

Two-time correlation function of an open quantum system in contact with a Gaussian reservoirMasashi Ban,^{*} Sachiko Kitajima, and Fumiaki Shibata*Graduate School of Humanities and Sciences, Ochanomizu University, 2-1-1 Otsuka, Bunkyo-ku, Tokyo 112-8610, Japan*

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An exact formula of a two-time correlation function is derived for an open quantum system which interacts with a Gaussian thermal reservoir. It is provided in terms of functional derivative with respect to fictitious fields. A perturbative expansion and its diagrammatic representation are developed, where the small expansion parameter is related to a correlation time of the Gaussian thermal reservoir. The two-time correlation function of the lowest order is equivalent to that calculated by means of the quantum regression theorem. The result clearly shows that the violation of the quantum regression theorem is caused by a finiteness of the reservoir correlation time. By making use of an exactly solvable model consisting of a two-level system and a set of harmonic oscillators, it is shown that the two-time correlation function up to the first order is a good approximation to the exact one.

DOI: [10.1103/PhysRevA.97.052101](https://doi.org/10.1103/PhysRevA.97.052101)**I. INTRODUCTION**

A quantum system in the real world is not isolated from its surrounding environment [1–6] which may be a quantum or classical system with a large number of degrees of freedom. It is referred to as a thermal reservoir. In this paper, we treat it as a quantum system. When we investigate properties of a relevant quantum system, we have to treat it as an open quantum system. A description of an open quantum system only in terms of system variables is called a reduced description which is derived by eliminating reservoir variables from a description of the whole system. A quantum state of the relevant system is given by a reduced density operator which is obtained by performing partial trace over the thermal reservoir for a total density operator. The time evolution of a reduced density operator is determined by the time-local or time-nonlocal quantum master equation which is derived by means of the projection operator method [6–13]. Solving the quantum master equation, we can obtain statistical properties of the relevant quantum system. Hence various methods for solving the quantum master equations have been developed. For instance, the phase-space method [14–18] and the algebraic method [19–22] are very useful and have been applied to many problems. Since it is very difficult to derive and solve the quantum master equations, the second-order approximation (the Born approximation) with respect to a coupling strength between the relevant quantum system and the thermal reservoir has usually been applied [6]. However, when a thermal reservoir is Gaussian, an exact reduced time evolution of an open quantum system has been found in the path-integral form [23] and in the canonical form [24,25]. The properties of the reduced dynamics have been investigated in detail for a Gaussian thermal reservoir [26–29].

A reduced density operator can explain all statistical properties associated with single time events of an open quantum system [30,31]. However, there are important phenomena that cannot be described as a single time event. For instance, the

second-order optical coherence and the fluorescence spectrum are described by a correlation function which depends on two different times [18,32]. The photon counting process is characterized by multitime joint probability [33]. Furthermore a linear response function that describes a response of a quantum system to a weak external field depends on two different times [34–39]. When dynamics of a quantum system is taken into account, a weak value of an observable [40–42] depends on two times at which an observable is weakly measured and the quantum system is postselected after the weak measurement [43–45]. Usually one calculates a two-time correlation function of an open quantum system by making use of the quantum regression theorem [6,17,18,32]. However, the theorem is no longer valid when reduced time evolution of a relevant quantum system is not Markovian [46–51]. Recently it has been shown that the projection operator technique can be applied for calculating correlation functions of open quantum systems [52–54]. These methods make it possible to calculate correlation functions in a perturbative way, where the expansion is carried out with respect to a coupling strength between the relevant quantum system and the thermal reservoir.

In this paper, we derive an exact expression of a two-time correlation function for an open quantum system interacting with a Gaussian thermal reservoir [24,25]. We also present an expansion formula for the correlation function and its diagrammatic representation, where the small expansion parameter is related to a correlation time of the Gaussian thermal reservoir. The correlation function of the lowest order is equal to that derived by means of the quantum regression theorem. Using an exactly solvable model [6,55], we examine whether the expansion formula becomes a good approximation to the exact one. The paper is organized as follows. In Sec. II, we briefly review the reduced time evolution of an open quantum system which is influenced by a Gaussian thermal reservoir. We introduce some notations used throughout this paper. In Sec. III, we derive an exact expression of a two-time correlation function for a relevant quantum system with the assistance of functional derivative with respect to fictitious fields. In Sec. IV, we provide an expansion formula for a two-time

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correlation function and its diagrammatic representation. We also obtain a two-time correlation function in the rotating wave approximation. In Sec. V, using an exactly solvable model which consists of a two-level system and harmonic oscillators, we examine whether the approximated correlation function up to the first order becomes a good approximation. In Sec. VI, we summarize the result obtained in this paper.

II. REDUCED TIME EVOLUTION OF AN OPEN QUANTUM SYSTEM

In this section, we briefly review the reduced time evolution of an open quantum system interacting with a Gaussian thermal reservoir [24]. Generalizing the result, we can derive an exact formula for a two-time correlation function of a relevant quantum system. To explain the reduced time evolution, we suppose that an interaction Hamiltonian between a relevant quantum system and a thermal reservoir is given by $H_{\text{int}}(t, t_0) = S(t, t_0) \otimes R(t, t_0)$ in the interaction picture with $S(t, t_0) = e^{(i/\hbar)H_S(t-t_0)} S e^{-(i/\hbar)H_S(t-t_0)}$ and $R(t, t_0) = e^{(i/\hbar)H_R(t-t_0)} R e^{-(i/\hbar)H_R(t-t_0)}$, where H_S (H_R) is a Hamiltonian of the relevant system (the thermal reservoir) and S (R) is an appropriate system (reservoir) operator. It is straightforward to extend the result to the case that the interaction Hamiltonian is given by $H_{SR}(t, t_0) = \sum_j S_j(t, t_0) \otimes R_j(t, t_0)$. In this paper, we explicitly denote the time at which the interaction picture coincides with the Schrödinger one. If there is no initial correlation between the relevant system and the thermal reservoir, an initial density operator of the whole system is given by $W = W_S \otimes W_R$. In this case, a thermal reservoir is Gaussian if the characteristic function is Gaussian [24,25], namely,

$$\begin{aligned} & \text{Tr}_R \left\{ T_R \exp \left[-i \int_{t_0}^t d\tau f(\tau) R(\tau, t_0) \right] W_R \right\} \\ &= \exp \left[-i \int_{t_0}^t d\tau f(\tau) \langle R(\tau, t_0) \rangle_R \right. \\ & \quad \left. - \int_{t_0}^t d\tau_2 \int_{t_0}^{\tau_2} d\tau_1 f(\tau_2) f(\tau_1) \langle R(\tau_2, t_0) R(\tau_1, t_0) \rangle_R^c \right], \quad (1) \end{aligned}$$

with $f(t)$ being an analytic function, $\langle \bullet \rangle_R = \text{Tr}_R(\bullet W_R)$, and

$$\begin{aligned} & \langle R(\tau_2, t_0) R(\tau_1, t_0) \rangle_R^c \\ &= \langle R(\tau_2, t_0) R(\tau_1, t_0) \rangle_R - \langle R(\tau_2, t_0) \rangle_R \langle R(\tau_1, t_0) \rangle_R. \quad (2) \end{aligned}$$

In Eq. (1), Tr_R stands for the trace operation over a Hilbert space of the thermal reservoir and T_R represents the time ordering in which the reservoir operators $R(t, t_0)$'s are placed from the right to the left in the chronological order.

To derive the reduced time evolution of the relevant quantum system, it is convenient to introduce Liouvillian superoperators [6]. We denote the total Hamiltonian as $H = H_0 + H_1$ with $H_0 = H_S + H_R$ and $H_1 = H_{SR} = S \otimes R$. The corresponding Liouvillian superoperators are $L_x \bullet = -(i/\hbar) H_x \bullet = -(i/\hbar) [H_x, \bullet]$. Furthermore, we define a superoperator \tilde{X} by $\tilde{X} \bullet = \bullet X$. Then we have a commutator $X^\times = X - \tilde{X}$ and an anticommutator $X^\circ = X + \tilde{X}$. These superoperators make it possible to calculate the reduced time evolution of the relevant quantum system in a systematic way. In fact, we can obtain the time evolution of the reduced density

operator of the relevant quantum system [24],

$$W_S(t) = V_S(t, t_0) W_S, \quad (3)$$

where the time-evolution operator $V_S(t, t_0)$ of the relevant quantum system is given by

$$V_S(t, t_0) = \text{Tr}_R [e^{L(t-t_0)} W_R] = e^{L_S(t-t_0)} \text{Tr}_R [V(t, t_0 | t_0) W_R], \quad (4)$$

with

$$V(t, t_1 | t_0) = T \exp \left[\int_{t_1}^t d\tau L_1(\tau, t_0) \right] \quad (5)$$

and

$$L_1(t, t_0) = e^{-L_0(t-t_0)} L_1 e^{L_0(t-t_0)}. \quad (6)$$

In Eq. (5), T represents the time ordering for the Liouvillian superoperators $L_1(t, t_0)$'s. If the thermal reservoir is Gaussian, $\text{Tr}_R [V(t, t_0 | t_0) W_R]$ is calculated to be

$$\begin{aligned} & \text{Tr}_R [V(t_2, t_1 | t_0) W_R] \\ &= T_{(t_2, t_0)}^S \exp \left[- \int_{t_1}^{t_2} d\tau_2 \int_{t_1}^{\tau_2} d\tau_1 \mathcal{G}(\tau_2, \tau_1 | t_0) \right], \quad (7) \end{aligned}$$

with

$$\begin{aligned} \mathcal{G}(\tau_2, \tau_1 | t_0) &= S^\times(\tau_2, t_0) [C_R(\tau_2 - \tau_1) S^\times(\tau_1, t_0) \\ & \quad + i C_I(\tau_2 - \tau_1) S^\circ(\tau_1, t_0)], \quad (8) \end{aligned}$$

where $C_R(\tau_2 - \tau_1)$ and $C_I(\tau_2 - \tau_1)$ are real and imaginary parts of the reservoir correlation function $\langle R(\tau_2, t_0) R(\tau_1, t_0) \rangle_R$. In deriving the result, we have assumed $\langle R(t, t_0) \rangle_R = 0$ without a loss of generality. In Eq. (7), the symbol $T_{(t_2, t_0)}^S$ means the time ordering that the system operators $S(t, t_0)$'s in the time region $t_2 > t > t_0$ are placed in the chronological order. A generalization of this result to the case that there is initial correlation between the relevant quantum system and the Gaussian thermal reservoir is briefly discussed in Appendix A.

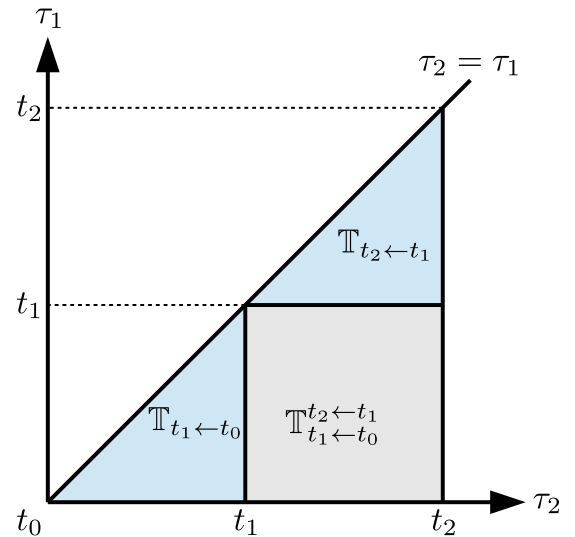


FIG. 1. Representation of the integral region $\mathbb{T}_{t_2 \leftarrow t_0} = \mathbb{T}_{t_2 \leftarrow t_1} + \mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1} + \mathbb{T}_{t_1 \leftarrow t_0}$, where there is no overlap of τ_2 integral and τ_1 integral in the gray-colored region $\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}$.

