Non-Markovian finite-temperature two-time correlation functions of system operators of a pure-dephasing model

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We evaluate the non-Markovian finite-temperature two-time correlation functions (CF’s) of system operators of a pure-dephasing spin-boson model in two different ways, one by the direct exact operator technique and the other by the recently derived evolution equations, valid to second order in the system-environment interaction Hamiltonian. This pure-dephasing spin-boson model that is exactly solvable has been extensively studied as a simple decoherence model. However, its exact non-Markovian finite-temperature two-time system operator CF’s, to our knowledge, have not been presented in the literature. This may be mainly due to the fact, illustrated in this article, that in contrast to the Markovian case, the time evolution of the reduced density matrix of the system (or the reduced quantum master equation) alone is not sufficient to calculate the two-time system operator CF’s of non-Markovian open systems. The two-time CF’s obtained using the recently derived evolution equations in the weak system-environment coupling case for this non-Markovian pure-dephasing model happen to be the same as those obtained from the exact evaluation. However, these results significantly differ from the non-Markovian two-time CF’s obtained by wrongly directly applying the quantum regression theorem (QRT), a useful procedure to calculate the two-time CF’s for weak-coupling Markovian open systems. This demonstrates clearly that the recently derived evolution equations generalize correctly the QRT to non-Markovian finite-temperature cases. It is believed that these evolution equations will have applications in many different branches of physics.

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I. INTRODUCTION

A quantum system is inevitably subject to the influence of its surroundings or environments [1–6]. An environment usually consists of a practically infinite number of degrees of freedom and acts statistically as a whole identity referred as a reservoir or bath of the open quantum system. Most often, one is concerned with only the system dynamics and the key quantity is the reduced system density matrix $\rho(t)$ defined as the partial trace of the total system-plus-reservoir density operator $\rho_T(t)$ over the reservoir degrees of freedom; i.e., $\rho(t) = \text{Tr}_R[\rho_T(t)]$. If the time evolution of the reduced density matrix that can be Markovian or non-Markovian is known, one is able to calculate the (one-time) expectation values or quantum average of the physical quantities of the system operators. But knowing the time evolution of the reduced density matrix is not sufficient to calculate the two-time (multiple-time) correlation functions (CF’s) of the system operators in the non-Markovian case [7–9].

In the Markovian case, an extremely useful procedure to calculate the two-time (multiple-time) CF’s is the so-called quantum regression theorem (QRT) [1–4] that gives a direct relation between the time evolution equation of the single-time expectation values and that of their corresponding two-time (multiple-time) CF’s. So knowing the time evolution of the system reduced density matrix allows one to calculate all of the two-time (multiple-time) Markovian CF’s. For the non-Markovian case, it is known that the QRT is not valid in general [10–13]. Recently, using the stochastic Schrödinger equation approach and the Heisenberg equation of system operator method, an evolution equation, valid to second order in system-environment coupling strength, for the two-time (multiple-time) CF’s of the system operators has been derived for an environment at the zero temperature and for a system in an initial pure state $[7,8]$. This evolution equation has been applied to calculate the emission spectra of a two-level atom placed in a structured non-Markovian environment (electromagnetic fields in a photonic band-gap material) [14]. In Ref. [8], an evolution equation for the reduced propagator of the system state vector, conditioned on an initial state of the environment differing from the vacuum, was derived using the stochastic Schrodinger equation approach. It is thus possible to use the reduced propagator to evaluate the expectation values and CF’s of the system observables for general environmental initial conditions, not necessarily an initial vacuum state for the environment [8]. By using another commonly used open quantum system technique, the quantum master equation approach [11–13], we are able to extend the two-time CF evolution equation to a non-Markovian finite-temperature environment for any initial system-environment separable state. The detailed derivation will be presented elsewhere [13] but the essential results will be summarized in Sec. II.
derived evolution equation that generalizes the QRT to the non-Markovian finite-temperature case is believed to have applications in many different branches of physics.

The purpose of this article is twofold: (a) We show that in general the time evolution of the reduced density matrix of the system (or the reduced quantum master equation) alone is not sufficient to calculate the two-time CF’s of the system operators of non-Markovian open systems, even in the weak system-environment coupling case. We present an evaluation of an exactly solvable non-Markovian model, i.e., a pure-dephasing spin-boson model [8–16, 21], to justify the statement. The exact non-Markovian finite-temperature two-time CF’s of the system operators of this model, to our knowledge, have not been presented in the literature. (b) This exactly solvable model allows us to test the validity of the derived non-Markovian finite-temperature evolution equation of two-time CF’s presented in Sec. III. It will be shown that the two-time CF’s obtained using the evolution equation in the weak system-environment coupling limit [15] in Sec. III for the exactly solvable non-Markovian model happen to be the same as those obtained from the exact evaluation. However, these results significantly differ from the non-Markovian CF’s obtained by wrongly applying directly the QRT. This demonstrates clearly that the derived evolution equations generalize correctly the QRT to non-Markovian finite-temperature cases.

The article is organized as follows. We first summarize the important results of the newly obtained evolution equations [15] that generalizes the QRT to the non-Markovian finite-temperature case in Sec. II. After brief description of the pure-dephasing spin-boson model in the beginning of Sec. III, we calculate the exact time evolution of the reduced density matrix of the system and one-time expectation values in Sec. IIIA. The exact two-time CF’s are evaluated in subsection IIIB. In Sec. IV, we use the derived evolution equations in Ref. [13] to calculate the one-time and two-time CF’s. It is shown that the results obtained in Sec. IV are the same as those by the exact evaluation in Sec. III. This demonstrates the validity and practical usage of the derived evolution equations in Ref. [13]. Numerical results and discussions are presented in Sec. V. A short conclusion is given in Sec. VI.

