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Unified projection operator formalism in nonequilibrium statistical mechanics

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The method of projection operators, which plays an important role in the field of nonequilibrium statistical mechanics, has been established with the use of the Liouville–von Neumann equation for a density matrix to eliminate irrelevant information from a whole system. We formulate a unified and general projection operator method for dynamical variables. The main features of our formalism parallel those for the Liouville–von Neumann equation. (1) Two types of basic equations, time-convolution and time-convolutionless decompositions, are systematically obtained without specifying a projection operator. (2) Expansion formulas for both decompositions are also obtained. (3) Problems incorporating a time-dependent Liouville operator can be flexibly treated. We apply the formulas to problems in random frequency modulation and low field resonance. In conclusion, our formalism yields a more direct and easier means of determining the average time evolution of an operator than the one for the Liouville–von Neumann equation. [S1063-651X(99)07009-9]

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I. INTRODUCTION

The relaxation dynamics of nonequilibrium systems have often been studied with models that have the system of interest in contact with a reservoir. A typical example is the phenomenon of nuclear magnetic (or spin) relaxation. NMR and muon spin resonance-relaxation-rotation (μSR) spectra are often analyzed on the assumption that the relaxation process is caused by random perturbations from the environment around a relevant spin. The environment is supposed to be a reservoir that is large enough to be kept in an equilibrium state of constant temperature. Simple models of this kind allow us to analyze relaxation processes in a systematic manner.

In recent years, intensive experimental studies of NMR and μSR have been carried out with the aim of clarifying the basic structure and characteristics of various materials. In particular, an increasing number of experiments for this purpose have been conducted in low or zero magnetic fields [1–10]. When the applied field is weak, we have to take fully into account the interaction between the relevant spin and the environment. In other words, we cannot neglect higher orders of perturbation. For this reason, a systematic method of evaluating the effects of perturbations up to infinite orders is necessary.

The projection operator method has demonstrated its usefulness in eliminating irrelevant information from a system and extracting only the information that is desired or relevant [11]. This is usually done with the use of the Liouville–von Neumann equation for a density matrix. The method was formulated in a straightforward way, and can be readily manipulated to derive a basic equation for the relevant (or necessary) parts of the density matrix [12–15]. Because the equation thus obtained includes a time-convolution term reflecting a memory effect, it is called a time-convolution (TC) equation in this paper. Although the TC equation was long considered unique, systematic methods of obtaining new types of time-convolutionless (TCL) equations were derived by renormalizing the memory kernel [16,17]. Earlier work and related papers on this subject deserve attention [18–22]. Expansion formulas have also been systematically derived [23,24]. The characteristic features of the formalism are as follows: (i) we need not specify the projection operator; (ii) problems with a time-dependent Liouville operator can also be addressed; (iii) a systematic formalism for derivation of a basic equation is established; and (iv) the original Liouville–von Neumann equation can be readily substituted by other basic equations, e.g., the Fokker-Planck equation [23,25]. It is possible to apply this formalism to nuclear magnetic (or spin) relaxation phenomena in weak (or zero) applied fields. However, long and complicated calculations are still required even if only a certain averaged quantity is desired [26].

To find a way out of this difficulty, focus was placed on the Heisenberg equation of motion, in which dynamic variables evolve directly with time. Since the temporal evolution of dynamic variables in these equations is determined by the Hamiltonian for the entire system, the projection procedure faces another kind of difficulty. This is because we must separate the systematic and fluctuating parts of the Heisenberg equation of motion even though we need an equation for the observables that evolve temporally with the Hamiltonian for the whole system. This difficulty has been overcome by Mori and Tokuyama and Mori. Specifying the projection operator, Mori has derived a TC equation [27] for a dynamic variable based on the Heisenberg equation of motion, and Tokuyama and Mori have proposed a TCL equation [28]. Additional derivations of the basic equations also depend on the choice of projection operator [29,30], and/or a seemingly arbitrary step that is performed to obtain a basic identity for a dynamic variable [28,30–32]. In other words, the projection operator method for the Heisenberg equation of motion cannot yet be fully characterized by the four points mentioned above.

Thus, our purpose in this paper is twofold: one is to propose a systematic and natural formalism for the Heisenberg equation of motion without specifying a projection operator, thus yielding a procedure paralleling that for the Liouville–

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von Neumann equation; the other is to set out explicit expansion formulas for the Heisenberg equation of motion which, again, are derived through a procedure that parallels the approach for the Liouville–von Neumann equation. We then apply this formalism to actual problems of random frequency modulation and low field resonance in a quantum environment; for the former, our calculations yield an exact result, and for the latter, which is a difficult problem for which to obtain solutions, we present explicit calculations for several finite orders of perturbation as an example of our formulas.