Introducing vector $\mathbf{S}(t, t_0)$ of superoperators and 2×2 matrix $\mathbf{C}(t)$ by

$$\mathbf{S}(t, t_0) = \begin{pmatrix} S^\times(t, t_0) \\ S^\circ(t, t_0) \end{pmatrix},$$

$$\mathbf{C}(\tau_2 - \tau_1) = \begin{pmatrix} C_R(\tau_2 - \tau_1) & iC_I(\tau_2 - \tau_1) \\ 0 & 0 \end{pmatrix}, \quad (9)$$

we can express the superoperator $\mathcal{G}(\tau_2, \tau_1 | t_0)$ as

$$\mathcal{G}(\tau_2, \tau_1 | t_0) = \sum_{\mu, \nu} \mathbf{C}_{\mu, \nu}(\tau_2 - \tau_1) \mathbf{S}_\mu(\tau_2, t_0) \mathbf{S}_\nu(\tau_1, t_0). \quad (10)$$

Before closing this section, we consider the integral with respect to τ_2 and τ_1 in Eq. (7). For any time t_1 with $t_2 > t_1 > t_0$, the integral decomposed into three parts as follows:

$$\begin{aligned} & \int_{t_0}^{t_2} d\tau_2 \int_{t_0}^{\tau_2} d\tau_1 \mathcal{G}(\tau_2, \tau_1 | t_0) \\ &= \left(\int_{t_1}^{t_2} d\tau_2 \int_{t_1}^{\tau_2} d\tau_1 + \int_{t_1}^{t_2} d\tau_2 \int_{t_0}^{t_1} d\tau_1 \right. \\ & \quad \left. + \int_{t_0}^{t_1} d\tau_2 \int_{t_0}^{\tau_2} d\tau_1 \right) \mathcal{G}(\tau_2, \tau_1 | t_0) \\ &\equiv \left(\iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{21} + \iint_{\mathbb{T}_{t_2 \leftarrow t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} + \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{21} \right) \\ & \quad \times \mathcal{G}(\tau_2, \tau_1 | t_0), \end{aligned} \quad (11)$$

where we set $d^2 \tau_{21} = d\tau_2 d\tau_1$ and the three time regions $\mathbb{T}_{t_2 \leftarrow t_1}$, $\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}$, and $\mathbb{T}_{t_1 \leftarrow t_0}$ are depicted in Fig. 1. It is important to note that there is no overlap of the two integrals in the region $\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}$. Hence this integral becomes negligible if the correlation time of the thermal reservoir is sufficiently small. In this case, we may approximate as

$$\begin{aligned} & \int_{t_0}^{t_2} d\tau_2 \int_{t_0}^{\tau_2} d\tau_1 \mathcal{G}(\tau_2, \tau_1 | t_0) \\ &\approx \left(\iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{21} + \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{21} \right) \mathcal{G}(\tau_2, \tau_1 | t_0). \end{aligned} \quad (12)$$

This result is essential for deriving an expansion formula for a two-time correlation function. In fact, the integral in the region $\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}$ plays a role of a small parameter of perturbative expansion. In the rest of this paper, we refer to $\iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21}$ as t_1 -disconnected integral since the two integrals are separated at time t_1 . Here it should be noted that the intermediate time t_1 will be chosen so that a systematic perturbative expansion is possible.

III. TWO-TIME CORRELATION FUNCTION OF AN OPEN QUANTUM SYSTEM

A two-time correlation function of the relevant quantum system that we consider in this paper is expressed in the following form [54]:

$$C_{X_2, X_1}(t_2, t_1) = \text{Tr}[X_2 e^{L(t_2 - t_1)} X_1 e^{L(t_1 - t_0)} W], \quad (13)$$

where X_j ($j = 1, 2$) is any system operator including a superoperator and quantum operation. Rewriting the Liouvillian superoperator L in terms of the Hamiltonian H and assuming observables X_2 and X_1 , we obtain the usual two-time correlation function $C_{X_2, X_1}(t_2, t_1) = \text{Tr}[X_2^{(H)}(t_2, t_0) X_1^{(H)}(t_1, t_0) W]$ [56] with the Heisenberg operator $X_j^{(H)}(t_j, t_0) = e^{(i/\hbar)H(t_j - t_0)} X_j e^{-(i/\hbar)H(t_j - t_0)}$. Our purpose is to derive the reduced description of the correlation function $C_{X_2, X_1}(t_2, t_1)$. Using the operators in the interaction picture, we can rewrite the correlation function into

$$C_{X_2, X_1}(t_2, t_1) = \text{Tr}[X_2(t_2, t_0) V(t_2, t_1 | t_0) X_1(t_1, t_0) V(t_1, t_0 | t_0) W], \quad (14)$$

where $X_j(t, t_0) = e^{-L_0(t - t_0)} X_j e^{L_0(t - t_0)} = e^{-L_S(t - t_0)} X_j e^{L_S(t - t_0)}$ and $V(t_2, t_1 | t_0)$ is given by Eq. (5). In deriving this equation, we have used the relation $e^{L(t - t_1)} e^{L_0(t_1 - t_0)} = e^{L_0(t - t_0)} V(t, t_1 | t_0)$.

To calculate the trace over the Hilbert space of the Gaussian thermal reservoir on the right-hand side of Eq. (14), we introduce a conditional time ordering $T_{(t_2, t_0)}^{S|X_1(t_1, t_0)}$. For any times τ_{n_i} ($i = 1, 2, \dots, N$) and τ_{m_j} ($j = 1, 2, \dots, M$) with $t_2 > \tau_{n_i} > t_0$ and $t_2 > \tau_{m_j} > t_0$, it is defined by

$$\begin{aligned} & T_{(t_2, t_0)}^{S|X_1(t_1, t_0)} [S^\times(\tau_{n_1}, t_0) \cdots S^\times(\tau_{n_N}, t_0) X_1(t_1, t_0) S^\times(\tau_{m_1}, t_0) \cdots S^\times(\tau_{m_M}, t_0)] \\ &= T_{(t_2, t_0)}^S [S^\times(\tau_{k_1}, t_0) \cdots S^\times(\tau_{k_K}, t_0)] X_1(t_1, t_0) T_{(t_1, t_0)}^S [S^\times(\tau_{\ell_1}, t_0) \cdots S^\times(\tau_{\ell_L}, t_0)], \end{aligned} \quad (15)$$

where τ_{k_i} ($i = 1, 2, \dots, K$) and τ_{ℓ_j} ($j = 1, 2, \dots, L$) satisfy $t_2 > \tau_{k_i} > t_1$ and $t_1 > \tau_{\ell_j} > t_0$ and the equality $M + N = K + L$ holds. It is obvious that if $X_1(t_1, t_0) = 1$, the conditional time ordering $T_{(t_2, t_0)}^{S|X_1(t_1, t_0)}$ reduces to the usual time ordering $T_{(t_2, t_0)}^S$. Thanks to the conditional time ordering, when we calculate the trace over the reservoir Hilbert space in Eq. (14), we can treat $\tilde{X}_1(t_1, t_0)$ and $S^\times(t, t_0)$'s as commutable variables under the ordering $T_{(t_2, t_0)}^{S|X_1(t_1, t_0)}$ and thus we can apply the method developed in Refs. [24, 25]. Hence, using the conditional time-ordering operation and Eq. (7), we can express the correlation function as

$$\begin{aligned} C_{X_2, X_1}(t_2, t_1) &= \text{Tr}_S [T_{(t_2, t_0)}^{S|X_1(t_1, t_0)} X_2(t_2, t_0) X_1(t_1, t_0) \text{Tr}_R (V(t_2, t_0 | t_0) W)] \\ &= \text{Tr}_S \left\{ X_2(t_2, t_0) T_{(t_2, t_0)}^{S|X_1(t_1, t_0)} X_1(t_1, t_0) \exp \left[- \iint_{\mathbb{T}_{t_2 \leftarrow t_0}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right] W_S \right\}. \end{aligned} \quad (16)$$

Furthermore, substituting Eqs. (11) and (15) into this equation, we obtain the correlation function,

$$C_{X_2, X_1}(t_2, t_1) = \text{Tr}_S \left\{ X_2(t_2, t_0) T_{(t_2, t_0)}^S \left[\exp \left(- \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right) \right. \right. \\ \left. \left. \times T_{(t_2, t_0)}^{S|X_1(t_1, t_0)} \left[X_1(t_1, t_0) \exp \left(- \iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right) \right] \exp \left(- \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right) \right] W_S \right\}. \quad (17)$$

Our next task is to rewrite the second term with the conditional time ordering in Eq. (17) into terms with the usual time ordering. First expanding the exponential and using Eqs. (10) and (15), we obtain

$$T_{(t_2, t_0)}^{S|X_1(t_1, t_0)} \left[X_1(t_1, t_0) \exp \left(- \iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right) \right] = T_{(t_2, t_0)}^{S|X_1(t_1, t_0)} \left[X_1(t_1, t_0) \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \left(\iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right)^n \right] \\ = X_1(t_1, t_0) + T_{(t_2, t_0)}^S \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21}^{(n)} \cdots \iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21}^{(1)} \\ \times \sum_{\mu_n, \nu_n} \cdots \sum_{\mu_1, \nu_1} \mathbf{C}_{\mu_n, \nu_n}(\tau_2^{(n)} - \tau_1^{(n)}) \cdots \mathbf{C}_{\mu_1, \nu_1}(\tau_2^{(1)} - \tau_1^{(1)}) \\ \times \mathbf{S}_{\mu_n}(\tau_2^{(n)}, t_0) \cdots \mathbf{S}_{\mu_1}(\tau_2^{(1)}, t_0) X_1(t_1, t_0) \mathbf{S}_{\nu_n}(\tau_1^{(n)}, t_0) \cdots \mathbf{S}_{\nu_1}(\tau_1^{(1)}, t_0). \quad (18)$$

Here it is convenient to introduce two superoperators,

$$K_{\mu_n \dots \mu_1}(t_2, t_1; \tau_2^{(n)}, \dots, \tau_2^{(1)} | t_0) = T_{(t_2, t_0)}^S \left[\mathbf{S}_{\mu_n}(\tau_2^{(n)}, t_0) \cdots \mathbf{S}_{\mu_1}(\tau_2^{(1)}, t_0) \exp \left(- \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right) \right], \quad (19)$$

$$L_{\nu_n \dots \nu_1}(t_1, t_0; \tau_1^{(n)}, \dots, \tau_1^{(1)} | t_0) = T_{(t_1, t_0)}^S \left[\mathbf{S}_{\nu_n}(\tau_1^{(n)}, t_0) \cdots \mathbf{S}_{\nu_1}(\tau_1^{(1)}, t_0) \exp \left(- \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right) \right] \quad (20)$$

for $n > 0$ and

$$K(t_2, t_1 | t_0) = \text{Tr}_R[V(t_2, t_1 | t_0) W_R], \quad L(t_1, t_0 | t_0) = \text{Tr}_R[V(t_1, t_0 | t_0) W_R], \quad (21)$$

for $n = 0$. Then the two-time correlation function $C_{X_2, X_1}(t_2, t_1)$ is given by

$$C_{X_2, X_1}(t_2, t_1) = C_{X_2, X_1}^{(0)}(t_2, t_1) + \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21}^{(n)} \cdots \iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21}^{(1)} \\ \times \sum_{\mu_n, \nu_n} \cdots \sum_{\mu_1, \nu_1} \mathbf{C}_{\mu_n, \nu_n}(\tau_2^{(n)} - \tau_1^{(n)}) \cdots \mathbf{C}_{\mu_1, \nu_1}(\tau_2^{(1)} - \tau_1^{(1)}) \\ \times \text{Tr}_S[X_2(t_2, t_0) K_{\mu_n \dots \mu_1}(t_2, t_1; \tau_2^{(n)}, \dots, \tau_2^{(1)} | t_0) X_1(t_1, t_0) L_{\nu_n \dots \nu_1}(t_1, t_0; \tau_1^{(n)}, \dots, \tau_1^{(1)} | t_0) W_S], \quad (22)$$

with

$$C_{X_2, X_1}^{(0)}(t_2, t_1) = \text{Tr}_S[X_2(t_2, t_0) K(t_2, t_1 | t_0) X_1(t_1, t_0) L(t_1, t_0 | t_0) W_S]. \quad (23)$$

To proceed further, we note that the following equality is established:

$$\mathbf{S}_{\mu_j}(\tau_2^{(j)}, t_0) \mathbf{S}_{\mu_k}(\tau_2^{(k)}, t_0) = e^{-L_S(t_1 - t_0)} \mathbf{S}_{\mu_j}(\tau_2^{(j)}, t_1) \mathbf{S}_{\mu_k}(\tau_2^{(k)}, t_1) e^{L_S(t_1 - t_0)}. \quad (24)$$

This equation relates $K_{\mu_n \dots \mu_1}(t_2, t_1; \tau_2^{(n)}, \dots, \tau_2^{(1)} | t_0)$ to $L_{\nu_n \dots \nu_1}(t_1, t_0; \tau_1^{(n)}, \dots, \tau_1^{(1)} | t_0)$ by the relation

$$K_{\mu_n \dots \mu_1}(t_2, t_1; \tau_2^{(n)}, \dots, \tau_2^{(1)} | t_0) = e^{-L_S(t_1 - t_0)} K_{\mu_n \dots \mu_1}(t_2, t_1; \tau_2^{(n)}, \dots, \tau_2^{(1)} | t_1) e^{L_S(t_1 - t_0)} \\ = e^{-L_S(t_1 - t_0)} L_{\mu_n \dots \mu_1}(t_2, t_1; \tau_2^{(n)}, \dots, \tau_2^{(1)} | t_1) e^{L_S(t_1 - t_0)}. \quad (25)$$

Then we obtain the correlation function from Eq. (22),

$$C_{X_2, X_1}(t_2, t_1) = C_{X_2, X_1}^{(0)}(t_2, t_1) + \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \left(\prod_{k=1}^n \iint_{\mathbb{T}_{t_1 \leftarrow t_0}^{t_2 \leftarrow t_1}} d^2 \tau_{21}^{(k)} \mathbf{C}_{\mu_k, \nu_k}(\tau_2^{(k)} - \tau_1^{(k)}) \right) \\ \times \text{Tr}_S[X_2 e^{L_S(t_2 - t_1)} L_{\mu_n \dots \mu_1}(t_2, t_1; \tau_2^{(n)}, \dots, \tau_2^{(1)} | t_1) X_1 e^{L_S(t_1 - t_0)} L_{\nu_n \dots \nu_1}(t_1, t_0; \tau_1^{(n)}, \dots, \tau_1^{(1)} | t_0) W_S], \quad (26)$$

with

$$C_{X_2, X_1}^{(0)}(t_2, t_1) = \text{Tr}_S[X_2 e^{L_S(t_2-t_0)} K(t_2, t_1 | t_0) X_1 e^{L_S(t_1-t_0)} L(t_1, t_0 | t_0) W_S]. \quad (27)$$