II. EVOLUTION EQUATION OF NON-MARKOVIAN FINITE-TEMPERATURE TWO-TIME CF’S

A class of systems considered in [7–9] is modeled by the Hamiltonian

\[ H = H_S + H_I + H_R = H_S + \sum_\lambda \hbar g_\lambda \left( L_\lambda^+ a_\lambda + L_\lambda a_\lambda^+ \right) + \sum_\lambda \hbar \omega_\lambda a_\lambda^+ a_\lambda, \]

where \( H_S \) and \( H_R \) are system and environment Hamiltonians, respectively, and \( H_I \) stands for the Hamiltonian that describes the interaction between the system and the environment. So \( L \) acts on the Hilbert space of the system, \( a_\lambda^+ \) and \( a_\lambda \) are creation and annihilation operators on the environment Hilbert space, and \( g_\lambda \) and \( \omega_\lambda \) are the coupling strength and the frequency of the \( \lambda \)th environment oscillator, respectively. The derivations of the non-Markovian evolution equations of the two-time (multitime) CF’s for the general Hamiltonian model [1] in Refs. [7–9] (Eq. (6) in Ref. [7], Eq. (31) in Ref. [8] and Eq. (60) in Ref. [9]) are presented for an environment at the zero temperature and for a system state in an initial pure state. It was mentioned in Ref. [8] that it is possible to use the reduced stochastic system propagator that corresponds to an initial state of the environment different from the vacuum to evaluate the single-time expectation values and multitime CF’s with more general initial conditions. But only a master equation that is conditioned on initial bath states and is capable of evaluating the single-time expectation values of system observables for general initial conditions, both for an initial pure state and mixed state, was derived [8]. In Refs. [7–9], calculations of the two-time CF’s of system observables for dissipative spin-boson models in thermal baths are, however, presented even though in their derivations of the two-time (multitime) evolution equations, the bath CF’s are given in its zero-temperature form. This is possible due to the reason that for a system-environment model with a Hermitian system operator \( L = L^\dagger \) coupled to the environment, the linear finite-temperature stochastic Schrödinger equation could be written in a simple form of the zero-temperature equation [19, 22] if the zero-temperature bath CF is replaced with its corresponding effective finite-temperature bath CF. As a result, the evolution equation of thermal two-time (multitime) CF’s for a Hermitian coupling operator \( L = L^\dagger \) also becomes equal to its zero-temperature counterpart with the replacement of the zero-temperature bath CF with its effective finite-temperature bath correlation kernel. It is for this reason that the dissipative spin-boson model with a thermal environment can be studied with the two-time (multitime) evolution equations derived in Refs. [7–9], since in that model \( L = \sigma_x = L^\dagger \). But this reduction of the finite-temperature evolution equation to its zero-temperature form [7–9] is not valid for more general non-Markovian finite-temperature cases where the system coupling operators are not Hermitian, i.e., \( L \neq L^\dagger \). In other words, if the system operator coupled to the environment is not Hermitian \( L \neq L^\dagger \), the two-time (multitime) differential evolution equations presented in Refs. [7–9] are valid for a zero-temperature environment only.

By using another commonly used open quantum system technique, the quantum master equation approach [4, 6], it is possible to obtain in the weak system-environment coupling limit a two-time evolution equation for non-Markovian finite-temperature environments with both Hermitian and non-Hermitian system coupling operators and for any initial system-environment separable states. The detailed derivation will be presented...
are known as the environment CF’s: 

\[ \sum_{\tau} \alpha(t_1 - \tau) \left\{ \tilde{L}^\dagger (\tau - t_1) [A, \hat{L}] \right\} (t_1) \rho(0) \]

and that of the two-time CF’s can be obtained as

\[ d \langle A(t_1) B(t_2) \rangle / dt_1 = (i/\hbar) \text{Tr}_S \left( \left[ [H_S, A] \right] (t_1) B(t_2) \rho(0) \right) \]

and

\[ + \int_0^{t_1} d\tau \text{Tr}_S \left( \alpha^* (t_1 - \tau) \left\{ \tilde{L}^\dagger (\tau - t_1) [A, \hat{L}] \right\} (t_1) B(t_2) \rho(0) \right) \]

\[ + \alpha (t_1 - \tau) \left\{ [\hat{L}^\dagger, A] \tilde{L} (\tau - t_1) \right\} (t_1) B(t_2) \rho(0) \]

\[ + \beta^* (t_1 - \tau) \left\{ \tilde{L} (\tau - t_1) [A, \hat{L}^\dagger] \right\} (t_1) B(t_2) \rho(0) \]

\[ + \beta (t_1 - \tau) \left\{ [L, A] \tilde{L} (\tau - t_1) \right\} (t_1) B(t_2) \rho(0) \]  

and

\[ + \int_0^{t_2} d\tau \text{Tr}_S \left( \alpha (t_1 - \tau) \left\{ [L^\dagger, A] \right\} (t_1) \left\{ [B, \tilde{L} (\tau - t_2)] \right\} (t_2) \rho(0) \right) \]

\[ + \beta (t_1 - \tau) \left\{ [L, A] \right\} (t_1) \left\{ [B, \tilde{L}^\dagger (\tau - t_2)] \right\} (t_2) \rho(0) \right) \right) \right) . \]  

Here \( \tilde{L}(t) = \exp (i H_S t/\hbar) L \exp (-i H_S t/\hbar) \) is the system operator in the interaction picture with respect to \( H_S \), and

\[ \alpha (t - s) = \sum_{\lambda} (\tilde{a}_{\lambda} + 1) |g_{\lambda}|^2 e^{-i \omega_\lambda (t - s)} , \]  

\[ \beta (t - s) = \sum_{\lambda} \tilde{a}_{\lambda} |g_{\lambda}|^2 e^{i \omega_\lambda (t - s)} , \]

are known as the environment CF’s: \( \alpha (t - s) = \sum_{\lambda} g_{\lambda} \tilde{a}_{\lambda} (s) \sum_{\lambda'} g_{\lambda'} \tilde{a}_{\lambda'}^\dagger (s) \) and \( \beta (t - s) = \sum_{\lambda} g_{\lambda} \tilde{a}_{\lambda}^\dagger (s) \sum_{\lambda'} g_{\lambda'} \tilde{a}_{\lambda'} (s) \), where \( \tilde{a}_{\lambda} (\tau) = a_{\lambda} e^{-i \omega_\lambda \tau} \) and \( \tilde{a}_{\lambda}^\dagger (\tau) = a_{\lambda}^\dagger e^{i \omega_\lambda \tau} \) are the reservoir operators in the interaction picture.

We note here that for a Hermitian coupling operator \( L = L^\dagger \) the finite-temperature evolution equations reduce, respectively, to their zero-temperature counterparts but with the effective bath CF given by \( \alpha (t_1 - \tau) + \beta (t_1 - \tau) \). This was pointed out to occur in general for N-time CF’s in Refs. \[ \text{[15–18]} \].

III. EXACT EVALUATIONS OF PURE DEPHASING SPIN-BOSON MODEL

Here we consider an exactly solvable pure dephasing model of

\[ H_S = (\hbar \omega_S / 2) \sigma_z , \quad L = \sigma_z = L^\dagger \]  

to test the evolution equations and \( \text{[3]} \). This pure dephasing spin-boson model in which \( [H_S, L] = 0 \) has been extensively studied as a simple decoherence model in the literature \( \text{[8–16]} \). But most of the studies focus on the discussion of the time evolution of the reduced density matrix of the spin, or other one-time expectation values of the spin system operators. Recently, the two-time CF’s of the system operators at the zero temperature for this model was reported in Ref. \( \text{[8]} \). Nevertheless, to demonstrate the validity and practical usage of the finite-temperature non-Markovian evolution equation of the two-time CF’s, we present a detailed evaluation of the exact finite-temperature two-time CF’s for this simple model. These exact non-Markovian finite-temperature two-time CF’s of the system operators, to our knowledge, have not been presented in the literature.

