mathcal{I}(t_1,\tau)$ is given by Eq. (58).

Summarizing the above results, we can obtain the change $\Delta\langle B(t)\rangle$ of the average value of an observable B,

$$\Delta\langle B(t)\rangle=\int_{t_{\rm in}}^{t} d\tau\,\phi_{BA}(t,\tau)F(\tau). \tag{75}$$

The linear response function $\phi_{BA}(t,\tau)$ consists of four parts,

$$\phi_{BA}(t,\tau)=\phi_{BA}^{(0)}(t,\tau)+\phi_{BA}^{(c)}(t,\tau)+\phi_{BA}^{(i,0)}(t,\tau)+\phi_{BA}^{(i,c)}(t,\tau), \tag{76}$$

where we have redefined $\phi_{BA}^{(i,0)}(t,\tau)\equiv\phi_{BA}^{(i,1)}(t,\tau)$ and $\phi_{BA}^{(i,c)}(t,\tau)\equiv\phi_{BA}^{(i,2)}(t,\tau)+\phi_{BA}^{(i,3)}(t,\tau)$. The function $\phi_{BA}^{(0)}(t,\tau)$ represents the linear response of the system initially prepared in the quantum state $W_S=\mathrm{Tr}_R W$ if we neglect the correlation between the states of the thermal reservoir before and after the application of the external field at the time τ and the inhomogeneous term of the quantum master equation. The function $\phi_{BA}^{(c)}(t,\tau)$ gives a correction due to the effect of the reservoir's correlation. The other two functions $\phi_{BA}^{(i,0)}(t,\tau)$ and

$\phi_{BA}^{(i,c)}(t,\tau)$ represent the effect of the system-reservoir initial correlation on the linear response. The former ignores the correlation between the reservoir's states before and after the application of the external field at τ. The latter is the correction due to the synthetic effect of the reservoir's correlation and the initial correlation. Note that $\phi_{BA}^{(c)}(t,\tau)$ and $\phi_{BA}^{(i,c)}(t,\tau)$ include the correlation function $\langle L_{SR}(t_j)[A^\times(\tau)]^{\times L_{SR}} L_{SR}(t_k)\rangle_R$ with $t_j > \tau > t_k$. This means that $\phi_{BA}^{(c)}(t,\tau)$ and $\phi_{BA}^{(i,2)}(t,\tau)$ become very small if the correlation time of the thermal reservoir is sufficiently small. Hence the existence of these functions is closely related to the non-Markovianity and the violation of the quantum regression theorem. This will be discussed in Sec. V.

In our approach, the reservoir variables are eliminated under the influence of the external field. If we first eliminate the reservoir variables and apply the external field after the elimination, we obtain the quantum master equation, instead of Eq. (59),

$$\frac{\partial}{\partial t} W_S(t) = \left[L_S + \frac{i}{\hbar} F(t) A^\times\right] W_S(t) + \Pi_0(t) W_S(t) + I_0(t), \tag{77}$$

which yields the linear response function $\phi'_{BA}(t,\tau) = \phi_{BA}^{(0)}(t,\tau) + \phi_{BA}^{(i,0)}(t,\tau)$. In this case, the functions $\phi_{BA}^{(c)}(t,\tau)$ and $\phi_{BA}^{(i,c)}(t,\tau)$ are missing in the linear response function. Thus the order of the elimination of the reservoir variables and the application of the external field is essential. Taking the Markovian limit and neglecting the inhomogeneous term in Eq. (77), we can reproduce the results given in Ref. [24].

C. Comparison with the Kubo formula

It is well known that the linear response function is given by the Kubo formula [1–3]. The formal solution of the Liouville–von Neumann equation, $(\partial/\partial t)W(t) = [L + L_{\rm ext}(t)]W(t)$, is given, up to the first order with respect to the external field, by

$$W(t) = e^{L(t-t_{\rm in})} W + \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau) e^{L(t-\tau)} A^\times e^{L(\tau - t_{\rm in})} W, \tag{78}$$

where L is the Liouvillian superoperator $L = -(i/\hbar)H^\times$ with $H = H_S + H_R + H_{SR}$. The effect of the external field on the average value of an observable B is obtained by calculating $\Delta\langle B(t)\rangle = \mathrm{Tr}_{SR}\{B[W(t) - e^{L(t-t_{\rm in})}W]\}$. Then the linear response function $\phi_{BA}(t,\tau)$ is defined by the relation $\Delta\langle B(t)\rangle = \int_{t_{\rm in}}^{t} d\tau\, \phi_{BA}(t,\tau)F(\tau)$, which yields

$$\phi_{BA}(t,\tau) = \frac{i}{\hbar}\langle [B(t), A(\tau)]\rangle_{SR}, \tag{79}$$

where $\langle\cdots\rangle_{SR} = \mathrm{Tr}_{SR}[\cdots W]$ is an initial average of the whole system and Tr_{SR} stands for the trace operation of the whole system. In Eq. (79), $A(\tau)$ and $B(t)$ are Heisenberg operators given by

$$A(\tau) = e^{(i/\hbar)H(\tau - t_{\rm in})} A e^{-(i/\hbar)H(\tau - t_{\rm in})},$$
$$B(t) = e^{(i/\hbar)H(t - t_{\rm in})} B e^{-(i/\hbar)H(t - t_{\rm in})}. \tag{80}$$

In the linear response theory developed by Kubo [1–3], it is assumed that $t_{\rm in} \to -\infty$ and W is the equilibrium state. In this case, the linear response function is a function of time difference $t - \tau$, namely, $\phi_{BA}(t,\tau) = \phi_{BA}(t-\tau)$. The Fourier-Laplace transformation yields a complex admittance [1–3].

To compare our result (76) with the Kubo formula (79), we rewrite the formula as follows:

$$\begin{aligned}\phi_{BA}(t,\tau) &= \frac{i}{\hbar}\mathrm{Tr}_{SR}[B e^{L(t-\tau)} A^\times e^{L(\tau - t_{\rm in})} W]\\ &= \frac{i}{\hbar}\mathrm{Tr}_{SR}[B e^{L(t-\tau)} A^\times e^{L(\tau - t_{\rm in})} W_S \rho_R]\\ &\quad + \frac{i}{\hbar}\mathrm{Tr}_{SR}[B e^{L(t-\tau)} A^\times e^{L(\tau - t_{\rm in})} \delta W],\end{aligned} \tag{81}$$

where we set $\delta W = W - W_S\rho_R$. Thus the linear response function given by the Kubo formula can be divided into four parts:

$$\begin{aligned}\phi_{BA}(t,\tau) &= \frac{i}{\hbar}\mathrm{Tr}_S[B\langle e^{L(t-\tau)}\rangle_R A^\times \langle e^{L(\tau - t_{\rm in})}\rangle_R W_S]\\ &\quad + \frac{i}{\hbar}\mathrm{Tr}_S[B\{\langle e^{L(t-\tau)} A^\times e^{L(\tau - t_{\rm in})}\rangle_R\\ &\quad - \langle e^{L(t-\tau)}\rangle_R A^\times \langle e^{L(\tau - t_{\rm in})}\rangle_R\} W_S]\\ &\quad + \frac{i}{\hbar}\mathrm{Tr}_S[B\langle e^{L(t-\tau)}\rangle_R A^\times \langle e^{L(\tau - t_{\rm in})}\rangle_{\delta W}]\\ &\quad + \frac{i}{\hbar}\mathrm{Tr}_S[B\{\langle e^{L(t-\tau)} A^\times e^{L(\tau - t_{\rm in})}\rangle_{\delta W}\\ &\quad - \langle e^{L(t-\tau)}\rangle_R A^\times \langle e^{L(\tau - t_{\rm in})}\rangle_{\delta W}\}],\end{aligned} \tag{82}$$

with $\langle\cdots\rangle_{\delta W} = \mathrm{Tr}_R[\cdots \delta W]$. It should be noted that the operator $\langle e^{L(t-t_{\rm in})}\rangle_R W_S$ satisfies the time-local quantum master equation without the inhomogeneous term. In the Born approximation, we find the equality $V(t,t_{\rm in}) = \langle e^{L(t-t_{\rm in})}\rangle_R$, where $V(t,t_{\rm in})$ is given by Eq. (60). Thus the first term on the right-hand side of Eq. (82) is equal to the function $\phi_{BA}^{(0)}(t,\tau)$ up to the second order with respect to the system-reservoir interaction. Furthermore, the second term of the right-hand side corresponds to the function $\phi_{BA}^{(c)}(t,\tau)$ since it represents the correlation between the states of the thermal reservoir before and after the external field is applied at the time τ. In the same way, it is found that the third and fourth terms correspond to the functions $\phi_{BA}^{(i,0)}(t,\tau)$ and $\phi_{BA}^{(i,c)}(t,\tau)$. Although it is very difficult to directly calculate the linear response function given by the Kubo formula for an open quantum system, we have found a systematic method for calculating the linear response function by making use of the time-local quantum master equation.

D. Simple example

As a simple example of the linear response of an open quantum system, we suppose that a thermal reservoir is modeled by a fluctuating classical field which obeys the stationary Gauss-Markov process [3,40]. Such a model has been originally proposed by Kubo [41] and Anderson [42] in order to investigate the absorption spectrum of a magnetic system. When the thermal reservoir is described by a classical stochastic process, the time evolution of the quantum system and the thermal reservoir is determined by the stochastic Liouville equation [39,43,44]. To obtain an exact solution, we assume that a relevant quantum system is a two-level system which has a dephasing coupling with the thermal reservoir. Then the total Hamiltonian of the system is given by

$$H(t) = \tfrac{1}{2}\hbar\omega\sigma_z + \tfrac{1}{2}\hbar\Omega(t)\sigma_z, \tag{83}$$

where ω is a transition frequency of the two-level system, and $\sigma_x,\sigma_y,\sigma_z$ are the Pauli matrices. In this equation, $\Omega(t)$ is a stochastic variable which obeys the stationary Gauss-Markov process with zero mean. Then the Doob theorem provides the two-time correlation function $\langle\Omega(t)\Omega(s)\rangle_R = \Delta^2 e^{-\gamma|t-s|}$ [3,40], where Δ represents a strength of the correlation and γ is its decay rate. For later convenience, we introduce

$$\Theta(t,s) = \int_s^t dt'\,\Omega(t'). \tag{84}$$

Furthermore, we assume that the observables A and B of the quantum system are given by

$$A = B = \tfrac{1}{2}(\sigma_z\cos\theta + \sigma_x\sin\theta). \tag{85}$$

Then it is easy to see that the commutation relation is given by

$$\begin{aligned}[B(t),A(\tau)] &= \tfrac{1}{2}(\sigma_+ e^{i\omega\tau+i\Theta(\tau,0)} - \sigma_- e^{-i\omega\tau-i\Theta(\tau,0)})\cos\theta\sin\theta \\ &\quad - \tfrac{1}{2}(\sigma_+ e^{i\omega t+i\Theta(t,0)} - \sigma_- e^{-i\omega t-i\Theta(t,0)})\cos\theta\sin\theta \\ &\quad + \tfrac{1}{4}\sigma_z(e^{i\omega(t-\tau)+i\Theta(t,\tau)} - e^{-i\omega(t-\tau)-i\Theta(t,\tau)})\sin^2\theta,\end{aligned} \tag{86}$$

where we set the initial time $t_{\text{in}} = 0$ and $\sigma_\pm = (\sigma_x \pm i\sigma_y)/2$. Assuming that $H_{\text{ext}}(t) = -\hbar F(t)A$, we can obtain the linear response function directly from the Kubo formula,

$$\begin{aligned}\phi_{BA}(t,\tau) &= -\tfrac{1}{2}(\langle\sigma_x\rangle\sin\omega\tau + \langle\sigma_y\rangle\cos\omega\tau)G(\tau,0)\cos\theta\sin\theta \\ &\quad + \tfrac{1}{2}(\langle\sigma_x\rangle\sin\omega t + \langle\sigma_y\rangle\cos\omega t)G(t,0)\cos\theta\sin\theta \\ &\quad - \tfrac{1}{2}\langle\sigma_z\rangle\sin\omega(t-\tau)G(t,\tau)\sin^2\theta,\end{aligned} \tag{87}$$

which is an exact result. In this equation, $G(t,\tau)$ is the characteristic function of the stochastic variable $\Omega(t)$,

$$\begin{aligned}G(t,\tau) &= \langle e^{-i\Theta(t,\tau)}\rangle_R \\ &= \exp\left\{-\left(\frac{\Delta}{\gamma}\right)^2[\gamma(t-\tau) - 1 + e^{-\gamma(t-\tau)}]\right\},\end{aligned} \tag{88}$$

where we have used the Gaussianity and the Doob theorem [3,40].

