Double-time correlation functions of two quantum operations in open systems

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A double-time correlation function of arbitrary two quantum operations is studied for a nonstationary open quantum system which is in contact with a thermal reservoir. It includes a usual correlation function, a linear response function, and a weak value of an observable. Time evolution of the correlation function can be derived by means of the time-convolution and time-convolutionless projection operator techniques. For this purpose, a quasidensity operator accompanied by a fictitious field is introduced, which makes it possible to derive explicit formulas for calculating a double-time correlation function in the second-order approximation with respect to a system-reservoir interaction. The derived formula explicitly shows that the quantum regression theorem for calculating the double-time correlation function cannot be used if a thermal reservoir has a finite correlation time. Furthermore, the formula is applied for a pure dephasing process and a linear dissipative process. The quantum regression theorem and the the Leggett-Garg inequality are investigated for an open two-level system. The results are compared with those obtained by exact calculation to examine whether the formula is a good approximation.

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I. INTRODUCTION

A study on a quantum system under the influence of a thermal reservoir, called an open quantum system, is of essential importance not only in the foundations of quantum mechanics [\[1\]](#page-18-0) but also in the applications of quantum mechanics to communication and computation, which are referred to as quantum information processing and make it possible to perform novel tasks that cannot be done by classical information processing [\[2\]](#page-18-0). In a realistic situation, a quantum system is inevitably influenced by a surrounding environmental system, which we refer to as a thermal reservoir in this paper. Since information that a quantum system possesses flows out to a thermal reservoir due to an interaction, a quantum system undergoes irreversible time evolution, during which quantumness of the system such as coherence, entanglement, and nonlocality is degraded [\[3–6\]](#page-18-0). Although a study on irreversibility has a long history in non-equilibrium statistical mechanics [\[7\]](#page-18-0), it has still attracted much attention since irreversibility plays an important role in controlling quantum states when performing quantum information processing and understanding coherence and quantum correlation of quantum states [\[8–11\]](#page-18-0).

A state of a quantum system is described by a density operator $\rho(t)$ which is a positive operator satisfying $Tr \rho(t) =$ 1, where Tr stands for a trace operation. The irreversible time evolution of a quantum system is governed by the equation of motion for a density operator $\rho(t)$. The reduced time evolution of an open quantum system can be derived by various methods including the phenomenological equation $[12,13]$, the stochastic equation $[14,15]$, the quantum master equation $[4,7]$, and the path integral $[16]$. When a thermal reservoir has a sufficiently short correlation time and there is no initial correlation between a relevant quantum system and a thermal reservoir, the reduced time evolution of a quantum system can be described by a dynamical semigroup [\[4–6\]](#page-18-0). In this case, a change from $\rho(t_0)$ to $\rho(t)$ with $t \geq t_0$ is given by a completely positive map and the time-evolution equation is given by the Markovian quantum master equation of the Lindblad form [\[4–6\]](#page-18-0). However, the Markovian time evolution of a density operator $\rho(t)$ is not sufficient in many systems treated in quantum information processing and coherent optical transient phenomena. In particular, the non-Markovian effects are also important to understand coherence and quantum correlation of quantum states [\[17–28\]](#page-18-0). To obtain quantum dynamics beyond the Markovian time evolution, an approximation method is necessary in many cases since an interaction between a quantum system and a thermal reservoir is too complicated to solve exactly. The systematic method for perturbative expansion for a reduced density operator has been developed by making use of the projection operator method $[4,29-31]$ with the assistance of cumulant expansion [\[14,32–36\]](#page-18-0). The results are given by the timelocal (time-convolutionless) quantum master equation and the time-nonlocal (time-convolution) quantum master equation, which are successfully applied to many quantum systems.

A density operator $\rho(t)$ can describe all properties of single-time events in a quantum system. Probability that projective measurement performed on a quantum system at time *t* yields an outcome *a* is given by $P(a; t) = \langle a | \rho(t) | a \rangle$ $[1,2,4]$, where *a* and $|a\rangle$ are eigenvalue and corresponding eigenstate of a measured observable *A*. This means that any statistical property of a single-time event is derive from the density operator $\rho(t)$. However, there are important quantities that are not single-time events. For instance, the second order coherence and the fluorescence spectrum in quantum optical systems are described by double-time correlation functions [\[37–39\]](#page-18-0). A linear response function that characterizes how a system responds to a weak external field depends on two times at which an external field is applied to the system and a system observable is measured [\[7,40–44\]](#page-18-0). Furthermore, a weak value of an observable *A* [\[45](#page-18-0)[–48\]](#page-19-0) is not a single-time quantity. It depends on two times $[49-51]$: One is a measurement time at which an observable *A* is weakly measured, and the other is a postselection time at which the weakly measured system is postselected in a certain quantum state. Usually double-correlation functions are calculated by making use of the quantum regression theorem [\[4,37–39\]](#page-18-0). However, it has been shown that the quantum regression theorem is valid only in restricted situations. When reduced time evolution of a relevant quantum system is non-Markovian, it cannot be no longer used for calculating correlation functions [\[52–58\]](#page-19-0). Therefore, it is of a great importance to formulate a systematic calculation method that is valid even if the reduced time evolution is non-Markovian.

In this paper, applying the projection operator method [\[4,29–31,34–36\]](#page-18-0) to a quasidensity operator with a fictitious field that is introduced here, we derive formulas for calculating double-time correlation functions of an open quantum system influenced by a thermal reservoir. The preliminary version of the method has been developed for formulating the linear response theory [\[59\]](#page-19-0) and the weak measurement process [\[60\]](#page-19-0) of open quantum systems. The projection operator method for calculating correlation functions has already been used in Ref. [\[58\]](#page-19-0). However, our method makes it possible to derive explicit forms of double-time correlation functions. The result is applied to exactly solvable models of an open two-level system to show how our formulas work well. The validity of quantum regression theorem [\[52–57\]](#page-19-0) and the violation of the Leggett-Garg inequality [\[61–71\]](#page-19-0) are also examined. In Sec. II, we define a double-time correlation function of two quantum operations for which we derive calculation formulas. The double-time correlation function becomes a usual correlation function $[4,72]$ $[4,72]$, a linear response function $[40,41]$, and a weak value of an observable [\[47,](#page-18-0)[48\]](#page-19-0) by setting the quantum operations appropriately.

This paper is organized as follows. In Sec. [III,](#page-2-0) we introduce a quasidensity operator accompanied with a fictitious field. The linear term with respect to the fictitious field yields the double-time correlation function. Using the projection operator method, we derive the time-local [\[4,29–31\]](#page-18-0) and timenonlocal equations [\[4,29–31\]](#page-18-0) for the reduced quasidensity operator. In Sec. [IV,](#page-4-0) we apply the second-order approximation (the Born approximation) [\[4\]](#page-18-0) to the time-local and timenonlocal equations [\[34–36\]](#page-18-0) and we derive the linearized solutions with respect to the fictitious field. Then we can obtain the double-time correlation function, which consists of two terms. One is derived by making use of the quantum regression theorem and the other is a correction term which is yielded by a finite correlation time of a thermal reservoir. The latter becomes negligible if the thermal reservoir has a sufficiently short correlation time. In Sec. [V,](#page-5-0) assuming a linear coupling between the relevant quantum system and the thermal reservoir, we derive the double-time correlation functions explicitly. In Sec. [VI,](#page-7-0) we treat two exactly solvable models for a two-level system in a pure dephasing [\[73,74\]](#page-19-0) and a linear dissipative process [\[4,19\]](#page-18-0) to show that the derived formulas work well. Furthermore, we investigate the validity of the quantum regression theorem [\[52–57\]](#page-19-0) and the violation of the Leggett-Garg inequality [\[61–71\]](#page-19-0). It will be found from the results that the formulas are good approximation for the double-time correlation functions. In Sec. [VII,](#page-12-0) we provide a brief summary of this paper.

II. DOUBLE-TIME CORRELATION FUNCTION OF QUANTUM OPERATIONS

Time evolution of a quantum system is described in terms of a Liouvillian superoperator \mathcal{L} [\[4\]](#page-18-0), which is defined by $\mathcal{L} \bullet = -(i/\hbar)H^{\times} \bullet$ with Kubo's notation $X^{\times} \bullet = [X, \bullet]$ [\[40\]](#page-18-0), where we denote a Hamiltonian of a quantum system as *H*. A quantum system that we consider in this paper consists of two parts: One is a relevant system and the other is a surrounding

environment or a thermal reservoir. Then the Hamiltonian is decomposed into $H = H_S + H_{SR} + H_R$ with H_S and H_R being the system and reservoir Hamiltonians and H_{SR} being the system-reservoir interaction Hamiltonian. Furthermore, we denote arbitrary quantum operations performed on the relevant system as K_1 and K_2 , where we do not require that these operations preserve a trace of a density operator. When the whole system is in a quantum state described by a density operator W at an initial time t_0 , we define a function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ by

$$
G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1) = \text{Tr}[\mathcal{K}_2 e^{\mathcal{L}(t_2 - t_1)} \mathcal{K}_1 e^{\mathcal{L}(t_1 - t_0)} W] \tag{1}
$$
\n
$$
(t_2 > t_1 > t_0).
$$

In the rest of this paper, we refer to this function as a doubletime correlation function of the two quantum operations \mathcal{K}_1 and K_2 . This function means that the two quantum operations \mathcal{K}_2 and \mathcal{K}_1 are performed on the relevant system at time t_2 and t_1 during the time evolution. As explained below, the double-time correlation function given by Eq. (1) includes various kinds of quantities as a special case.

First, we substitute $K_1 = A$ and $K_2 = B$ into Eq. (1), where *A* and *B* are observables of the relevant quantum system. Then $G(t_2,t_1|B,A)$ becomes a usual correlation function of A and *B* [\[4,](#page-18-0)[72\]](#page-19-0). In fact, we have

$$
G(t_2, t_1 | B, A)
$$

= Tr[$Be^{C(t_2 - t_1)} Ae^{C(t_1 - t_0)} W$]
= Tr[$Be^{-\frac{t}{\hbar}H(t_2 - t_1)}(Ae^{-\frac{t}{\hbar}H(t_1 - t_0)}We^{\frac{t}{\hbar}H(t_1 - t_0)})e^{\frac{t}{\hbar}H(t_2 - t_1)}]$
= Tr[$B(t_2)A(t_1)W$]
= $\langle B(t_2)A(t_1) \rangle \equiv C_{BA}(t_2, t_1),$ (2)

where $X(t) = e^{\frac{i}{\hbar}H(t-t_0)} X e^{-\frac{i}{\hbar}H(t_1-t_0)} (X = A, B)$ is a Heisenberg operator. If we substitute $K_1 = \tilde{A}$ and $K_2 = B$ with $\tilde{A} \bullet = \bullet A$ into Eq. (1), we obtain a correlation function $G(t_2,t_1|B,\tilde{A}) = \langle A(t_1)B(t_2) \rangle = C_{AB}(t_1,t_2)$ in the reverse order. Furthermore, by substituting $K_1 = iA^\times$ and $K_2 = B$, we can derive the linear response function [\[40,41\]](#page-18-0),

$$
G(t_2, t_1 | B, iA^{\times}) = \text{Tr}\Big[B e^{\mathcal{L}(t_2 - t_1)} i A^{\times} e^{\mathcal{L}(t_1 - t_0)} W\Big]
$$

= $i \text{Tr}\Big[B e^{\mathcal{L}(t_2 - t_1)} (A - \tilde{A}) e^{\mathcal{L}(t_1 - t_0)} W\Big]$
= $i \text{Tr}[B(t_2) A(t_1) W] - i \text{Tr}[A(t_1) B(t_2) W]$
= $i \langle [B(t_2), A(t_1)] \rangle \equiv \phi_{BA}(t_2, t_1).$ (3)

When $\mathcal{K}_1 = (A + \tilde{A})/2$ and $\mathcal{K}_2 = B$, it is obvious that Eq. (1) becomes a symmetrized correlation function, $G(t_2,t_1|B)$, $(A + \tilde{A})/2$ = $\frac{1}{2}$ $\langle B(t_2)A(t_1) + A(t_1)B(t_2) \rangle \equiv S_{BA}(t_2,t_1).$

The weak value of an observable *A* can be represented in terms of the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$. To see this, we denote a measurement operator as Π_f , which is an element of positive operator-valued measure (POVM) [\[2,](#page-18-0)[75\]](#page-19-0), satisfying positivity $\Pi_f \geq 0$ and normalization $\sum_f \Pi_f = 1$. Then, by substituting $K_1 = A$ ($K_1 = 1$) and $K_2 = \Pi_f$ into Eq. (1), we can derive the weak value of an observable *A* [\[47,](#page-18-0)[48,60\]](#page-19-0),

$$
\frac{G(t_2,t_1|\Pi_f,A)}{G(t_2,t_1|\Pi_f,1)} = \frac{\text{Tr}[\Pi_f e^{\mathcal{L}(t_2-t_1)} A e^{\mathcal{L}(t_1-t_0)} W]}{\text{Tr}[\Pi_f e^{\mathcal{L}(t_2-t_0)} W]} \n\equiv A_w(t_2,t_1|W),
$$
\n(4)

which is obtained by weak measurement of A at time t_1 , where the quantum system is preselected in *W* at initial time t_0 and postselected in Π_f at final time t_2 . Using the spectral decomposition $A = \sum_a a P_a$ with eigenvalue *a* and eigenprojector $P_a = |a\rangle\langle a|$, we have the weak conditional probability of the measurement outcome *a*,

$$
P_{\mathbf{w}}(a|\Pi_{f},W) = \frac{\text{Tr}[\Pi_{f}e^{\mathcal{L}(t_{2}-t_{1})}P_{a}e^{\mathcal{L}(t_{1}-t_{0})}W]}{\text{Tr}[\Pi_{f}e^{\mathcal{L}(t_{2}-t_{0})}W]},
$$
(5)

which is related to the weak value by $A_w(t_2,t_1|W) =$ $\sum_a a P_{\rm w}(a|\Pi_f, W)$.

Furthermore, we can obtain the joint probability $P(b,t_2; a,t_1|W)$ that sequential projective measurement of observables *A* and *B* performed on the relevant system at times t_1 and t_2 ($t_1 < t_2$) yields the outcomes *a* and *b*. To show this, we introduce two projective superoperator by $P_a \bullet = P_a \bullet P_a$ and $P_b \bullet = P_b \bullet P_b$, where $P_a = |a\rangle\langle a|$ and $P_b = |b\rangle\langle b|$ are projection operators with eigenstates $|a\rangle$ and $|b\rangle$ of the measured observables *A* and *B*. Then we can derive

$$
G(t_2, t_1 | \mathcal{P}_b, \mathcal{P}_a)
$$

= Tr[$\mathcal{P}_b e^{\mathcal{L}(t_2 - t_1)} \mathcal{P}_b e^{\mathcal{L}(t_1 - t_0)} W$]
= $|\langle b|e^{-\frac{i}{\hbar}H(t_2 - t_1)}|a\rangle|^2 \langle a|e^{-\frac{i}{\hbar}H(t_1 - t_0)} W e^{\frac{i}{\hbar}H(t_1 - t_0)}|a\rangle$
= $P(b, t_2 | a, t_1) P(a, t_1 | W)$
= $P(b, t_2; a, t_1 | W)$, (6)

where $P(b,t_2|a,t_1)$ represents the conditional probability that the measurement of B at the time t_2 yields the outcome b for given measurement outcome *a* at the time t_1 , and $P(a,t_1|W)$ is the probability that the measurement of A at the time t_1 yields the outcome *a* when the quantum system is initially prepared in the state W at the initial time t_0 . Here, we note that $C_{BA}(t_2,t_1) \neq \sum_a \sum_b abP(b,t_2;a,t_1|W)$ in general. If the two observables *A* and *B* are both dichotomous, the equality $\text{Re}C_{BA}(t_2,t_1) = \sum_a \sum_b abP(b,t_2;a,t_1|W)$ holds. The result is summarized in Table I. Since the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ includes various important quantities, it is very useful to provide a method for systematically calculating the double-time correlation function.