A. Reduced density matrix and one-time expectation values

Before we derive the two-time CF’s, we evaluate the exact time evolution of the reduced density matrix and one-time expectation values for the non-Markovian spin-boson model. In the interaction picture, the total density matrix of the combined (spin plus bath) system at time \( t \) is given by

\[ \tilde{\rho}_T (t) = \tilde{U}(t) \rho_T (0) \tilde{U}^\dagger (t) , \]  

where the time evolution operator is

\[ \tilde{U}(t) = e^{i H_0 t/\hbar} e^{-i H_1 t/\hbar} \]

\[ = T \left[ e^{-i H_1 (t)} \int_0^t d\tau H_1 (\tau) \right] . \]  

Here \( H_0 = H_S + H_R \), \( \tilde{H}_1 (t) = \exp (i H_0 t/\hbar) H_1 \exp (-i H_0 t/\hbar) \) and \( T \) is the time-ordering operator which arranges the operators with the earliest times to the right. From Eqs. \[ \text{(1)} \] and \[ \text{(6)} \], a simple calculation gives

\[ \tilde{H}_1 (t) = \sum_{\lambda} \hbar g_{\lambda} \sigma_z \left( e^{i \omega_\lambda t} a_{\lambda}^\dagger + e^{-i \omega_\lambda t} a_{\lambda} \right) . \]  

\[ \text{[19–21]} \]
This result allows us to calculate the time evolution operator to be (see Appendix A for details)

$$\tilde{U}(t) = \exp \left[ -i \int_0^t d\tau \sum_{\lambda} g_{\lambda} \bar{\sigma}_z \left( e^{i\omega_{\lambda}\tau} a_{\lambda}^\dagger + e^{-i\omega_{\lambda}\tau} a_{\lambda} \right) \right] \times \exp \left( \frac{1}{2} \int_0^t d\tau \int_0^t ds \sum_{\lambda} |g_{\lambda}|^2 e^{i\omega_{\lambda}(\tau-s)} \right) \times \exp \left( - \int_0^t d\tau \int_0^t ds \sum_{\lambda} |g_{\lambda}|^2 e^{-i\omega_{\lambda}(\tau-s)} \right).$$

The time integrations in the exponents in Eq. (10) can be easily and analytically carried out. But we keep them in those forms in Eq. (10) so it will be easier to identify them with the results in Ref. [15]. If the time-ordering operation in Eq. (3) for $\tilde{U}(t)$ were not performed, one could have just obtained the first term (line) of Eq. (10) for $\tilde{U}(t)$. Thus the second and third terms (lines) of Eq. (10) can be considered as the correction terms due to the time-ordering operation.

The reduced density matrix can be obtained by tracing over the reservoir’s degrees of freedom: $\rho(t) = \text{Tr}_R[\rho_T(t)]$. Suppose initially the state $\rho_T(0) = \tilde{\rho}_T(0) = \rho(0) \otimes R_0$ is factorized, where $\rho(0)$ and $R_0$ are initial system and thermal reservoir/environment density operators, respectively, and $R_0 = \exp(-H_R/k_BT)\text{Tr}_R[\exp(-H_R/k_BT)]$. Then the reduced density matrix elements in the interaction picture can be written as

$$\tilde{\rho}_{mn}(t) = \rho_{mn}(0) \text{Tr}_R\left[ \tilde{U}^{(n)}(t) \tilde{U}^{(m)}(t) R_0 \right], \quad (11)$$

where $\tilde{\rho}_{mn}(t) \equiv \langle m|\tilde{\rho}(t)|n \rangle$, $\tilde{U}^{(n)}(t) \equiv \langle n|\tilde{U}(t)|n \rangle$, $m, n = 0, 1$ and the states of the two-level system are defined as $\sigma_1|0\rangle = |0\rangle$, $\sigma_2|1\rangle = -|1\rangle$. To evaluate Eq. (11), the well known formula of

$$e^{A+B} = e^A e^B e^{[A,B]}, \quad (12)$$

valid for operators $A$ and $B$ both commuting with the commutator $[A,B]$, can be used to combine the evolution operators together. One then obtains

$$\tilde{U}^{(0)}(t_1)\tilde{U}^{(1)}(t_1) = \left[ \tilde{U}^{(1)}(t_1)\tilde{U}^{(0)}(t_1) \right]^\dagger$$

$$= \exp \left[ 2i \int_0^{t_1} d\tau \sum_{\lambda} g_{\lambda} \left( e^{i\omega_{\lambda}\tau} a_{\lambda}^\dagger + e^{-i\omega_{\lambda}\tau} a_{\lambda} \right) \right]. \quad (13)$$

Then a useful identity [23] for the average over the thermal reservoir (environment) density operator, $R_0$, can be employed:

$$\langle e^{\sum_{\lambda} c_{\lambda} a_{\lambda} + d_{\lambda} a_{\lambda}^\dagger} \rangle = e^{\frac{1}{2} \sum_{\lambda} c_{\lambda} d_{\lambda} (2\bar{n}_{\lambda} + 1)}, \quad (14)$$

where $c_{\lambda}, d_{\lambda}$ are complex numbers, and $\bar{n}_{\lambda} = [\exp(\hbar\omega_{\lambda}/k_BT) - 1]^{-1}$ stands for the thermal mean occupation number of the environment oscillators. As a result, we obtain

$$\text{Tr}_R \left[ \tilde{U}^{(0)}(t)\tilde{U}^{(1)}(t) R_0 \right] = \text{Tr}_R \left[ \tilde{U}^{(1)}(t)\tilde{U}^{(0)}(t) R_0 \right]$$

$$= \exp \left( - \int_0^t d\tau D(\tau) \right), \quad (15)$$

where

$$D(\tau) = 2 \int_0^\tau ds \{ \alpha_{\text{eff}}(\tau-s) + \alpha^*_{\text{eff}}(\tau-s) \}, \quad (16)$$

and $\alpha(t-\tau)$ and $\beta(t-\tau)$ are defined in Eqs. (4) and (5), respectively. It is easy to show that $\tilde{U}^{(n)}(t)\tilde{U}^{(n)}(t) R_0 = I$ and $\text{Tr}_R \left[ \tilde{U}^{(n)}(t)\tilde{U}^{(n)}(t) R_0 \right] = 1$. Thus, using these results for the reduced density matrix elements Eq. (11) in the interaction picture and then transforming them back to the Schrödinger picture $\rho(t) = \exp(-iH_st/\hbar)\tilde{\rho}(t)\exp(iH_st/\hbar)$, we obtain the exact reduced density operator in the matrix form of

$$\rho(t) = \begin{pmatrix} \rho_{00}(0) & \rho_{01}(0) e^{-F(t)} \\ \rho_{10}(0) e^{-F^*(t)} & \rho_{11}(0) \end{pmatrix}, \quad (18)$$

with $F(t) = i\omega_st + \int_0^t d\tau D(\tau)$. The same result was obtained in Ref. [19] using the stochastic Schrödinger equation approach.