This paper is organized as follows. The foundations of our formulation are laid down in Sec. II. Basic equations of our systematic formalism for the Heisenberg equation of motion are derived in Sec. III. We compare the formalism for the Heisenberg equation of motion with that for the Liouville–von Neumann equation in Sec. IV. Expansion formulas are presented in Sec. V. They are then applied to a model of random frequency modulation in Sec. VI, and to a model of low field resonance in a quantum environment in Sec. VII. Established results for the Liouville–von Neumann equation based on this method are briefly summarized in Appendixes A and B. Relationships between projection operators are discussed in Appendix C, and the characteristic features of the quantum environment applied in Sec. VII are defined in Appendix D. A preliminary outline of this paper has been published [33].

II. PRELIMINARIES

Let us consider the density operator $W(t)$ which evolves in time according to the Liouville–von Neumann equation:

$$\dot{W}(t) = -i\mathcal{L}(t)W(t), \tag{2.1}$$

where

$$\mathcal{L}(t)\cdot = \frac{1}{\hbar}[\mathcal{H}(t),\cdot]. \tag{2.2}$$

We assume that the total Hamiltonian $\mathcal{H}(t)$ consists of an unperturbed part $\mathcal{H}_0$ and a time-dependent perturbation $\mathcal{H}_1(t)$ giving the Liouville operator of the form

$$\mathcal{L}(t)\cdot = [\mathcal{L}_0 + \mathcal{L}_1(t)]\cdot . \tag{2.3}$$

Equation (2.1) is formally solved to give

$$W(t) = U_+(t,t_0)W(t_0), \tag{2.4}$$

where we defined $U_+(t,t_0)$ as

$$U_+(t,t_0) \equiv T_+[e^{-\int_{t_0}^{t}dt' i\mathcal{L}(t')}]. \tag{2.5}$$

In Eq. (2.5), the symbol T_+ indicates an increasing time ordering from the right to the left. Extracting the unperturbed part from the time evolution operator $U_+(t,t_0)$, we obtain

$$U_+(t,t_0) = e^{-i\mathcal{L}_0(t-t_0)}\hat{U}_+(t,t_0), \tag{2.6}$$

where

$$\hat{U}_+(t,t_0) \equiv T_+\left[\exp\left(-\int_{t_0}^{t}dt' i\hat{\mathcal{L}}_1(t')\right)\right] \tag{2.7}$$

and

$$\hat{\mathcal{L}}_1(t) = e^{i\mathcal{L}_0(t-t_0)}\mathcal{L}_1(t)e^{-i\mathcal{L}_0(t-t_0)}. \tag{2.8}$$

Noting a rule of a trace operation for two operators X and Y, $\mathrm{Tr}(XY) = \mathrm{Tr}(YX)$, we find a mean value of an operator A in two different ways:

$$\langle A\rangle_t = \mathrm{Tr}\, AW(t) = \mathrm{Tr}\; W(t_0)A(t) \equiv \langle A(t)\rangle, \tag{2.9}$$

where

$$A(t) = U_-(t,t_0)A, \tag{2.10}$$

with

$$U_-(t,t_0) \equiv T_-\left[\exp\left(\int_{t_0}^{t}dt' i\mathcal{L}(t')\right)\right]. \tag{2.11}$$

In the above expression, the symbol T_- indicates an increasing time ordering from the left to the right. For later convenience, we rewrite $U_-(t,t_0)$ as [see Eq. (2.6)]

$$U_-(t,t_0) = \hat{U}_-(t,t_0)e^{i\mathcal{L}_0(t-t_0)}, \tag{2.12}$$

where we defined

$$\hat{U}_-(t,t_0) \equiv T_-\left[\exp\left(\int_{t_0}^{t}dt' i\hat{\mathcal{L}}_1(t')\right)\right]. \tag{2.13}$$

When the unperturbed part is solved in the form

$$e^{i\mathcal{L}_0(t-t_0)}A = f(t,t_0)A, \tag{2.14}$$

where $f(t,t_0)$ is a c-number function, time evolution of the operator $A(t)$ is determined to be

$$A(t) = f(t,t_0)\hat{A}(t), \tag{2.15}$$

with

$$\hat{A}(t) \equiv \hat{U}_-(t,t_0)A. \tag{2.16}$$

Most of the existing projection operator methods use the time evolution operator $\hat{U}_+(t,t_0)$. We call them Schrödinger picture (SP) formalisms. In the following, we develop a method of projection operator based on $\hat{U}_-(t,t_0)$ and call it the Heisenberg picture (HP) formalism. We use different projection operator symbols for each picture: the projection operator used in the Heisenberg picture is called $\mathcal{P}$ and the one in the Schrödinger picture is $\tilde{\mathcal{P}}$. We discuss a relation between $\mathcal{P}$ and $\tilde{\mathcal{P}}$ in Appendix C.