If there is no external field, the reduced time evolution of the two-level system is determined by the time-local quantum master equation [39],

$$\frac{\partial}{\partial t}W_S(t) = \left[-\frac{i}{2}\omega\sigma_z^\times - \frac{1}{4}\dot g(t)(\sigma_z^\times)^2\right]W_S(t), \tag{89}$$

where we set $\dot g(t) = dg(t)/dt$ with $g(t) = (\Delta/\gamma)^2[\gamma t - 1 + e^{-\gamma t}]$. In this case, the superoperator $V(t,s)$ is given by

$$\begin{aligned}V(t,s) &= \exp\left\{\int_s^t dt'\left[-\frac{i}{2}\omega\sigma_z^\times - \frac{1}{4}\dot g(t')(\sigma_z^\times)^2\right]\right\} \\ &= \exp\left\{-\frac{i}{2}\omega(t-s)\sigma_z^\times - \frac{1}{4}[g(t)-g(s)](\sigma_z^\times)^2\right\}.\end{aligned} \tag{90}$$

Then the function $\phi_{BA}^{(0)}(t,\tau)$ given by Eq. (68) is calculated to be

$$\begin{aligned}\phi_{BA}^{(0)}(t,\tau) &= -\tfrac{1}{2}(\langle\sigma_x\rangle\sin\omega\tau + \langle\sigma_y\rangle\cos\omega\tau)G(\tau,0)\cos\theta\sin\theta \\ &\quad + \tfrac{1}{2}(\langle\sigma_x\rangle\sin\omega t + \langle\sigma_y\rangle\cos\omega t)G(t,0)\cos\theta\sin\theta \\ &\quad - \tfrac{1}{2}\langle\sigma_z\rangle\sin\omega(t-\tau)G(t,0)G^{-1}(\tau,0)\sin^2\theta,\end{aligned} \tag{91}$$

where $\langle\sigma_\mu\rangle$ stands for the initial average of the Pauli matrix σ_μ. Here we note that $\phi_{BA}^{(i,0)}(t,\tau) = \phi_{BA}^{(i,c)}(t,\tau) = 0$ since there is no initial correlation between the two-level system and the thermal reservoir. The correction term $\phi_{BA}^{(c)}(t,\tau)$ due to the reservoir correlation is calculated by the difference $\phi_{BA}(t,\tau) - \phi_{BA}^{(0)}(t,\tau)$,

$$\begin{aligned}\phi_{BA}^{(c)}(t,\tau) &= -\tfrac{1}{2}\langle\sigma_z\rangle[G(t,\tau) - G(t,0)G^{-1}(\tau,0)] \\ &\quad \times \sin\omega(t-\tau)\sin^2\theta.\end{aligned} \tag{92}$$

This correction is an exact result. It is found from Eq. (92) that if the thermal reservoir modulates the two-level system very fast and thus the reduced time evolution is Markovian, the function $\phi_{BA}^{(c)}(t,\tau)$ becomes negligible. In fact, we can approximate the characteristic function as $G(t,\tau) \approx e^{-(t-\tau)/T_2}$ for the fast-modulation limit or, equivalently, the narrowing limit [3], where $T_2 = \gamma/\Delta^2$ is a dephasing time of the two-level system. In this case, we have $G(t,\tau) \approx G(t,0)G^{-1}(\tau,0)$, which means $\phi_{BA}^{(c)}(t,\tau) \approx 0$.

Next, using the result in Sec. III B, we derive the linear response function in the Born approximation in order to see whether or not the Born approximation works well. Although the Born approximation provides the exact result without the external field due to the Gaussianity, it does not in the presence of the external field. Since the function given by (68) has already been calculated, we derive the correction term $\phi_{BA}^{(c)}(t,\tau)$ given by Eq. (69). After some calculation, we can obtain

$$\begin{aligned}&\mathrm{Tr}_S\big[BV(t,t_1)e^{L_S t_1}\langle L_{SR}(t_1)A^\times(\tau)L_{SR}(t_2)\rangle_R e^{-L_S t_1}V(t_1,0)W_S\big] \\ &\quad - \mathrm{Tr}_S[BV(t,t_1)e^{L_S t_1}\langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R A^\times(\tau) \\ &\quad \times e^{-L_S t_1}V(t_1,0)W_S] \\ &= \frac{i}{2}\langle\sigma_z\rangle\Delta^2 e^{-\gamma(t_1-t_2)-g(t)+g(t_1)}\sin\omega(t-\tau)\sin^2\theta,\end{aligned} \tag{93}$$

where the method for calculation is the same as that given in Ref. [15]. This result yields the correction term $\phi_{BA}^{(c)}(t,\tau)$ of the linear response function in the Born approximation,

$$\begin{aligned}\phi_{BA}^{(c)}(t,\tau) &= -\frac{1}{2}\langle\sigma_z\rangle\left[\frac{\Delta^2}{\gamma}(e^{\gamma\tau}-1)e^{-g(t)}\int_\tau^t dt_1\right. \\ &\quad \left.\times e^{-\gamma t_1+g(t_1)}\right]\sin\omega(t-\tau)\sin^2\theta,\end{aligned} \tag{94}$$

which is numerically calculated. Obviously this is not equal to the exact result given by Eq. (92). In the narrowing limit

$(\Delta/\gamma)^2 \ll 1$ [3], however, we have the approximation

$$\begin{aligned} G(t,\tau) &- G(t,0)G^{-1}(\tau,0) \\ &\approx \left(\frac{\Delta}{\gamma}\right)^2 [1 + e^{-\gamma t} - e^{-\gamma\tau} - e^{-\gamma(t-\tau)}] \\ &\approx \frac{\Delta^2}{\gamma}(e^{\gamma\tau} - 1)e^{-g(t)} \int_\tau^t dt_1\, e^{-\gamma t_1 + g(t_1)}. \end{aligned} \tag{95}$$

Thus Eq. (94) becomes identical to Eq. (92) in the limit of $(\Delta/\gamma)^2 \ll 1$.

Finally, we investigate the linear response in the case of $A = B = \frac{1}{2}\sigma_x (= S_x)$ $(\theta = \pi/2)$. In this case, the exact linear response function is

$$\phi_{BA}^{(\mathrm{ex})}(t,\tau) = -\langle S_z\rangle G(t,\tau)\sin\omega(t-\tau), \tag{96}$$

with $S_z = \frac{1}{2}\sigma_z$, and the Born approximation provides the approximated linear response function,

$$\begin{aligned} \phi_{BA}^{(\mathrm{2nd})}(t,\tau) = -\langle S_z\rangle \bigg[&\frac{G(t,0)}{G(\tau,0)} + \frac{\Delta^2}{\gamma}(e^{\gamma\tau} - 1)e^{-g(t)} \\ &\times \int_\tau^t dt_1\, e^{-\gamma t_1 + g(t_1)} \bigg] \sin\omega(t-\tau). \end{aligned} \tag{97}$$

If we neglect the correlation between the reservoir's states before and after the external field is applied at the time τ, the linear response function becomes

$$\phi_{BA}^{(0)}(t,\tau) = -\langle S_z\rangle \frac{G(t,0)}{G(\tau,0)}\sin\omega(t-\tau). \tag{98}$$

When we apply an extremely short pulse to the two-level system at $t = t_p$, we obtain the linear response of S_x,

$$\Delta\langle S_x(t)\rangle_\mu = \langle S_z\rangle \phi_{BA}^{(\mu)}(t,t_p)\theta(t-t_p) \quad (\mu = \mathrm{ex,2nd,0}). \tag{99}$$

On the other hand, when we apply a monochromatic field with angular frequency ω_0 to the two-level system, the linear response of S_x is given by

$$\Delta\langle S_x(t)\rangle_\mu = \langle S_z\rangle \int_0^t d\tau\, \phi_{BA}^{(\mu)}(t,\tau)\sin\omega_0\tau \quad (\mu = \mathrm{ex,2nd,0}). \tag{100}$$

The normalized linear response $\Delta\langle S_x(t)\rangle_\mu/\langle S_z\rangle$ of the two-level system is plotted in Fig. 2. It is found from the figure that the linear response in the Born approximation is nearly equal to the exact one, though it deviates a little from the exact linear response as $(\Delta/\gamma)^2$ and t/T_2 are large. Furthermore, the linear response obtained by discarding the reservoir correlation can approximate the exact linear response only if $(\Delta/\gamma)^2$ is sufficiently small.