In deriving Eq. (26), we have used $X_2(t_2, t_0) = e^{-L_S(t_2-t_0)} X_2 e^{L_S(t_2-t_0)}$ and $X_1(t_1, t_0) = e^{-L_S(t_1-t_0)} X_1 e^{L_S(t_1-t_0)}$. When we introduce fictitious fields $J_\mu^\pm(t)$'s and define superoperators $L(t_2, t_1 | J^\pm(t))$ by

$$L(t_2, t_1 | J^\pm(t)) = T_{(t_2, t_1)}^S \exp \left(- \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_1) + \sum_\mu \int_{t_1}^{t_2} d\tau J_\mu^\pm(\tau) \mathbf{S}_\mu(\tau, t_1) \right), \quad (28)$$

we can express the two-time correlation function $C_{X_2, X_1}(t_2, t_1)$ by means of functional derivative,

$$C_{X_2, X_1}(t_2, t_1) = \exp \left(- \sum_{\mu, \nu} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}}^{t_2 \leftarrow t_1} d^2 \tau_{21} \mathbf{C}_{\mu, \nu}(\tau_2 - \tau_1) \frac{\delta^2}{\delta J_\mu^+(\tau_2) \delta J_\nu^-(\tau_1)} \right) \times \text{Tr}_S[X_2 e^{L_S(t_2-t_1)} L(t_2, t_1 | J^+(t)) X_1 e^{L_S(t_1-t_0)} L(t_1, t_0 | J^-(t)) W_S] |_{J^\pm(t) \rightarrow 0}. \quad (29)$$

This is a main result of this paper. When we substitute $X_2 = 1$ or $X_1 = 1$ into this equation, we find that the average values,

$$\begin{aligned} C_{1, X_1}(t_2, t_1) &= \text{Tr}_S[X_1 W_S(t_1)], \\ C_{X_2, 1}(t_2, t_1) &= \text{Tr}_S[X_2 W_S(t_2)], \end{aligned} \quad (30)$$

are obtained (see Appendix B), where the reduced density operator $W_S(t)$ of the relevant quantum system is given by Eq. (3).

In deriving the result (29), we have assumed that the thermal reservoir is Gaussian, but we have not applied any approximation. Thus the two-time correlation function $C_{X_2, X_1}(t_2, t_1)$ that we have derived for the open quantum system is an exact and formal expression. Although the two-time correlation functions have been calculated perturbatively by means of the quantum master equation based on the projection operator technique [52–54], its exact expression has not been found. Of course, we may have to use some approximation to explicitly calculate the two-time correlation function (29). However, deriving an exact expression of a two-time correlation function means a lot to the theory of open quantum systems since it may be useful for investigating physical and mathematical properties of open quantum systems.

IV. EXPANSION OF THE TWO-TIME CORRELATION FUNCTION

In this section, we derive an expansion formula which provides the two-time correlation function up to the second order with respect to the disconnected integral. If the correlation time of the Gaussian thermal reservoir is sufficiently small, the disconnected integral becomes negligible (see Sec. II). The lowest-order correlation function $C_{X_2, X_1}^{(0)}(t_2, t_1)$ is obtained by discarding all the contributions from the t_1 - and $\tau_{1,2}$ -disconnected integrals in Eq. (26) or from the functional derivatives in Eq. (29). The n th-order correction $\Delta C_{X_2, X_1}^{(n)}(t_2, t_1)$ to the correlation function is obtained by taking into account the n th order of the t_1 - and $\tau_{1,2}$ -disconnected integral in Eq. (26). In the rest of this section, we assume that the system operator X_1 at the time t_1 in the correlation function is not an identity.

A. Lowest-order term and the quantum regression theorem

We derive the lowest-order correlation function $C_{X_2, X_1}^{(0)}(t_2, t_1)$ and discuss the quantum regression theorem [6, 17, 18, 32]. Since the disconnected integral comes out when the functional derivatives with respect to $J_\mu^\pm(t)$ are performed, we neglect the exponential in Eq. (29) when deriving the lowest-order correlation function. Then we obtain from Eq. (29)

$$C_{X_2, X_1}^{(0)}(t_2, t_1) = \text{Tr}_S[X_2 e^{L_S(t_2-t_1)} L(t_2, t_1 | J^+(t)) X_1 e^{L_S(t_1-t_0)} \times L(t_1, t_0 | J^-(t)) W_S] |_{J^\pm(t) \rightarrow 0}. \quad (31)$$

Here $L(t_1, t_0 | J^-(t)) |_{J^-(t) \rightarrow 0}$ is obtained from Eq. (28),

$$\begin{aligned} & e^{L_S(t_1-t_0)} L(t_1, t_0 | J^-(t)) |_{J^-(t) \rightarrow 0} \\ &= e^{L_S(t_1-t_0)} T_{(t_1, t_0)}^S \exp \left(- \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right) \\ &= e^{L_S(t_1-t_0)} \text{Tr}_R[V(t_1, t_0 | t_0) W_R] \\ &= \langle e^{L(t_1-t_0)} \rangle_R = V_S(t_1, t_0), \end{aligned} \quad (32)$$

where we set $\langle \bullet \rangle_R = \text{Tr}[\bullet W_R]$. In the same way, we obtain

$$e^{L_S(t_2-t_1)} L(t_2, t_1 | J^+(t)) |_{J^+(t) \rightarrow 0} = V_S(t_2, t_1). \quad (33)$$

Then the two-time correlation function of the lowest order is given by

$$C_{X_2, X_1}^{(0)}(t_2, t_1) = \text{Tr}_S[X_2 V_S(t_2, t_1) X_1 V_S(t_1, t_0) W_S]. \quad (34)$$

The lowest-order correlation function $C_{X_2, X_1}^{(0)}(t_2, t_1)$ can also be derived by applying the quantum regression theorem. To show this, we note that the reduced density operator of the relevant quantum system is given by $W_S(t) = \langle e^{L(t-t_0)} \rangle_R W_S = V_S(t, t_0) W_S$ if there is no initial correlation between the relevant quantum system and the thermal reservoir. Then the average value of a system observable X at time t_2 is $\langle X(t_2) \rangle = \text{Tr}_S[X V_S(t_2, t_0) W_S]$. Differentiating the average value with respect to t_2 , we obtain the equation of motion for the average value,

$$\begin{aligned} \frac{\partial}{\partial t_2} \langle X(t_2) \rangle &= \text{Tr}_S[X K_S(t_2, t_0) V_S(t_2, t_0) W_S] \\ &= \text{Tr}_S[(\tilde{K}_S(t_2, t_0) X) V_S(t_2, t_0) W_S], \end{aligned} \quad (35)$$

where $K_S(t_2, t_0) = \langle L e^{L(t_2-t_0)} \rangle_R / \langle e^{L(t_2-t_0)} \rangle_R$ and $\text{Tr}_S[(\tilde{K}_S(t_2, t_0)A)B] = \text{Tr}_S[AK_S(t_2, t_0)B]$. Here let $\{E_k\}$ be a complete set of system operators and we expand as $\tilde{K}_S(t_2, t_0)E_j = \sum_k G_{jk}(t_2, t_0)E_k$. Then the equation of motion for $\langle E_j(t_2) \rangle$ is given by

$$\frac{\partial}{\partial t_2} \langle E_j(t_2) \rangle = \sum_k G_{jk}(t_2, t_0) \langle E_k(t_2) \rangle. \quad (36)$$

On the other hand, differentiating Eq. (34) with respect to time t_2 and substituting $X_2 = E_j$ and $X_1 = E_l$, we obtain

$$\frac{\partial}{\partial t} C_{E_j, E_l}^{(0)}(t_2, t_1) = \sum_k G_{jk}(t_2, t_1) C_{E_k, E_l}^{(0)}(t_2, t_1). \quad (37)$$

Although $G_{jk}(t_2, t_1) \neq G_{jk}(t_2, t_0)$ in general, $G_{jk}(t_2, t_1)$ is obtained by replacing the initial time t_0 by the later time t_1 in $G_{jk}(t_2, t_0)$. Hence this result shows that the quantum regression theorem leads to the lowest-order correlation function $C_{X_2, X_1}^{(0)}(t_2, t_1) = \sum_{j,k} a_j b_k C_{jk}^{(0)}(t_2, t_1)$ with $X_2 = \sum_j a_j E_j$ and $X_1 = \sum_k b_k E_k$. However, the quantum regression theorem is not valid in general [46–51] due to the correction terms to $C_{X_2, X_1}(t_2, t_1)$.

B. First- and second-order correction terms

Next we obtain the first-order correction $\Delta C_{X_2, X_1}^{(1)}(t_2, t_1)$ to the two-time correlation function with respect to the disconnected integral. Picking up the term with $n = 1$ in the sum on the right-hand side of Eq. (26), we have

$$\Delta C_{X_2, X_1}(t_2, t_1) = - \sum_{\mu, \nu} \iint_{\mathbb{T}_{t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathbf{C}_{\mu, \nu}(\tau_2 - \tau_1) \text{Tr}_S[X_2 e^{L_S(t_2-t_1)} L_\mu(t_2, t_1; \tau_2 | t_1) X_1 e^{L_S(t_1-t_0)} L_\nu(t_1, t_0; \tau_1 | t_0) W_S], \quad (38)$$

with

$$L_\nu(t_1, t_0; \tau_1 | t_0) = T_{(t_1, t_0)}^S \left[\mathbf{S}_\nu(\tau_1, t_0) \exp \left(- \iint_{\mathbb{T}_{t_1}^{t_0 \leftarrow t_0}} d^2 \tau'_{21} \mathcal{G}(\tau'_2, \tau'_1 | t_0) \right) \right], \quad (39)$$

$$L_\mu(t_2, t_1; \tau_2 | t_1) = T_{(t_2, t_1)}^S \left[\mathbf{S}_\mu(\tau_2, t_1) \exp \left(- \iint_{\mathbb{T}_{t_2}^{t_1 \leftarrow t_1}} d^2 \tau'_{21} \mathcal{G}(\tau'_2, \tau'_1 | t_1) \right) \right]. \quad (40)$$

Here we note that the superoperator $L_\nu(t_1, t_0; \tau_1 | t_0)$ is decomposed into

$$L_\nu(t_1, t_0; \tau_1 | t_0) = T_{(t_1, t_0)}^S \left\{ \left[\exp \left(- \iint_{\mathbb{T}_{t_1}^{t_0 \leftarrow t_0}} d^2 \tau'_{21} \mathcal{G}(\tau'_2, \tau'_1 | t_0) \right) \right] \times \left[\mathbf{S}_\nu(\tau_1, t_0) \exp \left(- \iint_{\mathbb{T}_{t_1}^{t_0 \leftarrow t_0}} d^2 \tau'_{21} \mathcal{G}(\tau'_2, \tau'_1 | t_0) \right) \right] \left[\exp \left(- \iint_{\mathbb{T}_{t_1}^{t_0 \leftarrow t_0}} d^2 \tau'_{21} \mathcal{G}(\tau'_2, \tau'_1 | t_0) \right) \right] \right\}. \quad (41)$$

If we remove the τ_1 -disconnected integral from Eq. (41), we obtain

$$e^{L_S(t_1-t_0)} L_\nu(t_1, t_0; \tau_1 | t_0) \approx V_S(t_1, \tau_1) \mathbf{S}_\nu V_S(t_1, t_0). \quad (42)$$

In the same way, neglecting the τ_2 -disconnected integral in Eq. (40), we can also derive

$$e^{L_S(t_2-t_1)} L_\mu(t_2, t_1; \tau_2 | t_1) \approx V_S(t_2, \tau_2) \mathbf{S}_\mu V_S(t_2, t_1). \quad (43)$$

Since Eq. (38) has the t_1 -disconnected integral, substituting Eqs. (42) and (43) into Eq. (38), we obtain the first-order correction $\Delta C_{X_2, X_1}^{(1)}(t_2, t_1)$ to the correlation function,

$$\Delta C_{X_2, X_1}^{(1)}(t_2, t_1) = - \sum_{\mu, \nu} \iint_{\mathbb{T}_{t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathbf{C}_{\mu, \nu}(\tau_2 - \tau_1) \times \text{Tr}_S[X_2 V_S(t_2, \tau_2) \mathbf{S}_\mu V_S(t_2, t_1) X_1 V_S(t_1, \tau_1) \mathbf{S}_\nu V_S(t_1, t_0) W_S]. \quad (44)$$