With the exact time evolution of the reduced density matrix, the one-time expectation value of the system operators

$$\langle A(t_1) \rangle = \text{Tr}_{S\otimes R} [A(t_1) \rho_T(0)] = \text{Tr}_S [A(0) \rho(t_1)], \quad (19)$$

can be calculated exactly, where $A(t_1)$ represents a general system Heisenberg operator(s) and $\rho(t_1) = \text{Tr}_R[\rho_T(t_1)]$ is the reduced Schrödinger density matrix operator at time $t_1$. We may also write in the interaction picture,

$$\langle A(t_1) \rangle = \text{Tr}_{S\otimes R} \left[ \tilde{A}(t_1) \tilde{\rho}(t_1) \right] = \text{Tr}_S \left[ \tilde{A}(t_1) \tilde{\rho}(t_1) \right]$$

where $\tilde{\rho}$ is defined in Eq. (7), $\tilde{\rho}(t) = \text{Tr}_R[\tilde{\rho}(t)]$ and $\tilde{A}(t) = \exp(-iH_st/\hbar) A \exp(-iH_st/\hbar)$, and $A = A(0)$. For a general system operator $A = \begin{pmatrix} a & b \\ c & d \end{pmatrix}$, we obtain exactly from either Eq. (19) or Eq. (20)

$$\langle A(t_1) \rangle = e^{-\int_0^{t_1} d\tau D(\tau)} (a \rho_{00}(0) e^{-i\omega_st} + b \rho_{01}(0) e^{-i\omega_{st}(t)})$$

$$+ c \rho_{00}(0) + d \rho_{11}(0). \quad (21)$$

### B. Two-time correlation functions

In contrast to the Markovian case in which the QRT is valid, the time evolution of the reduced density matrix
of a non-Markovian open system alone is not sufficient to obtain the two-time system operator CF’s. This can be understood as follows. The two-time CF’s of system operators $A(t_1)B(t_2)$ for $t_1 > t_2$ can be written as

$$\langle A(t_1)B(t_2) \rangle = \text{Tr}_{S\otimes R}[U^\dagger(t_1)AU(t_1)0U^\dagger(t_2)0BU(t_2)0]\rho_T(0)$$

$$= \text{Tr}_{S\otimes R}[AU(t_1, t_2)BU(t_2, 0)\rho_T(0)U^\dagger(t_2, 0)U^\dagger(t_1, t_2)]$$

where the Heisenberg evolution operators $U(t_1, t_2) = U(t_1, 0)U^\dagger(t_2, 0)$ and $U(t, 0) = \exp(-iHt/\hbar)$. If the environment is Markovian so the environment operator CF at two different times is $\delta$ correlated in time, then we may regard that the environment operator in $U(t_1, t_2)$ is not correlated with that in $U(t_2, 0)$. So the trace over the environment degrees of freedom for operator $U(t_1, t_2)$ and operator $U(t_2, 0)$ can be performed independently or separately. Thus one may first trace $\rho_T(t_2) = U(t_2, 0)\rho_T(0)U^\dagger(t_2, 0)$ over the environment degrees of freedom to obtain the reduced density matrix $\rho(t_2) = \text{Tr}_R[\rho_T(t_2)]$. Equation (22) in this case can be written as

$$\langle A(t_1)B(t_2) \rangle = \text{Tr}_{S\otimes R}[AU(t_1, t_2)(B\rho(t_2) \otimes R_0)U^\dagger(t_1, t_2)]$$

$$= \text{Tr}_S[A\chi(t)]$$

where $\chi(t)$ is the effective reduced density matrix at time $\tau = t_1 - t_2$ with the initial condition $\chi(0) = B\rho_0$. Thus knowing the time evolution of the reduced density matrix in the Markovian case, one is able to calculate the two-time CF’s of the system operators. This is also the reason why the QRT works in the Markovian case. But the situation differs for a non-Markovian environment as the environment operator in $U(t_1, t_2)$ may, in general, be correlated with that in $U(t_2, 0)$.

The two-time CF’s of the system operators for the pure-dephasing spin-boson model can also be evaluated exactly. To evaluate the two-time CF of system operators $A(t_1)B(t_2)$ for $t_1 > t_2$, we express it in terms of the interaction picture operators as

$$\langle A(t_1)B(t_2) \rangle = \text{Tr}_{S\otimes R}[U^\dagger(t_1)AU(t_1)U^\dagger(t_2)BU(t_2)\rho_T(0)]$$

$$= \text{Tr}_{S\otimes R}[\tilde{U}^\dagger(t_1)\tilde{A}(t_1)\tilde{U}(t_1)\tilde{U}^\dagger(t_2)\tilde{B}(t_2)\tilde{U}(t_2)\rho_T(0)]$$

where again an operator with a tilde on the top indicates that it is an operator in the interaction picture with respect to the free Hamiltonian $H_0$. Compared with Eq. (20), Eq. (24) for general non-Markovian open systems can not be expressed as a product of the reduced density matrix and system operators. So again, the reduced density matrix alone is not sufficient to obtain the non-Markovian two-time system operator CF’s.

As we want to compare the results by the direct evaluation with those by the evolution equation (23), we calculate, in the following, the two-time CF’s $\langle A(t_1)B(t_2) \rangle$ for different cases of system operators $A$ and $B$. The structure of the evolution equations in Ref. [15] or Eqs. (3) and (5) in this article depends on the commutation relations of operator $A$ and operator $L$ (or $L^\dagger$), and on the commutation relations of operator $B$ and operator $\tilde{L}(\tau - t_2)$ (or $\tilde{L}^\dagger(\tau - t_2)$), where $\tilde{L}(t) = \exp(iH_St/\hbar)\exp(-iH_St/\hbar)$ is the system operator in the interaction picture with respect to $H_S$. For the pure-dephasing spin-boson model, $H_S = (i\hbar\omega_s/2)\sigma_z, L = \sigma_z = L^\dagger$, and then $\tilde{L}(t) = \sigma_z$. So we will discuss the two-time CF’s in the following three cases and the trivial case of $\langle \sigma_z(t_1)\sigma_z(t_2) \rangle = \langle \sigma_z(0)\sigma_z(0) \rangle = 1$ is obvious due to $\langle \sigma_z, H \rangle = 0$.