E. Linear response function based on the time-nonlocal master equation

We formulate the linear response theory of an open quantum system, the reduced time evolution of which is governed by the time-nonlocal quantum master equation [10–12,15,16]. In the Born approximation, the time-nonlocal quantum master equation with the external field is obtained from Eq. (15),

$$\frac{\partial}{\partial t}W_S(t) = [L_S + L_{\mathrm{ext}}(t)]W_S(t) + \int_{t_{\mathrm{in}}}^t dt_1\, \Phi^{(2)}(t,t_1)W_S(t_1) + J^{(1)}(t) + J^{(2)}(t), \tag{101}$$

with

$$\Phi^{(2)}(t,t_1) = U_S(t,t_{\mathrm{in}})\langle \hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\rangle_R U_S^{-1}(t_1,t_{\mathrm{in}}), \tag{102}$$

where we have assumed $\langle \hat{L}_{SR}(t)\rangle_R = 0$. In the Born approximation, the inhomogeneous terms are equal to those of the time-local quantum master equation, namely, $J^{(1)}(t) = I^{(1)}(t)$ and $J^{(2)}(t) = I^{(2)}(t)$, where $I^{(1)}(t)$ and $I^{(2)}(t)$ are given by Eqs. (25) and (26). Therefore, we obtain the equality $J^{(1)}(t) + J^{(2)}(t) = I_0(t) + I_1(t)$ up to the first order with respect to the external field, where $I_0(t)$ and $I_1(t)$ are given by Eqs. (56) and (57). Neglecting all the terms higher than the first order with respect to the external field, we can derive the memory kernel superoperator $\Phi^{(2)}(t,t_1)$ as follows:

$$\Phi^{(2)}(t,t_1) = \langle L_{SR}U_{SR}(t,t_1)L_{SR}U_{SR}(t_1,t_{\mathrm{in}})\rangle_R U_S^{-1}(t_1,t_{\mathrm{in}}) \equiv \Phi_0(t,t_1) + \Phi_1(t,t_1), \tag{103}$$

with

$$\Phi_0(t,t_1) = e^{L_S(t-t_{\mathrm{in}})}\langle L_{SR}(t)L_{SR}(t_1)\rangle_R e^{-L_S(t_1-t_{\mathrm{in}})}, \tag{104}$$

$$\Phi_1(t,t_1) = \frac{i}{\hbar}\int_{t_1}^t d\tau\, F(\tau)e^{L_S(t-t_{\mathrm{in}})}\langle L_{SR}(t)A^\times(\tau)L_{SR}(t_1)\rangle_R e^{-L_S(t_1-t_{\mathrm{in}})}, \tag{105}$$

where we have used Eqs. (27), (29), and (30). The first-order memory term $\Phi_1(t,t_1)$ is further calculated to be

$$\int_{t_{\mathrm{in}}}^t dt_1\, \Phi_1(t,t_1)W_S(t_1) = \frac{i}{\hbar}\int_{t_{\mathrm{in}}}^t d\tau\, F(\tau)\int_{t_{\mathrm{in}}}^\tau dt_1\, \Phi_1(t,t_1|\tau)W_S(t_1), \tag{106}$$

with

$$\Phi_1(t,t_1|\tau) = e^{L_S(t-t_{\mathrm{in}})}\langle L_{SR}(t)A^\times(\tau)L_{SR}(t_1)\rangle_R e^{-L_S(t_1-t_{\mathrm{in}})}. \tag{107}$$

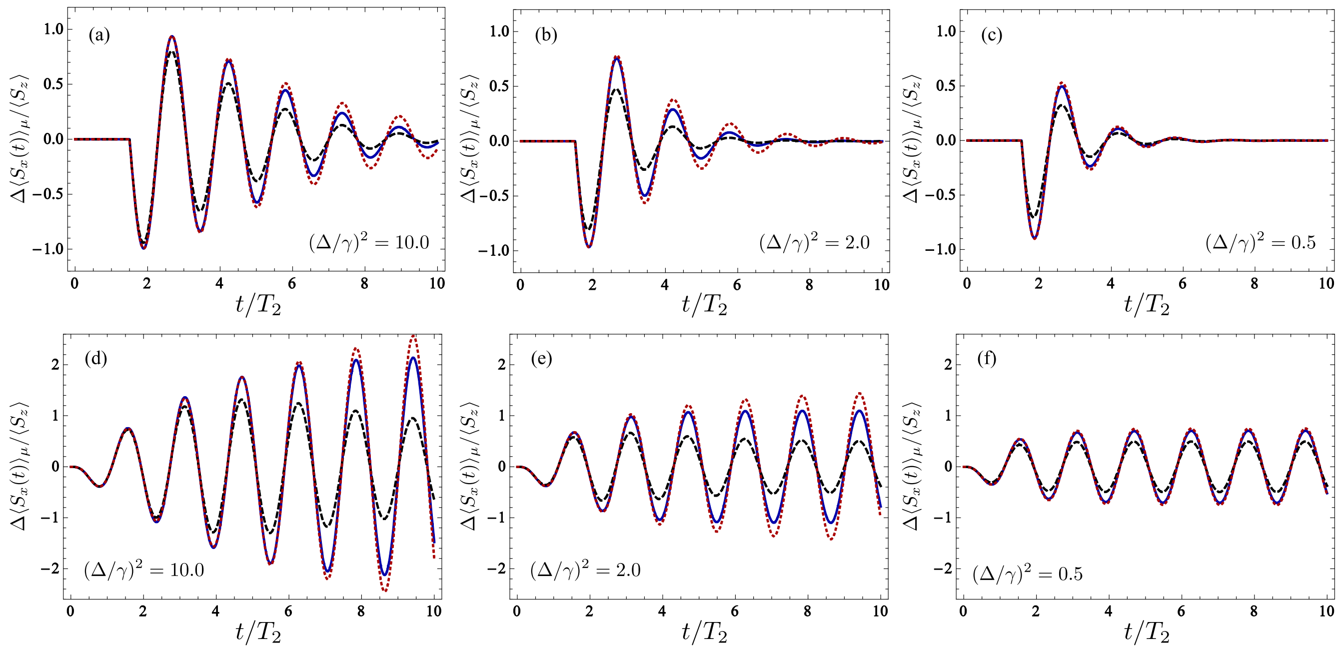

FIG. 2. The normalized linear response $\Delta\langle S_x(t)\rangle_\mu/\langle S_z\rangle$ of the two-level system influenced by the fluctuating environment, where the (a)–(c) short pulse and (d)–(f) monochromatic field are applied to the system. The blue solid line stands for the exact linear response, the red dotted line for the linear response in the Born approximation, and the black dashed line for the linear response obtained by neglecting the reservoir correlation. In the figure, we set $t_p/T_2 = 1.5$ and $\omega T_2 = \omega_0 T_2 = 4.0$, where $T_2 = \gamma/\Delta^2$ is a dephasing time of the two-level system. (a), (d) The two-level system is slowly modulated by the thermal reservoir; (c), (f) it is modulated very fast.

Therefore, up to the first order with respect to the external field $F(\tau)$, we obtain the time-nonlocal quantum master equation in the Born approximation,

$$\frac{\partial}{\partial t}W_S(t) = L_S W_S(t) + \int_{t_{\rm in}}^t dt_1\,\Phi_0(t,t_1)W_S(t_1) + I_0(t) + \frac{i}{\hbar}F(t)A^\times W_S(t) + \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\,F(\tau)\int_{t_{\rm in}}^\tau dt_1\,\Phi_1(t,t_1|\tau)W_S(t_1) + \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\,F(\tau)\mathcal{I}(t,\tau), \tag{108}$$

where $\mathcal{I}(t,\tau)$ is given by Eq. (58). The first three terms on the right-hand side of this equation correspond to the time-nonlocal quantum master equation in the absence of the external field and the others are the first-order correction due to the external field.

To derive the linear response function, we assume that the time-nonlocal quantum master equation can be solved when the inhomogeneous term is ignored and the external field is not applied. Then we define the superoperator $\bar{V}(t,t_{\rm in})$ by

$$\frac{\partial}{\partial t}\bar{V}(t,t_{\rm in}) = L_S\bar{V}(t,t_{\rm in}) + \int_{t_{\rm in}}^t dt_1\,\Phi_0(t,t_1)\bar{V}(t_1,t_{\rm in}), \tag{109}$$

with $\bar{V}(t_{\rm in},t_{\rm in}) = 1$. We denote the reduced density operator of the quantum system as $W_S(t) = W_{S,0}(t) + W_{S,1}(t)$, where $W_{S,0}(t)$ and $W_{S,1}(t)$ are the zeroth- and first-order terms with respect to the external field, which are determined by

$$\frac{\partial}{\partial t}W_{S,0}(t) = L_S W_{S,0}(t) + \int_{t_{\rm in}}^t dt_1\,\Phi_0(t,t_1)W_{S,0}(t_1) + I_0(t), \tag{110}$$

$$\frac{\partial}{\partial t}W_{S,1}(t) = L_S W_{S,1}(t) + \int_{t_{\rm in}}^t dt_1\,\Phi_0(t,t_1)W_{S,1}(t_1) + \frac{i}{\hbar}F(t)A^\times W_{S,0}(t) + \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\,F(\tau)\int_{t_{\rm in}}^\tau dt_1\,\Phi_1(t,t_1|\tau)W_{S,0}(t_1) + \frac{i}{\hbar}\int_{t_{\rm in}}^t d\tau\,F(\tau)\mathcal{I}(t,\tau). \tag{111}$$

Furthermore, we assume that the operator $\bar{V}(t,t_{\rm in})$ defined by Eq. (109) is invertible, though this assumption is not always true. Then we obtain, from Eq. (110),

$$W_{S,0}(t) = \bar{V}(t,t_{\rm in})W_S + \int_{t_{\rm in}}^t dt_1\,\bar{V}(t,t_1)I_0(t_1), \tag{112}$$

where we have defined $\bar{V}(t,s)$ by $\bar{V}(t,s) = \bar{V}(t,t_{\rm in})\bar{V}^{-1}(s,t_{\rm in})$ $(t > s)$. Substituting this equation into Eq. (111) and discarding all the terms higher than the first order with respect to the external field, we can obtain

$$W_{S,1}(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\bar{V}(t,\tau)A^{\times}\bar{V}(\tau,t_{\rm in})W_S + \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2\, \bar{V}(t,t_1)\Phi_1(t_1,t_2|\tau)\bar{V}(t_2,t_{\rm in})W_S$$
$$+ \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{t_{\rm in}}^{\tau} dt_1\, \bar{V}(t,\tau)A^{\times}\bar{V}(\tau,t_1)I_0(t_1) + \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, \bar{V}(t,t_1)\Phi_1(t_1,t_2|\tau)\bar{V}(t_2,t_3)I_0(t_3)$$
$$+ \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{\tau}^{t} dt_1\, \bar{V}(t,t_1)\mathcal{I}(t_1,\tau). \tag{113}$$

In deriving this equation, we have rearranged the order of integrations so that the integration including the external field $F(\tau)$ is carried out last.

The linear response $\Delta\langle B(t)\rangle$ of an observable B is calculated by $\Delta\langle B(t)\rangle = {\rm Tr}_S[BW_{S,1}(t)]$, which can be expressed as

$$\Delta\langle B(t)\rangle = \int_{t_{\rm in}}^{t} d\tau\, \bar{\phi}_{BA}(t,\tau)F(\tau). \tag{114}$$

Therefore, when the reduced time evolution of the quantum system is determined by the time-nonlocal quantum master equation, the linear response function $\bar{\phi}_{BA}(t,\tau)$ is given by

$$\bar{\phi}_{BA}(t,\tau) = \bar{\phi}_{AB}^{(0)}(t,\tau) + \bar{\phi}_{AB}^{(c)}(t,\tau) + \bar{\phi}_{BA}^{(i,0)}(t,\tau) + \bar{\phi}_{BA}^{(i,c)}(t,\tau), \tag{115}$$

with

$$\bar{\phi}_{BA}^{(0)}(t,\tau) = \frac{i}{\hbar}{\rm Tr}_S[B\bar{V}(t,\tau)A^{\times}\bar{V}(\tau,t_{\rm in})W_S], \tag{116}$$

$$\bar{\phi}_{BA}^{(c)}(t,\tau) = \frac{i}{\hbar}\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2\, {\rm Tr}_S[B\bar{V}(t,t_1)\Phi_1(t_1,t_2|\tau)\bar{V}(t_2,t_{\rm in})W_S], \tag{117}$$

$$\bar{\phi}_{BA}^{(i,0)}(t,\tau) = \frac{i}{\hbar}\int_{t_{\rm in}}^{\tau} dt_1\, {\rm Tr}_S[B\bar{V}(t,\tau)A^{\times}\bar{V}(\tau,t_1)I_0(t_1)], \tag{118}$$

$$\bar{\phi}_{BA}^{(i,c)}(t,\tau) = \frac{i}{\hbar}\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, {\rm Tr}_S[B\bar{V}(t,t_1)\Phi_1(t_1,t_2|\tau)\bar{V}(t_2,t_3)I_0(t_3)] + \frac{i}{\hbar}\int_{\tau}^{t} dt_1\, {\rm Tr}_S[B\bar{V}(t,t_1)\mathcal{I}(t_1,\tau)], \tag{119}$$

where $\Phi_1(t_1,t_2|\tau)$ is given by Eq. (107). The meaning of each part of the linear response function is the same as that obtained by making use of the time-local quantum master equation.