To obtain the second-order correction $\Delta C_{X_2, X_1}^{(2)}(t_2, t_1)$, we have to perform straightforward but lengthy calculation, which is given in Appendix C. Here we only provide the result,

$$\begin{aligned} \Delta C_{X_2, X_1}^{(2)}(t_2, t_1) = & \sum_{\mu_1, \mu_2, \mu_3, \mu_4} \iint_{\mathbb{T}_{t_2}^{t_2 \leftarrow t_1}} d^2 \tau_{43} \iint_{\mathbb{T}_{t_1}^{t_1 \leftarrow t_0}} d^2 \tau_{21} \mathbf{C}_{\mu_4, \mu_1}(\tau_4 - \tau_1) \mathbf{C}_{\mu_3, \mu_2}(\tau_3 - \tau_2) \\ & \times \text{Tr}_S[X_2 V_S(t_2, \tau_4) \mathbf{S}_{\mu_4} V_S(t_2, \tau_3) \mathbf{S}_{\mu_3} V_S(t_3, t_1) X_1 V_S(t_1, \tau_2) \mathbf{S}_{\mu_2} V_S(t_2, \tau_1) \mathbf{S}_{\mu_1} V_S(t_1, t_0) W_S] \\ & + \sum_{\mu_1, \mu_2, \mu_3, \mu_4} \iint_{\mathbb{T}_{t_2}^{t_2 \leftarrow t_1}} d^2 \tau_{43} \iint_{\mathbb{T}_{t_1}^{t_1 \leftarrow t_0}} d^2 \tau_{21} \mathbf{C}_{\mu_4, \mu_2}(\tau_4 - \tau_2) \mathbf{C}_{\mu_3, \mu_1}(\tau_3 - \tau_1) \\ & \times \text{Tr}_S[X_2 V_S(t_2, \tau_4) \mathbf{S}_{\mu_4} V_S(t_2, \tau_3) \mathbf{S}_{\mu_3} V_S(t_3, t_1) X_1 V_S(t_1, \tau_2) \mathbf{S}_{\mu_2} V_S(t_2, \tau_1) \mathbf{S}_{\mu_1} V_S(t_1, t_0) W_S] \end{aligned}$$

$$\begin{aligned}
& + \sum_{\mu_1, \mu_2, \mu_3, \mu_4} \iiint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^3 \tau_{432} \int_{\mathbb{T}_{t_1 \leftarrow t_0}} d\tau_1 \mathbf{C}_{\mu_4, \mu_2}(\tau_4 - \tau_2) \mathbf{C}_{\mu_3, \mu_1}(\tau_3 - \tau_1) \\
& \times \text{Tr}_S [X_2 V_S(t_2, \tau_4) \mathbf{S}_{\mu_4} V_S(\tau_4, \tau_3) \mathbf{S}_{\mu_3} V_S(\tau_3, \tau_2) \mathbf{S}_{\mu_2} V_S(\tau_2, \tau_1) X_1 V_S(t_1, \tau_1) \mathbf{S}_{\mu_1} V_S(\tau_1, t_0) W_S] \\
& + \sum_{\mu_1, \mu_2, \mu_3, \mu_4} \int_{\mathbb{T}_{t_2 \leftarrow t_1}} d\tau_4 \iiint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^3 \tau_{321} \mathbf{C}_{\mu_4, \mu_2}(\tau_4 - \tau_2) \mathbf{C}_{\mu_3, \mu_1}(\tau_3 - \tau_1) \\
& \times \text{Tr}_S [X_2 V_S(t_2, \tau_4) \mathbf{S}_{\mu_4} V_S(\tau_4, t_1) X_1 V_S(t_1, \tau_3) \mathbf{S}_{\mu_3} V_S(\tau_3, \tau_2) \mathbf{S}_{\mu_2} V_S(\tau_2, \tau_1) \mathbf{S}_{\mu_1} V_S(\tau_1, t_0) W_S], \quad (45)
\end{aligned}$$

where we have denoted the time-order integral as

$$\iint \cdots \iint_{\mathbb{T}_{t_b \leftarrow t_a}} d^n \tau_{n-1 \dots 21} f(\tau_n, \tau_{n-1}, \dots, \tau_2, \tau_1) = \int_{t_a}^{t_b} d\tau_n \int_{t_a}^{\tau_n} d\tau_{n-1} \cdots \int_{t_a}^{\tau_3} d\tau_2 \int_{t_a}^{\tau_2} d\tau_1 f(\tau_n, \tau_{n-1}, \dots, \tau_2, \tau_1). \quad (46)$$

Therefore, the two-time correlation function $C_{X_2, X_1}(t_2, t_1)$ up to the second order with respect to the disconnected integral is given by

$$\begin{aligned}
C_{X_2, X_1}(t_2, t_1) &= C_{X_2, X_1}^{(0)}(t_2, t_1) + \Delta C_{X_2, X_1}^{(1)}(t_2, t_1) \\
&+ \Delta C_{X_2, X_1}^{(2)}(t_2, t_1), \quad (47)
\end{aligned}$$

which is diagrammatically represented in Fig. 2. It is easy to see that the two-time arguments of the reservoir correlation function in the disconnected integral are separated by the times t_1 and τ_k . To roughly estimate the order of the disconnected integral, we assume that the reservoir correlation function is given by $\langle R_\mu(\tau_2, t_0) R_\nu(\tau_1, t_0) \rangle_R \sim g f(\tau_2 - \tau_1)$, where the

parameter g stands for a strength of the correlation and $f(\tau)$ a normalized function of time τ . For instance, it is given by $f(\tau) = (1/\tau_R) e^{-|\tau|/\tau_R}$, where τ_R is a correlation time of the Gaussian reservoir. Then, assuming that the reservoir correlation time is short in comparison with a characteristic time of the relevant system, we can see that the contribution of the reservoir correlation $\mathbf{C}_{\mu_j, \mu_k}(\tau_2 - \tau_1)$ function to the disconnected integral is of the order $g\tau_R$. The second order of the disconnected integral is found to be $(g\tau_R)^2$.

Before proceeding further, we briefly comment on the difference between our result and those given in Refs. [52–54], which have been derived by means of the quantum master equation or the projection operator method. It should be noted that the quantum master equation has not been used for deriving the two-time correlation function $C_{X_2, X_1}(t_2, t_1)$ given by Eq. (29). The two-time correlation function based on the projection operator method is calculated perturbatively with respect to the system-reservoir interaction. The approximated two-time correlation function $C_{X_2, X_1}(t_2, t_1)$ given by Eq. (47) can be calculated in terms of the reduced time-evolution operator $V_S(t_2, t_1)$ of the relevant quantum system, which is given by $V_S(t_2, t_1) = \langle e^{L(t_2 - t_1)} \rangle_R$. If we use the projection operator method, the reduced time-evolution operator appearing in the two-time correlation function is replaced by $\tilde{V}_S(t_2, t_1) = V_S(t_2, t_0) V_S^{-1}(t_1, t_0)$ [54] and correction terms to the lowest order are different from $\Delta C_{X_2, X_1}^{(1)}(t_2, t_1)$ and $\Delta C_{X_2, X_1}^{(2)}(t_2, t_1)$. The expansion formula (47) and those given in Refs. [52–54] become equivalent only if the reduced time evolution of the relevant system is described by a dynamical semigroup [4–6]. In this case, the equality $V_S(t_2, t_1) = \tilde{V}_S(t_2, t_1) = e^{\mathcal{L}_S(t_2 - t_1)}$ holds, where \mathcal{L}_S is a generator of the dynamical semigroup. Therefore, it should be considered that our result and those in Refs. [52–54] provide the different expansion formulas for the same two-time correlation function of an open quantum system. It depends on problems for which the expansion formula provides a good approximation.

C. Rotating wave approximation for the two-time correlation function

In this subsection, we introduce the rotating wave approximation [6] in the calculation of the two-time correlation function. In deriving Eq. (29), we have assumed that the interaction between the relevant quantum system and the

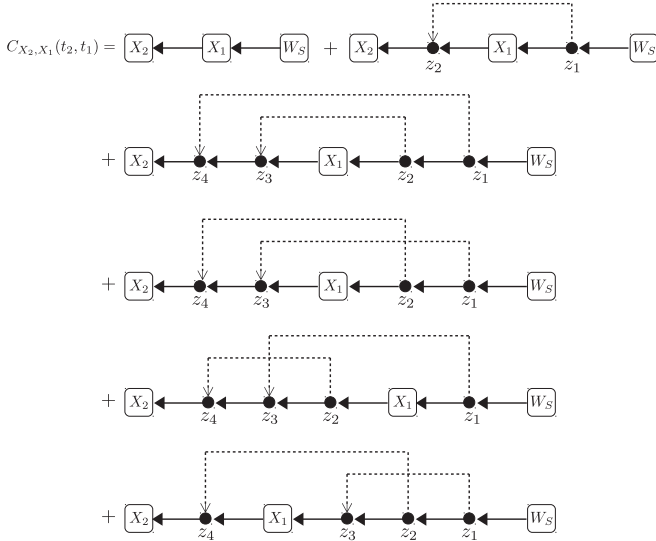


FIG. 2. Diagrammatic representation of the two-time correlation function $C_{X_2, X_1}(t_2, t_1)$ up to the second order with respect to the disconnected integral. In this figure, the arrowed thick and dashed lines from z_k to z_j respectively represent the reduced time evolution superoperator $V_S(t_j, t_k)$ and the reservoir correlation function $\mathbf{C}_{\mu_j, \mu_k}(\tau_j - \tau_k)$; the bullet \bullet with z_j is the system operator \mathbf{S}_{μ_j} . Here we set $z_j = (\tau_j, \mu_j)$. The sum is taken for all μ 's and the time-ordered integration is performed for all τ 's. The first and second terms are the lowest-order correlation function $C_{X_2, X_1}^{(0)}(t_2, t_1)$ and the first-order correction $\Delta C_{X_2, X_1}^{(1)}(t_2, t_1)$. The terms from the third to the sixth provide the second-order correction $\Delta C_{X_2, X_1}^{(2)}(t_2, t_1)$ to the correlation function.

Gaussian thermal reservoir is given by $\hat{H}_{\text{int}}(t, t_0) = S(t, t_0) \otimes R(t, t_0)$ in the interaction picture. Here we decompose the two operators $S(t, t_0)$ and $R(t, t_0)$ into positive and negative frequency parts [24], that is, $S(t, t_0) = S_+(t, t_0) + S_-(t, t_0)$ and

$R(t, t_0) = R_+(t, t_0) + R_-(t, t_0)$ with $S_+^\dagger(t, t_0) = S_-(t, t_0)$ and $R_+^\dagger(t, t_0) = R_-(t, t_0)$. For the sake of simplicity, we assume that $\langle R_+(t_2, t_0) R_+(t_1, t_0) \rangle_R = \langle R_-(t_2, t_0) R_-(t_1, t_0) \rangle_R = 0$. Then we have the equality for the reservoir correlation function,

$$C_R(\tau_2 - \tau_1) + iC_I(\tau_2 - \tau_1) = C_{+-}(\tau_2 - \tau_1) + C_{-+}(\tau_2 - \tau_1), \quad (48)$$

where we set $C_{\pm\mp}(\tau_2 - \tau_1) = \langle R_{\pm}(\tau_2, t_0) R_{\mp}(\tau_1, t_0) \rangle_R$. In this case, Eq. (8) becomes

$$\begin{aligned} \mathcal{G}(\tau_2, \tau_1 | t_0) &= [S_+^\times(\tau_2, t_0) + S_-^\times(\tau_2, t_0)][C_{+-}(\tau_2 - \tau_1) + C_{-+}(\tau_2 - \tau_1)][S_+(\tau_1, t_0) + S_-(\tau_1, t_0)] \\ &\quad - [S_+^\times(\tau_2, t_0) + S_-^\times(\tau_2, t_0)][C_{+-}^*(\tau_2 - \tau_1) + C_{-+}^*(\tau_2 - \tau_1)][\tilde{S}_+(\tau_1, t_0) + \tilde{S}_-(\tau_1, t_0)], \end{aligned} \quad (49)$$

with $\tilde{S}_j \bullet = \bullet S_j$. When applying the rotating wave approximation to Eq. (8), we obtain

$$\begin{aligned} \mathcal{G}(\tau_2, \tau_1 | t_0) &\stackrel{\text{RWA}}{\approx} S_+^\times(\tau_2, t_0) C_{-+}(\tau_2 - \tau_1) S_-(\tau_1, t_0) + S_-^\times(\tau_2, t_0) C_{+-}(\tau_2 - \tau_1) S_+(\tau_1, t_0) \\ &\quad - S_+^\times(\tau_2, t_0) C_{+-}^*(\tau_2 - \tau_1) \tilde{S}_-(\tau_1, t_0) - S_-^\times(\tau_2, t_0) C_{-+}^*(\tau_2 - \tau_1) \tilde{S}_+(\tau_1, t_0) \\ &\equiv \hat{\mathcal{G}}(\tau_2, \tau_1 | t_0). \end{aligned} \quad (50)$$

To proceed further, we introduce the 4×4 matrix,

$$\hat{\mathbf{C}}(t) = \begin{pmatrix} \frac{C_{-+}(t) + C_{+-}^*(t)}{2} & \frac{C_{-+}(t) - C_{+-}^*(t)}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & \frac{C_{+-}(t) + C_{-+}^*(t)}{2} & \frac{C_{+-}(t) - C_{-+}^*(t)}{2} \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad (51)$$

and the superoperator vectors,

$$\hat{\mathbf{S}}^+(t, t_0) = \begin{pmatrix} S_+^\times(t, t_0) \\ S_+^\circ(t, t_0) \\ S_-^\times(t, t_0) \\ S_-^\circ(t, t_0) \end{pmatrix}, \quad \hat{\mathbf{S}}^-(t, t_0) = \begin{pmatrix} S_-^\times(t, t_0) \\ S_-^\circ(t, t_0) \\ S_+^\times(t, t_0) \\ S_+^\circ(t, t_0) \end{pmatrix}. \quad (52)$$