Case 1. $[A, L] \neq 0$ and $[B, L(t)] = 0$. In this case, let $A = a\sigma_+ + b\sigma_-$, and $B = \sigma_z$. Then

$\tilde{A}(t) = a\sigma_+ e^{i\omega_st^1} + b\sigma_- e^{-i\omega_st^1}$ and $\tilde{B}(t) = \sigma_z$. It is easy to see from Eq. (10) that $\tilde{U}(t)$ commutes with $\tilde{B}(t)$ but anticommutes with $\tilde{A}(t)$, i.e., $[\tilde{U}(t), \tilde{B}(t)] = 0$ and $\{\tilde{U}(t), \tilde{A}(t)\} = 0$. Using these results and the fact that $U^{(n)}(t)U^{(n)}(t_1) = I$, we obtain from Eq. (24)

$$\langle A(t_1)B(t_2) \rangle$$

$$= -a\rho_{10}(0)e^{i\omega_st^1}\text{Tr}_R[U^\dagger(0)(t_1)\tilde{U}(1)(0)R_0]$$

$$+ b\rho_{01}(0)e^{-i\omega_st^1}\text{Tr}_R[U^\dagger(1)(0)\tilde{U}(0)(t_1)R_0].$$

Substituting the result of Eq. (15) into Eq. (25), we arrive at the exact two-time CF’s

$$\langle A(t_1)B(t_2) \rangle$$

$$= e^{-\int_0^\tau dt D(\tau)} (-a\rho_{10}(0)e^{i\omega_st^1} + b\rho_{01}(0)e^{-i\omega_st^1}).$$

(26)

Case 2. $[A, L] = 0$ and $[B, \tilde{L}(t)] \neq 0$. In this case, let $A = \sigma_z$, and $B = a\sigma_+ + b\sigma_-$. Similar to the calculations in Case 1, we obtain

$$\langle A(t_1)B(t_2) \rangle$$

$$= e^{-\int_0^\tau dt D(\tau)} (a\rho_{10}(0)e^{i\omega_st^1} - b\rho_{01}(0)e^{-i\omega_st^1}).$$

(27)

The exact two-time CF’s of Eqs. (26) and (27) depend on only one time variable, $t_1$ or $t_2$, respectively, since one of the system operator $\sigma_z(t) = \sigma_z(0)$ is time-independent.

Case 3. $[A, L] \neq 0$ and $[B, \tilde{L}(t)] \neq 0$. Suppose $A = a\sigma_+ + b\sigma_-$, and $B = a'\sigma_+ + b'\sigma_-$. In this case, both $A(t)$ and $B(t)$ anticommute with both $\tilde{U}(t)$ and $\tilde{U}^\dagger(t)$. Furthermore, $\tilde{A}(t_1)\tilde{B}(t_2) = ab'\sigma_+\sigma_-\exp[i\omega_s(t_1 - t_2)] + ba'\sigma_-\sigma_+\exp[-i\omega_s(t_1 - t_2)]$. Thus we can obtain from Eq. (24)

$$\langle A(t_1)B(t_2) \rangle$$

$$= ab'\rho_{00}(0)e^{i\omega_s(t_1 - t_2)}$$

$$\times \text{Tr}_R[U^\dagger(0)(t_1)\tilde{U}(1)(0)\tilde{U}(1)(1)(t_2)\tilde{U}(0)(0)(t_2)R_0]$$

$$+ ba'\rho_{11}(0)e^{-i\omega_s(t_1 - t_2)}$$

$$\times \text{Tr}_R[\tilde{U}(1)(0)\tilde{U}(0)(t_1)\tilde{U}(1)(0)(t_1)\tilde{U}(1)(1)(t_2)\tilde{U}(0)(0)(t_2)R_0].$$

(28)

It is obvious from Eq. (28) that to evaluate the general two-time CF, we need to take into account the correlations of the reservoir operators of the evolution operators.
between different time periods of $[0, t_2]$ and $[0, t_1]$ before the trace over the environment is performed. Using Eqs. (12), (13), and (14), we get

$$\text{Tr}_R [\tilde{U}^{(0)}(t_1) \tilde{U}^{(1)}(t_1) \tilde{U}^{(1)}(t_2) \tilde{U}^{(0)}_1(t_2) R_0] = \exp \left[ - \int_0^{t_1} d\tau D(\tau) - \int_0^{t_2} d\tau D(\tau) + \int_0^{t_1} d\tau D(\tau, t_2) \right],$$

(29)

where

$$D(\tau, t_2) = 4 \int_0^{t_2} ds \alpha_{\text{eff}}(\tau - s).$$

(30)

The term $\int_0^{t_1} d\tau D(\tau, t_2)$ in Eq. (29) describes the cross-time contribution of the environment CF’s of the reservoir operators in the evolution operators $\tilde{U}^{(n)}(t_1)$ and $\tilde{U}^{(n)}(t_2)$ (or $\tilde{U}^{(n)}(t_1)$ and $\tilde{U}^{(n)}(t_2)$) of the two different time periods $[0, t_1]$ and $[0, t_2]$. We can see this from $D(\tau, t_2)$ of Eq. (30) and in Eq. (29) that the environment CF $\alpha_{\text{eff}}(\tau - s)$, defined in Eq. (17), has the time variable $\tau$ in $[0, t_1]$ and the time variable $s$ in $[0, t_2]$. On the other hand, the time evolution of the reduced density matrix $\rho(t)$ is involved with the reservoir operator CF’s in the evolution operators of only the same time interval. As a result, it, alone, cannot provide us with the full information to evaluate the non-Markovian two-time CF even if the interaction strength $\alpha(t - \tau)$ of previous terms, and an operator with a tilde on the top indicates that it is an operator in the interaction picture. For the pure-dephasing spin-boson model, Eq. (32) gives the master equation of the reduced system density matrix

$$\frac{d\rho(t)}{dt} = \frac{1}{\hbar} \left[ H_S, \rho(t) \right] - \int_0^t d\tau \left[ \alpha(t - \tau) [\hat{L}^\dagger(\tau - t) \rho(t) - \hat{L}(\tau - t) \rho(t) \hat{L}^\dagger], \beta(t - \tau) [\hat{L}^\dagger(\tau - t) \rho(t) - \hat{L}(\tau - t) \rho(t) \hat{L}^\dagger] + \text{H.c.} \right],$$

(32)

where $\alpha(t - \tau)$ and $\beta(t - \tau)$ are defined in Eqs. (11) and (5) respectively. H.c. indicates the Hermitian conjugate of previous terms, and an operator with a tilde on the top indicates that it is an operator in the interaction picture. For the pure-dephasing spin-boson model, Eq. (32) gives the master equation of the reduced system density matrix

$$\frac{d\rho(t)}{dt} = -\frac{i\omega_S}{2} [\sigma_z, \rho(t)] - \frac{D(t)}{2} [\rho(t) - \sigma_z \rho(t) \sigma_z],$$