IV. LINEAR RESPONSE BEYOND THE BORN APPROXIMATION

In Sec. III, we have developed the linear response theory, using the time-local and time-nonlocal quantum master equations in the Born approximation which is valid only if the interaction between the quantum system and the thermal reservoir is sufficiently weak. In this section, using the general form of the quantum master equation, we extend the linear response theory beyond the Born approximation. We assume that there is no initial correlation between the quantum system and the thermal reservoir to avoid mathematical complexity. Hence the inhomogeneous terms do not appear in the quantum master equations. However, the method for deriving the linear response function is still valid even in the presence of the initial correlation.

A. Linear response function based on the time-local quantum master equation

It is well known that the time-local quantum master equation can be expanded in terms of time-ordered cumulants of the Liouvillian superoperators $\hat{L}_{SR}(t)$ [10,13–16],

$$\frac{\partial}{\partial t}W_S(t) = [L_S + L_{\rm ext}(t)]W_S(t) + U_S(t,t_{\rm in})\langle\hat{L}_{SR}(t)\rangle_R U_S^{-1}(t,t_{\rm in})W_S(t)$$
$$+ \sum_{n=2}^{\infty}\int_{t_{\rm in}}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \cdots \int_{t_{\rm in}}^{t_{n-2}} dt_{n-1} U_S(t,t_{\rm in})\langle\hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\cdots\hat{L}_{SR}(t_{n-1})\rangle_R^{\rm o.c.}U_S^{-1}(t,t_{\rm in})W_S(t), \tag{120}$$

where $\langle\cdots\rangle_R^{\rm o.c.}$ stands for the time-ordered cumulant [10,13–16] and we set $t_0 = t$. In this equation, we have not assumed the equality $\langle\hat{L}_{SR}(t)\rangle_R = 0$. To obtain the linear response function, we have to extract the terms up to the first order with respect to the external field from Eq. (120). For this purpose, we expand the superoperators $U_{SR}(t,t_{\rm in})$ and $U_S(t,t_{\rm in})$ as follows:

$$e^{-L_0(t_j-t_{\rm in})}U_{SR}(t_j,t_k)e^{L_0(t_k-t_{\rm in})} = 1 + \Delta u(t_j,t_k), \tag{121}$$

$$U_S^{-1}(t,t_{\rm in})e^{L_S(t-t_{\rm in})} = 1 - \Delta u(t,t_{\rm in}), \tag{122}$$

with

$$\Delta u(t_j,t_k) = \frac{i}{\hbar}\int_{t_k}^{t_j} d\tau\, F(\tau)A^{\times}(\tau). \tag{123}$$

In deriving these equations, we have discarded all the terms higher than the first order with respect to $F(\tau)$. Substituting Eqs. (121) and (122) into Eq. (120), we can derive the linear response function in any order with respect to the system-reservoir interaction.

It is easy to see from Eqs. (121) and (122) that the first moment M_1 does not depend on the external field up to the first order. In fact, we obtain

$$\begin{aligned} M_1 &= U_S(t,t_{\rm in})\langle \hat{L}_{SR}(t)\rangle_R U_S^{-1}(t,t_{\rm in}) \\ &= e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)[1+\Delta u(t,t_{\rm in})]\rangle_R[1-\Delta u(t,t_{\rm in})]e^{-L_S(t-t_{\rm in})} \\ &= e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)\rangle_R[1+\Delta u(t,t_{\rm in})][1-\Delta u(t,t_{\rm in})]e^{-L_S(t-t_{\rm in})} \\ &= e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)\rangle_R e^{-L_S(t-t_{\rm in})}, \end{aligned} \tag{124}$$

where we have used Eq. (27) and $L_{SR}(t)$ is given by Eq. (38). In the same way, we can calculate the nth moment up to the first order with respect to the external field,

$$\begin{aligned} M_n &= U_S(t,t_{\rm in})\langle \hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\hat{L}_{SR}(t_2)\cdots\hat{L}_{SR}(t_{n-1})\rangle_R U_S^{-1}(t,t_{\rm in}) \\ &= e^{L_S(t-t_{\rm in})}\{\langle L_{SR}(t)e^{-L_0(t-t_{\rm in})}U(t,t_1)e^{L_0(t_1-t_{\rm in})}L_{SR}(t_1) \\ &\quad\times e^{-L_0(t_1-t_{\rm in})}U(t_1,t_2)e^{L_0(t_2-t_{\rm in})}L_{SR}(t_2)\times\cdots \\ &\quad\times e^{-L_0(t_{n-2}-t_{\rm in})}U(t_{n-2},t_{n-1})e^{L_0(t_{n-1}-t_{\rm in})}L_{SR}(t_{n-1})\times\cdots \\ &\quad\times e^{-L_0(t_{n-1}-t_{\rm in})}U(t_{n-1},t_{\rm in})\rangle_R U_S^{-1}(t,t_{\rm in})e^{L_S(t-t_{\rm in})}\}e^{-L_S(t-t_{\rm in})} \\ &= e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\rangle_R e^{-L_S(t-t_{\rm in})} \\ &\quad+ e^{L_S(t-t_{\rm in})}\{\langle L_{SR}(t)\Delta u(t,t_1)L_{SR}(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\rangle_R \\ &\quad+ \langle L_{SR}(t)L_{SR}(t_1)\Delta u(t_1,t_2)L_{SR}(t_2)\cdots L_{SR}SR(t_{n-1})\rangle_R \\ &\quad+ \langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\Delta u(t_2,t_3)\cdots L_{SR}(t_{n-1})\rangle_R \\ &\quad+ \langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\cdots\Delta u(t_{n-2},t_{n-1})L_{SR}(t_{n-1})\rangle_R \\ &\quad+ \langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\Delta u(t_{n-1},t_{\rm in})\rangle_R \\ &\quad- \langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\rangle_R\Delta u(t,t_{\rm in})\}e^{-L_S(t-t_{\rm in})}. \end{aligned} \tag{125}$$

To proceed further, we decompose $\Delta u(t,t_{\rm in})$ into

$$\Delta u(t,t_{\rm in}) = \Delta u(t,t_1) + \Delta u(t_1,t_2) + \cdots + \Delta u(t_{n-1},t_{\rm in}), \tag{126}$$

and we use the fact that the equality $\langle\bullet\rangle_R\Delta u(t,t_{\rm in}) = \langle\bullet\Delta u(t,t_{\rm in})\rangle_R$ holds since $\Delta u(t,t_{\rm in})$ is a system operator. Then we can express Eq. (125) in a concise form,

$$\begin{aligned} M_n &= e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\rangle_R e^{-L_S(t-t_{\rm in})} \\ &\quad+ e^{L_S(t-t_{\rm in})}\left[\sum_{k=1}^{n-1}\mathcal{R}_{k-1,k}\langle L_{SR}(t)L_{SR}(t_1)\cdots L_{SR}(t_{n-1})\rangle_R\right]e^{-L_S(t-t_{\rm in})}. \end{aligned} \tag{127}$$

In this equation, the symbol $\mathcal{R}_{j,k}$ means that the product $L_{SR}(t_j)L_{SR}(t_k)$ is replaced by $L_{SR}(t_j)[\Delta u(t_j,t_k)]^{\times L_{SR}}L_{SR}(t_k)$. For instance, we have

$$\mathcal{R}_{k-1,k}\langle L_{SR}(t)L_{SR}(t_1)\cdots L_{SR}(t_{n-1})\rangle_R = \langle L_{SR}(t)\cdots L_{SR}(t_{k-1})[\Delta u(t_{k-1},t_k)]^{\times L_{SR}}L_{SR}(t_k)\cdots L_{SR}(t_{n-1})\rangle_R, \tag{128}$$

where the symbol $X^{\times L_{SR}}$ is explained below Eq. (45). Using the same method that we have used to derive Eq. (127), we can obtain, for a product of the two moments,

$$\begin{aligned} &U_S(t,t_{\rm in})\langle \hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\cdots\hat{L}_{SR}(t_{m-1})\rangle_R\langle\hat{L}_{SR}(t_\ell)\cdots\hat{L}_{SR}(t_{\ell+n-1})\rangle_R U_S^{-1}(t,t_{\rm in}) \\ &\quad= e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)\cdots L_{SR}(t_{m-1})\rangle_R\langle L_{SR}(t_\ell)\cdots L_{SR}(t_{\ell+n-1})\rangle_R e^{-L_S(t-t_{\rm in})} \end{aligned}$$

$$+ e^{L_S(t-t_{\rm in})}\Bigg[\sum_{k=1}^{m-1}\mathcal{R}_{k-1,k}\langle L_{SR}(t)L_{SR}(t_1)\cdots L_{SR}(t_{m-1})\rangle_R\langle L_{SR}(t_\ell)\cdots L_{SR}(t_{\ell+n-1})\rangle_R$$
$$+\mathcal{R}_{m-1,\ell}\langle L_{SR}(t)L_{SR}(t_1)\cdots L_{SR}(t_{m-1})\rangle_R\langle L_{SR}(t_\ell)\cdots L_{SR}(t_{\ell+n-1})\rangle_R$$
$$+\sum_{k=\ell+1}^{\ell+n-1}\mathcal{R}_{k-1,k}\langle L_{SR}(t)L_{SR}(t_1)\cdots L_{SR}(t_{m-1})\rangle_R\langle L_{SR}(t_\ell)\cdots L_{SR}(t_{\ell+n-1})\rangle_R\Bigg]e^{-L_S(t-t_{\rm in})}, \tag{129}$$

where we have discarded all the terms higher than the first order of $F(\tau)$. Finally, it is found that the nth time-ordered cumulant of the Liouvillian superoperators $\hat{L}_{SR}(t)$ can be expressed, up to the first order with respect to the external field, by

$$U_S(t,t_{\rm in})\langle\hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\cdots\hat{L}_{SR}(t_{n-1})\rangle_R^{\rm o.c.}\hat{U}_S^{-1}(t,t_{\rm in}) = e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_1(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\rangle_R^{\rm o.c.}e^{-L_S(t-t_{\rm in})}$$
$$+e^{L_S(t-t_{\rm in})}\hat{\mathcal{R}}\big[\langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\rangle_R^{\rm o.c.}\big]e^{-L_S(t-t_{\rm in})}. \tag{130}$$

In this equation, the symbol $\hat{\mathcal{R}}$ means as follows: First one successive pair $L_{SR}(t_j)L_{SR}(t_k)$ is replaced by $L_{SR}(t_j)[\Delta u(t_j,t_k)]^{\times L_{SR}}L_{SR}(t_k)$, regardless of the partial average over the thermal reservoir, and then the summation is taken over all the possible successive pairs.