III. PROJECTION OPERATOR METHOD FOR DOUBLE-TIME CORRELATION FUNCTIONS

To drive the time evolution of the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ for an open quantum system under the influence of a thermal reservoir, we introduce a fictitious external field $g(t)$, which is linearly coupled to the quantum operation K_1 of the relevant quantum system. The Liouvillian superoperator is replaced by $\mathcal{L} + g(t)\mathcal{K}_1$. Then the time evolution operator of the whole system is given by

$$
U(t,t_0|g) = \exp_{\leftarrow} \left(\int_{t_0}^t d\tau \left[\mathcal{L} + g(\tau) \mathcal{K}_1 \right] \right), \tag{7}
$$

where \exp_{max} stands for the time-ordered exponential, in which operators are placed from right to left in the chronological order. This superoperator is not unitary in general. We have up to the first order with respect to the fictitious field,

$$
\mathcal{U}(t,t_0|g) = e^{\mathcal{L}(t-t_0)} \exp_{\leftarrow} \left[\int_{t_0}^t d\tau \ g(\tau) \mathcal{K}_1(\tau) \right] \approx e^{\mathcal{L}(t-t_0)} + \int_{t_0}^t d\tau \ g(\tau) e^{\mathcal{L}(t-\tau)} \mathcal{K}_1 e^{\mathcal{L}(\tau-t_0)}.
$$
 (8)

A quasidensity operator of the whole system interacting with the fictitious field $g(t)$ is introduced by $W(t|g) = U(t,t_0|g)W$. The relevant quantum system is described by the reduced quasidensity operator $W_S(t|g) = Tr_R W(t|g)$, where Tr_R is a trace operation over a Hilbert space of the thermal reservoir. Thus, using Eq. (8) , we can express the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ as follows:

$$
G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1) = \text{Tr}_S \big[\mathcal{K}_2 \text{Tr}_R \big(e^{\mathcal{L}(t_2 - t_1)} \mathcal{K}_1 e^{\mathcal{L}(t_1 - t_0)} W \big) \big]
$$

= $\text{Tr}_S [\mathcal{K}_2 W_S(t_2, t_1 | \mathcal{K}_1)]$ (9)

with

$$
W_S(t_2, t_1 | \mathcal{K}_1) = \left. \frac{\delta W_S(t_2 | g)}{\delta g(t_1)} \right|_{g(t) \to 0}, \tag{10}
$$

where Tr_S is a trace operation of the relevant quantum system. It is found from this result that the first-order term of the reduced quasidensity operator $W_S(t|g)$ with respect to the fictitious field $g(t)$ provides the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$. Therefore, our task is to obtain the time evolution of the first-order term of the reduced quasidensity operator $W_S(t|g)$.

First, using the projection operator method [\[4,29–31,34–](#page-18-0) [36\]](#page-18-0), we obtain the time-evolution equation for the reduced

TABLE I. The double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ of two quantum operations \mathcal{K}_1 and \mathcal{K}_2 with $t_2 > t_1$, where *A* and *B* are system observables, Π_f is POVM, and $P_{a,b}$ is a projective superoperator.

Time t_1	Time t_2	$G(t_2,t_1 \mathcal{K}_2,\mathcal{K}_1)$
$\mathcal{K}_1 = A$	$\mathcal{K}_2 = B$	Correlation function $C_{BA}(t_2,t_1)$
$\mathcal{K}_1 = \tilde{A}$	$\mathcal{K}_2 = B$	Correlation function $C_{AB}(t_1,t_2)$
$\mathcal{K}_1 = \frac{1}{2}(A + \tilde{A})$	$\mathcal{K}_2 = B$	Symmetrized correlation function $S_{BA}(t_2,t_1)$
$\mathcal{K}_1 = i A^{\times}$	$\mathcal{K}_2 = B$	Linear response function $\phi_{BA}(t_2,t_1)$
$\mathcal{K}_1 = A$	$\mathcal{K}_2 = \Pi_f$	Weak value $A_w(t_2,t_1 W)$
$\mathcal{K}_1 = P_a$	$\mathcal{K}_2 = \Pi_f$	Weak probability $P_w(a \Pi_f, W)$
$\mathcal{K}_1 = \mathcal{P}_a$	$\mathcal{K}_2 = \mathcal{P}_h$	Joint probability $P(b,t_2;a,t_1 W)$

quasidensity operator $W_S(t|g)$ and then linearize it with respect to the fictitious field $g(t)$. When we differentiate the quasidensity operator $W(t|g) = U(t,t_0|g)W$ of the whole system with respect to time *t*, we obtain the equation of motion, $\partial W(t|g)/\partial t = [\mathcal{L} + g(t)\mathcal{K}_1]W(t|g)$. Introducing a projection operator $\mathcal{P} \bullet = \rho_R \text{Tr}_R \bullet$ with a density operator ρ_R of the thermal reservoir [\[4\]](#page-18-0), we can derive two different time-evolution equations for the reduced quasidensity operator $W_S(t|g)$, where the derivation is the same as that for the non-Markovian quantum master equation. One is a time-local equation [\[34–36\]](#page-18-0),

$$
\frac{\partial}{\partial t}W_S(t|g) = [\mathcal{L}_S + g(t)\mathcal{K}_1]W_S(t|g) \n+ \mathcal{K}_S(t|g)W_S(t|g) + \mathcal{I}_S(t|g), \quad (11)
$$

where \mathcal{L}_s , $\mathcal{K}_s(t|g)$ and $\mathcal{I}_s(t|g)$ are given by Eqs. [\(A1\)](#page-12-0), [\(A16\)](#page-13-0), and $(A17)$. The other is a time-nonlocal equation,

$$
\frac{\partial}{\partial t}W_S(t|g) = [\mathcal{L}_S + g(t)\mathcal{K}_1]W_S(t|g) + \Phi_S(t|g)W_S(t|g)
$$

$$
+ \int_{t_0}^t dt_1 \Phi_S(t,t_1|g)W_S(t_1|g) + \mathcal{J}_S(t|g), \quad (12)
$$

where $\Phi_S(t|g)$, $\Phi_S(t,t_1|g)$, and $\mathcal{J}_S(t|g)$ are given by Eqs. $(A20)$ – $(A22)$. The derivation of Eqs. (11) and (12) is briefly summarized in Appendix [A.](#page-12-0)

We can derive the time-evolution equation for the doubletime correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ from Eqs. (11) or (12). To do this, we calculate a functional derivative of Eq. (11) with respect to the fictitious field $g(t_1)$ and take the limit $g(t) \rightarrow 0$. Then, we obtain the equation for the operator $W_S(t_2,t_1|\mathcal{K}_1)$ of the relevant quantum system from Eq. [\(10\)](#page-2-0),

$$
\frac{\partial}{\partial t_2} W_S(t_2, t_1 | \mathcal{K}_1)
$$
\n
$$
= [\mathcal{L}_S + \mathcal{K}_S(t_2 | 0)] W_S(t_2, t_1 | \mathcal{K}_1) + \mathcal{I}_S(t_2 | 0) + \Delta \mathcal{K}_S(t_2, t_1 | \mathcal{K}_1) W_S(t_2 | 0) + \Delta \mathcal{I}_S(t_2, t_1 | \mathcal{K}_1), \quad (13)
$$

with

$$
\Delta \mathcal{K}_S(t_2, t_1 | \mathcal{K}_1) = \left. \frac{\delta \mathcal{K}_S(t_2 | g)}{\delta g(t_1)} \right|_{g \to 0},\tag{14}
$$

$$
\Delta \mathcal{I}_S(t_2, t_1 | \mathcal{K}_1) = \left. \frac{\delta \mathcal{I}_S(t_2 | g)}{\delta g(t_1)} \right|_{g \to 0} - \mathcal{I}_S(t_2 | 0), \qquad (15)
$$

where we have used the fact that the equality $\delta g(t_2)/\delta g(t_1) = 0$ holds due to $t_2 > t_1$. The double-time correlation function is given by $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1) = \text{Tr}_S[\mathcal{K}_2 W_S(t_2,t_1|\mathcal{K}_1)]$. In Eq. (13), $W_S(t₂|0)$ is a reduced density operator of the relevant quantum system, the time evolution of which is governed by the timelocal quantum master equation [\[34–36\]](#page-18-0),

$$
\frac{\partial}{\partial t_2} W_S(t_2|0) = [\mathcal{L}_S + \mathcal{K}_S(t_2|0)] W_S(t_2|0) + \mathcal{I}_S(t_2|0). \quad (16)
$$

The average value of the quantum operation K_2 is given by $G(t_2|\mathcal{K}_2) = \text{Tr}_S[\mathcal{K}_2 W_S(t_2|0)]$. Therefore, comparing Eq. (13) with Eq. (16) , we find that the time evolution of double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ and the average value $G(t_2|\mathcal{K}_2)$ are determined by the same equation if the last two terms on the right-hand side of Eq. (13) can be ignored. In

this case, it is found that the quantum regression theorem for the double-time correlation function is established [\[4,37–39\]](#page-18-0). In the absence of the initial correlation between the relevant quantum system and the thermal reservoir, the similar result has been obtained in the second-order approximation by a different approach [\[56\]](#page-19-0).

In the same way, calculating a functional derivative of Eq. (12) with respect to the fictitious field $g(t_1)$ and taking the limit $g(t) \rightarrow 0$, we can derive

$$
\frac{\partial}{\partial t_2} W_S(t_2, t_1 | \mathcal{K}_1) \n= \mathcal{L}_S W_S(t_2, t_1 | \mathcal{K}_1) + \Phi_S(t_2 | 0) W_S(t_2, t_1 | \mathcal{K}_1) \n+ \int_{t_0}^{t_2} d\tau \, \Phi_S(t_2, \tau | 0) W_S(\tau, t_1 | \mathcal{K}_1) + \mathcal{J}_S(t_2 | 0) \n+ \Delta \Phi_S(t_2, t_1 | \mathcal{K}_1) W_S(t_2 | 0) \n+ \int_{t_0}^{t_2} d\tau \, \Delta \Phi_S(t_2, t_1, \tau | \mathcal{K}_1) W_S(\tau | 0) + \Delta \mathcal{J}_S(t_2, t_1 | \mathcal{K}_1),
$$
\n(17)

with

$$
\Delta \Phi_S(t_2, t_1 | \mathcal{K}_1) = \left. \frac{\delta \Phi_S(t_2 | g)}{\delta g(t_1)} \right|_{g \to 0},\tag{18}
$$

$$
\Delta \Phi_S(t_2, t_1, \tau | \mathcal{K}_1) = \left. \frac{\delta \Phi_S(t_2, t_1 | g)}{\delta g(t_1)} \right|_{g \to 0},\tag{19}
$$

$$
\Delta \mathcal{J}_S(t_2, t_1 | \mathcal{K}_1) = \left. \frac{\delta \mathcal{J}_S(t_2 | g)}{\delta g(t_1)} \right|_{g \to 0} - \mathcal{J}_S(t_2 | 0), \quad (20)
$$

where the reduced density operator $W_S(t₂|0)$ of the relevant quantum system is determined by the time-nonlocal quantum master equation [\[29–31\]](#page-18-0),

$$
\frac{\partial}{\partial t_2} W_S(t_2|0) = \mathcal{L}_S W_S(t_2|0) + \Phi_S(t_2|0) W_S(t_2|0) \n+ \int_{t_0}^{t_2} d\tau \, \Phi_S(t_2, \tau|0) W_S W_S(\tau|0) + \mathcal{J}_S(t_2|0).
$$
\n(21)

It is seen from Eqs. (17) and (21) that if the last three terms on the right-hand side of Eq. (17) vanish, the quantum regression theorem is established for the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$. As will be show later, the quantum regression theorem is valid only if the correlation time of the thermal reservoir is sufficiently short.

Before closing this section, we briefly mention the result given in Ref. [\[58\]](#page-19-0), which is closely related to Eq. (13). We can rewrite it into

$$
\frac{\partial}{\partial t_2} W_S(t_2, t_1 | \mathcal{K}_1) \n= [\mathcal{L}_S + \mathcal{K}_S(t_2 | 0)] W_S(t_2, t_1 | \mathcal{K}_1) \n+ \Delta \mathcal{K}_S(t_2, t_1 | \mathcal{K}_1) W_S(t_2 | 0) + \frac{\delta \mathcal{I}_S(t_2 | g)}{\delta g(t_1)} \bigg|_{g \to 0}.
$$
\n(22)

Here we have $W_S(t_2,t_1|\mathcal{K}_1) = \text{Tr}_R[e^{\mathcal{L}(t_2-t_1)}\mathcal{K}_1e^{\mathcal{L}(t_1-t_0)}W]$ from Eq. [\(10\)](#page-2-0), which is equivalent to the system operator $\Lambda(t_1 - t_0, t_2 - t_1)$ introduced in Ref. [\[58\]](#page-19-0). Then the first line on the right-hand side of Eq. [\(22\)](#page-3-0) corresponds to the second term on the right-hand side of Eq. (10) in Ref. [\[58\]](#page-19-0) and the second line to the first term on the right-hand side of Eq. (10). The relation between the results in this paper and in Ref. [\[58\]](#page-19-0) becomes more clear when we apply the second-order approximation with respect to the system-reservoir interaction in the case that there is no initial correlation between the relevant quantum system and the thermal reservoir (see Appendix [B\)](#page-13-0). Furthermore, we note that calculation in terms of the quasidensity operator $W_S(t|g)$ is easier than that using the system operator *W_S*($t_2, t_1 | K_1$) or $\Lambda(t_1 - t_0, t_2 - t_1)$. In fact, we can obtain explicit formulas for the double-time correlation function.

IV. DOUBLE-TIME CORRELATION FUNCTIONS IN THE SECOND-ORDER APPROXIMATION

In this section, in order to derive the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$, we assume that the interaction between the relevant quantum system and the thermal reservoir is so weak that we can apply the second-order approximation (the Born approximation) [\[4\]](#page-18-0) to the time-local and time-nonlocal equations (11) and (12) for the reduced quasidensity operator $W_S(t|g)$. In the rest of this paper, we assume that the equality $\langle \hat{\mathcal{L}}_{SR}(t) \rangle_R = 0$ holds for the sake of simplicity.