Then $\hat{\mathcal{G}}(\tau_2, \tau_1 | t_0)$ can be expressed as

$$\hat{\mathcal{G}}(\tau_2, \tau_1 | t_0) = \sum_{\mu, \nu} \hat{\mathbf{C}}_{\mu, \nu}(\tau_2 - \tau_1) \hat{\mathbf{S}}_\mu^+(\tau_2, t_0) \hat{\mathbf{S}}_\nu^-(\tau_1, t_0). \quad (53)$$

Thus, in the rotating wave approximation, Eqs. (28) and (29) are replaced by

$$\begin{aligned} C_{X_2, X_1}(t_2, t_1) &= \exp \left(- \sum_{\mu, \nu} \iint_{\mathbb{T}_{t_2 \leftarrow t_0}} d^2 \tau_{21} \hat{\mathbf{C}}_{\mu, \nu}(\tau_2 - \tau_1) \frac{\delta^2}{\delta J_\mu^+(\tau_2) \delta J_\nu^-(\tau_1)} \right) \\ &\quad \times \text{Tr}_S [X_2 e^{L_S(t_2 - t_1)} L^+(t_2, t_1 | J^+(t)) X_1 e^{L_S(t_1 - t_0)} L^-(t_1, t_0 | J^-(t)) W_S] |_{J^\pm(t) \rightarrow 0}, \end{aligned} \quad (54)$$

with

$$L^\pm(t_j, t_k | J^\pm(t)) = T_{(t_j, t_k)}^S \exp \left(- \iint_{\mathbb{T}_{t_j \leftarrow t_k}} d^2 \tau_{21} \hat{\mathcal{G}}(\tau_2, \tau_1 | t_k) + \sum_\mu \int_{t_k}^{t_j} d\tau J_\mu^\pm(\tau) \mathbf{S}_\mu^\pm(\tau, t_k) \right). \quad (55)$$

Furthermore, the expansion formula for the two-time correlation function in the rotating wave approximation can be derived from Eqs. (34), (44), and (45) as follows. (i) The matrix element $\mathbf{C}_{\mu_j \mu_k}(\tau_j, \tau_k)$ is replaced by $\hat{\mathbf{C}}_{\mu_j, \mu_k}(\tau_j, \tau_k)$. (ii) The system operators $\mathbf{S}_{\mu_k}(\tau_k)$ and $\mathbf{S}_{\mu_j}(\tau_j)$, which have the earlier time τ_k and the later one τ_j in the matrix element $\hat{\mathbf{C}}_{\mu_j, \mu_k}(\tau_j, \tau_k)$, are replaced respectively by $\hat{\mathbf{S}}_{\mu_k}^-(\tau_k)$ and $\hat{\mathbf{S}}_{\mu_j}^+(\tau_j)$. (iii) The upper limit of the summation is extended to four.

V. SIMPLE EXAMPLE OF THE CORRELATION FUNCTION

In this section, we calculate the two-time correlation function up to the first order and compare the result with the exact correlation function. For this purpose, we consider an exactly solvable model consisting of a two-level system with transition frequency ω and a set of independent harmonic oscillators with

frequency ω_k [6,55]. The total Hamiltonian is given by

$$H = \frac{1}{2} \hbar \omega \sigma_z + \sum_k \hbar \omega_k a_k^\dagger a_k + \sum_k \hbar (g_k \sigma_+ a_k + g_k^* \sigma_- a_k^\dagger), \quad (56)$$

where σ_z is the Pauli operator, σ_\pm is the raising (lowering) operator, and a_k (a_k^\dagger) is an annihilation (creation) operator of the

k th reservoir oscillator and g_k represents a coupling strength between the two-level system and the reservoir oscillator. Note that the rotating wave approximation is applied to the Hamiltonian (56). The system operator S_{\pm} and the reservoir operator R_{\pm} are respectively given by

$$S_{\pm} = \sigma_{\pm}, \quad R_{-} = R_{+}^{\dagger} = \sum_k \hbar g_k a_k. \quad (57)$$

Furthermore, we assume that the thermal reservoir is initially in the vacuum state so that it becomes Gaussian and the spectral density $J(\Omega)$ of the system-reservoir coupling is given by the Lorentzian distribution with detuning δ ,

$$J(\Omega) = \left(\frac{\gamma}{2\pi}\right) \frac{\lambda^2}{(\Omega - \omega - \delta)^2 + \lambda^2}, \quad (58)$$

where γ is a system-reservoir coupling strength and λ is an inverse of the reservoir correlation time. In the previous work [54,57], the exact two-time correlation function $C_{zz}(t_2, t_1) = \langle \sigma_z(t_2) \sigma_z(t_1) \rangle$ has been obtained,

$$C_{zz}(t_2, t_1) = 1 - [|A(t_2)|^2 + |A(t_1)|^2 - 2A^*(t_2)A(t_2 - t_1)A(t_1)](1 + \langle \sigma_z \rangle), \quad (59)$$

where the time-dependent parameter $A(t)$ is given by

$$A(t) = e^{-i\omega t - \frac{1}{2}(\lambda + i\delta)t} \times \left[\cosh\left(\frac{\lambda + i\delta}{2}at\right) + \frac{1}{a} \sinh\left(\frac{\lambda + i\delta}{2}at\right) \right], \quad (60)$$

with $a = \sqrt{1 - 2\gamma\lambda/(\lambda + i\delta)^2}$. In Eq. (59), we set the initial time $t_0 = 0$ and $\langle \sigma_z \rangle$ stands for an initial average of the Pauli operator σ_z .

To evaluate our expansion formula, we obtain the lowest-order two-time correlation function $C_{zz}^{(0)}(t_2, t_1)$ and the first-

order correction $\Delta C_{zz}^{(1)}(t_2, t_1)$. To calculate these quantities, we need to find the reduced time-evolution operator $V_S(t_j, t_k) = \langle e^{L(t_j - t_k)} \rangle_R$ of the two-level system, where the average is taken with the vacuum state of the reservoir. The exact reduced time-evolution operator $V_S(t_j, t_k)$ can be derived from the reduced density operator $W_S(t)$ of the two-level system, which is obtained by solving the Schrödinger equation of the whole system [6,55]. The application of $V_S(t_j, t_k)$ to σ_z , σ_{\pm} , and 1 is given by

$$V_S(t_j, t_k) \sigma_z = |A(t_j - t_k)|^2 \sigma_z, \quad (61)$$

$$V_S(t_j, t_k) \sigma_{-} = A^*(t_j - t_k) \sigma_{-}, \quad (62)$$

$$V_S(t_j, t_k) \sigma_{+} = A(t_j - t_k) \sigma_{+}, \quad (63)$$

$$V_S(t_j, t_k) 1 = 1 - (1 - |A(t_j - t_k)|^2) \sigma_z, \quad (64)$$

which yields the lowest-order correlation function,

$$C_{zz}^{(0)}(t_2, t_1) = \text{Tr}_S[\sigma_z V_S(t_2, t_1) \sigma_z V_S(t_1, 0) W_S] = 1 - |A(t_1)|^2 (1 - |A(t_2 - t_1)|^2) (1 + \langle \sigma_z \rangle). \quad (65)$$

Different from the exact correlation function $C_{zz}(t_2, t_1)$, the lowest-order correlation function $C_{zz}^{(0)}(t_2, t_1)$ is a real-valued function. It is easy to check that, substituting $t_1 = 0$ into Eqs. (59) and (65), we obtain the equality $C_{zz}(t_2, 0) = C_{zz}^{(0)}(t_2, 0)$ for any γ and λ . In Appendix D, we show that the necessary and sufficient condition that the equality $C_{zz}(t_2, t_1) = C_{zz}^{(0)}(t_2, t_1)$ is satisfied for any t_2 and t_1 with $t_2 > t_1 \geq 0$ is that the time-dependent parameter $A(t)$ satisfies $A(t_1)A(t_2) = A(t_1 + t_2)$. Since $A(t)$ is given by Eq. (60), the condition is equivalent to $\lambda \gg \gamma$. In this case, the reduced time evolution of the two-level system is described by a dynamical semigroup.

Next we obtain the first-order correction $\Delta C_{zz}^{(1)}(t_2, t_1)$ to the correlation function. Since the Hamiltonian is given in the rotating wave approximation, the first-order correction is

$$\Delta C_{zz}^{(1)}(t_2, t_1) = - \sum_{\mu, \nu=1}^4 \iint_{\mathbb{T}_{t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \hat{C}_{\mu, \nu}(\tau_2 - \tau_1) \text{Tr}_S[\sigma_z V_S(t_2, \tau_2) \hat{S}_{\mu}^{+} V_S(\tau_2, t_1) \sigma_z V_S(t_1, \tau_1) \hat{S}_{\nu}^{-} V_S(\tau_1, 0) W_S]. \quad (66)$$

In this equation, the matrix $\hat{C}(\tau_2 - \tau_1)$ and the vectors \hat{S}^{\pm} of superoperators are given respectively by

$$\hat{C}(\tau_2 - \tau_1) = \frac{1}{2} \begin{pmatrix} f(\tau_2 - \tau_1) & f(\tau_2 - \tau_1) & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & f^*(\tau_2 - \tau_1) & -f^*(\tau_2 - \tau_1) \\ 0 & 0 & 0 & 0 \end{pmatrix} \quad (67)$$

and

$$\hat{S}^{+} = \begin{pmatrix} \sigma_{+}^{\times} \\ \sigma_{+}^{\circ} \\ \sigma_{-}^{\times} \\ \sigma_{-}^{\circ} \end{pmatrix}, \quad \hat{S}^{-} = \begin{pmatrix} \sigma_{-}^{\times} \\ \sigma_{-}^{\circ} \\ \sigma_{+}^{\times} \\ \sigma_{+}^{\circ} \end{pmatrix}, \quad (68)$$

with $f(t) = \frac{1}{2} \gamma \lambda e^{-i(\omega + \delta)t - \lambda t}$. Then $\Delta C_{zz}^{(1)}(t_2, t_1)$ becomes

$$\Delta C_{zz}^{(1)}(t_2, t_1) = - \iint_{\mathbb{T}_{t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} [f(\tau_2 - \tau_1) G_1(\tau_2, \tau_1) - f^*(\tau_2 - \tau_1) G_2(\tau_2, \tau_1)], \quad (69)$$

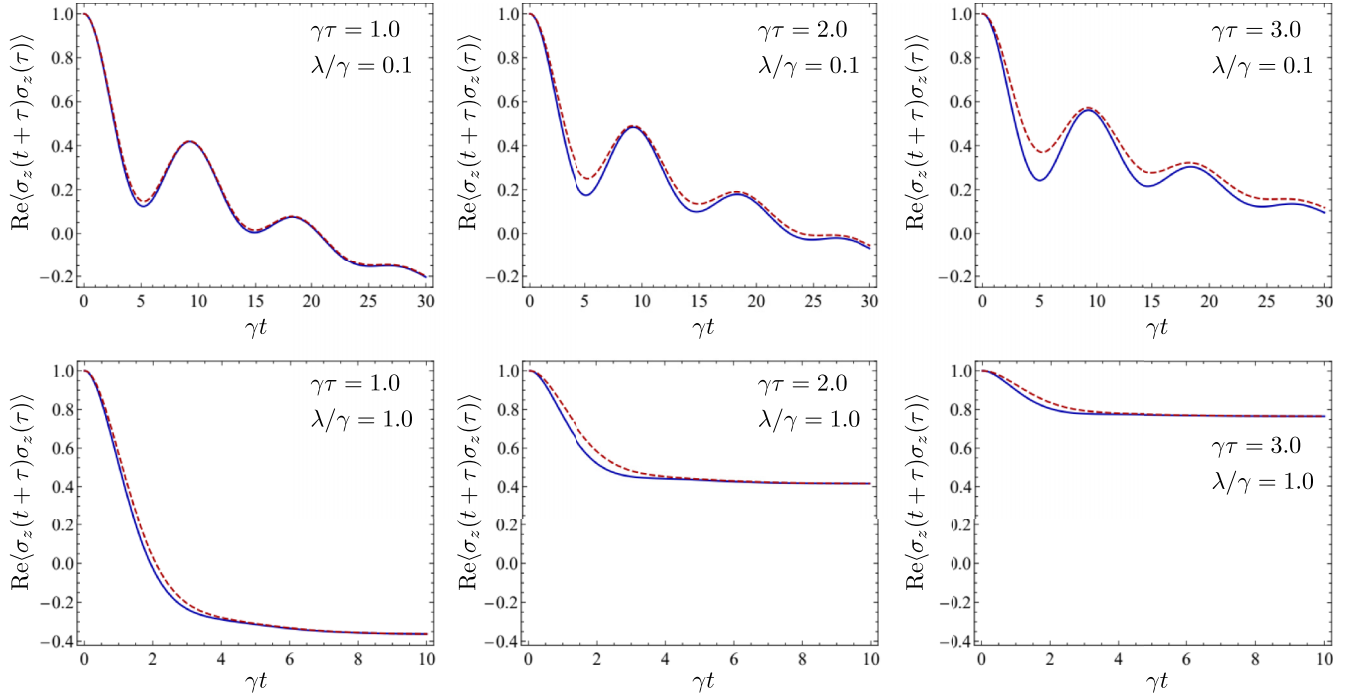


FIG. 3. Real part of the two-time correlation function $\langle \sigma_z(t+\tau)\sigma_z(\tau) \rangle$ of the Pauli operator σ_z , where the solid (blue) line stands for the exact correlation function and the dashed (red) line for the approximated one up to the first order. In the figure, we set $\delta/\gamma = 0.5$ and $\langle \sigma_z \rangle = 1$.