Summarizing the above results, we obtain the time-local quantum master equation up to the first order with respect to the external field,

$$\frac{\partial}{\partial t}W_S(t) = \left[L_S + \frac{i}{\hbar}F(t)A^\times\right]W_S(t) + \Pi_0(t)W_S(t) + \Pi_1(t)W_S(t), \tag{131}$$

with

$$\Pi_0(t) = e^{L_0(t-t_{\rm in})}\langle L_{SR}(t)\rangle_R e^{-L_0(t-t_{\rm in})} + \sum_{n=2}^{\infty}\int_{t_{\rm in}}^{t}dt_1\int_{t_{\rm in}}^{t_1}dt_2\cdots\int_{t_{\rm in}}^{t_{n-2}}dt_{n-1}$$
$$\times e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\rangle_R^{\rm o.c.}e^{-L_S(t-t_{\rm in})}, \tag{132}$$

$$\Pi_1(t) = \sum_{n=2}^{\infty}\int_{t_{\rm in}}^{t}dt_1\int_{t_{\rm in}}^{t_1}dt_2\cdots\int_{t_{\rm in}}^{t_{n-2}}dt_{n-1}e^{L_S(t-t_{\rm in})}\hat{\mathcal{R}}\big[\langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)\cdots L_{SR}(t_{n-1})\rangle_R^{\rm o.c.}\big]e^{-L_S(t-t_{\rm in})}, \tag{133}$$

where $\Pi_0(t)$ is the damping operator of the quantum master equation in the absence of the external field and $\Pi_1(t)$ is the first-order correction to the damping operator. The solution of the quantum master equation (131) up to the first order of the external field $F(\tau)$ is given by

$$W_S(t) = V(t,t_{\rm in})W_S + \Delta W_S(t), \tag{134}$$

with

$$\Delta W_S(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^{t}d\tau\ F(\tau)V(t,\tau)A^\times V(\tau,t_{\rm in})W_S + \int_{t_{\rm in}}^{t}dt'\ V(t,t')\Pi_1(t')V(t',t_{\rm in})W_S, \tag{135}$$

$$V(t,s) = \exp_{\leftarrow}\left\{\int_s^t dt'\ [L_S + \Pi_0(t')]\right\}. \tag{136}$$

The linear response function of the quantum system can be derived from the correction term $\Delta W_S(t)$.

The change of the average value of an observable B caused by the external field is calculated to be

$$\Delta\langle B(t)\rangle = {\rm Tr}_S[B\Delta W_S(t)] = \int_{t_{\rm in}}^{t}d\tau\ \phi_{BA}^{(0)}(t,\tau)F(\tau) + C_{BA}(t). \tag{137}$$

The first term on the right-hand side of this equation represents the linear response when we ignore the correlation between the states of the thermal reservoir before and after the external field is applied at time τ. The linear response function $\phi_{BA}^{(0)}(t,\tau)$ is given by

$$\phi_{BA}^{(0)}(t,\tau) = \frac{i}{\hbar}{\rm Tr}_S[BV(t,\tau)A^\times V(\tau,t_{\rm in})W_S]. \tag{138}$$

The second term provides the correction to the response function due to the reservoir's correlation,

$$C_{BA}(t) = \int_{t_{\rm in}}^{t}dt'\ {\rm Tr}_S[BV(t,t')\Pi_1(t')V(t',t_{\rm in})W_S]. \tag{139}$$

Rearranging the order of integrations so that the integration including the external field $F(\tau)$ is performed last, we can express the correction term as

$$C_{BA}(t) = \int_{t_{\rm in}}^{t} d\tau\, \phi_{BA}^{(c)}(t,\tau)F(\tau), \tag{140}$$

where $\phi_{BA}^{(c)}(t,\tau)$ is the correction term to the linear response function $\phi_{BA}^{(0)}(t,\tau)$. The correction term derived from the second time-ordered cumulant has already been given in Sec. III [see Eq. (69)]. In the Appendix, we will obtain the correction term from the fourth time-ordered cumulant.

B. Linear response function based on the time-nonlocal quantum master equation

Next we obtain the linear response function when the reduced time evolution of the quantum system is determined by the time-nonlocal quantum master equation [10–12,15,16]. Since we assume that there is no inhomogeneous term, the general form of the time-nonlocal quantum master equation is

$$\frac{\partial}{\partial t}W_S(t) = [L_S + L_{\rm ext}(t)]W_S(t) + e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)\rangle_R e^{-L_S(t-t_{\rm in})}W_S(t) + \int_{t_{\rm in}}^{t} dt'\, \Phi(t,t')W_S(t'), \tag{141}$$

where we have used Eq. (124). The memory term can be expressed in terms of the partial cumulants of the Liouvillian superoperators $\hat{L}_{SR}(t)$ [15,16],

$$\int_{t_{\rm in}}^{t} dt'\, \Phi(t,t')W_S(t') = \sum_{n=2}^{\infty}\int_{t_{\rm in}}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \cdots \int_{t_{\rm in}}^{t_{n-2}} dt_{n-1} U_S(t,t_{\rm in})\langle \hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\cdots \hat{L}_{SR}(t_{n-1})\rangle_R^{\rm p.c.} U_S^{-1}(t_{n-1},t_{\rm in})W_S(t_{n-1}). \tag{142}$$

The partial cumulant is expressed in terms of the projection operator P as

$$\langle \hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\cdots \hat{L}_{SR}(t_{n-1})\rangle_R^{\rm p.c.} = \langle \hat{L}_{SR}(t)(1-P)\hat{L}_{SR}(t_1)(1-P)\cdots(1-P)\hat{L}_{SR}(t_{n-1})\rangle_R. \tag{143}$$

Here we note that the Liouvillian superoperators are placed in order of $\hat{L}_{SR}(t)$, $\hat{L}_{SR}(t_1)$, ..., $\hat{L}_{SR}(t_{n-1})$ in the partial cumulant. This fact makes the extraction of the first-order terms from the partial cumulants much easier than that from the time-ordered cumulants. Then, using Eqs. (27), (121) and (122), we calculate the partial cumulant up to the first order with respect to the external field,

$$\begin{aligned}
&U_S(t,t_{\rm in})\langle \hat{L}_{SR}(t)\hat{L}_{SR}(t_1)\cdots \hat{L}_{SR}(t_{n-1})\rangle_S^{\rm p.c.} U_S^{-1}(t_{n-1},t_{\rm in})\\
&= e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)[1+\Delta u(t,t_1)]L_{SR}(t_1)[1+\Delta u(t_1,t_2)]\cdots[1+\Delta u(t_{n-2},t_{n-1})]L_{SR}(t_{n-1})[1+\Delta u(t_{n-1},t_{\rm in})]\rangle_R^{\rm p.c.}\\
&\quad\times[1-\Delta u(t_{n-1},t_{\rm in})]e^{-L_S(t_{n-1}-t_{\rm in})}\\
&= e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)\cdots L_{SR}(t_{n-1})\rangle_R^{\rm p.c.} e^{-L_S(t_{n-1}-t_{\rm in})}\\
&\quad+\sum_{k=1}^{n-1} e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)\cdots L(t_{k-1})\Delta u(t_{k-1},t_k)L_{SR}(t_k)\cdots L_{SR}(t_{n-1})\rangle_R^{\rm p.c.} e^{-L_S(t_{n-1}-t_{\rm in})},
\end{aligned} \tag{144}$$

with $t_0 = t$. Substituting this equation into Eq. (142), we can derive the time-nonlocal quantum master equation up to the first order with respect to the external field,

$$\begin{aligned}
\frac{\partial}{\partial t}W_S(t) = &\left[L_S + \frac{i}{\hbar}F(t)A^{\times}\right]W_S(t) + e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)\rangle_R e^{-L_S(t-t_{\rm in})}W_S(t)\\
&+\int_{t_{\rm in}}^{t} dt'\, \Phi_0(t,t')W_S(t') + \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{t_{\rm in}}^{t} dt'\, \Phi_1(t,t'|\tau)W_S(t'),
\end{aligned} \tag{145}$$

where the two memory terms are, respectively, given by

$$\int_{t_{\rm in}}^{t} dt'\, \Phi_0(t,t')W_S(t') = \sum_{n=2}^{\infty}\int_{t_{\rm in}}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \cdots \int_{t_{\rm in}}^{t_{n-2}} dt_{n-1}\, e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)\cdots L_{SR}(t_{n-1})\rangle_R^{\rm p.c.} e^{-L_S(t_{n-1}-t_{\rm in})}W_S(t_{n-1}), \tag{146}$$

and

$$\begin{aligned}
\int_{t_{\rm in}}^{t} dt'\, \Phi_1(t,t'|\tau)W_S(t') = &\sum_{n=2}^{\infty}\sum_{k=1}^{n-1}\int_{\tau}^{t} dt_1 \int_{\tau}^{t_1} dt_2 \cdots \int_{\tau}^{t_{k-2}} dt_{k-1}\int_{t_{\rm in}}^{\tau} dt_k \int_{t_{\rm in}}^{t_k} dt_{k+1}\cdots\int_{t_{\rm in}}^{t_{n-2}} dt_{n-1}\\
&\times e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)\cdots L(t_{k-1})A^{\times}(\tau)L_{SR}(t_k)\cdots L_{SR}(t_{n-1})\rangle_R^{\rm p.c.} e^{-L_S(t_{n-1}-t_{\rm in})}W_S(t_{n-1})
\end{aligned}$$

$$= \int_{t_{\rm in}}^{\tau} dt_1\, e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)A^{\times}(\tau)L_{SR}(t_1)\rangle_R^{\rm p.c.}e^{-L_S(t_1-t_{\rm in})}W_S(t_1)$$
$$+ \int_{t_{\rm in}}^{\tau} dt_1 \int_{t_{\rm in}}^{t_1} dt_2\, e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)A^{\times}(\tau)L_{SR}(t_1)L_{SR}(t_2)\rangle_R^{\rm p.c.}e^{-L_S(t_2-t_{\rm in})}W_S(t_2)$$
$$+ \int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2\, e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)A^{\times}(\tau)L_{SR}(t_2)\rangle_R^{\rm p.c.}e^{-L_S(t_2-t_{\rm in})}W_S(t_2) + \cdots . \quad (147)$$

In deriving Eq. (147), we have rearranged the order of integrations so that the integration including the external field $F(\tau)$ is performed last.