A. Double-time correlation function from the time-local equation

In the second-order approximation with respect to the system-reservoir interaction, using the relation $G(t_2,t_1|K_2,K_1)$ $Tr_S[\mathcal{K}_2 \delta W_{S,1}(t_2|g)/\delta g(t_1)]_{g\to 0}$, we can obtain the double-time correlation function from Eq. [\(11\)](#page-3-0),

$$
G(t_2, t_1 | K_2, K_1) = \text{Tr}_S[K_2U_S(t_2, t_1)K_1U_S(t_1, t_0)W_S] + \int_{t_1}^{t_2} d\tau_1 \int_{t_0}^{t_1} d\tau_2 \text{Tr}_S[K_2U_S(t_2, \tau_1) \times e^{\mathcal{L}_S(\tau_1 - t_0)} \langle \mathcal{L}_{SR}(\tau_1) [K_1(t_1), \mathcal{L}_{SR}(\tau_2)] \rangle_R e^{-\mathcal{L}_S(\tau_1 - t_0)} U_S(\tau_1, t_0)W_S]
$$

+
$$
\int_{t_0}^{t_1} d\tau_1 \text{Tr}_S[K_2U_S(t_2, t_1)K_1U_S(t_1, \tau_1)I_{S,0}(\tau_1)] + \int_{t_1}^{t_2} d\tau_1 \int_{t_0}^{\tau_1} d\tau_2 \int_{t_0}^{t_1} d\tau_3 \text{Tr}_S[K_2U_S(t_2, \tau_1) e^{\mathcal{L}_S(\tau_1 - t_0)} \times \langle \mathcal{L}_{SR}(\tau_1) [K_1(t_1), \mathcal{L}_{SR}(\tau_3)] \rangle_R e^{-\mathcal{L}_S(\tau_1 - t_0)} U_S(\tau_1, \tau_2)I_{S,0}(\tau_2)] + \int_{t_1}^{t_2} d\tau_1 \text{Tr}_S[K_2U_S(t_2, \tau_1)I_{S,1}(\tau_1, t_1)], \quad (23)
$$

where the derivation is given in Appendix [B.](#page-13-0) In this equation, $\mathcal{L}_{SR}(t)$, $\mathcal{K}_1(t)$, $I_{S,0}(t)$, $I_{S,1}(t,\tau)$, and $\mathcal{U}_S(t,\tau)$ are respectively given by Eqs. [\(B3\)](#page-13-0), [\(B7\)](#page-13-0), [\(B15\)](#page-13-0), [\(B17\)](#page-13-0), and [\(B26\)](#page-14-0). When we use the reduced density operator $W_S(t) = W_{S,0}(t)$ of the relevant quantum system, which is a solution of the inhomogeneous time-local master equation [\(B24\)](#page-14-0), the expression of the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ is simplified as

$$
G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1) = \text{Tr}_S[\mathcal{K}_2 \mathcal{U}_S(t_2, t_1) \mathcal{K}_1 W_S(t_1)] + \int_{t_1}^{t_2} d\tau_1 \int_{t_0}^{t_1} d\tau_2 \text{Tr}_S[\mathcal{K}_2 \mathcal{U}_S(t_2, \tau_1) \times e^{\mathcal{L}_S(\tau_1 - t_0)} \langle \mathcal{L}_{SR}(\tau_1) | \mathcal{K}_1(t_1), \mathcal{L}_{SR}(\tau_2)] \rangle_R e^{-\mathcal{L}_S(\tau_1 - t_0)} W_S(\tau_1)] + \int_{t_1}^{t_2} d\tau_1 \text{Tr}_S[\mathcal{K}_2 \mathcal{U}_S(t_2, \tau_1) I_S(\tau_1, t_1 | \mathcal{K}_1)], \tag{24}
$$

with $I_S(\tau_1, t_1 | \mathcal{K}_1) = I_{S,1}(\tau_1, t_1)$. Furthermore, when we introduce $\mathcal{K}_1(t,t')$ and $J(t,t' | \mathcal{K}_1)$ by

$$
\mathcal{K}_1(t,t') = \delta(t-t')\mathcal{K}_1 + \theta(t-t')\mathcal{K}_{S,1}(t,t'),
$$
\n(25)

$$
J(t,t'|\mathcal{K}_1) = \theta(t-t')I_S(t,t'|\mathcal{K}_1),
$$
\n(26)

where $K_{S,1}(t,t')$ is given by Eq. [\(B13\)](#page-13-0), the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ can be expressed as

$$
G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1) = \int_{t_0}^{t_2} d\tau \left\{ \text{Tr}_S[\mathcal{K}_2 \mathcal{U}_S(t_2, \tau) \mathcal{K}_1(\tau, t_1) W_S(\tau)] + \text{Tr}_S[\mathcal{K}_2 \mathcal{U}_S(t_2, \tau) J(\tau, t_1 | \mathcal{K}_1)] \right\}.
$$
 (27)

When there is no initial correlation between the relevant quantum system and the thermal reservoir, that is, $W = W_S \otimes W_R$, the second term on the right-hand side of this equation vanishes if we set $\rho_R = W_R$ in the definition of the projection operator P .

For instance, setting $K_1 = A$ and $K_2 = B$, we obtain the usual double-time correlation function of two observables A and B,

$$
\langle B(t_2)A(t_1) \rangle = \text{Tr}_S[B\mathcal{U}_S(t_2, t_1)A W_S(t_1)] + \int_{t_1}^{t_2} d\tau_1 \int_{t_0}^{t_1} d\tau_2 \text{Tr}_S[B\mathcal{U}_S(t_2, \tau_1) \times e^{\mathcal{L}_S(\tau_1 - t_0)} \langle \mathcal{L}_{SR}(\tau_1)[A(t_1), \mathcal{L}_{SR}(\tau_2)] \rangle_R e^{-\mathcal{L}_S(\tau_1 - t_0)} W_S(\tau_1) + \int_{t_1}^{t_2} d\tau_1 \text{Tr}_S[B\mathcal{U}_S(t_2, \tau_1)I_S(\tau_1, t_1|A)],
$$
 (28)

with

$$
I_S(t,\tau|A) = e^{\mathcal{L}_S(t-t_0)} \text{Tr}_R[\mathcal{L}_{SR}(t)A(\tau)\delta W] + \int_{t_0}^t dt_1 e^{\mathcal{L}_S(t-t_0)} \text{Tr}_R[\mathcal{L}_{SR}(t)T\{\mathcal{L}_{SR}(t_1)A(\tau)\}\delta W].
$$
\n(29)

When we substitute $K_1 = iA^\times$ and $K_2 = B$ into Eq. [\(24\)](#page-4-0), we have the linear response function $\phi_{BA}(t_2, t_1) = i\langle [B(t_2), A(t_1)] \rangle$ of an open quantum system,

$$
\phi_{BA}(t_2, t_1) = i \text{Tr}_S[BU_S(t_2, t_1)A^\times W_S(t_1)] + i \int_{t_1}^{t_2} d\tau_1 \int_{t_0}^{t_1} d\tau_2 \text{Tr}_S[BU_S(t_2, \tau_1) \times e^{C_S(\tau_1 - t_0)} \langle \mathcal{L}_{SR}(\tau_1) [A^\times(t_1), \mathcal{L}_{SR}(\tau_2)] \rangle_R e^{-C_S(\tau_1 - t_0)} W_S(\tau_1) + \int_{t_1}^{t_2} d\tau_1 \text{Tr}_S[BU_S(t_2, \tau_1) I_S(\tau_1, t_1 | i A^\times)], \tag{30}
$$

with

$$
I_S(t,\tau|iA^{\times}) = ie^{\mathcal{L}_S(t-t_0)} \text{Tr}_R[\mathcal{L}_{SR}(t)A^{\times}(\tau)\delta W] + i \int_{t_0}^t dt_1 e^{\mathcal{L}_S(t-t_0)} \text{Tr}_R[\mathcal{L}_{SR}(t)T\{\mathcal{L}_{SR}(t_1)A^{\times}(\tau)\}\delta W].
$$
 (31)

Furthermore, the weak value of an observable *A* in a postselected open quantum system, where the postselection is described by a measurement operator Π_f , is provided by [\[60\]](#page-19-0)

$$
A_{W}(t_{2},t_{1}|\Pi_{f}) = \frac{\text{Tr}_{S}[\Pi_{f}U_{S}(t_{2},t_{1})A W_{S}(t_{1})]}{\text{Tr}_{S}[\Pi_{f}W_{S}(t_{2})]} + \frac{1}{\text{Tr}_{S}[\Pi_{f}W_{S}(t_{2})]} \int_{t_{1}}^{t_{2}} d\tau_{1} \int_{t_{0}}^{t_{1}} d\tau_{2} \text{Tr}_{S}[\Pi_{f}U_{S}(t_{2},\tau_{1})] \times e^{\mathcal{L}_{S}(\tau_{1}-t_{0})} \langle \mathcal{L}_{SR}(\tau_{1})[A(t_{1}),\mathcal{L}_{SR}(\tau_{2})] \rangle_{R} e^{-\mathcal{L}_{S}(\tau_{1}-t_{0})} W_{S}(\tau_{1})] + \frac{1}{\text{Tr}_{S}[\Pi_{f}W_{S}(t_{2})]} \int_{t_{1}}^{t_{2}} d\tau_{1} \text{Tr}_{S}[\Pi_{f}U_{S}(t_{2},\tau_{1})I_{S}(\tau_{1},t_{1}|A)], \qquad (32)
$$

where $I_S(t, \tau | A)$ is given by Eq. (29). In deriving Eq. (32), we have used the equality $I_S(t, \tau | 1) = I_{S,0}(t)$. Furthermore, by substituting $K_1 = \mathcal{P}_a$ and $K_2 = \mathcal{P}_b$ into Eq. [\(24\)](#page-4-0), we can obtain the joint probability of two projective measurements performed on the relevant quantum system at times t_1 and t_2 .

B. Double-time correlation function from time-nonlocal equation

Using the same method for the time-local equation, we can obtain the double-time correlation function from the time-nonlocal equation [\(12\)](#page-3-0),

$$
G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1) = \text{Tr}_S[\mathcal{K}_2 \bar{\mathcal{U}}_S(t, t_1) \mathcal{K}_1 \bar{W}_S(t_1)] + \int_{t_1}^t d\tau_1 \int_{t_0}^{t_1} d\tau_2 \text{Tr}_S[\mathcal{K}_2 \bar{\mathcal{U}}_S(t_2, \tau_1)] \times e^{\mathcal{L}_S(\tau_1 - t_0)} \langle \mathcal{L}_{SR}(\tau_1) \mathcal{K}_1(t_1) \mathcal{L}_{SR}(\tau_2) \rangle_R e^{-\mathcal{L}_S(\tau_2 - t_0)} \bar{W}_S(\tau_2)] + \int_{t_1}^{t_2} d\tau_1 \text{Tr}_S[\mathcal{K}_2 \bar{\mathcal{U}}_S(t, \tau_1) I_{S,1}(\tau_1, t_1 | \mathcal{K}_1)], \tag{33}
$$

the derivation of which is given in Appendix [C.](#page-14-0) In this equation, $\bar{W}_S(t_1)$ and $\bar{U}_S(t,t_1)$ are defined by Eqs. [\(C11\)](#page-15-0) and [\(C12\)](#page-15-0). Comparing this result with Eq. [\(24\)](#page-4-0), we can derive the correlation function $\langle B(t_2)A(t_1) \rangle$, the linear response function $\phi_{BA}(t_2,t_1)$, and the weak value $A_W(t_2,t_1|\Pi_f)$ by replacing $U_S(t,t')$, $W_S(t)$, and

$$
e^{\mathcal{L}_S(\tau_1-t_0)}\langle \mathcal{L}_{SR}(\tau_1)[\mathcal{K}_1(t_1),\mathcal{L}_{SR}(\tau_2)]\rangle_R e^{-\mathcal{L}_S(\tau_1-t_0)}W_S(\tau_1)
$$

in Eqs. [\(28\)](#page-4-0), (30), and (32) by $\bar{U}_S(t,t')$, $\bar{W}_S(t)$ and

$$
e^{\mathcal{L}_S(\tau_1-t_0)}\langle \mathcal{L}_{SR}(\tau_1)\mathcal{K}_1(t_1)\mathcal{L}_{SR}(\tau_2)\rangle_R e^{-\mathcal{L}_S(\tau_2-t_0)}\bar{W}_S(\tau_2),
$$

where $K_1 = A$ for the correlation function and the weak value and $K_1 = iA^{\times}$ for the linear response function.

V. EXPLICIT FORMS OF THE DOUBLE-TIME CORRELATION FUNCTIONS

To obtain explicit expressions of the double-time correlation function in the second-order approximation, we assume that the interaction Hamiltonian between the relevant quantum system and the thermal reservoir is given by $H_{SR} = \hbar \sum_{\mu} S_{\mu} R_{\mu}$, where S_μ and R_μ are operators of the relevant quantum system and the thermal reservoir. The corresponding Liouvillian superoperator is $\mathcal{L}_{SR} = -i \sum_{\mu} (S_{\mu} R_{\mu})^{\times}$. For the sake of simplicity, we assume that the initial time is $t_0 = 0$ and there is no initial correlation between them. So, we can write the initial state $W = W_S W_R$. In this case, setting $\rho_R = W_R$ in the definition of the projection operator P, we have the equality $\delta W = 0$ and thus the inhomogeneous terms in the time-local and time-nonlocal equations for the reduced quasidensity operator disappear. It is easy to find that

$$
e^{\mathcal{L}_{S}t}\langle\mathcal{L}_{SR}(t)\mathcal{L}_{SR}(t_1)\rangle_{R}e^{-\mathcal{L}_{S}t}W_{S}(t) = \sum_{\mu,\nu}\langle R_{\mu}(t)R_{\nu}(t_1)\rangle_{R}[S_{\nu}(t_1-t)W_{S}(t),S_{\mu}] + \sum_{\mu,\nu}\langle R_{\nu}(t_1)R_{\mu}(t)\rangle_{R}[S_{\mu},W_{S}(t)S_{\nu}(t_1-t)], \quad (34)
$$

where $S_{\mu}(t) = e^{iH_S t/\hbar} S_{\mu} e^{-iH_S t/\hbar}$ and $R_{\mu}(t) = e^{iH_R t/\hbar} R_{\mu} e^{-iH_R t/\hbar}$. Then the time-local quantum master equation is given by

$$
\frac{\partial}{\partial t}W_S(t) = [\mathcal{L}_S + K_{S,0}(t)]W_S(t)
$$
\n(35)

with

$$
K_{S,0}(t)W_S(t) = \sum_{\mu,\nu} \int_0^t dt_1 \langle R_{\mu}(t)R_{\nu}(t_1) \rangle_R[S_{\nu}(t_1 - t)W_S(t), S_{\mu}] + \sum_{\mu,\nu} \int_0^t dt_1 \langle R_{\nu}(t_1)R_{\mu}(t) \rangle_R[S_{\mu}, W_S(t)S_{\nu}(t_1 - t)]. \tag{36}
$$

On the other hand, since we obtain

$$
e^{\mathcal{L}_{S}t} \langle \mathcal{L}_{SR}(t) \mathcal{L}_{SR}(t_1) \rangle_R e^{-\mathcal{L}_{S}t_1} \bar{W}_S(t_1) = e^{\mathcal{L}_{S}t} \langle \mathcal{L}_{SR}(t) \mathcal{L}_{SR}(t_1) \rangle_R e^{-\mathcal{L}_{S}t} e^{\mathcal{L}_S(t-t_1)} \bar{W}_S(t_1)
$$

\n
$$
= \sum_{\mu,\nu} \langle R_{\mu}(t) R_{\nu}(t_1) \rangle_R \Big[S_{\nu}(t_1-t) \Big(e^{\mathcal{L}_S(t-t_1)} \bar{W}_S(t_1) \Big), S_{\mu} \Big]
$$

\n
$$
+ \sum_{\mu,\nu} \langle R_{\nu}(t_1) R_{\mu}(t) \rangle_R \Big[S_{\mu}, \Big(e^{\mathcal{L}_S(t-t_1)} \bar{W}_S(t_1) \Big) S_{\nu}(t_1-t) \Big], \tag{37}
$$

the time-nonlocal quantum master equation of the relevant quantum system is given by

$$
\frac{\partial}{\partial t}\bar{W}_S(t) = \mathcal{L}_S \bar{W}_S(t) + \int_0^t dt_1 \Phi_{S,0}(t,t_1) \bar{W}_S(t_1),\tag{38}
$$

with

$$
\int_{0}^{t} dt_{1} \, \Phi_{S,0}(t,t_{1}) \bar{W}_{S}(t_{1}) = \sum_{\mu,\nu} \int_{0}^{t} dt_{1} \, \langle R_{\mu}(t) R_{\nu}(t_{1}) \rangle_{R} \Big[S_{\nu}(t_{1} - t) \big(e^{\mathcal{L}_{S}(t-t_{1})} \bar{W}_{S}(t_{1}) \big), S_{\mu} \Big] + \sum_{\mu,\nu} \int_{0}^{t} dt_{1} \, \langle R_{\nu}(t_{1}) R_{\mu}(t) \rangle_{R} \Big[S_{\mu}, \big(e^{\mathcal{L}_{S}(t-t_{1})} \bar{W}_{S}(t_{1}) \big) S_{\nu}(t_{1} - t) \Big].
$$
\n(39)