(33)

where $D(t)$ is defined in Eq. (10). It is not difficult to show that the exact expression of the density matrix is the solution of the master equation (33) although the master equation is derived perturbatively. Non-Markovian dynamics usually means that the current time evolution of the system state depends on its history, and the memory effects typically enter through integrals over the past state history. However, the non-Markovian system dynamics of some class of open quantum system models may be summed up and expressed as a time-local, convolutionless form (29) where the dynamics is determined by the system state at the current time $t$ only. This time-local, convolutionless class of open quantum systems may be treated exactly without any approximation. The quantum Brownian motion model or the damped harmonic oscillator bilinearly coupled to a bosonic bath of harmonic oscillators is a famous example of this class. The pure-dephasing spin-boson model considered here also belongs to this class, and the non-Markovian effect in the master equation (33) is taken into account by the time-dependent coefficient $D(t)$ instead of memory integral. This time-local, convolutionless property and the fact of $[\hat{L}, H_S] = 0$ allow the exact system density matrix Eq. (18) to be obtained from Eq. (33).

Since the exact solution of the system density matrix can be calculated from the perturbatively derived master equation (33), one may expect that the exact non-Markovian finite-temperature one-time expectation values and two-time CF’s of the pure-dephasing model can be obtained from the evolution equation (33). We show
below that this is indeed the case, and at the same time
the agreement of the results demonstrates the validity
and practical usage of the evolution equation (3). For
the pure-dephasing spin-boson model, \( H_S = \frac{\hbar \omega S}{2} \sigma_z \), \( L = \sigma_x = L^\dagger \), and we have \( L(t) = \sigma_z \).
Taking \( A = \sigma_i \), \( i = x, y, z \), in Eq. (2), we obtain straight-
forwardly the equation of the single-time expectation
values as
\[
\begin{align*}
\frac{d}{dt} \langle \sigma_x(t_1) \rangle / dt_1 &= -D(t_1) \langle \sigma_x(t_1) \rangle - \omega_S \langle \sigma_y(t_1) \rangle, \quad (34) \\
\frac{d}{dt} \langle \sigma_z(t_1) \rangle / dt_1 &= -D(t_1) \langle \sigma_y(t_1) \rangle + \omega_S \langle \sigma_x(t_1) \rangle, \quad (35) \\
\frac{d}{dt} \langle \sigma_z(t_1) \rangle / dt_1 &= 0 \quad (36)
\end{align*}
\]
with \( D(t_1) \) defined in Eq. (16). With proper chosen values
for \( a, b, c, \) and \( d \) of a general operator \( A \) for \( \sigma_i \), one
can verify that the exact expression of the expectation
value of \( \sigma_i(t_1) \) in Eq. (21) satisfies Eqs. (34)–(36).

B. Two-time correlation functions

Before using Eq. (3) to calculate the non-Markovian
finite-temperature two-time CF’s, we discuss briefly
below the relation between the QRT and the evolution
equation (3). If the last two terms of Eq. (3) vanish,
then the single-time and two-time evolution equations
(2) and (3) will have the same form with the same evolu-
tion coefficients and thus the QRT will be applicable.
The last two terms of Eq. (2) or more generally the last
term of Eq. (17) in Ref. [12] involve(s) the propagation
from \( \tau = 0 \) to \( \tau = t_2 \), and these terms would vanish
for the CF’s \( \langle A(t_1)B(0) \rangle \) as \( t_2 = 0 \) in this case. So
the QRT is valid to calculate the CF’s \( \langle A(t_1)B(0) \rangle \) of both
Markovian and non-Markovian open systems, where the
system-environment density matrix is separable at \( t = 0 \).
The QRT is also valid and is often applied to calcula-
te, in the Markovian weak system-environment coupl-
ing case, more general CF’s \( \langle A(t_2 + \tau)B(t_2) \rangle \) or equiv-
antly \( \langle A(t_1)B(t_2) \rangle \) with \( t_2 \neq 0 \). For example, the QRT is
used to calculate the Markovian steady-state CF’s, and
in this case \( t_2 \) is set to any of the large times when the
steady state is reached. This is because in the Markov-
ian case, the last two terms of Eq. (2) vanish since the
time integration of the corresponding \( \delta \)-correlated reserv-
voir CF’s, \( \alpha(t_1 - \tau) \propto \delta(t_1 - \tau) \) and \( \beta(t_1 - \tau) \propto \delta(t_1 - \tau) \),
over the variable \( \tau \) in the domain from 0 to \( t_2 \) is zero as
\( t_1 > t_2 \). On the other hand, the QRT cannot be blindly
applied to calculate \( \langle A(t_1)B(t_2) \rangle \) with \( t_2 \neq 0 \) in a general
non-Markovian open system due to the non-vanishing
contributions of the cross correlation of the reservoir oper-
ators at two different times: a later time \( t_1 \) and an ear-
lier time in the period between 0 and \( t_2 \) (see the last
two terms of Eq. (3) and also Fig. 1). In other words,
in contrast to the Markovian case, not only the initial
condition \( \langle A(t_2)B(t_2) \rangle \) for the two-time evolution
equation (2) but also the equation (3) itself may depend on
the choice of the starting time \( t_2 \) of the non-Markovian
finite-temperature two-time CF’s. In the steady state,
where $\tilde{D}(t_1, t_2)$ is defined in Eq. (39). The evolution equations, Eqs. (41)–(44), have different forms as those of single-time expectation values due to the existence of $\tilde{D}(t_1, t_2)$ terms. As a result, the QRT does not hold in this case. Again, taking the derivative of Eq. (41) with respect to $t_1$ with properly chosen values for $a$, $b$, $a'$ and $b'$, we arrive at the same evolution equations as those from Eqs. (11) to (14). Alternatively, solving the coupled equations, Eqs. (41)–(44), one would obtain the solutions in a form as Eq. (61).

The agreement between the results obtained by the direct operator evaluation and those obtained by solving the coupled evolution equations demonstrates clearly the validity of the equations (2) and (3). In addition, the easiness to obtain Eqs. (41)–(44) from the evolution equation (2), and to obtain Eqs. (37)–(38), Eqs. (39) and (40), and Eqs. (41)–(44) from the evolution equation (3) illustrates the practical usage of the non-Markovian finite-temperature evolution equations (2) and (3).