In order to obtain the linear response function when the reduced time evolution is described by the time-nonlocal quantum master equation, we assume that the time-nonlocal quantum master equation can be solved in the absence of the external field. Then we define the operator $\bar{V}(t,t_{\rm in})$ by

$$\frac{\partial}{\partial t}\bar{V}(t,t_{\rm in}) = \left[L_S + e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)\rangle_R e^{-L_S(t-t_{\rm in})}\right]\bar{V}(t,t_{\rm in}) + \int_{t_{\rm in}}^{t} dt'\, \Phi_0(t,t')\bar{V}(t',t_{\rm in}), \quad (148)$$

with the initial condition $\bar{V}(t_{\rm in},t_{\rm in}) = 1$. Furthermore, we assume that the superoperator $\bar{V}(t,t_{\rm in})$ is invertible. Then up to the first order with respect to the external field, the solution of the time-nonlocal quantum master equation (145) is given by

$$W_S(t) = \bar{V}(t,t_{\rm in})W_S + \Delta W_S(t), \quad (149)$$

with

$$\Delta W_S(t) = \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\bar{V}(t,\tau)A^{\times}\bar{V}(\tau,t_{\rm in})W_S + \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2\, \bar{V}(t,t_1)\Phi_1(t_1,t_2|\tau)\bar{V}(t_2,t_{\rm in})W_S. \quad (150)$$

In this equation, we set $\bar{V}(t,s) = \bar{V}(t,t_{\rm in})\bar{V}^{-1}(s,t_{\rm in})$ $(t \geqslant s)$. The linear response function is derived by calculating the change of the average value $\Delta\langle B(t)\rangle = {\rm Tr}_S[B\Delta W_S(t)]$,

$$\Delta\langle B(t)\rangle = \int_{t_{\rm in}}^{t} d\tau\, \bar{\phi}_{BA}(t,\tau)F(\tau). \quad (151)$$

The response function $\bar{\phi}_{BA}(t,\tau) = \bar{\phi}_{BA}^{(0)}(t,\tau) + \bar{\phi}_{BA}^{(c)}(t,\tau)$ consists of two part. One represents the linear response that is obtained when the correlation of the reservoir's states is discarded,

$$\bar{\phi}_{BA}^{(0)}(t,\tau) = \frac{i}{\hbar}{\rm Tr}_S[B\bar{V}(t,\tau)A^{\times}\bar{V}(\tau,t_{\rm in})W_S], \quad (152)$$

and the other is the correction term due to the correlation,

$$\bar{\phi}_{BA}^{(c)}(t,\tau) = \frac{i}{\hbar}\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2\, {\rm Tr}_S[B\bar{V}(t,t_1)\Phi_1(t_1,t_2|\tau)\bar{V}(t_2,t_{\rm in})W_S]. \quad (153)$$

In this section, we have assumed that there is no initial correlation between the quantum system and the thermal reservoir. Thus we have treated the homogeneous quantum master equation in the derivation of the linear response function. However, our method is still useful even if there are inhomogeneous terms in the quantum master equation since the inhomogeneous term can be expanded into a combination of the partial averages such as

$$\left\langle \hat{L}_{SR}(t_i)\cdots\hat{L}_{RS}(t_j)\rangle_R \cdots \langle \hat{L}_{SR}(t_k)\cdots\hat{L}_{RS}(t_\ell)\rangle_R \langle \hat{L}_{SR}(t_m)\cdots\hat{L}_{RS}(t_n)\right\rangle_R',$$

with $\langle\bullet\rangle_R = {\rm Tr}_R[\bullet\rho_R]$ and $\langle\bullet\rangle_R' = {\rm Tr}_R[\bullet(W - W_S\rho_R)]$. The structure is similar to that of the time-ordered or partial cumulants. Therefore, our method developed in this section can be used for extracting the first-order terms from the inhomogeneous terms.

V. CORRELATION FUNCTION AND QUANTUM REGRESSION THEOREM

In Secs. III and IV, we have developed the linear response theory for open quantum systems when the reduced time evolution is described by the time-local and time-nonlocal quantum master equations. In this section, using the method for deriving the linear response function, we derive an explicit expression of a two-time correlation function of an open quantum system. Furthermore, using the result, we discuss the violation of the quantum regression theorem for open quantum systems in the Born approximation [28–37].

A two-time correlation function of observables A and B of an open quantum system is given by $\langle B(t)A(\tau)\rangle = {\rm Tr}_{SR}[B(t)A(\tau)W]$ with $t \geqslant \tau$, where $A(\tau)$ and $B(t)$ are the Heisenberg operators given by $A(\tau) = e^{(i/\hbar)H(\tau-t_{\rm in})}Ae^{-(i/\hbar)H(\tau-t_{\rm in})}$ and $B(t) = e^{(i/\hbar)H(t-t_{\rm in})}Be^{-(i/\hbar)H(t-t_{\rm in})}$ with $H = H_S + H_R + H_{SR}$. Then, using the Liouvillian superoperator $L = -(i/\hbar)H^{\times}$, we can rewrite the correlation function into the following form:

$$\langle B(t)A(\tau)\rangle = \mathrm{Tr}_S[BW_S(t,\tau|A)], \tag{154}$$

with

$$W_S(t,\tau|A) = \mathrm{Tr}_R[e^{L(t-\tau)}Ae^{L(\tau-t_{\rm in})}W]. \tag{155}$$

Here we introduce the nonunitary superoperator $U(t,t_{\rm in}|A)$ by

$$U(t,t_{\rm in}|A) = \exp_{\leftarrow}\left\{\int_{t_{\rm in}}^{t} d\tau\,[L + g(\tau)A]\right\}, \tag{156}$$

where $g(t)$ is a test function of time t. Setting $W_S^{(g)}(t|A) = \mathrm{Tr}_R[U(t,t_{\rm in}|A)W]$ and expanding it with respect to the test function $g(t)$, we can derive

$$W_S^{(g)}(t|A) = \mathrm{Tr}_R[e^{L(t-t_{\rm in})}W] + \int_{t_{\rm in}}^{t} d\tau\, g(\tau)\mathrm{Tr}_R[e^{L(t-\tau)}Ae^{L(\tau-t_{\rm in})}W] + O(g^2). \tag{157}$$

Multiplying the system observable B from the left and taking the trace over the Hilbert space of the quantum system, we obtain

$$\mathrm{Tr}_S\big[BW_S^{(g)}(t|A)\big] = \langle B(t)\rangle + \int_{t_{\rm in}}^{t} d\tau\, g(\tau)\langle B(t)A(\tau)\rangle + O(g^2), \tag{158}$$

which means that the linear term with respect to the test function yields the two-time correlation function. More explicitly, we have the relation

$$\frac{\delta}{\delta g(\tau)}\mathrm{Tr}_S\big[BW_S^{(g)}(t|A)\big]\bigg|_{g(t)\to 0} = \langle B(t)A(\tau)\rangle. \tag{159}$$

We can see that $U(t,t_{\rm in}|A)$ is a time-evolution operator of the whole system when a fictitious field $g(t)$ is applied to the quantum system, where the coupling between the system and field is given by a system operator A. Then the two-time correlation function $\langle B(t)A(\tau)\rangle$ represents the linear response of the quantum system to the fictitious field. It is obvious that if we replace $(i/\hbar)F(\tau)A^{\times}$ by $g(\tau)A$ in the linear response theory, the linear response function $\phi_{BA}(t,\tau)$ becomes the two-time correlation function $\langle B(t)A(\tau)\rangle$. Although the replacement makes the time-evolution superoperators such as $U_{SR}(t,t_{\rm in})$ and $U_S(t,t_{\rm in})$ nonunitary, we have not used the unitarity in the derivation of the linear response function. Therefore, the results are still valid under the replacement.

When the time-local quantum master equation in the Born approximation is used, the two-time correlation function $\langle B(t)A(\tau)\rangle$ of the quantum system is obtained in the following form:

$$\langle B(t)A(\tau)\rangle = C_{BA}^{(0)}(t,\tau) + C_{BA}^{(c)}(t,\tau), \tag{160}$$

where $C_{BA}^{(0)}(t,\tau)$ and $C_{BA}^{(c)}(t,\tau)$ are given by

$$C_{BA}^{(0)}(t,\tau) = \mathrm{Tr}_S[BV(t,\tau)AV(\tau,t_{\rm in})W_S] + \int_{t_{\rm in}}^{\tau} dt_1\,\mathrm{Tr}_S[BV(t,\tau)AV(\tau,t_1)I_0(t_1)], \tag{161}$$

$$\begin{aligned} C_{BA}^{(c)}(t,\tau) = {} & \int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2\,\mathrm{Tr}_S[BV(t,t_1)e^{L_S(t_1-t_{\rm in})}\langle L_{SR}(t_1)[A(\tau),L_{SR}(t_2)]\rangle_R e^{-L_S(t_1-t_{\rm in})}V(t_1,t_{\rm in})W_S] \\ & + \int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \int_{t_{\rm in}}^{\tau} dt_3\,\mathrm{Tr}_S[BV(t,t_1)e^{L_S(t_1-t_{\rm in})}\langle L_{SR}(t_1)[A(\tau),L_{SR}(t_3)]\rangle_R e^{-L_S(t_1-t_{\rm in})}V(t_1,t_2)I_0(t_2)] \\ & + \int_{\tau}^{t} dt_1\,\mathrm{Tr}_S[BV(t,t_1)\mathcal{J}(t_1,\tau)], \end{aligned} \tag{162}$$

with

$$\begin{aligned} \mathcal{J}(t,\tau) = {} & e^{L_S(t-t_{\rm in})}\mathrm{Tr}_R[L_{SR}(t)A(\tau)\delta W] + \int_{t_{\rm in}}^{\tau} dt_1 e^{L_S(t-t_{\rm in})}\mathrm{Tr}_R[L_{SR}(t)A(\tau)L_{SR}(t_1)\delta W] \\ & + \int_{\tau}^{t} dt_1 e^{L_S(t-t_{\rm in})}\mathrm{Tr}_R[L_{SR}(t)L_{SR}(t_1)A(\tau)\delta W]. \end{aligned} \tag{163}$$

In these equations, $V(t,t_{\rm in})$ and $I_0(t)$ are given by Eqs. (60) and (56). On the other hand, the average value of the observable B is calculated by the time-local quantum master equation,

$$\frac{\partial}{\partial t}W_S(t) = [L_S + \Pi_0(t)]W_S(t) + I_0(t), \tag{164}$$

which yields

$$\langle B(t)\rangle = \mathrm{Tr}_S[BV(t,t_{\rm in})W_S] + \int_{t_{\rm in}}^{t} dt_1\,\mathrm{Tr}_S[BV(t,t_1)I_0(t_1)]. \tag{165}$$

It is obvious from this equation that the time evolution of the average value $\langle B(t)\rangle$ is determined by the two operators

$V(t,t_{\rm in})$ and $I_0(t)$. However, these operators only determine the first term $C^{(0)}_{BA}(t,\tau)$ of the two-time correlation function. This means the violation of the quantum regression theorem for an open quantum system. In particular, if there is no initial correlation between the quantum system and the thermal reservoir, the average value and the two-time correlation function are simplified as follows:

$$\langle B(t)\rangle = {\rm Tr}_S[BV(t,t_{\rm in})W_S], \tag{166}$$

$$\begin{aligned}\langle B(t)A(\tau)\rangle &= {\rm Tr}_S[BV(t,\tau)AV(\tau,t_{\rm in})W_S]\\ &\quad+\int_\tau^t dt_1\int_{t_{\rm in}}^\tau dt_2\,{\rm Tr}_S\{BV(t,t_1)e^{L_S(t_1-t_{\rm in})}\\ &\quad\times\langle L_{SR}(t_1)[A(\tau),L_{SR}(t_2)]\rangle_R e^{-L_S(t_1-t_{\rm in})}V(t_1,t_{\rm in})W_S\}.\end{aligned} \tag{167}$$

If the second term on the right-hand side of Eq. (167) vanishes, the quantum regression theorem is established. Here we note that there is no overlap between the two integrations with respect to t_1 and t_2 in Eq. (167). Hence, the second term becomes negligible if the correlation time of the thermal reservoir is sufficiently small. This explains that the quantum regression theorem is fulfilled if the reduced time evolution of the quantum system is Markovian. This result is equivalent to that provided by a different approach in Ref. [35].