Next, we obtain the second term on the right-hand side of Eq. [\(24\)](#page-4-0). After straightforward calculation, we have

$$
e^{\mathcal{L}_S(\tau_1 - t_0)} \langle \mathcal{L}_S(\tau_1)[\mathcal{K}_1(t_1), \mathcal{L}_{SR}(\tau_2)] \rangle_R e^{-\mathcal{L}_S(\tau_1 - t_0)} W_S(\tau_1) = \sum_{\mu, \nu} \langle R_{\mu}(\tau_1) R_{\nu}(\tau_2) \rangle_R \{ [\mathcal{K}_1(t_1 - \tau_1), S_{\nu}(\tau_2 - \tau_1)] W_S(\tau_1), S_{\mu} \} + \sum_{\mu, \nu} \langle R_{\nu}(\tau_2) R_{\mu}(\tau_1) \rangle_R \{ S_{\mu}, [\mathcal{K}_1(t_1 - \tau_1), \tilde{S}_{\nu}(\tau_2 - \tau_1)] W_S(\tau_1) \}, \tag{40}
$$

where we have defined the operator $\tilde{S}_v(\tau_2)$ by

$$
\tilde{S}_{\nu}(\tau_2)W_S(\tau_1) = W_S(\tau_1)S_{\nu}(\tau_2). \tag{41}
$$

Then the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ is given by

$$
G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1) = \text{Tr}_S[\mathcal{K}_2\mathcal{U}_S(t_2,t_1)\mathcal{K}_1\mathcal{U}_S(t_1,0)W_S] + \Delta G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1). \tag{42}
$$

The first term of the right-hand side is derived by making use of the quantum regression theorem. The second term is given by

$$
\Delta G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1) = \sum_{\mu, \nu} \int_{t_1}^{t_2} d\tau_1 \int_0^{t_1} d\tau_2 \langle R_{\mu}(\tau_1) R_{\nu}(\tau_2) \rangle_R \text{Tr}_S[\mathcal{K}_2 \mathcal{U}_S(t_2, \tau_1) \{ [\mathcal{K}_1(t_1 - \tau_1), S_{\nu}(\tau_2 - \tau_1)] W_S(\tau_1), S_{\mu} \}]
$$

+
$$
\sum_{\mu, \nu} \int_{t_1}^{t_2} d\tau_1 \int_0^{t_1} d\tau_2 \langle R_{\nu}(\tau_2) R_{\mu}(\tau_1) \rangle_R \text{Tr}_S[\mathcal{K}_2 \mathcal{U}_S(t_2, \tau_1) \{ S_{\mu}, [\mathcal{K}_1(t_1 - \tau_1), \tilde{S}_{\nu}(\tau_2 - \tau_1)] W_S(\tau_1) \}]. \tag{43}
$$

Here, it is important to note that there is no overlap between the two integrations with respect to τ_1 and τ_2 . If the thermal reservoir has a sufficiently short correlation time, the reservoir correlation function can be approximated as $\langle R_\mu(\tau_1)R_\nu(\tau_2)\rangle_R \approx$ $\langle R_{\mu}(\tau_1)R_{\nu}(\tau_1) \rangle_R \delta(\tau_1 - \tau_2)$. In this case, the correction term becomes negligible and thus the quantum regression theorem is established.

To derive the correction term of the double-time correlation function from the time-nonlocal inhomogeneous equation, we fist calculate

$$
e^{\mathcal{L}_{S}\tau_{1}}\langle\mathcal{L}_{S}(\tau_{1})\mathcal{K}_{1}(t_{1})\mathcal{L}_{SR}(\tau_{2})\rangle_{R}e^{-\mathcal{L}_{S}\tau_{2}}\bar{W}_{S}(\tau_{2}) = e^{\mathcal{L}_{S}\tau_{1}}\langle\mathcal{L}_{S}(\tau_{1})\mathcal{K}_{1}(t_{1})\mathcal{L}_{SR}(\tau_{2})\rangle_{R}e^{-\mathcal{L}_{S}\tau_{1}}e^{\mathcal{L}_{S}(\tau_{1}-\tau_{2})}\bar{W}_{S}(\tau_{2})
$$
\n
$$
= \sum_{\mu,\nu}\langle R_{\mu}(\tau_{1})R_{\nu}(\tau_{2})\rangle_{R}\big\{\mathcal{K}_{1}(t_{1}-\tau_{1})S_{\nu}(\tau_{2}-\tau_{1})\big[e^{\mathcal{L}_{S}(\tau_{1}-\tau_{2})}\bar{W}_{S}(\tau_{2})\big],S_{\mu}\big\}
$$
\n
$$
+ \sum_{\mu,\nu}\langle R_{\nu}(\tau_{2})R_{\mu}(\tau_{1})\rangle_{R}\big\{S_{\mu},\mathcal{K}_{1}(t_{1}-\tau_{1})\tilde{S}_{\nu}(\tau_{2}-\tau_{1})\big[e^{\mathcal{L}_{S}(\tau_{1}-\tau_{2})}\bar{W}_{S}(\tau_{2})\big]\big\}.\tag{44}
$$

Then, we can obtain the double-time correlation function in the second-order approximation,

$$
G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1) = \text{Tr}_S[\mathcal{K}_2 \bar{\mathcal{U}}_S(t_2, t_1) \mathcal{K}_1 \bar{\mathcal{U}}_S(t_1, 0) W_S] + \Delta G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1),\tag{45}
$$

with

$$
\Delta G(t_2, t_1 | \mathcal{K}_2, \mathcal{K}_1) = \sum_{\mu, \nu} \int_{t_1}^{t_2} d\tau_1 \int_0^{t_1} d\tau_2 \langle R_{\mu}(\tau_1) R_{\nu}(\tau_2) \rangle_R \text{Tr}_S \big[\mathcal{K}_2 \bar{\mathcal{U}}_S(t_2, \tau_1) \big\{ \mathcal{K}_1(t_1 - \tau_1) S_{\nu}(\tau_2 - \tau_1) \big[e^{\mathcal{L}_S(\tau_1 - \tau_2)} \bar{W}_S(\tau_2) \big], S_{\mu} \big\} \big] + \sum_{\mu, \nu} \int_{t_1}^{t_2} d\tau_1 \int_0^{t_1} d\tau_2 \langle R_{\nu}(\tau_2) R_{\mu}(\tau_1) \rangle_R \text{Tr}_S \big[\mathcal{K}_2 \bar{\mathcal{U}}_S(t_2, \tau_1) \big\{ S_{\mu}, \mathcal{K}_1(t_1 - \tau_1) \tilde{S}_{\nu}(\tau_2 - \tau_1) \big[e^{\mathcal{L}_S(\tau_1 - \tau_2)} \bar{W}_S(\tau_2) \big] \big\} \big].
$$
\n(46)

Comparing Eqs. [\(43\)](#page-6-0) and (46), we find the correspondence between the double-time correlation functions derived from the time-local and time-nonlocal equations,

$$
\mathcal{U}_S(t,t') \Leftrightarrow \bar{\mathcal{U}}_S(t,t'),
$$

\n
$$
[\mathcal{K}_1(t_1 - \tau_1), S_\nu(\tau_2 - \tau_1)] \Leftrightarrow \mathcal{K}_1(t_1 - \tau_1)S_\nu(\tau_2 - \tau_1),
$$

\n
$$
[\mathcal{K}_1(t_1 - \tau_1), \tilde{S}_\nu(\tau_2 - \tau_1)] \Leftrightarrow \mathcal{K}_1(t_1 - \tau_1) \tilde{S}_\nu(\tau_2 - \tau_1),
$$

\n
$$
W_S(\tau_1) \Leftrightarrow e^{\mathcal{L}_S(\tau_1 - \tau_2)} \bar{W}_S(\tau_2).
$$

Since the range of the τ_1 integration in Eq. (46) does not have an overlap with that of the τ_2 integration, the correction term $\Delta G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ becomes negligible if the thermal reservoir has a sufficiently short correlation time.

VI. SIMPLE EXAMPLES

In this section, we calculate the double-time correlation functions of an open two-level system interacting with a thermal reservoir, where a pure dephasing process and a linear dissipative process are assumed. Using the formula [\(42\)](#page-6-0), we explicitly obtain the double-time correlation functions of two observables *A* and *B*. Furthermore, we examine the quantum regression theorem [\[52–57\]](#page-19-0) and the Leggett-Garg inequality [\[61–71\]](#page-19-0). The results are compared with those obtained by exact calculation.

A. Pure dephasing of a two-level system

For a pure dephasing of a two-level system, the interaction Hamiltonian H_{SR} between the two-level system and the thermal reservoir is assumed to be $H_S = \sigma_z \otimes R$, where σ_z is the Pauli operator of the *z* component of a spin-1*/*2 and *R* is a Hermitian operator of the thermal reservoir. When we denote the ground and excited states of the two-level system as $|g\rangle$ and $|e\rangle$, we have $\sigma_z = |e\rangle\langle e| - |g\rangle\langle g|$. Then the time-local quantum master equation for the reduced density operator $W_S(t)$ of the two-level system is obtained from Eq. [\(B24\)](#page-14-0),

$$
\frac{\partial}{\partial t}W_S(t) = -\frac{i}{2}\omega[\sigma_z, W_S(t)] + 2\text{Re}\phi(t)[\sigma_z W_S(t)\sigma_z - W_S(t)],
$$
\n(47)

where ω represents a transition frequency between the ground and excited states and the time-dependent function $\phi(t)$ is given by

$$
\phi(t) = \int_0^t d\tau \ \langle R(\tau)R(0)\rangle_R. \tag{48}
$$

When we express an initial density operator as

$$
W_S(t) = \frac{1}{2} + c\sigma_+ + c^*\sigma_- + \frac{1}{2}c_z\sigma_z,
$$
 (49)

with $|c|^2 + (c_z/2)^2 \le 1$ and ladder operators $\sigma_+ = |e\rangle\langle g|$ and $\sigma_{-} = |g\rangle\langle e|$, the solution of the quantum master equation is

$$
W_S(t) = \frac{1}{2} + u(t,0)c\sigma_+ + u^*(t,0)c^*\sigma_- + \frac{1}{2}c_z\sigma_z, \quad (50)
$$

where
$$
u(t,s)
$$
 $(t \geq s)$ is given by

$$
u(t,s) = \exp\left(-\int_{s}^{t} d\tau \left[i\omega + \gamma(\tau)\right]\right),\tag{51}
$$

with

$$
\gamma(\tau) = 4\text{Re}\int_0^{\tau} d\tau' \left\langle R(\tau')R(0)\right\rangle_R. \tag{52}
$$

Then the map defined by the relation $W_S(t) = U_S(t,0)W_S(0)$ for any initial state $W_S(0)$ is determined by $U_S(t,0)|i\rangle\langle i|$ = $|i\rangle\langle i|$ ($i = g,e$) and

$$
\mathcal{U}_S(t,0)|e\rangle\langle g| = u(t,0)|e\rangle\langle g|, \quad \mathcal{U}_S(t,0)|g\rangle\langle e| = u^*(t,0)|g\rangle\langle e|.
$$
\n(53)

It is easy to see that $U_S(t,s) = U_S(t,0)U_S^{-1}(s,0)$ is provided by $\mathcal{U}_S(t,s)|i\rangle\langle i|=|i\rangle\langle i|$ and

$$
\mathcal{U}_S(t,s)|e\rangle\langle g| = u(t,s)|e\rangle\langle g|, \quad \mathcal{U}_S(t,0)|g\rangle\langle e| = u^*(t,s)|g\rangle\langle e|.
$$
\n(54)

Substituting $K_1 = \sigma_{\pm}$ and $K_2 = \sigma_{\mp}$ into Eq. [\(42\)](#page-6-0), we obtain the first term on the right-hand side,

$$
\langle \sigma_{-}(t_2)\sigma_{+}(t_1)\rangle_0 = \langle \sigma_{-}\sigma_{+}\rangle e^{-i\omega(t_2-t_1)}e^{-g(t_2)+g(t_1)},\tag{55}
$$

$$
\langle \sigma_+(t_2)\sigma_-(t_1) \rangle_0 = \langle \sigma_+\sigma_- \rangle e^{i\omega(t_2 - t_1)} e^{-g(t_2) + g(t_1)}, \tag{56}
$$

and $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_0 = 1$. In the right-hand side of these equations, $\langle \cdots \rangle$ stands for the initial average $Tr_S[\cdots W_S]$ of the two-level system and

$$
g(t) = \int_0^t d\tau \, \gamma(\tau). \tag{57}
$$

These results are also obtained by making use of the quantum regression theorem.

Next we obtain the correction term (43) of the doubletime correlation function. Since we obtain the relations from Eq. [\(50\)](#page-7-0) and $\sigma_{\pm}(t) = e^{-\mathcal{L}_S t} \sigma_{\pm} e^{\mathcal{L}_S t} = \sigma_{\pm} e^{i\omega t}$,

$$
\{[\sigma_+(t_1-\tau_1),\sigma_z]W_S(\tau_1),\sigma_z\} = 2e^{i\omega(t_1-\tau_1)}(1-c_z)\sigma_+, \qquad (58)
$$

$$
\{[\sigma_{-}(t_1-\tau_1),\sigma_{z}]\,W_{S}(\tau_1),\sigma_{z}\} = 2e^{-i\omega(t_1-\tau_1)}(1+c_z)\sigma_{-},\quad (59)
$$

$$
\{[\sigma_z(t_1 - \tau_1), \sigma_z]W_S(\tau_1), \sigma_z\} = 0,\tag{60}
$$

we can derive from Eq. [\(43\)](#page-6-0)

$$
\Delta \langle \sigma_{-}(t_2) \sigma_{+}(t_1) \rangle = 4 \langle \sigma_{-} \sigma_{+} \rangle \int_{t_1}^{t_2} d\tau_1 \int_0^{t_1} d\tau_2
$$

$$
\times \langle R(\tau_1 - \tau_2) R(0) \rangle_R e^{i\omega(t_1 - \tau_1)} u(t_2, \tau_1), \tag{61}
$$

$$
\Delta \langle \sigma_{+}(t_2) \sigma_{-}(t_1) \rangle = 4 \langle \sigma_{+} \sigma_{-} \rangle \int_{t_1} d\tau_1 \int_0^{\tau_2} d\tau_2
$$

$$
\times \langle R(\tau_1 - \tau_2) R(0) \rangle_R e^{-i\omega(t_1 - \tau_1)} u^*(t_2, \tau_1), \tag{62}
$$

where we have used the relation $\langle \sigma_{\pm} \sigma_{\mp} \rangle = \frac{1}{2} (1 \pm c_z)$. Here we introduce the function $f(t_2,t_1)$ by

$$
f(t_2,t_1) = 4 \int_{t_1}^{t_2} d\tau_1 \int_0^{t_1} d\tau_2 \langle R(\tau_1 - \tau_2)R(0) \rangle_R e^{-g(t_2) + g(\tau_1)}.
$$
\n(63)

Then, the double-time correlation functions of the two-level system with the correction term in the second-order approximation are given by

$$
\langle \sigma_{-}(t_2)\sigma_{+}(t_1)\rangle_c = \langle \sigma_{-}\sigma_{+}\rangle e^{-i\omega(t_2-t_1)} [e^{-g(t_2)+g(t_1)}+f(t_2,t_1)],
$$
\n(64)

$$
\langle \sigma_+(t_2)\sigma_-(t_1) \rangle_c = \langle \sigma_+\sigma_- \rangle e^{i\omega(t_2 - t_1)} [e^{-g(t_2) + g(t_1)} + f(t_2, t_1)], \tag{65}
$$

and $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_c = 1$. For the pure dephasing, we can easily obtain the exact double-time correlation functions,

$$
\langle \sigma_{-}(t_2)\sigma_{+}(t_1)\rangle_{\text{ex}} = \langle \sigma_{-}\sigma_{+}\rangle e^{-i\omega(t_2 - t_1)} F(t_2, t_1),\tag{66}
$$

$$
\langle \sigma_+(t_2)\sigma_-(t_1) \rangle_{\text{ex}} = \langle \sigma_+\sigma_- \rangle e^{i\omega(t_2 - t_1)} F(t_2, t_1), \qquad (67)
$$

and $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_{\text{ex}} = 1$, where $F(t_2, t_1)$ is given by

$$
F(t_2, t_1) = \left\{ \exp_{\leftarrow} \left(-2i \int_{t_1}^{t_2} ds \ R(s) \right) \right\}_R.
$$
 (68)

If the thermal reservoir is Gaussian with respect to the Hermitian operator *R*, the function $F(t_2,t_1)$ becomes

$$
F(t_2,t_1) = \exp\left(-2\int_{t_1}^{t_2} ds_1 \int_{t_1}^{t_2} ds_2 \langle \text{TR}(s_1)R(s_2) \rangle\right), \quad (69)
$$

where T stands for the time-ordered product.