V. RESULTS AND DISCUSSIONS

To calculate the two important functions $D(t)$ and $\tilde{D}(t_1, t_2)$, we need to evaluate the environment CF $\langle \sigma_x(t_1)\sigma_y(t_2) \rangle$ in case 1 and Eq. (31) in case 3 are

$\langle \sigma_x(t_1)\sigma_y(t_2) \rangle = 4\gamma \omega \exp(-i\omega t_2) \langle \sigma_x(t_1) \rangle + 4\gamma \omega \exp(i\omega t_2) \langle \sigma_x(t_1) \rangle$.

where $J(\omega) = \sum \delta(\omega - \omega_\lambda)$ is the spectral density of the environment. We may consider any spectral density to characterize the environment, but for simplicity we consider an ohmic bath with exponential cut-off function as

$J(\omega) = \lambda \omega \exp(-\omega/\Lambda)$.

where $\Lambda$ is the cut-off frequency and $\gamma$ is a dimensionless constant characterizing the interaction strength to the environment. At the zero temperature, the function $D(t)$ and $\tilde{D}(t_1, t_2)$ have simple analytical forms:

$D(t_1) = 4\gamma \lambda^2 t_1 \left[ 1 + \lambda^2 (t_1 - t_2) \right]$

$\tilde{D}(t_1, t_2) = 4\gamma \lambda^2 t_2 \left[ 1 - \lambda^2 (t_1 - t_2) \right]$

Consequently, the one-time expectation values and the two-time CF’s also have simple analytical expressions. For example, the zero-temperature two-time CF’s of Eq. (29) in case 1 and Eq. (61) in case 3 are

$\langle A(t_1)B(t_2) \rangle = \left[ 1 + \lambda^2 (t_1 - t_2) \right] \left[ \lambda^2 (t_1 - t_2) \right]$

$\langle A(t_1)B(t_2) \rangle = \left[ 1 + \lambda^2 (t_1 - t_2) \right] \left[ \lambda^2 (t_1 - t_2) \right]$

and

$\langle A(t_1)B(t_2) \rangle = \left[ 1 + \lambda^2 (t_1 - t_2) \right] \left[ \lambda^2 (t_1 - t_2) \right]$

respectively.

Figure 1 shows the the time evolutions of the real part of the system operator CF $\langle \sigma_x(t_1)\sigma_y(t_2) \rangle$ for different times: Markovian (solid line), non-Markovian using the QRT (dashed line) and non-Markovian using the QRT (dotted line) using Eq. (3) and exact operator evaluation (dot-dashed line). Other parameters used are $\omega_S = 1$, $k_BT = 0.1$, $\Lambda = 5$, $\gamma = 0.1$, and $t_2 = 0.2$.

FIG. 1. (Color online) Time evolutions of the real part of the system operator CF $\langle \sigma_x(t_1)\sigma_y(t_2) \rangle$ for different times: Markovian (solid line), non-Markovian using the QRT (dashed line) and non-Markovian using the QRT (dotted line) using Eq. (3) and exact operator evaluation (dot-dashed line). Other parameters used are $\omega_S = 1$, $k_BT = 0.1$, $\Lambda = 5$, $\gamma = 0.1$, and $t_2 = 0.2$. 

and

$\langle A(t_1)B(t_2) \rangle = \left[ 1 + \lambda^2 (t_1 - t_2) \right] \left[ \lambda^2 (t_1 - t_2) \right]$

respectively.

Figure 1 shows the the time evolutions of the real part of the system operator CF $\langle \sigma_x(t_1)\sigma_y(t_2) \rangle$ for different times: Markovian (solid line), non-Markovian using the QRT (dashed line) and non-Markovian using the QRT (dotted line) using Eq. (3) and exact operator evaluation (dot-dashed line). Other parameters used are $\omega_S = 1$, $k_BT = 0.1$, $\Lambda = 5$, $\gamma = 0.1$, and $t_2 = 0.2$. 

and

$\langle A(t_1)B(t_2) \rangle = \left[ 1 + \lambda^2 (t_1 - t_2) \right] \left[ \lambda^2 (t_1 - t_2) \right]$

respectively.

Figure 1 shows the the time evolutions of the real part of the system operator CF $\langle \sigma_x(t_1)\sigma_y(t_2) \rangle$ for different times: Markovian (solid line), non-Markovian using the QRT (dashed line) and non-Markovian using the QRT (dotted line) using Eq. (3) and exact operator evaluation (dot-dashed line). Other parameters used are $\omega_S = 1$, $k_BT = 0.1$, $\Lambda = 5$, $\gamma = 0.1$, and $t_2 = 0.2$. 

and

$\langle A(t_1)B(t_2) \rangle = \left[ 1 + \lambda^2 (t_1 - t_2) \right] \left[ \lambda^2 (t_1 - t_2) \right]$

respectively.

Figure 1 shows the the time evolutions of the real part of the system operator CF $\langle \sigma_x(t_1)\sigma_y(t_2) \rangle$ for different times: Markovian (solid line), non-Markovian using the QRT (dashed line) and non-Markovian using the QRT (dotted line) using Eq. (3) and exact operator evaluation (dot-dashed line). Other parameters used are $\omega_S = 1$, $k_BT = 0.1$, $\Lambda = 5$, $\gamma = 0.1$, and $t_2 = 0.2$. 

and

$\langle A(t_1)B(t_2) \rangle = \left[ 1 + \lambda^2 (t_1 - t_2) \right] \left[ \lambda^2 (t_1 - t_2) \right]$

respectively.

Figure 1 shows the the time evolutions of the real part of the system operator CF $\langle \sigma_x(t_1)\sigma_y(t_2) \rangle$ for different times: Markovian (solid line), non-Markovian using the QRT (dashed line) and non-Markovian using the QRT (dotted line) using Eq. (3) and exact operator evaluation (dot-dashed line). Other parameters used are $\omega_S = 1$, $k_BT = 0.1$, $\Lambda = 5$, $\gamma = 0.1$, and $t_2 = 0.2$. 

and

$\langle A(t_1)B(t_2) \rangle = \left[ 1 + \lambda^2 (t_1 - t_2) \right] \left[ \lambda^2 (t_1 - t_2) \right]$

respectively.
in Eqs. (44) and (45) by its long-time limit value. At a finite temperature, the Markovian (time-independent) coefficient from Eqs. (10), (15) and (16) can be written as

$$D_{\infty} = \lim_{t \to \infty} D(t) = 4\gamma \pi k_B T / h.$$  \hfill (51)

We may see that $D_{\infty} \to 0$ as the temperature $T \to 0$. This is because in the Markovian limit, the decoherence or dephasing is strongly dependent on the infrared behavior ($\omega \to 0$ modes) of the environment in the pure dephasing model. Since the spectral density considered in Eq. (40) is Ohmic, we then have $J(\omega \to 0) = 0$, and thus $D_{\infty} \to 0$ at $T = 0$. This is in contrast to other quantum open system models with a resonant type of system-environment coupling, in which the environment modes near the system resonance frequency are relevant to the relaxation and decoherence. We can see from Fig. 1 that the difference between the results of the Markovian QRT case and the non-Markovian QRT case is visible, while the two-time CF’s obtained by the non-Markovian evolution equation (3) and by the exact operator evaluation are identical for the pure-dephasing spin-boson model. The perfect agreement of the results between the non-Markovian evolution equation case and the exact operator evaluation case, and the significant difference in the short time region between the non-Markovian evolution equation case and the wrong non-Markovian QRT case demonstrate clearly the validity and practical usage of the evolution equation (3). All of the four cases approach one another to zero in the long time region.