VI. SUMMARY

In this paper, using the time-local and time-nonlocal quantum master equations, we have formulated the linear response theory for open quantum systems, where a relevant quantum system interacts not only with an external field, but also with a thermal reservoir. In the previous works [17,18,24,25], it is assumed that the whole system or the relevant quantum system is in a stationary state before applying an external field. So the linear response function only explains the deviation of an observable from its stationary value [see Fig. 1(b)]. In the present work, however, we have not assumed such a stationarity of the system. The linear response function derived in this paper can explain how an observable is influenced by the external field during the irreversible time evolution from an initial value to its stationary value [see Fig. 1(c)]. We have provided the explicit expressions for the linear response function when the reduced time evolution of the quantum system is determined by the time-local and time-nonlocal quantum master equations. First we have obtained the linear response functions when we apply the Born approximation to the quantum master equations, and then we have extended the result beyond the Born approximation. We have found that the linear response function consists of four parts. The first part represents the linear response when the initial correlation between the quantum system and the thermal reservoir and the finite correlation of the thermal reservoir are discarded. Here the reservoir correlation means the correlation between the reservoir states before and after the application of the external field. The second one is a correction that is obtained by taking into account only the reservoir correlation, and the third one is a correction obtained by the effect of the system-reservoir initial correlation. The last one is a correction due to the synthetic effect of the two correlations. If we introduce an external field into the derived quantum master equation [24], we obtain only the first and third parts of the linear response function. The second and fourth parts can be obtained when we eliminate the reservoir variables from the Liouville–von Neumann equation under the influence of the external field. The linear response function has been compared with the Kubo formula of the usual linear response theory [1–3]. The result has been examined by making use of an exactly solvable model, in which the thermal reservoir is modeled by a stochastically fluctuating field which has a dephasing coupling with the quantum system. We have compared the Born approximation with the exact solution. In this model, it has been found that the Born approximation works very well in the linear response theory. Finally, using the method for obtaining the linear response function, we have derived a two-time correlation function of an open quantum system. We have discussed the violation of the quantum regression theorem when the reduced time evolution of the quantum system is non-Markovian [35]. The method that we have developed in this paper to derive the linear response function is also useful for investigating statistical properties of weak measurement performed on postselected quantum systems [45].

APPENDIX: THE FOURTH-ORDER CORRECTION TO THE LINEAR RESPONSE FUNCTION

In this appendix, we derive the contribution from the fourth time-ordered cumulant to the linear response function. To reduce the number of terms to be calculated, we assume that the equality $\langle \hat{L}_{SR}(t)\rangle_R = \langle L_{SR}(t)\rangle_R = 0$ holds. Furthermore, we temporarily neglect the superoperator $e^{\pm L_S(t-t_{\rm in})}$ outside the partial average $\langle\bullet\rangle_R$ of the thermal reservoir since it does not include the integral variables. Then, the fourth-order cumulant in the damping operator $\Pi_1(t)$ is given by

$$\begin{aligned}&\int_{t_{\rm in}}^t dt_1\int_{t_{\rm in}}^{t_1} dt_2\int_{t_{\rm in}}^{t_2} dt_3\,\hat{\mathcal{R}}\langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)L_{SR}(t_3)\rangle_R^{\rm o.c.}\\ &=\int_{t_{\rm in}}^t dt_1\int_{t_{\rm in}}^{t_1} dt_2\int_{t_{\rm in}}^{t_2} dt_3\,\hat{\mathcal{R}}\langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)L_{SR}(t_3)\rangle_R-\int_{t_{\rm in}}^t dt_1\int_{t_{\rm in}}^{t_1} dt_2\int_{t_{\rm in}}^{t_2} dt_3\,\hat{\mathcal{R}}\langle L_{SR}(t)L_{SR}(t_1)\rangle_R\langle L_{SR}(t_2)L_{SR}(t_3)\rangle_R\\ &\quad-\int_{t_{\rm in}}^t dt_1\int_{t_{\rm in}}^{t_1} dt_2\int_{t_{\rm in}}^{t_2} dt_3\,\hat{\mathcal{R}}\langle L_{SR}(t)L_{SR}(t_2)\rangle_R\langle L_{SR}(t_1)L_{SR}(t_3)\rangle_R-\int_{t_{\rm in}}^t dt_1\int_{t_{\rm in}}^{t_1} dt_2\int_{t_{\rm in}}^{t_2} dt_3\,\hat{\mathcal{R}}\langle L_{SR}(t)L_{SR}(t_3)\rangle_R\langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R.\end{aligned} \tag{A1}$$

For instance, we can calculate the fourth term, denoted as A_4, on the right-hand side of this equation. From the definition of the symbol $\hat{\mathcal{R}}$, we obtain

$$\begin{aligned}A_4 =& -\int_{t_{\rm in}}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, \langle L_{SR}(t)[\Delta u(t,t_3)]^{\times_{L_{SR}}} L_{SR}(t_3)\rangle_R \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R \\ &- \int_{t_{\rm in}}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, \langle L_{SR}(t)L_{SR}(t_3)\rangle_R [\Delta u(t_3,t_1)]^{\times_{L_{SR}}} \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R \\ &- \int_{t_{\rm in}}^{t} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, \langle L_{SR}(t)L_{SR}(t_3)\rangle_R \langle L_{SR}(t_1)[\Delta u(t_1,t_2)]^{\times_{L_{SR}}} L_{SR}(t_2)\rangle_R \\ \equiv& A_{41} + A_{42} + A_{43}. \end{aligned} \tag{A2}$$

In the calculation of A_{41}, we divide the integration in $\Delta u(t,t_3)$ as $\int_{t_3}^{t} d\tau = \int_{t_1}^{t} d\tau + \int_{t_2}^{t_1} d\tau + \int_{t_3}^{t_2} d\tau$ and rearrange the order of integrations so that the integration including the external field $F(\tau)$ is performed last. Then, we obtain

$$A_{41} = -\frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\big[A_{41}^{(1)}(\tau) + A_{41}^{(2)}(\tau) + A_{41}^{(3)}(\tau)\big], \tag{A3}$$

with

$$A_{41}^{(1)}(\tau) = \int_{t_{\rm in}}^{\tau} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, \langle L_{SR}(t)[A^{\times}(\tau)]^{\times_{L_{SR}}} L_{SR}(t_3)\rangle_R \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R, \tag{A4}$$

$$A_{41}^{(2)}(\tau) = \int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, \langle L_{SR}(t)[A^{\times}(\tau)]^{\times_{L_{SR}}} L_{SR}(t_3)\rangle_R \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R, \tag{A5}$$

$$A_{41}^{(3)}(\tau) = \int_{\tau}^{t} dt_1 \int_{\tau}^{t_1} dt_2 \int_{t_{\rm in}}^{\tau} dt_3\, \langle L_{SR}(t)[A^{\times}(\tau)]^{\times_{L_{SR}}} L_{SR}(t_3)\rangle_R \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R. \tag{A6}$$

Since the integration in $\Delta u(t_3,t_1)$ is rewritten as $\int_{t_1}^{t_3} d\tau = -\int_{t_2}^{t_1} d\tau - \int_{t_3}^{t_2} d\tau$, we can derive, for A_{42},

$$A_{42} = \frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\big[A_{42}^{(1)}(\tau) + A_{42}^{(2)}(\tau)\big], \tag{A7}$$

with

$$A_{42}^{(1)}(\tau) = \int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, \langle L_{SR}(t)L_{SR}(t_3)\rangle_R [A^{\times}(\tau)]^{\times_{L_{SR}}} \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R, \tag{A8}$$

$$A_{42}^{(2)}(\tau) = \int_{\tau}^{t} dt_1 \int_{\tau}^{t_1} dt_2 \int_{t_{\rm in}}^{\tau} dt_3\, \langle L_{SR}(t)L_1(t_3)\rangle_R [A^{\times}(\tau)]^{\times_{L_{SR}}} \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R. \tag{A9}$$

Furthermore, rearranging the order of integrations in A_{43}, we obtain

$$A_{43} = -\frac{i}{\hbar}\int_{t_{\rm in}}^{t} d\tau\, F(\tau)\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, \langle L_{SR}(t)L_{SR}(t_3)\rangle_R \langle L_{SR}(t_1)[A^{\times}(\tau)]^{\times_{L_{SR}}} L_{SR}(t_2)\rangle_R. \tag{A10}$$

Summarizing the above results, we can obtain the part of the correction to the response function from the fourth term A_4 in Eq. (A1),

$$\begin{aligned}\Delta_4^{\rm 4th}\phi_{BA}^{(c)}(t,\tau) =& -\frac{i}{\hbar}\left(\int_{t_{\rm in}}^{\tau} dt_1 \int_{t_{\rm in}}^{t_1} dt_2 \int_{t_{\rm in}}^{t_2} dt_3 + \int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2 \int_{t_{\rm in}}^{t_2} dt_3 + \int_{\tau}^{t} dt_1 \int_{\tau}^{t_1} dt_2 \int_{t_{\rm in}}^{\tau} dt_3\right) \\ &\times {\rm Tr}_S\big\{B V(t,t_1) e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)[A^{\times}(\tau)]^{\times_{L_{SR}}} L_{SR}(t_3)\rangle_R \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R e^{-L_S(t-t_{\rm in})} V(t_1,t_{\rm in}) W_S\big\} \\ &+ \frac{i}{\hbar}\left(\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2 \int_{t_{\rm in}}^{t_2} dt_3 + \int_{\tau}^{t} dt_1 \int_{\tau}^{t_1} dt_2 \int_{t_{\rm in}}^{\tau} dt_3\right){\rm Tr}_S\big\{B V(t,t_1) e^{L_S(t-t_{\rm in})} \\ &\times \langle L_{SR}(t)L_{SR}(t_3)\rangle_R [A^{\times}(\tau)]^{\times_{L_{SR}}} \langle L_{SR}(t_1)L_{SR}(t_2)\rangle_R e^{-L_S(t-t_{\rm in})} V(t_1,t_{\rm in}) W_S\big\} \\ &- \frac{i}{\hbar}\int_{\tau}^{t} dt_1 \int_{t_{\rm in}}^{\tau} dt_2 \int_{t_{\rm in}}^{t_2} dt_3\, {\rm Tr}_S\big\{B V(t,t_1) e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_3)\rangle_R \langle L_{SR}(t_1)[A^{\times}(\tau)]^{\times_{L_{SR}}} L_{SR}(t_2)\rangle_R \\ &\times\, e^{-L_S(t-t_{\rm in})} V(t_1,t_{\rm in}) W_S\big\}. \end{aligned} \tag{A11}$$