We next compare the correlation functions to examine whether our formula is a good approximation and whether the quantum regression theorem is valid. For this purpose, we assume that the thermal reservoir is Gaussian and the reservoir correlation function $\langle R(t_2)R(t_1) \rangle_R$ is given by $\langle R(t_2)R(t_1) \rangle =$ $\frac{1}{4}R_0^2e^{-\lambda|t_2-t_1|}$. In this case, the exact time-time correlation functions become

$$
\langle \sigma_{\mp}(t_2)\sigma_{\pm}(t_1)\rangle_{\text{ex}} = \langle \sigma_{\mp}\sigma_{\pm}\rangle e^{\mp i\omega(t_2 - t_1)} e^{-g(t_2 - t_1)},\tag{70}
$$

with

$$
g(t) = \left(\frac{R_0}{\lambda}\right)^2 [\lambda t - 1 + e^{-\lambda t}]. \tag{71}
$$

When the correlation functions are calculated by means of the quantum regression theorem, we obtain

$$
\langle \sigma_{\mp}(t_2)\sigma_{\pm}(t_1)\rangle_0 = \langle \sigma_{\mp}\sigma_{\pm}\rangle e^{\mp i\omega(t_2-t_1)}e^{-g(t_2)+g(t_1)}.\tag{72}
$$

The result clearly shows that the quantum regression theorem is valid if and only if the equality $g(t_2 + t_1) = g(t_2) + g(t_1)$ holds. Thus the quantum regression theorem is established when the thermal reservoir has a sufficiently short correlation time. On the other hand, Eq. (63) is calculated to be

$$
f(t_2, t_1)
$$

= $e^{-g(t_2)} \left[\frac{R_0^2}{\lambda} (e^{\lambda t_1} - 1) \int_{t_1}^{t_2} ds \ e^{-\lambda s + g(s)} \right]$
= $\left(\frac{R_0}{\lambda} \right)^2 \left[e^{-(R_0/\lambda)^2} e^{-g(t_2)} (e^{\lambda t_1} - 1) \right]$
 $\times \sum_{n=0}^{\infty} \frac{1}{n!} \left(\frac{R_0}{\lambda} \right)^{2n} \frac{e^{-[1+n-(R_0/\lambda)^2]\lambda t_1} - e^{-[1+n-(R_0/\lambda)^2]\lambda t_2}}{1+n-(R_0/\lambda)^2} \right].$ (73)

Substituting this equation into Eqs. (64) and (65) , we obtain the correlation functions with the correction term. In this model, the dephasing time T_2 of the two-level system is given by $T_2 = \lambda/R_0^2$ and the correlation time T_R of the thermal reservoir is $T_R = 1/\lambda$. It is obvious from Eq. (73) that the second-order approximation is valid only when $(R_0^2/\lambda)^2 = T_R/T_2 < 1$. The time dependences of the correlation functions are depicted in Fig. [1.](#page-9-0) Since the second-order approximation is valid if λT_2 = 2*.*0, the correlation function calculated from our formula is

FIG. 1. The time evolution of the normalized correlation functions $\langle \sigma_{\pm}(t_2) \sigma_{\mp}(t_1) \rangle_{0,c_1}$, $\langle \sigma_{\pm} \sigma_{\mp} \rangle$ with $t_1/T_2 = 1.0$, $t_1/T_2 = 4.0$ and $t_1/T_2 = 1$ 7*.*0. The black solid line stands for the exact correlation function, the red dashed line for the correlation function derived our formula, and the blue dash-dotted line for one by means of the quantum regression theorem. In this figure, we set $\lambda T_2 = 2.0$ in panel (a) and $\lambda T_2 = 0.8$ in panel (b).

much closer to the exact one than the correlation function derived by the quantum regression theorem [Fig. $1(a)$]. On the other hand, when $\lambda T_2 = 0.8$, it deviates from the exact correlation function as $|t_2 - t_1|/T_2$ is larger. However, our result is better than the quantum regression theorem. We note that the quantum regression theorem is established only in the limit $\lambda T_2 \rightarrow \infty$.

Noninvasive measurability and macrorealism lead to the Leggett-Garg inequality, which is considered as a temporal version of the Bell inequality $[61–71]$. Let us denote as $Q(t)$ a dichotomous observable which takes the values ± 1 at time *t*. Then assuming macroscopic realism and noninvasive measurability, we can derive the the Leggett-Garg inequality, $\langle Q(t_2)Q(t_1)\rangle + \langle Q(t_3)Q(t_2)\rangle - \langle Q(t_3)Q(t_1)\rangle \leq 1$ with *t*₃ > $t_2 > t_1$. For the Pauli operator σ_x of a two-level system, the inequality is replaced by [\[65,69\]](#page-19-0)

$$
K_3(t_3,t_2,t_1) = \text{Re}\langle\sigma_x(t_2)\sigma_x(t_1)\rangle + \text{Re}\langle\sigma_x(t_3)\sigma_x(t_2)\rangle
$$

-\text{Re}\langle\sigma_x(t_3)\sigma_x(t_1)\rangle \le 1. (74)

The correlation function $\langle \sigma_x(t_2) \sigma_x(t_1) \rangle$ with $\sigma_x = |e\rangle \langle g| +$ $|g\rangle\langle e|$ is given by

$$
\langle \sigma_x(t_2)\sigma_x(t_1) \rangle = \cos \omega(t_2 - t_1)G(t_2, t_1) + i \langle \sigma_z \rangle \sin \omega(t_2 - t_1)G(t_2, t_1), \qquad (75)
$$

with

$$
G(t_2, t_1)
$$

=
$$
\begin{cases} e^{-g(t_2-t_1)} & \text{for the exact calculation} \\ e^{-g(t_2)+g(t_1)} + f(t_2, t_1) & \text{for the derived formula} \\ e^{-g(t_2)+g(t_1)} & \text{for the quantum regression} \\ \text{theorem} & \text{theorem} \end{cases}
$$

(76)

Note that the real part of the correlation function $G(t_2,t_1)$ does not depend on the initial condition. To investigate whether the Leggett-Garg inequality is violated, we introduce

$$
K_3(\tau,t) = \text{Re}\langle\sigma_x(t+2\tau)\sigma_x(t+\tau)\rangle + \text{Re}\langle\sigma_x(t+\tau)\sigma_x(t)\rangle
$$

- \text{Re}\langle\sigma_x(t+2\tau)\sigma_x(t)\rangle, (77)

where the inequality $K_3(\tau,t) > 1$ means the violation of the Leggett-Garg inequality. In Fig. [2,](#page-10-0) the parameter $K_3(\tau,t)$ is plotted as a function of τ . It is found from the figure that our formula for the double-time correlation function is a good approximation for the exact calculation of the parameter $K_3(\tau,t)$. Furthermore, one can see that the Leggett-Garg inequality is violated in the initial time region for all the cases. For small values of λT_2 , the revival of the violation can be observed.

B. Linear dissipative process of a two-level system

Next, we consider a linear dissipative process of a two-level system to obtain the double-time correlation function, where the interaction Hamiltonian between the two-level system and the thermal reservoir is given by $H_{SR} = \sigma_- R^{\dagger} + \sigma_+ R$. In the interaction picture, it becomes $H_{SR}(t) = \sigma_- R^{\dagger}(t) e^{-i\omega t}$ + $\sigma_+ R(t)e^{i\omega t}$ with $R(t) = e^{iH_R t/\hbar}Re^{-iH_R t/\hbar}$. In this case, the time-local quantum master equation for the reduced density operator of the two-level system is given by

$$
\frac{\partial}{\partial t}W_S(t) = -\frac{i}{\hbar}[H_S, W_S(t)] + \phi_{+-}(t)[\sigma_+W_S(t), \sigma_-] \n+ \phi_{+-}^*(t)[\sigma_+, W_S(t)\sigma_-] + \phi_{-+}(t)[\sigma_-W_S(t), \sigma_+] \n+ \phi_{-+}^*(t)[\sigma_-, W_S(t)\sigma_+],
$$
\n(78)

with

$$
\phi_{-+}(t) = \int_0^t dt_1 \langle R(t_1)R^{\dagger}(0)\rangle_R e^{i\omega t_1},\tag{79}
$$

$$
\phi_{+-}(t) = \int_0^t dt_1 \langle R^\dagger(t_1)R(0)\rangle_R e^{-i\omega t_1}.
$$
 (80)

When we express the reduced density operator as $W_S(t) =$ $\frac{1}{2} + c(t)\sigma_+ + c^*(t)\sigma_- + \frac{1}{2}c_z(t)\sigma_z$, we obtain the differential equations of the coefficients $c(t)$ and $c_z(t)$,

$$
\frac{d}{dt}c(t) = -[\gamma(t) + i\delta(t) + i\omega]c(t),\tag{81}
$$

$$
\frac{d}{dt}c_z(t) = -2\gamma(t)[c_z(t) - w(t)],\tag{82}
$$

FIG. 2. The dependence of the parameter $K_3(\tau,t)$ on time τ with (a) $t/T_2 = 0.0$ and $\lambda T_2 = 2.0$, (b) $t/T_2 = 2.0$ and $\lambda T_2 = 2.0$, (c) $t/T_2 = 0.0$ and $\lambda T_2 = 0.8$, and (d) $t/T_2 = 2.0$ and $\lambda T_2 = 0.8$. In the figure, we set $\omega T_2 = 5.0$. The black solid line stands for the exact calculation, the red dashed line for our formula, and the blue dash-dotted line for the quantum regression theorem. The inequality $K_3(\tau,t) > 1$ means the violation of the Leggett-Garg inequality.

where the time-dependent parameters $\gamma(t)$, $\delta(t)$, and $w(t)$ are given by

$$
\gamma(t) = \text{Re}[\phi_{-+}(t) + \phi_{+-}^*(t)],\tag{83}
$$

$$
\delta(t) = \text{Im}[\phi_{-+}(t) + \phi_{+-}^*(t)],\tag{84}
$$

$$
w(t) = -\frac{\text{Re}[\phi_{-+}(t) - \phi_{+-}(t)]}{\text{Re}[\phi_{-+}(t) + \phi_{+-}(t)]}.
$$
 (85)

Then we obtain the solutions,

$$
c(t) = u(t)c(0), \quad c_z(t) = u_z(t)c_z(0) + \sigma(t), \tag{86}
$$

with

$$
u(t) = e^{-i\omega t} \exp\left(-\int_0^t d\tau \left[\gamma(\tau) + i\delta(\tau)\right]\right),\qquad(87)
$$

$$
u_z(t) = \exp\left(-2\int_0^t d\tau \, \gamma(\tau)\right),\tag{88}
$$

$$
\sigma(t) = w(t) - w(0) \exp\left(-2 \int_0^t d\tau \gamma(\tau)\right)
$$

$$
- \int_0^t d\tau \, \dot{w}(\tau) \exp\left(-2 \int_0^{\tau} d\tau' \gamma(\tau')\right), \quad (89)
$$

where we set $\dot{w}(t) = \frac{dw(t)}{dt}$.

To obtain an explicit form of the correlation function, we assume that the thermal reservoir consists of independent harmonic oscillators in the ground state and it has the Lorentzian spectrum $[4,19]$. Then, we can obtain the double-time correlation functions,

$$
\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_0 = 1 + \big(e^{-g(t_2)} - e^{-g(t_1)} \big) (1 + \langle \sigma_z \rangle), \tag{90}
$$

$$
\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_c = 1 + \left[e^{-g(t_2)} - e^{-g(t_1)} + \left(\frac{\gamma}{\lambda} \right) \left(1 - e^{-\lambda(t_2 - t_1)} \right) \right]
$$

$$
\times (1 - e^{-\lambda t_1})e^{-g(t_2)}\bigg](1 + \langle \sigma_z \rangle), \tag{91}
$$

with $g(t) = \left(\frac{y}{\lambda}\right) (\lambda t - 1 + e^{-\lambda t})$. These correlation functions are derived in Appendix [D.](#page-15-0) On the other hand, we can derive the exact correlation function [\[76\]](#page-19-0),

$$
\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_{\text{ex}}
$$

= 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), (92)

where $A(t)$ is given by

$$
A(t) = e^{-\frac{1}{2}\lambda t} \left[\cosh\left(\frac{a}{2}\lambda t\right) + \frac{1}{a}\sinh\left(\frac{a}{2}\lambda t\right) \right],\qquad(93)
$$

with $a = \sqrt{1 - 2\gamma/\lambda}$. The derivation of the exact correlation function is briefly summarized in Appendix [E.](#page-16-0) It is easy to check that $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_{\text{ex}} \approx \langle \sigma_z(t_2) \sigma_z(t_1) \rangle_{\text{c}} \approx \langle \sigma_z(t_2) \sigma_z(t_1) \rangle_0$ is established if the condition $\gamma/\lambda \ll 1$ is fulfilled. In this case, the quantum regression theorem is valid. It is important

FIG. 3. The time evolution of the correlation functions $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_{0,c,\text{ex}}$ with (a) $\lambda/\gamma = 4.0$ and (b) $\lambda/\gamma = 0.2$. The black solid line stands for the exact correlation function, the red dashed line for one derived from our formula, and the blue dash-dotted line for one by making use of the quantum regression theorem. We set from the left to the right $\gamma t_1 = 1.0, 1.5, 3.0$ in panel (a) and $\gamma t_1 = 1.0, 2.5, 4.0$ in panel (b). In this figure, we set $\langle \sigma_z \rangle = 1.0$.

to note that this condition is much stronger than the Markovian condition. In fact, the reduced time evolution is Markovian if and only if $\lambda > 2\gamma$ [\[77–80\]](#page-19-0). The correlation functions are depicted in Fig. 3. It is found from the figure that the correlation function given by our formula is a good approximation in the time region of $\gamma(t_2 - t_1) \lesssim 1$ not only for $\lambda/\gamma > 1$ but also for λ/γ < 1. However, it approaches the correlation function derived by the quantum regression theorem and deviates from the exact one as the time difference $t_2 - t_1$ is large.

Finally, using the correlation function $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle$, we investigate the violation of the Leggett-Garg inequality. The inequality is given by

$$
\operatorname{Re}\langle\sigma_z(t_3)\sigma_z(t_2)\rangle + \operatorname{Re}\langle\sigma_z(t_2)\sigma_z(t_1)\rangle - \operatorname{Re}\langle\sigma_z(t_3)\sigma_z(t_1)\rangle \leq 1,
$$
\n(94)

with $t_3 > t_2 > t_1$. First, we note that the correlation function derived from the quantum regression theorem satisfies the Leggett-Garg inequality. In fact, using Eq. [\(90\)](#page-10-0),

FIG. 4. The dependence of the parameter $K_3(\tau, t)$ on time τ with (a) $\gamma t = 0.0$ and $\lambda/\gamma = 4.0$, (b) $\gamma t = 1.0$ and $\lambda/\gamma = 4.0$, (c) $\gamma t = 0.0$ and $\lambda/\gamma = 0.2$, and (d) $\gamma t = 1.0$ and $\lambda/\gamma = 0.2$. The black solid line stands for the exact correlation function, the red dashed line for one with the correction term, and the blue dash-dotted line for one derived by the quantum regression theorem. In this figure, we set $\langle \sigma_z \rangle = 1.0$. The inequality $K_3(\tau,t) > 1$ means the violation of the Leggett-Garg inequality.

we obtain the equality $\text{Re}\langle\sigma_z(t_3)\sigma_z(t_2)\rangle + \text{Re}\langle\sigma_z(t_2)\sigma_z(t_1)\rangle \text{Re}\langle\sigma_z(t_3)\sigma_z(t_1)\rangle = 1.$ To examine the violation of the Leggett-Garg inequality, we introduce the parameter $K_3(\tau,t)$ by

$$
K_3(\tau,t) = \text{Re}\langle\sigma_z(t+2\tau)\sigma_z(t+\tau)\rangle + \text{Re}\langle\sigma_z(t+\tau)\sigma_z(t)\rangle
$$

- \text{Re}\langle\sigma_z(t+2\tau)\sigma_z(t)\rangle, (95)

which is plotted as a function of τ in Fig. [4.](#page-11-0) The figure clearly shows the violation of the Leggett-Garg inequality. Furthermore, it is found that the correlation function $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_c$ is a good approximation for the exact correlation function $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_{\text{ex}}$ if λ/γ is not small.