Figure 2 investigates the dependence of the exact two-time system operator CF on the time variable $t_2$. We see that the time evolutions of the real part of the CF $\langle \sigma_x(t_1)\sigma_y(t_2) \rangle$ as a function of $t = t_1 - t_2$ for the values of $t_2 \leq 1$ behave quite differently, but they approach one another for $t_2 \geq 2$. When $t_2 \geq 5$, the steady state is reached as indicated in the time evolutions of the expectation values $\langle \sigma_x(t) \rangle$ and $\langle \sigma_y(t) \rangle$ shown in the insets of Fig. 2. In this case, the time evolutions of the two-time CF are independent of the choices of the starting time of $t_2$ in the steady state and depend only on the time difference $t = t_1 - t_2$ for the parameters used in Fig. 2. This can also be seen from the analytical expression of the exact zero-temperature CF (50). The zero-temperature CF (50) is a function of variables $t_1$ and $t_2$, but for a large value of the cut-off frequency $\Lambda$, when $t_2$ is reasonably large, the CF depends almost only on $t = t_1 - t_2$. Figure 3 shows the time evolutions of the real part of the steady-state $(t_2 = 10)$ system operator CF $(\sigma_x(t_1)\sigma_y(t_2))$ obtained in four different cases as in Fig. 1. As expected, the non-Markovian evolution equation case coincides with the exact operator evaluation case. They are, however, significantly different from the wrong non-Markovian QRT case and Markovian QRT case, even though the time evolutions of the steady-state two-time CF’s depend only on the time difference $t = t_1 - t_2$. One can also see that the CF’s of the Markovian QRT and the non-Markovian QRT cases approach each other much more closely in the steady state than in Fig. 1.

VI. CONCLUSION

We have evaluated the exact non-Markovian finite-temperature one-time expectation values and two-time CF’s of the system operators for the exactly solvable pure-dephasing spin-boson model. The evaluation has been performed in two ways, one by exact direct operator technique without any approximation and the other
by the evolution equations (2) and (3) valid to second order in the system-environment interaction Hamiltonian. Since the non-Markovian dynamics of the pure-dephasing spin-boson model can be cast into a time-local, convolutionless form and \([L, H_s] = 0\), the results obtained by the second-order evolution equations (2) and (3) turn out to be exactly the same as the exact results obtained by the exact direct operator evaluation. The agreement of the results between the two different approaches demonstrates clearly the validity of the evolution equations (2) and (3). Furthermore, it is easy to obtain Eqs. (57), (58), Eqs. (39) and (40), and Eqs. (41)–(43) from the evolution equation (3). Other non-Markovian open quantum system models that are not exactly solvable can be proceeded in a similar way to obtain the time evolutions of their two-time system operator CF’s valid to second order in the system-environment interaction Hamiltonian. This illustrates the practical usage of the evolution equations. It is thus believed that the evolution equations (2) and (3), which generalize the QRT to the non-Markovian finite-temperature case will have applications in many different branches of physics.

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Appendix A: Derivation of time evolution operator

To show the time evolution operator of Eq. (10), we begin from Eq. (8) with \(\tilde{H}_I(t)\) given by Eq. (9). Since \(\tilde{H}_I(t)\) in Eq. (9) contains only two major terms, which are, respectively, proportional to \(a_\lambda\) and \(a_\lambda^\dagger\), one is tempting to evaluate the time-ordered exponent by the reverse operator identity of Eq. (12)

\[ e^{A+B} = e^A e^B e^{-\frac{i}{2}[A,B]}, \tag{A1} \]

valid for the commutator \([A, B]\) commuting with both \(A\) and \(B\). As the exponent operates at different times and \([\tilde{H}_I(t), \tilde{H}_I(\tau)] \neq 0\), it is not correct to use the precise form of Eq. (A1). The proper procedure done in [20,32] is to separate the two terms of Eq. (9) in the time-ordered exponent of Eq. (8) by

\[
\tilde{U}(t) = \exp \left\{ -i \int_0^t d\tau \sum_\lambda g_\lambda \sigma_\lambda e^{i\omega_\lambda \tau} a_\lambda^\dagger \right\} 
\times T \left\{ \exp \left[ -i \int_0^t d\tau e^{i\omega_\lambda \tau} \sum_\lambda g_\lambda \sigma_\lambda e^{i\omega_\lambda \tau} a_\lambda^\dagger \times \left( \sum_\lambda g_\lambda \sigma_\lambda e^{-i\omega_\lambda \tau} a_\lambda \right) e^{-i \int_0^\tau ds \sum_\lambda g_\lambda \sigma_\lambda e^{i\omega_\lambda \tau} a_\lambda^\dagger} \right] \right\}, \tag{A2}
\]

where \(T\) is the time-ordering operator. Then using the identity \(e^{-\phi a_\lambda^\dagger a_\lambda} e^{\phi a_\lambda^\dagger a_\lambda} = a_\lambda + \phi\) in the exponent of the time-ordered term in Eq. (A2), we obtain

\[
\tilde{U}(t) = \exp \left\{ -i \int_0^t d\tau \sum_\lambda g_\lambda \sigma_\lambda e^{i\omega_\lambda \tau} a_\lambda^\dagger \right\} 
\times \exp \left( -i \int_0^t d\tau \sum_\lambda g_\lambda \sigma_\lambda e^{-i\omega_\lambda \tau} a_\lambda \right) 
\times \exp \left( -i \int_0^t d\tau \int_0^\tau ds \sum_\lambda |g_\lambda|^2 e^{-i\omega_\lambda (\tau-s)} \right). \tag{A3}
\]

We have dropped the time-ordering operator in Eq. (A3). Using the operator identity of Eq. (12) to combine the first two terms in Eq. (A3), we then obtain Eq. (10). Note that the first term in Eq. (10) is just\(\exp[-i(\hbar/2)\int_0^t d\tau \tilde{H}_I(\tau)]\) if the time-ordering operator for \(\tilde{U}(t)\) is not performed. The correct time-ordering procedure generates extra phase factors in Eq. (A3) and thus in Eq. (10).

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