In the same way, we can derive the corrections to the response function from the first, second, and third terms on the right-hand side of Eq. (A1). The results are summarized as follows:

$$\begin{aligned}\Delta_4^{\rm 1st}\phi_{BA}^{(c)}(t,\tau)\\ &= \frac{i}{\hbar}\int_{t_{\rm in}}^{\tau}dt_1\int_{t_{\rm in}}^{t_1}dt_2\int_{t_{\rm in}}^{t_2}dt_3\,{\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)[A^\times(\tau)]^{\times_{L_{SR}}}L_{SR}(t_1)\\ &\quad\times L_{SR}(t_2)L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\}\\ &\quad+\frac{i}{\hbar}\int_{\tau}^{t}dt_1\int_{t_{\rm in}}^{\tau}dt_2\int_{t_{\rm in}}^{t_2}dt_3\,{\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)[A^\times(\tau)]^{\times_{L_{SR}}}L_{SR}(t_2)L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\}\\ &\quad+\frac{i}{\hbar}\int_{\tau}^{t}dt_1\int_{\tau}^{t_1}dt_2\int_{t_{\rm in}}^{\tau}dt_3\,{\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)L_{SR}(t_2)[A^\times(\tau)]^{\times_{L_{SR}}}L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\},\end{aligned} \tag{A12}$$

$$\begin{aligned}\Delta_4^{\rm 2nd}\phi_{BA}^{(c)}(t,\tau)\\ &= -\frac{i}{\hbar}\int_{t_{\rm in}}^{\tau}dt_1\int_{t_{\rm in}}^{t_1}dt_2\int_{t_{\rm in}}^{t_2}dt_3\,{\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)[A^\times(\tau)]^{\times_{L_{SR}}}L_{SR}(t_1)\rangle_R\langle L_{SR}(t_2)L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\}\\ &\quad-\frac{i}{\hbar}\int_{\tau}^{t}dt_1\int_{t_{\rm in}}^{\tau}dt_2\int_{t_{\rm in}}^{t_2}dt_3\,{\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)\rangle_R[A^\times(\tau)]^{\times_{L_{SR}}}\langle L_{SR}(t_2)L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\}\\ &\quad-\frac{i}{\hbar}\int_{\tau}^{t}dt_1\int_{\tau}^{t_1}dt_2\int_{t_{\rm in}}^{\tau}dt_3\,{\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_1)\rangle_R\langle L_{SR}(t_2)[A^\times(\tau)]^{\times_{L_{SR}}}L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\},\end{aligned} \tag{A13}$$

$$\begin{aligned}\Delta_4^{\rm 3rd}\phi_{BA}^{(c)}(t,\tau) &= -\frac{i}{\hbar}\left(\int_{t_{\rm in}}^{\tau}dt_1\int_{t_{\rm in}}^{t_1}dt_2\int_{t_{\rm in}}^{t_2}dt_3+\int_{\tau}^{t}dt_1\int_{t_{\rm in}}^{\tau}dt_2\int_{t_{\rm in}}^{t_2}dt_3\right){\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\\ &\quad\times\langle L_{SR}(t)[A^\times(\tau)]^{\times_{L_{SR}}}L_{SR}(t_2)\rangle_R\langle L_{SR}(t_1)L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\}\\ &\quad-\frac{i}{\hbar}\left(\int_{\tau}^{t}dt_1\int_{t_{\rm in}}^{\tau}dt_2\int_{t_{\rm in}}^{t_2}dt_3+\int_{\tau}^{t}dt_1\int_{\tau}^{t_1}dt_2\int_{t_{\rm in}}^{\tau}dt_3\right){\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\\ &\quad\times\langle L_{SR}(t)L_{SR}(t_2)\rangle_R\langle L_{SR}(t_1)[A^\times(\tau)]^{\times_{L_{SR}}}L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\}\\ &\quad+\frac{i}{\hbar}\int_{\tau}^{t}dt_1\int_{t_{\rm in}}^{\tau}dt_2\int_{t_{\rm in}}^{t_2}dt_3\,{\rm Tr}_S\{BV(t,t_1)e^{L_S(t-t_{\rm in})}\langle L_{SR}(t)L_{SR}(t_2)\rangle_R\\ &\quad\times[A^\times(\tau)]^{\times_{L_{SR}}}\langle L_{SR}(t_1)L_{SR}(t_3)\rangle_R e^{-L_S(t-t_{\rm in})}V(t_1,t_{\rm in})W_S\}.\end{aligned} \tag{A14}$$

Therefore, we finally obtain the fourth-order correction to the linear response function,

$$\Delta_4\phi_{BA}^{(c)}(t,\tau)=\Delta_4^{\rm 1st}\phi_{BA}^{(c)}(t,\tau)+\Delta_4^{\rm 2nd}\phi_{BA}^{(c)}(t,\tau)+\Delta_4^{\rm 3rd}\phi_{BA}^{(c)}(t,\tau)+\Delta_4^{\rm 4th}\phi_{BA}^{(c)}(t,\tau), \tag{A15}$$

which stems from the fourth time-ordered cumulant of the system-reservoir interaction. The calculation performed in this appendix can be applied for obtaining the correction from the inhomogeneous term of the quantum master equation in the presence of the initial correlation between the quantum system and the thermal reservoir.

[1] R. Kubo, Statistical-mechanical theory of irreversible processes I: General theory and simple applications to magnetic and conduction problems, J. Phys. Soc. Jpn. **12**, 570 (1957).

[2] R. Kubo, The fluctuation-dissipation theorem, Rep. Prog. Phys. **29**, 255 (1966).

[3] R. Kubo, M. Toda, and N. Hashitsume, *Statistical Physics II* (Springer, Berlin, 1985).

[4] T. Matsubara, A new approach to quantum-statistical mechanics, Prog. Theor. Phys. **14**, 351 (1955).

[5] D. N. Zubarev, Double-time Green functions in statistical physics, Usp. Fiz. Nauk. **71**, 71 (1960) [Sov. Phys. Usp. **3**, 320 (1960)].

[6] A. Einstein, Über die von der molekularkinetischen Theorie der Wärme geforderte Bewegung von in ruhenden Flüssigkeiten suspendierten Teilchen, Ann. Phys. **322**, 549 (1905).

[7] H. Nyquist, Thermal agitation of electric charge in conductors, Phys. Rev. **32**, 110 (1928).

[8] L. Onsager, Reciprocal relations in irreversible processes. I, Phys. Rev. **37**, 405 (1931).

[9] L. Onsager, Reciprocal relations in irreversible processes. II, Phys. Rev. **38**, 2265 (1931).

[10] H. P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, Oxford, 2006).

[11] S. Nakajima, On quantum theory of transport phenomena steady diffusion, Prog. Theor. Phys. **20**, 948 (1958).

[12] R. Zwanzig, Ensemble method in the theory of irreversibility, J. Chem. Phys. **33**, 1338 (1960).

[13] F. Shibata, Y. Takahashi, and N. Hashitsume, A generalized stochastic Liouville equation. Non-Markovian versus memoryless master equations, J. Stat. Phys. **17**, 171 (1977).

[14] S. Chaturvedi and F. Shibata, Time-convolutionless projection operator formalism for elimination of fast variables, Applications to Brownian motion, Z. Phys. B **35**, 297 (1979).

[15] F. Shibata and T. Arimitsu, Expansion formulas in nonequilibrium statistical mechanics, J. Phys. Soc. Jpn. **49**, 891 (1980).

[16] C. Uchiyama and F. Shibata, Unified projection operator formalism in nonequilibrium statistical mechanics, Phys. Rev. E **60**, 2636 (1999).

[17] C. Uchiyama, M. Aihara, M. Saeki, and S. Miyashita, Master equation approach to line shape in dissipative systems, Phys. Rev. E **80**, 021128 (2009).

[18] M. Saeki, C. Uchiyama, T. Mori, and S. Miyashita, Comparison among various expressions of complex admittance for quantum systems, in contact with a heat reservoir, Phys. Rev. E **81**, 031131 (2010).

[19] C. Uchiyama and M. Aihara, Role of initial quantum correlation in transient linear response, Phys. Rev. A **82**, 044104 (2010).

[20] C. Uchiyama, Exploring initial correlations in a Gibbs state by application of external field, Phys. Rev. A **85**, 052104 (2012).

[21] J. E. Avron, M. Fraas, G. M. Graf, and O. Kenneth, Quantum response of dephasing open systems, New J. Phys. **13**, 053042 (2011).

[22] J. E. Avron, M. Fraas, and G. M. Graf, Adiabatic response for Lindblad dynamics, J. Stat. Phys. **148**, 800 (2012).

[23] H. Z. Shen, M. Qin, X. Q. Shao, and X. X. Yi, General response formula and application to topological insulator in quantum open system, Phys. Rev. E **92**, 052122 (2015).

[24] L. C. Venuti and P. Zanardi, Dynamical response theory for driven-dissipative quantum systems, Phys. Rev. A **93**, 032101 (2016).

[25] H. Z. Shen, D. X. Li, and X. X. Yi, Non-Markovian linear response theory for quantum open systems and its applications, Phys. Rev. E **95**, 012156 (2017).

[26] M. Ban, Linear response of a pre-and post-selected system to an external field, Phys. Lett. A **379**, 284 (2015).

[27] M. Ban, Linear response theory for open quantum systems within the framework of the ABL formalism, Quantum Stud.: Math. Found. **2**, 51 (2015).

[28] S. Swain, Master equation derivation of quantum regression theorem, J. Phys. A **14**, 2577 (1981).

[29] G. W. Ford and R. F. O'Connell, There is No Quantum Regression Theorem, Phys. Rev. Lett. **77**, 798 (1996).

[30] D. Alonso and I. Vega, Hierarchy of equations of multiple-time correlation functions, Phys. Rev. A **75**, 052108 (2007).

[31] G. W. Ford and R. F. O'Connell, Driven systems and the Lax formula, Opt. Commun. **179**, 451 (2000).

[32] I. Vega and D. Alonso, Non-Markovian reduced propagator, multiple-time correlation functions, and master equations with general initial conditions in the weak-coupling limit, Phys. Rev. A **73**, 022102 (2006).

[33] A. A. Budini, Operator correlations and quantum regression theorem in non-Markovian Lindblad rate equations, J. Stat. Phys. **131**, 51 (2008).

[34] H. Goan, C. Jian, and P. Chen, Non-Markovian finite-temperature two-time correlation functions of system operators of a pure-dephasing model, Phys. Rev. A **82**, 012111 (2010).

[35] H. Goan, P. Chen, and C. Jian, Non-Markovian finite-temperature two-time correlation functions of system operators: Beyond the quantum regression theorem, J. Chem. Phys. **134**, 124112 (2011).

[36] G. Guarnieri, A. Smirne, and B. Vacchini, Quantum regression theorem and non-Markovianity of quantum dynamics, Phys. Rev. A **90**, 022110 (2014).

[37] D. P. S. McCutcheon, Optical signatures of non-Markovian behavior in open quantum systems, Phys. Rev. A **93**, 022119 (2016).

[38] M. Ban, S. Kitajima, and F. Shibata, Decoherence of quantum information of qubits by stochastic dephasing, Phys. Lett. A **349**, 415 (2006).

[39] M. Ban, S. Kitajima, and F. Shibata, Relaxation process of quantum system: Stochastic Liouville equation and initial correlation, Phys. Rev. A **82**, 022111 (2010).

[40] N. G. van Kampen, *Stochastic Processes in Physics and Chemistry* (Elsevier, Amsterdam, 1983).

[41] R. Kubo, Note on the stochastic theory of resonance absorption, J. Phys. Soc. Jpn. **9**, 935 (1954).

[42] P. W. Anderson, A mathematical model for the narrowing of spectral lines by exchange or motion, J. Phys. Soc. Jpn. **9**, 316 (1954).

[43] R. Kubo, Stochastic Liouville equation, J. Math. Phys. **4**, 174 (1962).

[44] R. Kubo, A stochastic theory of line shape, in *Advances in Chemical Physics, Volume 15: Stochastic Processes in Chemical Physics*, edited by J. Shuler (John Wiley & Sons, New York, 1969), pp. 101–127.

[45] M. Ban, Weak measurement on a quantum system in contact with a thermal reservoir: Projection operator method (unpublished).