with

$$G_1(\tau_2, \tau_1) = \text{Tr}_S[\sigma_z V_S(t_2, \tau_2) \sigma_+^\times V_S(\tau_2, t_1) \sigma_z V_S(t_1, \tau_1) \sigma_- V_S(\tau_1, 0) W_S], \quad (70)$$

$$G_2(\tau_2, \tau_1) = \text{Tr}_S[\sigma_z V_S(t_2, \tau_2) \sigma_-^\times V_S(\tau_2, t_1) \sigma_z V_S(t_1, \tau_1) \tilde{\sigma}_+ V_S(\tau_1, 0) W_S], \quad (71)$$

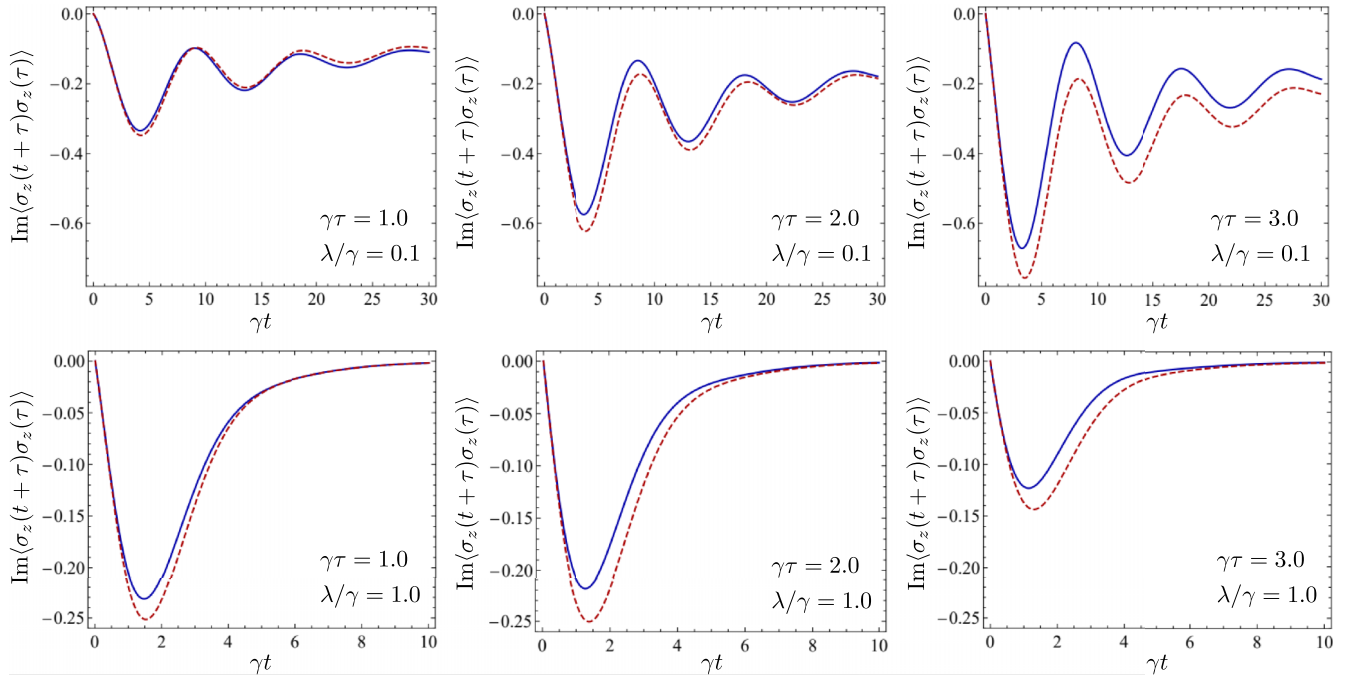


FIG. 4. Imaginary part of the two-time correlation function $\langle \sigma_z(t+\tau)\sigma_z(\tau) \rangle$ of the Pauli operator σ_z with $\delta/\gamma = 0.5$ and $\langle \sigma_z \rangle = 1$. In the figure, the solid (blue) line stands for the exact correlation function and the dashed (red) line for the approximated one up to the first order.

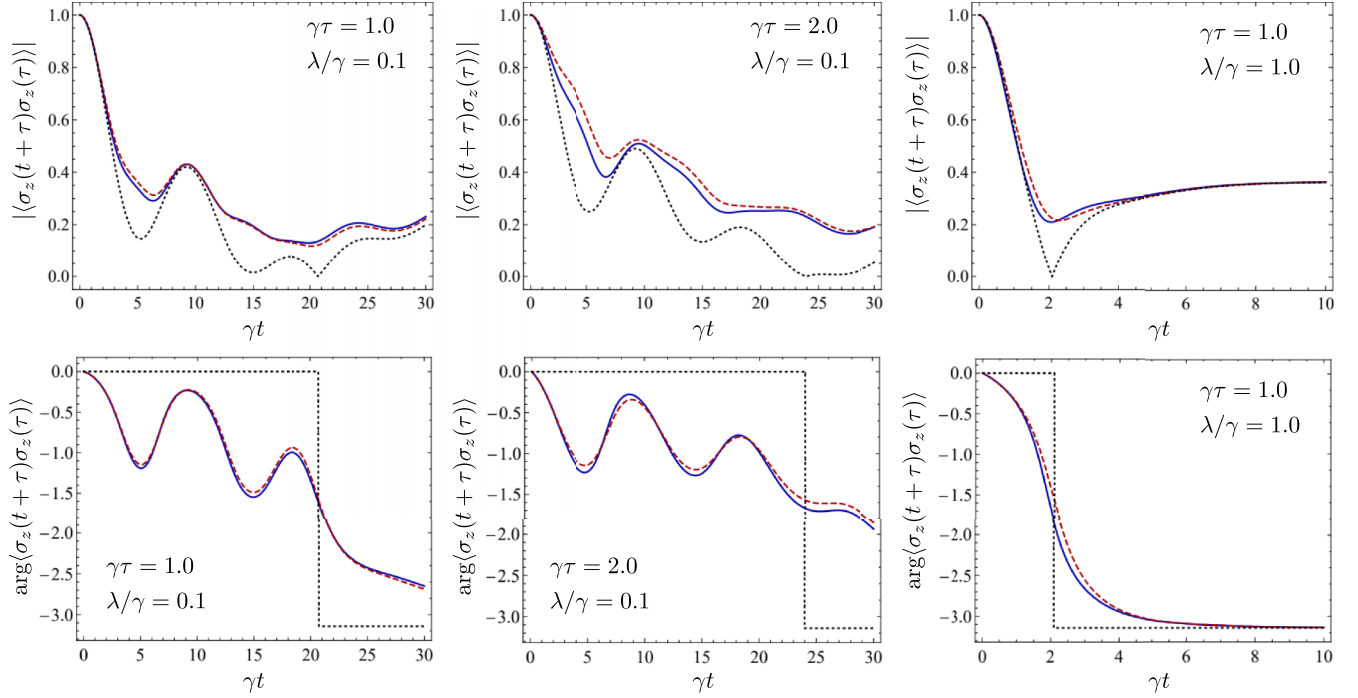


FIG. 5. Absolute value and argument of the two-time correlation function $\langle\sigma_z(t+\tau)\sigma_z(\tau)\rangle$ of the Pauli operator σ_z , where the solid (blue) line stands for the exact correlation function, the dashed (red) line for the one including the first-order correction, and the dotted (black) line for the lowest-order correlation function. In the figure, we set $\delta/\gamma = 0.5$ and $\langle\sigma_z\rangle = 1$.

which are calculated to be

$$G_1(\tau_2, \tau_1) = -(1 + \langle\sigma_z\rangle)|A(\tau_1)|^2 A^*(t_1 - \tau_1) A^*(\tau_2 - t_1) |A(t_2 - \tau_2)|^2, \quad (72)$$

$$G_2(\tau_2, \tau_1) = -(1 + \langle\sigma_z\rangle)|A(\tau_1)|^2 A(t_1 - \tau_1) A(\tau_2 - t_1) |A(t_2 - \tau_2)|^2. \quad (73)$$

Thus we obtain the first-order correction to the two-time correlation function,

$$\Delta C_{zz}^{(1)}(t_2, t_1) = -i\gamma\lambda(1 + \langle\sigma_z\rangle) \iint_{\substack{T_1^{\tau_2} \leftarrow t_1 \\ T_1^{\tau_1} \leftarrow 0}} d^2\tau_{21} e^{-\lambda(\tau_2 - \tau_1)} |A(\tau_1)|^2 \text{Im}[e^{i(\omega + \delta)(\tau_2 - \tau_1)} A(t_1 - \tau_1) A(\tau_2 - t_1)] |A(t_2 - \tau_2)|^2. \quad (74)$$

It is found that, although the lowest-order correlation function $C_{zz}^{(0)}(t_2, t_1)$ is real, the first-order correction $\Delta C_{zz}^{(1)}(t_2, t_1)$ yields an imaginary part of the correlation function. Then we compare the approximated correlation function up to the first order, $C_{zz}^{(0)}(t_2, t_1) + \Delta C_{zz}^{(1)}(t_2, t_1)$, with the exact correlation function $C_{zz}(t_2, t_1)$. The result is depicted in Figs. 3 and 4. It is found from the figures that the correlation function provided by our formula is a good approximation even if the Gaussian thermal reservoir has a finite correlation time. Furthermore, to compare the exact, the first-order, and the lowest-order correlation functions, we plot the amplitude $|\langle\sigma_z(t+\tau)\sigma_z(\tau)\rangle|$ and the argument $\arg(\langle\sigma_z(t+\tau)\sigma_z(\tau)\rangle)$ in Fig. 5.

VI. SUMMARY

In this paper, we have considered a two-time correlation function of an open quantum system interacting with a Gaussian thermal reservoir. We have derived the exact formula for the correlation function which is given by making use of the functional derivative with respect to fictitious fields.

We have also developed the perturbative expansion, where a small expansion parameter is related to the correlation time of the thermal reservoir via the disconnected integral and thus the higher-order terms become negligible if the correlation time of the thermal reservoir is sufficiently small. We have explicitly obtained the correlation function up to the second order with respect to the disconnected integral. The diagrammatic representation of the perturbative expansion has also been presented. Using a model consisting of a two-level system and independent harmonic oscillators, which is exactly solvable if the total system does not have more than a single excitation, we have calculated the two-time correlation function of the Pauli operator σ_z up to the first order. Comparing the approximated correlation function with the exact one, we have found that our expansion formula provides a good approximation. In this paper, we have assumed that there is no initial correlation between a relevant quantum system and a Gaussian thermal reservoir. However, our result may be generalized to the case that a relevant quantum system has initial correlation as long as a thermal reservoir is Gaussian.

APPENDIX A: REDUCED TIME EVOLUTION WITH INITIAL CORRELATION

In this Appendix, we briefly consider the reduced density operator of a relevant quantum system which is initially correlated with a Gaussian thermal reservoir. Assuming the interaction Hamiltonian $H_{SR}(t, t_0) = S(t, t_0) \otimes R(t, t_0)$ in the interaction picture, we have the time-evolved state $W(t) = U(t, t_0) W U^\dagger(t, t_0)$ of the whole system. The unitary operator $U(t, t_0)$ is given by

$$U(t, t_0) = T_S T_R \exp \left[-i \int_{t_0}^t d\tau S(\tau, t_0) \otimes R(\tau, t_0) \right], \quad (\text{A1})$$

where T_S (T_R) stands for the usual time ordering of the system (reservoir) operators $S(t, t_0)$'s [$R(t, t_0)$'s]. Furthermore, we assume the commutation relation $[R(t_2, t_0), R(t_1, t_0)] = 2i f(t_2, t_1)$, where $f(t_2, t_1)$ is some c -number function. Then we can remove the time ordering T_R from the unitary operator $U(t, t_0)$,

$$U(t, t_0) = T_S \exp \left[-i \int_{t_0}^t d\tau S(\tau, t_0) \otimes R(\tau, t_0) - i \iint_{\mathbb{T}_{t \leftarrow t_0}} d^2 \tau_{21} f(\tau_2, \tau_1) S(\tau_2, t_0) S(\tau_1, t_0) \right], \quad (\text{A2})$$

where we have used the operator identity [58],

$$T \exp \left[-i \int_{t_1}^{t_2} d\tau X(\tau, t_0) \right] = \exp \left[-i \int_{t_1}^{t_2} d\tau X(\tau, t_0) - \frac{1}{2} \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{21} [X(\tau_2, t_0), X(\tau_1, t_0)] \right], \quad (\text{A3})$$

with $[X(\tau_2, t_0), X(\tau_1, t_0)]$ being a c -number function. Hence we can derive the reduced density operator $W_S(t) = \text{Tr}_R W(t)$ of the relevant quantum system from Eq. (A2),

$$W_S(t) = T_S \left\{ \exp \left[-i \iint_{\mathbb{T}_{t \leftarrow t_0}} d^2 \tau_{21} f(\tau_2, \tau_1) S^\times(\tau_2, t_0) S^\circ(\tau_1, t_0) \right] \text{Tr}_R \left[\exp \left(-i \int_{t_0}^t d\tau S^\times(\tau, t_0) \otimes R(\tau, t_0) \right) W \right] \right\}. \quad (\text{A4})$$