VII. SUMMARY

In this paper, using the projection operator method, we have studied the double-time correlation functions of two quantum operations performed on an open quantum system which is placed under the influence of a thermal reservoir. The two quantum operations are done sequentially during the irreversible time evolution of the relevant quantum system. The double-time correlation function considered in this paper includes a usual correlation function, a linear response function, a weak value of an observable, and joint probability of sequential measurement as a special case. Here, it is important to note that the quantum regression theorem cannot be used for calculating double-time correlation functions when the thermal reservoir has a finite correlation time. To obtain the double-time correlation function, we introduce a fictitious field and a quasidensity operator. The double-time correlation function is provided by the first-order term of the reduced quasidensity operator with respect to the fictitious field. Applying the projection operator method, we have derived the timelocal and time-nonlocal equations for the reduced quasidensity operator. By solving these equations, we have obtained the formulas for calculating the double-time correlation function. Furthermore, applying the second-order approximation with respect to the system-reservoir interaction, we have obtained the explicit form of the double-time correlation function. The derived correlation function consists of two parts. One is obtained by means of the quantum regression theorem and the other is a correction term that takes care of finite correlation time of the thermal reservoir. The latter causes the violation of the quantum regression theorem and vanishes only if correlation time of the thermal reservoir is sufficiently short. This condition is stronger than that of the Markovianity for the reduced time evolution of the relevant quantum system. As an example, we have calculated the double-time correlation functions for a pure dephasing process and a linear dissipative process of an open two-level system. Comparing the results with the exact calculation, we have shown that the derived formulas are a good approximation for the exact double-time correlation functions. Furthermore, using the results, we have investigated the violation of the Leggett-Garg inequality.

APPENDIX A: TIME-LOCAL AND TIME-NONLOCAL EQUATIONS FOR THE REDUCED QUASIDENSITY OPERATOR

To derive the time-evolution equation for the reduced quasidensity operator $W_S(t|g) = Tr_R W(t|g)$ of the relevant system from $\partial W(t|g)/\partial t = [\mathcal{L} + g(t)\mathcal{K}_1]W(t|g)$, we decompose the

Liouvillian superoperator as follows:

$$
\mathcal{L} + g(t)\mathcal{K}_1 = \mathcal{L}_S + g(t)\mathcal{K}_1 + \mathcal{L}_R + \mathcal{L}_{SR} \qquad (A1)
$$

$$
= \mathcal{L}_S(t) + \mathcal{L}_R + \mathcal{L}_{SR} \tag{A2}
$$

$$
= \mathcal{L}(t) + \mathcal{L}_{SR} \tag{A3}
$$

$$
= \mathcal{L}_0 + g(t)\mathcal{K}_1 + \mathcal{L}_{SR}, \tag{A4}
$$

with $\mathcal{L}_S = -(i/\hbar)H_S^{\times}$, $\mathcal{L}_R = -(i/\hbar)H_R^{\times}$, $\mathcal{L}_{SR} = -(i/\hbar)H_{SR}^{\times}$, $\mathcal{L}_0 = \mathcal{L}_S + \mathcal{L}_R$, and $\mathcal{L}_0(t) = \mathcal{L}_0 + g(t)\mathcal{K}_1$. Then in the interaction picture, we have $\partial \hat{W}(t|g)/\partial t = \hat{\mathcal{L}}_{SR}(t) \hat{W}(t|g)$, where we set $\hat{W}(t|g) = \mathcal{V}^{-1}(t,t_0|g)W(t|g)$ and $\hat{\mathcal{L}}_{SR}(t) =$ $V^{-1}(t,t_0|g) \mathcal{L}_{SR} V(t,t_0|g)$ with

$$
\mathcal{V}(t,t_0|g) = \exp_{\leftarrow} \left(\int_{t_0}^t d\tau \, \mathcal{L}(t) \right). \tag{A5}
$$

Using the projection operator method [\[4\]](#page-18-0), where a projection operator is defined by $\mathcal{P} \bullet = \rho_R \text{Tr}_R \bullet$ with a density operator ρ_R of the thermal reservoir, we can derive a time-local time-evolution equation for the reduced quasidensity operator $W_S(t|g)$ of the relevant system [\[34–36\]](#page-18-0),

$$
\frac{\partial}{\partial t}\hat{W}_S(t|g) = \hat{\mathcal{K}}_S(t|g)\hat{W}_S(t|g) + \hat{\mathcal{I}}_S(t|g),\tag{A6}
$$

with

$$
\hat{\mathcal{K}}_S(t|g) = \langle \hat{\mathcal{L}}_{SR}(t)[1 - \hat{\Sigma}(t|g)]^{-1} \rangle_R, \tag{A7}
$$

$$
\hat{\mathcal{I}}_S(t|g) = \text{Tr}_R\{\hat{\mathcal{L}}_{SR}(t)[1-\hat{\Sigma}(t|g)]^{-1}\hat{\mathcal{G}}_P(t,t_0|g)(1-\mathcal{P})W\}.
$$
\n(A8)

In these equations, we set $\langle \bullet \rangle_R = \text{Tr}_R[\bullet \rho_R]$ and

$$
\hat{\Sigma}(t|g) = \int_{t_0}^t d\tau \,\hat{\mathcal{G}}_P(t,\tau|g)(1-\mathcal{P})\hat{\mathcal{L}}_{SR}(\tau)\mathcal{P}\hat{\mathcal{G}}^{-1}(t,\tau|g),
$$
\n(A9)

$$
\hat{\mathcal{G}}_P(t,\tau|g) = \exp_{\leftarrow}\left(\int_{\tau}^t d\tau'(1-\mathcal{P})\hat{\mathcal{L}}_{SR}(\tau')\right),\tag{A10}
$$

$$
\hat{\mathcal{G}}(t,\tau|g) = \exp_{\leftarrow} \left(\int_{\tau}^{t} d\tau' \, \hat{\mathcal{L}}_{SR}(\tau') \right). \tag{A11}
$$

We can also derive a time-nonlocal equation [\[29–31\]](#page-18-0),

$$
\frac{\partial}{\partial t} \hat{W}_S(t|g) = \langle \hat{\mathcal{L}}_{SR}(t) \rangle_R \hat{W}_S(t|g) \n+ \int_{t_0}^t dt_1 \hat{\Phi}_S(t, t_1|g) \hat{W}_S(t_1|g) + \hat{\mathcal{J}}_S(t|g),
$$
\n(A12)

where $\hat{\Phi}_S(t,t_1|g)$ and $\hat{\mathcal{J}}_S(t|g)$ are given by

$$
\hat{\Phi}_S(t,t_1|g) = \langle \hat{\mathcal{L}}_{SR}(t)\hat{\mathcal{G}}_P(t,t_1|g)(1-\mathcal{P})\hat{\mathcal{L}}_{SR}(t_1)\rangle_R, \text{ (A13)}
$$

$$
\hat{\mathcal{J}}_S(t|g) = \text{Tr}_R[\hat{\mathcal{L}}_{SR}(t)\hat{\Phi}_S(t,t_0|g)(1-\mathcal{P})W]. \text{ (A14)}
$$

In the Schrödinger picture, the time-local equation becomes

$$
\frac{\partial}{\partial t} W_S(t|g) = [\mathcal{L}_S + g(t)\mathcal{K}_1]W_S(t|g) + \mathcal{K}_S(t|g)W_S(t|g) + \mathcal{I}_S(t|g), \quad \text{(A15)}
$$

with

$$
\mathcal{K}_S(t|g) = \mathcal{V}_S(t,t_0)\hat{\mathcal{K}}_S(t|g)\mathcal{V}_S^{-1}(t,t_0),\tag{A16}
$$

$$
\mathcal{I}_S(t|g) = \mathcal{V}_S(t,t_0)\hat{\mathcal{I}}_S(t|g),\tag{A17}
$$

$$
\mathcal{V}_S(t,t_0) = \exp_{\leftarrow} \left(\int_{t_0}^t d\tau \left[\mathcal{L}_S + g(\tau) \mathcal{K}_1 \right] \right), \quad \text{(A18)}
$$

and the time-nonlocal equation is given by

$$
\frac{\partial}{\partial t}W_S(t|g) = [\mathcal{L}_S + g(t)\mathcal{K}_1]W_S(t|g) + \Phi_S(t|g)W_S(t|g)
$$

$$
+ \int_{t_0}^t dt_1 \Phi_S(t, t_1|g)W_S(t_1|g) + \mathcal{J}_S(t|g), \quad (A19)
$$

with

$$
\Phi_S(t|g) = \mathcal{V}_S(t,t_0)\langle \hat{\mathcal{L}}_{SR}(t) \rangle_R \mathcal{V}_S^{-1}(t,t_0), \tag{A20}
$$

$$
\Phi_S(t,t_1|g) = V_S(t,t_0)\hat{\Phi}_S(t,t_1|g)V_S^{-1}(t_1,t_0), \quad (A21)
$$

$$
\mathcal{J}_S(t|g) = \mathcal{V}_S(t,t_0)\hat{\mathcal{J}}_S(t|g). \tag{A22}
$$

APPENDIX B: DERIVATION OF THE DOUBLE-TIME CORRELATION FUNCTION FROM THE TIME-LOCAL EQUATION IN THE SECOND-ORDER APPROXIMATION

In this appendix, applying the second-order approximation to the time-local equation (11) , we derive the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ given by Eq. [\(23\)](#page-4-0). First, it is easy to see that $\mathcal{K}_S(t|g)$ and $\mathcal{I}_S(t|g)$, given by Eqs. (A16) and $(A17)$, become up to second order with respect to the system-reservoir interaction,

$$
\mathcal{K}_S(t|g) = \int_{t_0}^t d\tau \, \mathcal{V}_S(t,t_0) \langle \hat{\mathcal{L}}_{SR}(t) \hat{\mathcal{L}}_{SR}(\tau) \rangle_R \mathcal{V}_S^{-1}(t,t_0)
$$
\n
$$
= \int_{t_0}^t d\tau \, e^{\mathcal{L}_S(t-t_0)} \langle \mathcal{L}_{SR}(t) e^{-\mathcal{L}_0(t-\tau)} \mathcal{V}(t,\tau|g) e^{\mathcal{L}_0(t-\tau)}
$$
\n
$$
\times \mathcal{L}_{SR}(\tau) e^{-\mathcal{L}_0(\tau-t_0)} \mathcal{V}(\tau,t_0|g) \rangle_R \mathcal{V}_S^{-1}(t,t_0), \quad (B1)
$$

$$
\mathcal{I}_S(t|g) = \mathcal{V}_S(t,t_0) \left\{ \text{Tr}_R[\hat{\mathcal{L}}_{SR}(t)\delta W] + \int_{t_0}^t d\tau \, \text{Tr}_R[\hat{\mathcal{L}}_{SR}(t)\hat{\mathcal{L}}_{SR}(\tau)\delta W] \right\},\tag{B2}
$$

where we set $\delta W = W - W_S \otimes \rho_R$ and

$$
\mathcal{L}_{SR}(t) = e^{-\mathcal{L}_0(t-t_0)} \mathcal{L}_{SR} e^{\mathcal{L}_0(t-t_0)}.
$$
 (B3)

Since we only need the terms up to the first order with respect to the fictitious field $g(t)$, we can use the approximated relations,

$$
e^{-\mathcal{L}_0(t-t_0)} \mathcal{V}(t,t_1|g) e^{\mathcal{L}_0(t_1-t_0)} = 1 + \int_{t_1}^t d\tau \ g(\tau) \mathcal{K}_1(\tau)
$$

$$
\equiv 1 + \Delta \mathcal{V}(t,t_1), \qquad (B4)
$$

$$
e^{-\mathcal{L}_0(t_1 - t_0)} \mathcal{V}(t_1, t_0 | g) = 1 + \int_{t_0}^{t_1} d\tau \ g(\tau) \mathcal{K}(\tau)
$$

$$
= 1 + \Delta \mathcal{V}(t_1, t_0) \tag{B5}
$$

$$
= 1 + \Delta V(t_1, t_0),
$$
 (B3)

$$
V_S^{-1}(t, t_0)e^{\mathcal{L}_S(t - t_0)} = 1 - \int_{t_0}^t d\tau g(\tau) \mathcal{K}_1(\tau)
$$

$$
\equiv 1 - \Delta \mathcal{V}(t, t_0), \tag{B6}
$$

with

$$
\mathcal{K}_1(t) = e^{-\mathcal{L}_S(t-t_0)} \mathcal{K}_1 e^{\mathcal{L}_S(t-t_0)},
$$
 (B7)

$$
\Delta \mathcal{V}(t, t_1) = \int_{t_1}^t d\tau \ g(\tau) \mathcal{K}_1(\tau). \tag{B8}
$$

Here we note that $\Delta V(t,t_1)$ is a superoperator of the relevant quantum system.

The superoperator $\mathcal{K}_S(t|g)$ given by Eq. (B1) is further calculated to be

$$
\mathcal{K}_S(t|g) = \mathcal{K}_{S,0}(t) + \mathcal{K}_{S,1}(t|g),
$$
 (B9)

where the zeroth- and first-order terms are given respectively by

$$
\mathcal{K}_{S,0}(t) = \int_{t_0}^t dt_1 e^{\mathcal{L}_S(t-t_0)} \langle \mathcal{L}_{SR}(t) \mathcal{L}_{SR}(t_1) \rangle_R e^{-\mathcal{L}_S(t-t_0)}, \quad \text{(B10)}
$$

$$
\mathcal{K}_{S,1}(t|g) = \int_{t_0}^t dt_1 e^{\mathcal{L}_S(t-t_0)} \langle \mathcal{L}_{SR}(t)[\Delta \mathcal{V}(t,t_1), \mathcal{L}_{SR}(t_1)] \rangle_R e^{-\mathcal{L}_S(t-t_0)}.
$$

In deriving the last equation, we have used the equality $\Delta V(t, t_0) = \Delta V(t, t_1) + \Delta V(t_1, t_0)$ with $t > t_1 > t_0$. Then by rearranging the order of integration, we can rewrite the first-order superoperator $\mathcal{K}_{S,1}(t|g)$ into

$$
\mathcal{K}_{S,1}(t|g) = \int_{t_0}^t d\tau \ g(\tau) \mathcal{K}_{S,1}(t,\tau), \tag{B12}
$$

with

$$
\mathcal{K}_{S,1}(t,\tau) = \int_{t_0}^{\tau} dt_1 e^{\mathcal{L}_S(t-t_0)} \langle \mathcal{L}_{SR}(t) [\mathcal{K}_1(\tau),
$$

$$
\mathcal{L}_{SR}(t_1)] \rangle_R e^{-\mathcal{L}_S(t-t_0)}.
$$
 (B13)

The inhomogeneous term $\mathcal{I}_S(t|g)$, given by Eq. (B2), is calculated up to the first order with respect to the fictitious field $g(t)$,

$$
\mathcal{I}_S(t|g) = I_{S,0}(t) + I_{S,0}(t|g),
$$
 (B14)

where the zeroth- and first-order terms are given respectively by

$$
I_{S,0}(t) = e^{\mathcal{L}_S(t-t_0)} \text{Tr}_R[\mathcal{L}_{SR}(t)\delta W] + \int_{t_0}^t dt_1 e^{\mathcal{L}_S(t-t_0)} \text{Tr}_R[\mathcal{L}_{SR}(t)\mathcal{L}_{SR}(t_1)\delta W], \quad (B15)
$$

$$
I_{S,1}(t|g) = \int_{t_0}^t d\tau \ g(\tau) I_{S,1}(t,\tau), \tag{B16}
$$

with

$$
I_{S,1}(t,\tau) = e^{\mathcal{L}_S(t-t_0)} \text{Tr}_R[\mathcal{L}_{SR}(t) \mathcal{K}_1(\tau) \delta W] + \int_{t_0}^t dt_1 e^{\mathcal{L}_S(t-t_0)} \text{Tr}_R[\mathcal{L}_{SR}(t) \text{T} \{\mathcal{L}_{SR}(t_1) \mathcal{K}_1(\tau)\} \delta W].
$$
\n(B17)

Here we note that $\text{Tr}_R[\mathcal{L}_{SR}(t)\delta W]$ and $\text{Tr}_R[\mathcal{L}_{SR}(t)\mathcal{K}_1(\tau)\delta W]$ do not necessarily become zero even if $\langle \hat{\mathcal{L}}_{SR}(t) \rangle_R = 0$.