In terms of complete orthonormal set $\{|\psi_k\rangle | k \in \mathcal{S}\}$ of the relevant quantum system, we express the initial state W as $W = \sum_{j,k \in \mathcal{S}} p_{jk} |\psi_j\rangle \langle \psi_k| \otimes W_R^{jk}$, where $p_{jk} = \text{Tr}_R \langle \psi_j | W | \psi_k \rangle$ and $W_R^{jk} = (1/p_{jk}) \langle \psi_j | W | \psi_k \rangle$. Substituting this into Eq. (A4), we obtain

$$\begin{aligned} W_S(t) &= \sum_{j,k \in \mathcal{S}} p_{jk} T_S \left\{ \exp \left[-i \iint_{\mathbb{T}_{t \leftarrow t_0}} d^2 \tau_{21} f(\tau_2, \tau_1) S^\times(\tau_2, t_0) S^\circ(\tau_1, t_0) \right] \right. \\ &\quad \left. \times \text{Tr}_R \left[\exp \left(-i \int_{t_0}^t d\tau S^\times(\tau, t_0) \otimes R(\tau, t_0) \right) W_R^{jk} \right] \right\} |\psi_j\rangle \langle \psi_k|. \end{aligned} \quad (\text{A5})$$

Here we note that, although W_R^{jk} is not a density operator, the cumulant expansion technique [59,60] can be applied since the equality $\text{Tr}_R W_R^{jk} = 1$ is satisfied. Assuming that the quasidensity operator W_R^{jk} is still Gaussian, we can calculate the trace in Eq. (A5),

$$\begin{aligned} &\text{Tr}_R \left[\exp \left(-i \int_{t_0}^t d\tau S^\times(\tau, t_0) \otimes R(\tau, t_0) \right) W_R^{jk} \right] \\ &= \exp \left[-i \int_{t_0}^t d\tau \langle R(\tau, t_0) \rangle_{jk} S^\times(\tau, t_0) - \iint_{\mathbb{T}_{t \leftarrow t_0}} d^2 \tau_{21} \text{Re} \langle R(\tau_2, t_0) R(\tau_1, t_0) \rangle_{jk}^c S^\times(\tau_2, t_0) S^\times(\tau_1, t_0) \right], \end{aligned} \quad (\text{A6})$$

with $\langle R(\tau, t_0) \rangle_{jk} = \text{Tr}_R [R(\tau, t_0) W_R^{jk}]$ and

$$\langle R(\tau_2, t_0) R(\tau_1, t_0) \rangle_{jk}^c = \langle R(\tau_2, t_0) R(\tau_1, t_0) \rangle_{jk} - \langle R(\tau_2, t_0) \rangle_{jk} \langle R(\tau_1, t_0) \rangle_{jk}. \quad (\text{A7})$$

Substituting this equation into Eq. (A5) and using $f(\tau_2, \tau_1) = \text{Im} \langle R(\tau_2, t_0) R(\tau_1, t_0) \rangle_{jk}^c$, we obtain the reduced density operator of the relevant quantum system,

$$W_S(t) = \sum_{\mu, \nu \in \mathcal{S}} p_{\mu\nu} T_S \exp \left[- \int_{t_0}^t d\tau \bar{\mathcal{G}}_{\mu\nu}(\tau, t_0) \right] |\psi_\mu\rangle \langle \psi_\nu|, \quad (\text{A8})$$

with

$$\bar{\mathcal{G}}_{jk}(t, t_0) = i \langle R(t, t_0) \rangle_{jk} S^\times(t, t_0) + \int_{t_0}^t d\tau S^\times(t, t_0) [C_R^{jk}(t, \tau) S^\times(\tau, t_0) + i C_I^{jk}(t, \tau) S^\circ(\tau, t_0)]. \quad (\text{A9})$$

In this equation, we set $C_{jk}(t_2, t_1) = \langle R(t_2, t_0) R(t_1, t_0) \rangle_{jk}^c = C_R^{jk}(t_2, t_1) + i C_I^{jk}(t_2, t_1)$. If there is no initial correlation, $\bar{\mathcal{G}}_{jk}(t, t_0)$ becomes independent of j and k and thus Eq. (A8) reduces to Eq. (3). The perturbative expansion for the time-local and time-nonlocal quantum master equations with initial correlation has been developed in Ref. [61].

APPENDIX B: DERIVATION OF EQ. (30)

In this Appendix, we show that the formula (29) yields the average value of an observable with the reduced density operator. First substituting $X_2 = 1$ into Eq. (29), we obtain

$$C_{1,X_1}(t_2, t_1) = \exp \left(- \sum_{\mu, v} \iint_{\mathbb{T}_{t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathbf{C}_{\mu, v}(\tau_2 - \tau_1) \frac{\delta^2}{\delta J_{\mu}^+(\tau_2) \delta J_{\nu}^-(\tau_1)} \right) \times \text{Tr}_S [e^{L_S(t_2-t_1)} L(t_2, t_1 | J^+(t)) X_1 e^{L_S(t_1-t_0)} L(t_1, t_0 | J^-(t)) W_S] |_{J^{\pm}(t) \rightarrow 0}. \quad (\text{B1})$$

Here we note that $\text{Tr}_S L_S \bullet = \text{Tr}_S S^{\times}(t, t') \bullet = 0$ and $\mathbf{C}_{2,1}(\tau_2 - \tau_1) = \mathbf{C}_{2,2}(\tau_2 - \tau_1) = 0$. Then this equation becomes

$$C_{1,X_1}(t_2, t_1) = \text{Tr}_S [X_1 e^{L_S(t_1-t_0)} L(t_1, t_0 | 0) W_S] = \text{Tr}_S [X_1 V_S(t_1, t_0) W_S] = \text{Tr}_S [X_1 W_S(t_1)], \quad (\text{B2})$$

where we have used Eqs. (3)–(5), (7), and (28). Next substituting $X_1 = 1$ into Eq. (29), we obtain

$$\begin{aligned} C_{X_2,1}(t_2, t_1) &= \exp \left(- \sum_{\mu, v} \iint_{\mathbb{T}_{t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathbf{C}_{\mu, v}(\tau_2 - \tau_1) \frac{\delta^2}{\delta J_{\mu}^+(\tau_2) \delta J_{\nu}^-(\tau_1)} \right) \\ &\quad \times \text{Tr}_S [X_2 e^{L_S(t_2-t_1)} L(t_2, t_1 | J^+(t)) e^{L_S(t_1-t_0)} L(t_1, t_0 | J^-(t)) W_S] |_{J^{\pm}(t) \rightarrow 0} \\ &= \exp \left(- \sum_{\mu, v} \iint_{\mathbb{T}_{t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathbf{C}_{\mu, v}(\tau_2 - \tau_1) \frac{\delta^2}{\delta J_{\mu}^+(\tau_2) \delta J_{\nu}^-(\tau_1)} \right) \\ &\quad \times \text{Tr}_S [X_2 e^{L_S(t_2-t_0)} L(t_2, t_0 | J^+(t)) L(t_1, t_0 | J^-(t)) W_S] |_{J^{\pm}(t) \rightarrow 0} \\ &= \exp \left(- \sum_{\mu, v} \iint_{\mathbb{T}_{t_1}^{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathbf{C}_{\mu, v}(\tau_2 - \tau_1) \frac{\delta^2}{\delta J_{\mu}^+(\tau_2) \delta J_{\nu}^-(\tau_1)} \right) \\ &\quad \times \text{Tr}_S \left\{ X_2 e^{L_S(t_2-t_0)} T_{(t_2, t_0)}^S \exp \left[- \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) - \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right. \right. \\ &\quad \left. \left. + \sum_{\mu} \int_{t_1}^{t_2} d\tau J_{\mu}^+(\tau) \mathbf{S}_{\mu}(\tau, t_0) + \sum_{\nu} \int_{t_0}^{t_1} d\tau J_{\nu}^-(\tau) \mathbf{S}_{\nu}(\tau, t_0) \right] W_S \right\} |_{J^{\pm}(t) \rightarrow 0} \\ &= \text{Tr}_S \left[X_2 e^{L_S(t_2-t_0)} T_{(t_2, t_0)}^S \exp \left(- \iint_{\mathbb{T}_{t_2 \leftarrow t_0}} d^2 \tau_{21} \mathcal{G}(\tau_2, \tau_1 | t_0) \right) W_S \right] \\ &= \text{Tr}_S [X_2 V_S(t_2, t_0) W_S] = \text{Tr}_S [X_2 W_S(t_2)]. \end{aligned} \quad (\text{B3})$$

In deriving this equation, we have used $\mathbb{T}_{t_2 \leftarrow t_0} = \mathbb{T}_{t_2 \leftarrow t_1} + \mathbb{T}_{t_1}^{t_2 \leftarrow t_1} + \mathbb{T}_{t_1 \leftarrow t_0}$ (see Fig. 1). Therefore, we have derived Eq. (30).

APPENDIX C: CALCULATION OF THE SECOND-ORDER CORRECTION TERMS

We derive the second-order correction $\Delta C_{X_2, X_1}^{(2)}(t_2, t_1)$ to the two-time correlation function, which is given by Eq. (45). The second-order correction consists of two parts. (i) One is obtained from Eq. (38), in which the first order with respect to the disconnected integral is taken into account in Eqs. (39) and (40). (ii) The other comes from the term with $n = 2$ in the sum on the right-hand side of Eq. (26).

We first obtain the contribution (i). Thanks to the time-ordering operation, expanding the exponential, we can obtain the first-order term of $L_{\nu}(t_1, t_0; \tau_1 | t_0)$ with respect to the t_1 -disconnected integral,

$$\begin{aligned} \Delta L_{\nu}^{(1)}(t_1, t_0; \tau_1 | t_0) &= -T_{(t_1, t_0)}^S \left[\iint_{\mathbb{T}_{t_1}^{t_1 \leftarrow \tau_1}} d^2 \tau'_{21} \mathbf{S}_{\nu}(\tau_1, t_0) \mathcal{G}(\tau'_2, \tau'_1 | t_0) \right. \\ &\quad \left. \times \exp \left(- \iint_{\mathbb{T}_{t_1 \leftarrow \tau_1}} d^2 \tau''_{21} \mathcal{G}(\tau''_2, \tau''_1 | t_0) \right) \exp \left(- \iint_{\mathbb{T}_{\tau_1 \leftarrow t_0}} d^2 \tau''_{21} \mathcal{G}(\tau''_2, \tau''_1 | t_0) \right) \right]. \end{aligned} \quad (\text{C1})$$

Note that the two exponentials in this equation include the τ'_2 - and τ'_1 -disconnected integrals, where these times appear in $\mathcal{G}(\tau'_2, \tau'_1|t_0)$. Then we can approximate up to the first order as

$$\begin{aligned} \iint_{\mathbb{T}_{t_1 \leftarrow \tau_1}} d^2 \tau''_{21} \bullet &= \iint_{\mathbb{T}_{t_1 \leftarrow \tau'_2}} d^2 \tau''_{21} \bullet + \iint_{\mathbb{T}_{\tau'_2 \leftarrow \tau_1}} d^2 \tau''_{21} \bullet + \iint_{\mathbb{T}_{\tau'_2 \leftarrow \tau_1}} d^2 \tau''_{21} \bullet \\ &\approx \iint_{\mathbb{T}_{t_1 \leftarrow \tau'_2}} d^2 \tau''_{21} \bullet + \iint_{\mathbb{T}_{\tau'_2 \leftarrow \tau_1}} d^2 \tau''_{21} \bullet, \end{aligned} \quad (C2)$$

$$\begin{aligned} \iint_{\mathbb{T}_{\tau_1 \leftarrow t_0}} d^2 \tau''_{21} \bullet &= \iint_{\mathbb{T}_{\tau_1 \leftarrow \tau'_1}} d^2 \tau''_{21} \bullet + \iint_{\mathbb{T}_{\tau'_1 \leftarrow t_0}} d^2 \tau''_{21} \bullet + \iint_{\mathbb{T}_{\tau'_1 \leftarrow t_0}} d^2 \tau''_{21} \bullet \\ &\approx \iint_{\mathbb{T}_{\tau_1 \leftarrow \tau'_1}} d^2 \tau''_{21} \bullet + \iint_{\mathbb{T}_{\tau'_1 \leftarrow t_0}} d^2 \tau''_{21} \bullet, \end{aligned} \quad (C3)$$

where we have used the fact that $\tau''_2, \tau''_1 > \tau'_1$ in Eq. (C2) and $\tau'_1 > \tau''_2, \tau''_1$ in Eq. (C3). Substituting these approximations into Eq. (C1), after some calculation we obtain the first-order term of $L_v(t_1, t_0; \tau_1|t_0)$:

$$\Delta L_v^{(1)}(t_1, t_0; \tau_1|t_0) = -e^{-L_S(t_1-t_0)} \sum_{\mu', \nu'} \iint_{\mathbb{T}_{\tau_1 \leftarrow t_0}} d^2 \tau'_{21} \mathbf{C}_{\mu', \nu'}(\tau'_2 - \tau'_1) V_S(t_1, \tau'_2) \mathbf{S}_{\mu'} V_S(\tau'_2, \tau_1) \mathbf{S}_{\nu'} V_S(\tau_1, \tau'_1) \mathbf{S}_{\nu'} V_S(\tau'_1, t_0). \quad (C4)$$

Then, up to the first order, $e^{L_S(t_1-t_0)} L_v(t_1, t_0; \tau_1|t_0)$ is given by

$$\begin{aligned} e^{L_S(t_1-t_0)} L_v(t_1, t_0; \tau_1|t_0) &\approx V_S(t_1, \tau_1) \mathbf{S}_{\nu} V_S(\tau_1, t_0) - \sum_{\mu', \nu'} \iint_{\mathbb{T}_{\tau_1 \leftarrow t_0}} d^2 \tau'_{21} \mathbf{C}_{\mu', \nu'}(\tau'_2 - \tau'_1) V_S(t_1, \tau'_2) \mathbf{S}_{\mu'} V_S(\tau'_2, \tau_1) \mathbf{S}_{\nu} V_S(\tau_1, \tau'_1) \mathbf{S}_{\nu'} V_S(\tau'_1, t_0). \end{aligned} \quad (C5)$$

In the same way, we can derive up to the first order

$$\begin{aligned} e^{L_S(t_2-t_1)} L_{\mu}(t_2, t_1; \tau_2|t_1) &\approx V_S(t_2, \tau_2) \mathbf{S}_{\mu} V_S(\tau_2, t_1) - \sum_{\mu', \nu'} \iint_{\mathbb{T}_{\tau_2 \leftarrow t_1}} d^2 \tau'_{21} \mathbf{C}_{\mu', \nu'}(\tau'_2 - \tau'_1) V_S(t_2, \tau'_2) \mathbf{S}_{\mu'} V_S(\tau'_2, \tau_2) \mathbf{S}_{\mu} V_S(\tau_2, \tau'_1) \mathbf{S}_{\nu'} V_S(\tau'_1, t_1). \end{aligned} \quad (C6)$$

Substituting Eqs. (C5) and (C6) into Eq. (38) and picking up the second-order terms with respect to the disconnected integral, we obtain the contribution (i),

$$\begin{aligned} \Delta^{(i)} C_{X_2, X_1}^{(2)}(t_2, t_1) &= \sum_{\mu, \nu} \sum_{\mu', \nu'} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{21} \iint_{\mathbb{T}_{\tau_1 \leftarrow t_0}} d^2 \tau'_{21} \mathbf{C}_{\mu, \nu}(\tau_2 - \tau_1) \mathbf{C}_{\mu', \nu'}(\tau'_2 - \tau'_1) \\ &\quad \times \text{Tr}_S[X_2 V_S(t_2, \tau_2) \mathbf{S}_{\mu} V_S(\tau_2, t_1) X_1 V_S(t_1, \tau'_2) \mathbf{S}_{\mu'} V_S(\tau'_2, \tau_1) \mathbf{S}_{\nu} V_S(\tau_1, \tau'_1) \mathbf{S}_{\nu'} V_S(\tau'_1, t_0) W_S] \\ &\quad + \sum_{\mu, \nu} \sum_{\mu', \nu'} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{21} \iint_{\mathbb{T}_{\tau_2 \leftarrow t_1}} d^2 \tau'_{21} \mathbf{C}_{\mu, \nu}(\tau_2 - \tau_1) \mathbf{C}_{\mu', \nu'}(\tau'_2 - \tau'_1) \\ &\quad \times \text{Tr}_S[X_2 V_S(t_2, \tau'_2) \mathbf{S}_{\mu'} V_S(\tau'_2, \tau_2) \mathbf{S}_{\mu} V_S(\tau_2, \tau'_1) \mathbf{S}_{\nu'} V_S(\tau'_1, t_1) X_1 V_S(t_1, \tau_1) \mathbf{S}_{\nu} V_S(\tau_1, t_0) W_S]. \end{aligned} \quad (C7)$$

Next, in order to obtain the contribution (ii), picking up the term with $n = 2$ in the sum on the right-hand side of Eq. (26), we have

$$\begin{aligned} \Delta^{(ii)} C_{X_2, X_1}^{(2)}(t_2, t_1) &= \frac{1}{2} \sum_{\mu, \nu} \sum_{\mu', \nu'} \iint_{\mathbb{T}_{t_2 \leftarrow t_0}} d^2 \tau_{21} \iint_{\mathbb{T}_{t_2 \leftarrow t_0}} d^2 \tau'_{21} \mathbf{C}_{\mu, \nu}(\tau_2 - \tau_1) \mathbf{C}_{\mu', \nu'}(\tau'_2 - \tau'_1) \\ &\quad \times \text{Tr}_S[X_2 e^{L_S(t_2-t_1)} L_{\mu, \mu'}(t_2, t_1; \tau_2, \tau'_2|t_1) X_1 e^{L_S(t_1-t_0)} L_{\nu, \nu'}(t_1, t_0; \tau_1, \tau'_1|t_0) W_S], \end{aligned} \quad (C8)$$

with

$$L_{\nu, \nu'}(t_1, t_0; \tau_1, \tau'_1|t_0) = T_{(t_1, t_0)}^S \left[\mathbf{S}_{\nu}(\tau_1, t_0) \mathbf{S}_{\nu'}(\tau'_1, t_0) \exp \left(- \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau''_{21} \mathcal{G}(\tau''_2, \tau''_1|t_0) \right) \right], \quad (C9)$$

$$L_{\mu, \mu'}(t_2, t_1; \tau_2, \tau'_2|t_1) = T_{(t_2, t_1)}^S \left[\mathbf{S}_{\mu}(\tau_2, t_1) \mathbf{S}_{\mu'}(\tau'_2, t_1) \exp \left(- \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau''_{21} \mathcal{G}(\tau''_2, \tau''_1|t_1) \right) \right]. \quad (C10)$$

Using the fact that the integral on the right-hand side of Eq. (C8) is decomposed into

$$\begin{aligned} \iint_{\mathbb{T}_{t_2 \leftarrow t_0}} d^2 \tau_{43} \iint_{\mathbb{T}_{t_2 \leftarrow t_0}} d^2 \tau_{21} \bullet &= \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{42} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{31} \bullet + \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{24} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{13} \bullet \\ &+ \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{42} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{13} \bullet + \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{24} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{31} \bullet, \end{aligned} \quad (\text{C11})$$

we can rewrite Eq. (C8) into

$$\begin{aligned} \Delta^{(\text{ii})} C_{X_2, X_1}^{(2)}(t_2, t_1) &= \sum_{\mu_4, \mu_3, \mu_2, \nu_1} \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{42} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{31} \mathbf{C}_{\mu_4, \mu_3}(\tau_4 - \tau_3) \mathbf{C}_{\mu_2, \nu_1}(\tau_2 - \tau_1) \\ &\times \text{Tr}_S [X_2 e^{L_S(t_2 - t_1)} L_{\mu_4, \mu_2}(t_2, t_1; \tau_4, \tau_2 | t_1) X_1 e^{L_S(t_1 - t_0)} L_{\mu_3, \nu_1}(t_1, t_0; \tau_3, \tau_1 | t_0) W_S] \\ &+ \sum_{\mu_4, \mu_3, \mu_2, \nu_1} \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{42} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{13} \mathbf{C}_{\mu_4, \mu_3}(\tau_4 - \tau_3) \mathbf{C}_{\mu_2, \mu_1}(\tau_2 - \tau_1) \\ &\times \text{Tr}_S [X_2 e^{L_S(t_2 - t_1)} L_{\mu_4, \mu_2}(t_2, t_1; \tau_4, \tau_2 | t_1) X_1 e^{L_S(t_1 - t_0)} L_{\mu_3, \mu_1}(t_1, t_0; \tau_3, \tau_1 | t_0) W_S]. \end{aligned} \quad (\text{C12})$$

Up to the lowest order with respect to the $\tau_{2,1}$ - and $\tau'_{2,1}$ -disconnected integrals, the superoperators $L_{\nu, \nu'}(t_1, t_0; \tau_1, \tau'_1 | t_0)$ and $L_{\mu, \mu'}(t_2, t_1; \tau_2, \tau'_2 | t_1)$ are approximated as

$$L_{\nu, \nu'}(t_1, t_0; \tau_1, \tau'_1 | t_0) \approx \begin{cases} e^{-L_S(t_1 - t_0)} V_S(t_1, \tau_1) \mathbf{S}_{\nu'} V_S(\tau_1, \tau'_1) \mathbf{S}_{\nu} V_S(\tau'_1, t_0) & (\tau_1 > \tau'_1), \\ e^{-L_S(t_1 - t_0)} V_S(t_1, \tau'_1) \mathbf{S}_{\nu'} V_S(\tau'_1, \tau_1) \mathbf{S}_{\nu} V_S(\tau_1, t_0) & (\tau_1 < \tau'_1), \end{cases} \quad (\text{C13})$$

$$L_{\mu, \mu'}(t_2, t_1; \tau_2, \tau'_2 | t_1) \approx \begin{cases} e^{-L_S(t_2 - t_1)} V_S(t_2, \tau_2) \mathbf{S}_{\mu'} V_S(\tau_2, \tau'_2) \mathbf{S}_{\mu} V_S(\tau'_2, t_1) & (\tau_2 > \tau'_2), \\ e^{-L_S(t_2 - t_1)} V_S(t_2, \tau'_2) \mathbf{S}_{\mu'} V_S(\tau'_2, \tau_2) \mathbf{S}_{\mu} V_S(\tau_2, t_1) & (\tau_2 < \tau'_2). \end{cases} \quad (\text{C14})$$

Then we obtain the terms from the contribution (ii),

$$\begin{aligned} \Delta^{(\text{ii})} C_{X_2, X_1}^{(2)}(t_2, t_1) &= \sum_{\mu_4, \mu_3, \mu_2, \mu_1} \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{42} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{31} \mathbf{C}_{\mu_4, \mu_3}(\tau_4 - \tau_3) \mathbf{C}_{\mu_2, \mu_1}(\tau_2 - \tau_1) \\ &\times \text{Tr}_S [X_2 V_S(t_2, \tau_4) \mathbf{S}_{\mu_4} V_S(\tau_4, \tau_2) \mathbf{S}_{\mu_2} V_S(\tau_2, t_1) X_1 V_S(t_1, \tau_3) \mathbf{S}_{\mu_3} V_S(\tau_3, \tau_1) \mathbf{S}_{\mu_1} V_S(\tau_1, t_0) W_S] \\ &+ \sum_{\mu_4, \mu_3, \mu_2, \mu_1} \iint_{\mathbb{T}_{t_2 \leftarrow t_1}} d^2 \tau_{42} \iint_{\mathbb{T}_{t_1 \leftarrow t_0}} d^2 \tau_{13} \mathbf{C}_{\mu_4, \mu_3}(\tau_4 - \tau_3) \mathbf{C}_{\mu_2, \mu_1}(\tau_2 - \tau_1) \\ &\times \text{Tr}_S [X_2 V_S(t_2, \tau_4) \mathbf{S}_{\mu_4} V_S(\tau_4, \tau_2) \mathbf{S}_{\mu_2} V_S(\tau_2, t_1) X_1 V_S(t_1, \tau_1) \mathbf{S}_{\mu_1} V_S(\tau_1, \tau_3) \mathbf{S}_{\mu_3} V_S(\tau_3, t_0) W_S]. \end{aligned} \quad (\text{C15})$$

Therefore, the second-order correction $\Delta C_{X_2, X_1}^{(2)}(t_2, t_1)$ to the two-time correlation function is given by

$$\Delta C_{X_2, X_1}^{(2)}(t_2, t_1) = \Delta^{(\text{i})} C_{X_2, X_1}^{(2)}(t_2, t_1) + \Delta^{(\text{ii})} C_{X_2, X_1}^{(2)}(t_2, t_1). \quad (\text{C16})$$

After rearranging the order of integration and changing the integral variables, we finally obtain Eq. (45).

APPENDIX D: PROOF OF THE NECESSARY AND SUFFICIENT CONDITION

In this Appendix, we prove that the equality $A(t_1)A(t_2) = A(t_1 + t_2)$ is a necessary and sufficient condition that the lowest-order correlation function is exact, namely, $C_{zz}(t_2, t_1) = C_{zz}^{(0)}(t_2, t_1)$. First, when $A(t_1)A(t_2) = A(t_1 + t_2)$, it is obvious from Eqs. (59) and (65) that the equality $C_{zz}(t_2, t_1) = C_{zz}^{(0)}(t_2, t_1)$ is established. Next we assume that the equality $C_{zz}(t_2, t_1) = C_{zz}^{(0)}(t_2, t_1)$ holds. Then we obtain from Eqs. (59) and (65)

$$2A^*(t_2)A(t_2 - t_1)A(t_1) = |A(t_2)|^2 + |A(t_2 - t_1)|^2 |A(t_1)|^2, \quad (\text{D1})$$

the real and imaginary parts of which are given by

$$2 \text{Re} A^*(t_2)A(t_2 - t_1)A(t_1) = |A(t_2)|^2 + |A(t_2 - t_1)|^2 |A(t_1)|^2, \quad (\text{D2})$$

$$2 \text{Im} A^*(t_2)A(t_2 - t_1)A(t_1) = 0. \quad (\text{D3})$$

Then we obtain

$$\begin{aligned} |A(t_2) - A(t_2 - t_1)A(t_1)|^2 &= |A(t_2)|^2 + |A(t_2 - t_1)|^2 |A(t_1)|^2 - 2 \text{Re} A^*(t_2)A(t_2 - t_1)A(t_1) \\ &= 0, \end{aligned} \quad (\text{D4})$$

which yields the equality $A(t_2) = A(t_1)A(t_2 - t_1)$. Thus the proof is completed.

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