Therefore, we finally obtain the time-local equation for the reduced quasidensity operator $W_S(t|g)$ in the second-order approximation,

$$
\frac{\partial}{\partial t} W_S(t|g) = [\mathcal{L}_S + g(t)\mathcal{K}_1]W_S(t|g) + \mathcal{K}_{S,0}(t)W_S(t|g) \n+ \mathcal{K}_{S,1}(t|g)W_S(t|g) + I_{S,0}(t) + I_{S,1}(t|g).
$$
\n(B18)

The formal solution of this equation is given by

$$
W_S(t|g) = U_S(t,t_0)U'_S(t,t_0|g)W_S
$$

+ $\int_{t_0}^t dt_1 U_S(t,t_0)U'_S(t,t_1|g)U_S^{-1}(t_1,t_0)I_{S,0}(t_1)$
+ $\int_{t_0}^t dt_1 U_S(t,t_0)U'_S(t,t_1|g)U^{-1}(t_1,t_0)I_{S,1}(t_1|g),$
(B19)

with

$$
\mathcal{U}_{S}(t,t_{0}) = \exp_{\leftarrow} \left(\int_{t_{0}}^{t} dt_{1} \left[\mathcal{L}_{S} + \mathcal{K}_{S,0}(t_{1}) \right] \right), \qquad (B20)
$$

$$
\mathcal{U}'_{S}(t,t_{0}|g) = \exp_{\leftarrow} \left(\int_{0}^{t} dt_{1} \mathcal{U}_{S}^{-1}(t_{1},t_{0}) \left[g(t_{1}) \mathcal{K}_{1} + \mathcal{K}_{S,1}(t_{1}|g) \right] \mathcal{U}_{S}(t_{1},t_{0}) \right). \qquad (B21)
$$

By discarding all the terms higher than the first order with respect to the fictitious field $g(t)$, we can obtain the reduced quasidensity operator $W_S(t|g)$ of the relevant quantum system,

$$
W_S(t|g) = W_{S,0}(t) + W_{S,1}(t|g). \tag{B22}
$$

The zeroth-order term $W_{S,0}(t)$ is nothing but the reduced density operator of the relevant quantum system, that is,

$$
W_{S,0}(t) = U_S(t,t_0)W_S + \int_{t_0}^t dt_1 U_S(t,t_1)I_{S,0}(t_1), \quad (B23)
$$

which is a solution of the inhomogeneous time-local quantum master equation in the second-order approximation [\[4,34–36\]](#page-18-0),

$$
\frac{\partial}{\partial t}W_{S,0}(t) = [\mathcal{L}_S + \mathcal{K}_{S,0}(t)]W_{S,0}(t) + I_{S,0}(t).
$$
\n(B24)

After some calculation, we can obtain the first-order correction term $W_{S,1}(t|g)$ of the reduced quasidensity operator,

$$
W_{S,1}(t|g) = \int_{t_0}^t d\tau g(\tau)U_S(t,\tau)K_1U_S(\tau,t_0)W_S + \int_{t_0}^t d\tau g(\tau)\int_{\tau}^t dt_1 \int_{t_0}^{\tau} dt_2 U_S(t,t_1)e^{\mathcal{L}_S(t_1-t_0)}\langle \mathcal{L}_{SR}(t_1)[K_1(\tau),\mathcal{L}_{SR}(t_2)]\rangle_R
$$

\n
$$
\times e^{-\mathcal{L}_S(t_1-t_0)}U_S(t_1,t_0)W_S + \int_{t_0}^t d\tau g(\tau)\int_{t_0}^{\tau} dt_1 U_S(t,\tau)K_1U_S(\tau,t_1)I_{S,0}(t_1)
$$

\n
$$
+ \int_{t_0}^t d\tau g(\tau)\int_{\tau}^t dt_1 \int_{t_0}^{t_1} dt_2 \int_{t_0}^{\tau} dt_3 U_S(t,t_1)e^{\mathcal{L}_S(t_1-t_0)}\langle \mathcal{L}_{SR}(t_1)[K_1(\tau),\mathcal{L}_{SR}(t_3)]\rangle_R e^{-\mathcal{L}_S(t_1-t_0)}U_S(t_1,t_2)I_{S,0}(t_2)
$$

\n
$$
+ \int_{t_0}^t d\tau g(\tau)\int_{\tau}^t dt_1 U_S(t,t_1)I_{S,1}(t_1,\tau),
$$
\n(B25)

with

$$
U_S(t,\tau) = U_S(t,t_0) U_S^{-1}(\tau,t_0) \quad (t > \tau > t_0).
$$
 (B26)

Then by using the relation $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1) =$ $Tr_S[\mathcal{K}_2 \delta W_{S,1}(t_2|g)/\delta g(t_1)]_{g\to 0}$, we can obtain the correlation function (23) .

Finally, we briefly consider the relation to the result given in Ref. [\[58\]](#page-19-0). For this purpose, we assume that there is no initial correlation between the relevant quantum system and the thermal reservoir. In this case, when we set $\rho_R = W_R$ in the definition of the projection operator P with the initial reservoir state W_R , Eq. (B18) becomes

$$
\frac{\partial}{\partial t}W_S(t|g) = [\mathcal{L}_S + g(t)\mathcal{K}_1]W_S(t|g) + \mathcal{K}_{S,0}(t)W_S(t|g) \n+ \mathcal{K}_{S,1}(t|g)W_S(t|g).
$$
\n(B27)

Performing the functional derivative with respect to $g(\tau)$ and taking the limit $g(t) \rightarrow 0$, we obtain

$$
\frac{\partial}{\partial t} W_S(t, \tau | \mathcal{K}_1) = \mathcal{L}_S W_S(t, \tau | \mathcal{K}_1) + \mathcal{K}_{S,0}(t) W_S(t, \tau | \mathcal{K}_1) \n+ \mathcal{K}_{S,1}(t, \tau) W_{S,0}(t).
$$
\n(B28)

The system operators $\Lambda(t,t')$ and $\rho(t,t')$ introduced in Ref. [\[58\]](#page-19-0) are related to $W_S(t, \tau | \mathcal{K}_1)$ and $W_{S,0}(t)$ by $\Lambda(\tau - t_0, t \mathcal{L}_1 = W_S(t, \tau | \mathcal{K}_1)$ with $\mathcal{K}_1 = B$ and $\rho(\tau - t_0, t - \tau) =$ $e^{\mathcal{L}_S(t-\tau)}W_{S,0}(t)$. Then we find that the second and third terms on the right-hand side of Eq. (B28) are equivalent to $\mathcal{D}(\Lambda(\tau - t_0, t - \tau))$ and $\mathcal{C}(\rho(\tau - t_0, t - \tau))$ in Eq. (16) of Ref. [\[58\]](#page-19-0).

APPENDIX C: DERIVATION OF THE DOUBLE-TIME CORRELATION FUNCTION FROM THE TIME-NONLOCAL EQUATION IN THE SECOND-ORDER APPROXIMATION

We drive the time evolution of the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ given by Eq. [\(33\)](#page-5-0) from the time-nonlocal equation of motion [\(12\)](#page-3-0) in the second-order approximation. Since the equality $\langle \hat{\mathcal{L}}_{SR}(t) \rangle_R = 0$ is assumed, it is obvious from Eq. [\(A20\)](#page-13-0) that the equality $\Phi_S(t|g) = 0$ holds. Up to the first order with respect to the fictitious field $g(t)$, we can obtain the integral kernel superoperator $\Phi_S(t,t_1|g)$ in the second-order approximation,

$$
\Phi_S(t,t_1|g) = \Phi_{S,0}(t,t_1) + \Phi_{S,1}(t,t_1|g),
$$
 (C1)

with

∂

$$
\Phi_{S,0}(t,t_1) = e^{\mathcal{L}_S(t-t_0)} \langle \mathcal{L}_{SR}(t) \mathcal{L}_{SR}(t_1) \rangle_R e^{-\mathcal{L}_S(t_1-t_0)}, \quad (C2)
$$

$$
\Phi_{S,1}(t,t_1|g)
$$
\n
$$
= \int_{t_1}^t d\tau \ g(\tau) e^{\mathcal{L}_S(t-t_0)} \langle \mathcal{L}_{SR}(t) \mathcal{K}_1(\tau) \mathcal{L}_{SR}(t_1) \rangle_R e^{-\mathcal{L}_S(t_1-t_0)}.
$$
\n(C3)

In this approximation, we can readily verify that the inhomogeneous term of the time-nonlocal equation is identical with that of the time-local equation, and then we have $\mathcal{J}_S(t|g) = I_{S,0}(t) + I_{S,1}(t|g)$, where $I_{S,0}(t)$ and $I_{S,1}(t|g)$ are given respectively by Eqs. $(B15)$ and $(B16)$. The time-nonlocal equation for the reduced quasidensity operator $W_S(t|g)$ is given by

$$
\frac{\partial}{\partial t} W_S(t|g) = [\mathcal{L}_S + g(t)\mathcal{K}_1]W_S(t|g) \n+ \int_{t_0}^t dt_1 [\Phi_{S,0}(t,t_1) + \Phi_{S,1}(t,t_1|g)]W_S(t_1|g) \n+ I_{S,0}(t) + I_{S,1}(t|g).
$$
\n(C4)

To solve this equation, we denote as $\bar{U}_S(t,t_0)$ a solution of the following equation:

$$
\frac{\partial}{\partial t}\overline{\mathcal{U}}_S(t,t_0) = \mathcal{L}_S \overline{\mathcal{U}}_S(t,t_0) + \int_{t_0}^t dt_1 \, \Phi_{S,0}(t,t_1) \overline{\mathcal{U}}_S(t_1,t_0), \quad \text{(C5)}
$$

with the initial condition $\overline{U}_s(t_0,t_0) = 1$. Here we assume that $\overline{U}_S(t,t_0)$ is invertible, though this is not always true. Then $W'_{S}(t|g) = \overline{U}_{S}^{-1}(t,t_{0})W_{S}(t|g)$ satisfies

$$
\frac{\partial}{\partial t} W'_{S}(t|g) = \bar{\mathcal{U}}_{S}^{-1}(t,t_{0}) I_{S,0}(t) + \bar{\mathcal{U}}_{S}^{-1}(t,t_{0}) I_{S,1}(t|g) \n+ g(t) \bar{\mathcal{U}}_{S}^{-1}(t,t_{0}) \mathcal{K}_{1} \bar{\mathcal{U}}_{S}(t,t_{0}) W'_{S}(t|g) \n+ \int_{t_{0}}^{t} dt_{1} \bar{\mathcal{U}}_{S}^{-1}(t,t_{0}) \Phi_{S,1}(t,t_{1}|g) \bar{\mathcal{U}}_{S}(t_{1},t_{0}) W'_{S}(t_{1}|g).
$$
\n(C6)

We decompose the system operator $W'_{S}(t|g)$ into the zerothand first-order terms with respect to the fictitious field *g*(*t*),

$$
W'_{S}(t|g) = W'_{S,0}(t) + W'_{S,1}(t|g). \tag{C7}
$$

Discarding all the terms higher than the first order, we obtain from Eq. $(C6)$

$$
W'_{S,0}(t) = W_S + \int_{t_0}^t dt_1 \,\overline{\mathcal{U}}_S^{-1}(t_1, t_0) I_{S,0}(t_1),\tag{C8}
$$

$$
W'_{S,1}(t|g) = \int_{t_0}^t dt_1 \bar{U}_S^{-1}(t_1, t_0) I_{S,1}(t_1|g)
$$

+
$$
\int_{t_0}^t dt_1 g(t_1) \bar{U}_S^{-1}(t_1, t_0) \mathcal{K}_1 \bar{U}_S(t_1, t_0) W'_{S,0}(t_1)
$$

+
$$
\int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \bar{U}_S^{-1}(t_1, t_0) \Phi_{S,1}(t_1, t_2|g)
$$

×
$$
\bar{U}_S(t_2, t_0) W'_{S,0}(t_2).
$$
 (C9)

We denote as $\bar{W}_S(t)$ the reduced density operator of the relevant quantum system, where $\overline{W}_S(t)$ is a solution of the inhomogeneous time-nonlocal quantum master equation [\[4,29–31\]](#page-18-0),

$$
\frac{\partial}{\partial t}\bar{W}_{S}(t) = \mathcal{L}_{S}\bar{W}_{S}(t) + \int_{t_{0}}^{t} dt_{1} \Phi_{S,0}(t,t_{1})\bar{W}_{S}(t_{1}) + I_{S,0}(t),
$$
\n(C10)

the solution of which is given by

$$
\bar{W}_S(t) = \bar{\mathcal{U}}_S(t, t_0) W_S + \int_{t_0}^t dt_1 \, \bar{\mathcal{U}}_S(t, t_1) I_{S,0}(t_1), \quad \text{(C11)}
$$

with

$$
\bar{U}_S(t,t_1) = \bar{U}_S(t,t_0)\bar{U}_S^{-1}(t_1,t_0) \quad (t \geq t_1 \geq t_0). \tag{C12}
$$

Note that $\overline{W}_S(t)$ is nothing but $W_{S,0}(t) = \overline{U}_S(t,t_0)W'_{S,0}(t)$. After some calculation, we obtain the first-order term from Eq. (C9),

$$
W_{S,1}(t|g) = \bar{U}_S(t,t_0)W'_{S,1}(t|g)
$$

= $\int_{t_0}^t d\tau g(\tau)\bar{U}_S(t,\tau)K_1\bar{W}_S(\tau)$
+ $\int_{t_0}^t d\tau g(\tau)\int_{\tau}^t d\tau_1\int_{t_0}^{\tau} d\tau_2\bar{U}_S(t,\tau_1)e^{\mathcal{L}_S(\tau_1-t_0)}$
 $\times \langle \mathcal{L}_{SR}(\tau_1)K_1(\tau)\mathcal{L}_{SR}(\tau_2)\rangle_R e^{-\mathcal{L}_S(\tau_2-t_0)}\bar{W}_S(\tau_2)$
+ $\int_{t_0}^t d\tau g(\tau)\int_{\tau}^t d\tau_1\bar{U}_S(t,\tau_1)I_{S,1}(\tau_1,\tau)$. (C13)

Since the double-time correlation function $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1)$ is derived from the relation $G(t_2,t_1|\mathcal{K}_2,\mathcal{K}_1) = Tr_S[\mathcal{K}_2\delta W_{S,1}(t|g)]$ $\delta g(t_1)|_{g\to 0}$, we finally obtain Eq. [\(33\)](#page-5-0).

APPENDIX D: CORRELATION FUNCTIONS OF THE QUBIT IN THE SECOND-ORDER APPROXIMATION

In this appendix, we derive the correlation functions [\(90\)](#page-10-0) and (91) . When the time evolution of the qubit given by Eq. [\(86\)](#page-10-0), the map $\mathcal{U}_S(t,0)$ defined by the relation $W_S(t)$ = $U_S(t,0)W_S(0)$ for any initial state $W_S(0)$ is determined by

$$
\mathcal{U}_S(t,0)|e\rangle\langle e| = \frac{1}{2}[1 + \sigma(t) + u_z(t)]|e\rangle\langle e|
$$

$$
+ \frac{1}{2}[1 - \sigma(t) - u_z(t)]|g\rangle\langle g|, \quad (D1)
$$

$$
\mathcal{U}_S(t,0)|g\rangle\langle g| = \frac{1}{2}[1+\sigma(t)-u_z(t)]|e\rangle\langle e|
$$

$$
+\tfrac{1}{2}[1-\sigma(t)+u_z(t)]|g\rangle\langle g|, (D2)
$$

$$
U_S(t,0)|e\rangle\langle g| = u(t)|e\rangle\langle g|,\tag{D3}
$$

$$
\mathcal{U}_S(t,0)|g\rangle\langle e| = u^*(t)|g\rangle\langle e|.\tag{D4}
$$

The map $U_S(t,0)$ is invertible since $|u(t)|^2 = u_z(t) > 0$. In fact, the inverse map is given by

$$
\mathcal{U}_S^{-1}(t,0)|e\rangle\langle e| = \frac{1}{2} \left[1 + \frac{1 - \sigma(t)}{u_z(t)} \right] |e\rangle\langle e| + \frac{1}{2} \left[1 - \frac{1 - \sigma(t)}{u_z(t)} \right] |g\rangle\langle g|, \quad (D5)
$$

$$
\mathcal{U}_S^{-1}(t,0)|g\rangle\langle g| = \frac{1}{2} \left[1 \frac{1+\sigma(t)}{u_z(t)} \right] |e\rangle\langle e| + \frac{1}{2} \left[1 + \frac{1+\sigma(t)}{u_z(t)} \right] |g\rangle\langle g|, \quad (D6)
$$

$$
\mathcal{U}_S^{-1}(t,0)|e\rangle\langle g| = \frac{1}{u(t)}|e\rangle\langle g|,\tag{D7}
$$

$$
\mathcal{U}_S^{-1}(t,0)|g\rangle\langle e| = \frac{1}{u^*(t)}|g\rangle\langle e|.\tag{D8}
$$

Thus, we obtain the map $\mathcal{U}_S(t,s) = \mathcal{U}_S(t,0)\mathcal{U}_S^{-1}(s,0)$,

$$
\mathcal{U}_S(t,s)|e\rangle\langle e| = \frac{1}{2}[1 + \sigma(t,s)u_z(0,s) + u_z(t,s)]|e\rangle\langle e|
$$

+
$$
\frac{1}{2}[1 - \sigma(t,s)u_z(0,s) - u_z(t,s)]|g\rangle\langle g|,
$$
 (D9)

$$
\mathcal{U}_S(t,s)|g\rangle\langle g| = \frac{1}{2}[1 + \sigma(t,s)u_z(0,s) - u_z(t,s)]|e\rangle\langle e|
$$

$$
+ \frac{1}{2}[1 - \sigma(t,s)u_z(0,s) + u_z(t,s)]|g\rangle\langle g|,
$$

(D10)

$$
\mathcal{U}_S(t,s)|e\rangle\langle g| = u(t,s)|e\rangle\langle g|,\tag{D11}
$$

$$
\mathcal{U}_S(t,s)|g\rangle\langle e| = u^*(t,s)|g\rangle\langle e|,\tag{D12}
$$

or equivalently

$$
\mathcal{U}_S(t,s)1 = 1 + \sigma(t,s)u_z(0,2)\sigma_z,
$$

$$
\mathcal{U}_S(t,s)\sigma_z = u_z(t,s)\sigma_z,
$$
 (D13)

$$
\mathcal{U}_S(t,s)\sigma_+ = u(t,s)\sigma_+,
$$

$$
U_S(t,s)\sigma_- = u^*(t,s)\sigma_-.
$$
 (D14)

In these equation, we set

$$
u(t,s) = \exp\left(-\int_s^t d\tau \left[\gamma(\tau) + i\delta(\tau)\right]\right), \quad (D15)
$$

$$
u_z(t,s) = \exp\left(-2\int_s^t d\tau \, \gamma(\tau)\right),\tag{D16}
$$

$$
\sigma(t,s) = \sigma(t) - \sigma(s). \tag{D17}
$$

To make the model analytically tractable, we assume that the thermal reservoir is in the ground state so that the equality $\langle R^{\dagger}(t)R(t')\rangle_R = 0$ holds. Since we can obtain $w(t) = -1$ from Eq. (85) , the map $\mathcal{U}_s(t,s)$ given by Eqs. (D13) and (D14) becomes

$$
\mathcal{U}_S(t,s)1 = 1 - [1 - u_z(t,s)]\sigma_z,
$$

\n
$$
\mathcal{U}_S(t,s) = u_z(t,s)\sigma_z,
$$
 (D18)

$$
\mathcal{U}_S(t,s)\sigma_+ = u(t,s)\sigma_+,
$$

$$
U_S(t,s)\sigma_- = u^*(t,s)\sigma_-.
$$
 (D19)

Then the quantum regression theorem yields the correlation function,

$$
\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_0 = \text{Tr}_S[\sigma_z \mathcal{U}_S(t_2, t_1) \sigma_z \mathcal{U}_S(t_1, 0) W_S]
$$

= 1 + [u_z(t_2, 0) - u_z(t_1, 0)](1 + \langle \sigma_z \rangle). (D20)

Next we derive the correction term (43) to the double-time correlation function. After some calculation, we can derive

$$
\mathcal{U}_{S}(t_{2},\tau_{1})e^{\mathcal{L}_{S}\tau_{1}}\langle\mathcal{L}_{SR}(\tau_{1})[\sigma_{z},\mathcal{L}_{SR}(\tau_{2})]\rangle_{R}e^{-\mathcal{L}_{S}\tau_{1}}W_{S}(\tau_{1})
$$
\n
$$
= e^{i\omega(\tau_{1}-\tau_{2})}\langle R(\tau_{1})R^{\dagger}(\tau_{2})\rangle_{R}\{u_{z}(t_{2},\tau_{1})u_{z}(\tau_{1},0)[1+c_{z}(0)]\sigma_{z}
$$
\n
$$
+ u^{*}(t_{2},\tau_{1})2u(\tau_{1},0)c(0)\sigma_{-}\}.
$$
\n(D21)

Then the correction term is obtained from Eq. (43) :

$$
\Delta \langle \sigma_z(t_2) \sigma_z(t_1) \rangle = 2 \int_{t_1}^{t_2} d\tau_1 \int_0^{t_1} d\tau \ e^{i\omega(\tau_1 - \tau_2)} \times \langle R(\tau_1) R^{\dagger}(\tau_2) \rangle_R u_z(t_2, 0) (1 + \langle \sigma_z \rangle).
$$
\n(D22)

Thus, the double-time correlation function with the correlation term is $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle_c = \langle \sigma_z(t_2) \sigma_z(t_1) \rangle_0 + \Delta \langle \sigma_z(t_2) \sigma_z(t_1) \rangle.$

To proceed further, we suppose that the thermal reservoir is a set of independent harmonic oscillators [\[4,19\]](#page-18-0). The reservoir operator *R* is assumed to be $R = \sum_{k} g_k a_k$, where a_k is an annihilation operator of the *k*th mode of the reservoir oscillator and *gk* is a coupling constant. Then we obtain the correlation function of the thermal reservoir,

$$
\langle R(\tau_1)R^{\dagger}(\tau_2)\rangle_R = \sum_k |g_k|^2 e^{-i\omega_k(\tau_2 - \tau_1)}
$$

=
$$
\int d\Omega J(\Omega) e^{-i\Omega(\tau_2 - \tau_1)}, \quad (D23)
$$

where $J(\Omega)$ represents the spectral density of the systemreservoir interaction. Furthermore, we assume that $J(\Omega)$ is a Lorentzian function with the resonance condition

$$
J(\Omega) = \left(\frac{\gamma}{2\pi}\right) \frac{\lambda^2}{(\Omega - \omega)^2 + \lambda^2},
$$
 (D24)

where λ is an inverse of the reservoir correlation time and γ is a coupling strength. Then the correlation function becomes

$$
\langle R(\tau_1)R^{\dagger}(\tau_2)\rangle_R = \frac{1}{2}\gamma\lambda e^{-(\lambda + i\omega)(\tau_2 - \tau_1)}.
$$
 (D25)

In this case, the time-dependent parameter $u_7(t,0)$ is given by

$$
u_z(t,0) = \exp\bigg[-\bigg(\frac{\gamma}{\lambda}\bigg)(\lambda t - 1 + e^{-\lambda t})\bigg].
$$
 (D26)

By substituting this equation and Eq. (D25) into Eqs. (D20) and $(D22)$, we obtain Eqs. (90) and (91) .

APPENDIX E: DERIVATION OF THE EXACT CORRELATION FUNCTION OF THE QUBIT

In this appendix, we briefly explain the derivation of the exact correlation function $\langle \sigma_z(t_2) \sigma_z(t_1) \rangle$ given by Eq. [\(92\)](#page-10-0). We suppose that a two-level system is placed under the influence of a thermal reservoir. Furthermore, we assume that there is at most a single excitation in the whole system at an initial time and the Hamiltonian *H* conserves the total excitation number. Then the whole system can be described by a Hilbert space spanned by three basis vectors $|e\rangle$ (0), $|g\rangle$ $|\mathbf{1}_k\rangle$, and $|e\rangle$ $|\mathbf{0}\rangle$, where $|\mathbf{0}\rangle$ is the ground state of the thermal reservoir and $|\mathbf{1}_k\rangle$ is a single-excited state in which only the *k*th mode has one excitation. Since the total excitation number is conserved during the time evolution, we can express the time

evolution as

$$
e^{-iHt/\hbar}|g\rangle|\mathbf{0}\rangle = |g\rangle|\mathbf{0}\rangle,\tag{E1}
$$

$$
e^{-iHt/\hbar}|e\rangle|\mathbf{0}\rangle = A(t)|g\rangle|\mathbf{0}\rangle + \sum_{k} A_k(t)|g\rangle|\mathbf{1}_k\rangle, \quad \text{(E2)}
$$

$$
e^{-iHt/\hbar}|g\rangle|\mathbf{1}_k\rangle = B_k(t)|e\rangle|\mathbf{0}\rangle + \sum_l B_{kl}(t)|g\rangle|\mathbf{1}_l\rangle, \quad \text{(E3)}
$$

with the initial condition $A(0) = 1$, $A_k(0) = B_k(0) = 0$ and $B_{kl}(0) = \delta_{kl}$. The normalization condition yields the relations

$$
|A(t)|^{2} + \sum_{k} |A_{k}(t)|^{2} = 1,
$$

$$
|B_{k}(t)|^{2} + \sum_{l} |B_{kl}(t)|^{2} = 1.
$$
 (E4)

Using the trivial equalities $U(t_1+t_2)|e\rangle|\mathbf{0}\rangle=U(t_2)U(t_1)|e\rangle$ $|0\rangle$ and $U(t_1 + t_2)|g\rangle|1_k\rangle = U(t_2)U(t_2)|g\rangle|1_k\rangle$ with $U(t) =$ $e^{-iHt/\hbar}$, we can obtain the relations among the parameters

$$
A(t_1 + t_2) = A(t_1)A(t_2) + \sum_k A_k(t_1)B_k(t_2),
$$
 (E5)

$$
A_k(t_1 + t_2) = A(t_1)A_k(t_2) + \sum_l A_l(t_1)B_{lk}(t_2), \quad \text{(E6)}
$$

$$
B_k(t_1 + t_2) = B_k(t_1)A(t_2) + \sum_m B_{km}(t_1)B_m(t_2), \quad (E7)
$$

$$
B_{kl}(t_1 + t_2) = B_k(t_1)A_l(t_2) + \sum_m B_{km}(t_1)B_{ml}(t_2).
$$
 (E8)

We assume that the initial state of the whole system is given by $|\Psi(0)\rangle = (c_g|g\rangle + c_e|e\rangle)|0\rangle$ with $|c_g|^2 + |c_e|^2 = 1$. In this case, we have the relations

$$
U(t - s)\sigma_z U(s)|\Psi(0)\rangle
$$

= $-c_g|g\rangle|\mathbf{0}\rangle + c_e[2A(s)A(t - s) - A(t)]|e\rangle|\mathbf{0}\rangle$
+ $c_e \sum_k [2A(s)A_k(t - s) - A_k(t)]|g\rangle|\mathbf{1}_k\rangle$ (E9)

and

 \langle

$$
U(t)|\Psi(0)\rangle = c_g|g\rangle|\mathbf{0}\rangle + c_e A(t)|e\rangle|\mathbf{0}\rangle + c_e \sum_k A_k(t)|g\rangle|\mathbf{1}_k\rangle.
$$
\n(E10)

Then we obtain for any operator *X* of the two-level system

$$
\Psi(0)|U^{\dagger}(t)XU(t-s)\sigma_{z}U(s)|\Psi(0)\rangle
$$
\n
$$
= (c_{g}^{*}\langle g| + c_{e}^{*}A^{*}(t)\langle e|)
$$
\n
$$
\times X(-c_{g}|g) + c_{e}[2A(s)A(t-s) - A(t)]|e\rangle)
$$
\n
$$
+ |c_{e}|^{2} \sum_{k} A_{k}^{*}(t)[2A(s)A_{k}(t-s) - A_{k}(t)]\langle g|X|g\rangle.
$$
\n(E11)

Then by substituting $X = 1$ into this equation, we derive $\langle \Psi(0) | U^{\dagger}(s) \sigma_z U(s) | \Psi(0) \rangle$

$$
= -1 + 2|c_e|^2 \left[A^*(t)A(t-s) + \sum_k A_k^*(t)A_k(t-s) \right] A(s).
$$
\n(E12)

On the other hand, the direct calculation yields

$$
\langle \Psi(0)|U^{\dagger}(s)\sigma_z U(s)|\Psi(0)\rangle = -1 + 2|c_e|^2 A^*(s)A(s). \quad (E13)
$$

Since this equation has to be identical to Eq. $(E12)$, we obtain the relation between the parameters $A(t)$ and $A_k(t')$,

$$
A(s) = A(t)A^*(t - s) + \sum_{k} A_k(t)A_k^*(t - s), \quad \text{(E14)}
$$

with $t > s$. This relation together with $|A(t)|^2 + \sum_k |A_k(t)|^2 =$ 1 provides

$$
\sum_{k} A_{k}^{*}(t)[2A(s)A_{k}(t-s) - A_{k}(t)]
$$

= 2|A(s)|² + |A(t)|² - 1 - 2A^{*}(t)A(t-s)A(s). (E15)

Substituting this equation into Eq. $(E11)$, we can obtain the correlation function,

$$
\langle X(t)\sigma_z(s)\rangle
$$

= $\{-1 + |c_e|^2 [2|A(s)|^2 + |A(t)|^2 - 2A^*(t)A(t - s)A(s)]\}$
 $\times \langle g|X|g\rangle + |c_e|^2 [2A^*(t)A(t - s)A(s) - |A(t)|^2] \langle e|X|e\rangle$
 $- c_e^* c_g A^*(t) \langle e|X|g\rangle + c_g^* c_e [2A(t - s)A(s) - A(t)] \langle g|X|e\rangle.$ (E16)

Here, we note that the correlation function can be expressed only in terms of the single time-dependent parameter $A(t)$ if there is no initial correlation between the two-level system and the thermal reservoir. Setting $X = \sigma_z$ in this equation yields Eq. [\(92\)](#page-10-0). This result can be extended in the presence of the initial correlation. The details are given in Ref. [\[76\]](#page-19-0).

To find an explicit form of the time-dependent parameter $A(t)$, we assume that the Hamiltonian *H* of the whole system is

$$
H = \frac{1}{2}\hbar\omega\sigma_z + \sum_k \hbar\omega_k a_k^\dagger a_k + \sum_k \hbar(\lambda_k \sigma_+ a_k + \lambda_k^* \sigma_- a_k^\dagger).
$$
\n(E17)

By differentiating both sides of Eq. (E2) with respect to time *t*, we obtain

$$
\frac{d}{dt}A(t) = -i\omega A(t) - i\sum_{k} \lambda_k A_k(t),
$$
 (E18)

$$
\frac{d}{dt}A_k(t) = -i\lambda_k^*A(t) - i\omega_k A_k(t),
$$
\n(E19)

which yields

$$
\frac{d}{dt}A(t) = -i\omega A(t) - \sum_{k} |\lambda_k|^2 \int_0^t ds \, e^{-i\omega_k(t-s)} A(s). \quad (E20)
$$

Thus, by assuming the Lorentzian spectral density [\(D24\)](#page-16-0) and solving this integrodifferential equation under the initial condition $A(0) = 1$, we can obtain Eq. [\(93\)](#page-10-0).

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