III. BASIC DECOMPOSITION FORMULAS

In this section, we develop a projection operator method for $\hat{U}_-(t,t_0)$ and give the basic equations in the HP. Time evolution of the operator $\hat{U}_-(t,t_0)$ is determined by

$$\frac{\partial}{\partial t}\hat{U}_-(t,t_0) = \hat{U}_-(t,t_0)i\hat{\mathcal{L}}_1(t). \tag{3.1}$$

Comparing Eq. (3.1) with the equation for $\hat{U}_+(t,t_0)$, Eq. (A1), we find a reversed order of the operator $\hat{\mathcal{L}}_1(t)$ in the right hand side of the equations. Due to the difference, we can formulate a projection operator method [33].

We use a projection operator $\mathcal{P}$ in order to eliminate irrelevant variables. The operator $\mathcal{P}$ must satisfy an idempotent relation, $\mathcal{P}^2=\mathcal{P}$. Operating the projection operator $\mathcal{P}$ and $\mathcal{Q}(=1-\mathcal{P})$ in Eq. (3.1) from the right, we obtain

$$\frac{d}{dt}\hat{x}_-(t)=\hat{x}_-(t)i\hat{\mathcal{L}}_1(t)\mathcal{P}+\hat{y}_-(t)i\hat{\mathcal{L}}_1(t)\mathcal{P}, \tag{3.2}$$

$$\frac{d}{dt}\hat{y}_-(t)=\hat{x}_-(t)i\hat{\mathcal{L}}_1(t)\mathcal{Q}+\hat{y}_-(t)i\hat{\mathcal{L}}_1(t)\mathcal{Q}, \tag{3.3}$$

where we set

$$\hat{x}_-(t)=\hat{U}_-(t,t_0)\mathcal{P} \tag{3.4}$$

and

$$\hat{y}_-(t)=\hat{U}_-(t,t_0)\mathcal{Q}. \tag{3.5}$$

(i) Time-convolution decomposition. Equation (3.3) is solved to give

$$\hat{y}_-(t)=\mathcal{Q}\hat{u}_-(t,t_0)+\int_{t_0}^{t}d\tau\,\hat{x}_-(\tau)i\hat{\mathcal{L}}_1(\tau)\mathcal{Q}\hat{u}_-(t,\tau), \tag{3.6}$$

where

$$\hat{u}_-(t,\tau)=T_-\left[\exp\left(\int_{\tau}^{t}d\tau'\,i\hat{\mathcal{L}}_1(\tau')\mathcal{Q}\right)\right]. \tag{3.7}$$

With the use of Eqs. (3.2) and (3.6), we have a time-convolution type of decomposition:

$$\begin{aligned}\frac{d}{dt}\hat{x}_-(t)=&\hat{x}_-(t)i\hat{\mathcal{L}}_1(t)\mathcal{P}\\&+\int_{t_0}^{t}d\tau\,\hat{x}_-(\tau)i\hat{\mathcal{L}}_1(\tau)\mathcal{Q}\hat{u}_-(t,\tau)i\hat{\mathcal{L}}_1(t)\mathcal{P}\\&+\mathcal{Q}\hat{u}_-(t,t_0)i\hat{\mathcal{L}}_1(t)\mathcal{P}.\end{aligned} \tag{3.8}$$

For actual problems, almost all of $\mathcal{P}$ satisfies the relation $\mathcal{P}A=A$. Then we have, from Eqs. (2.16), (3.4), and the condition $\mathcal{P}A=A$,

$$\hat{x}_-(t)A=\hat{U}_-(t,t_0)\mathcal{P}A=\hat{A}(t). \tag{3.9}$$

Thus Eq. (3.8) gives

$$\begin{aligned}\frac{d}{dt}\hat{A}(t)=&\hat{x}_-(t)i\hat{\mathcal{L}}_1(t)A\\&+\int_{t_0}^{t}d\tau\,\hat{x}_-(\tau)i\hat{\mathcal{L}}_1(\tau)\mathcal{Q}\hat{u}_-(t,\tau)i\hat{\mathcal{L}}_1(t)A\\&+\mathcal{Q}\hat{u}_-(t,t_0)i\hat{\mathcal{L}}_1(t)A\end{aligned} \tag{3.10}$$

for a dynamic variable $\hat{A}(t)$.

(ii) Time-convolutionless decomposition. Since we have the following relation:

$$\begin{aligned}\hat{x}_-(\tau)&=\hat{U}_-(\tau,t_0)\mathcal{P}=\hat{U}_-(t,t_0)(\mathcal{P}+\mathcal{Q})\hat{U}_+(t,\tau)\mathcal{P}\\&=\hat{x}_-(t)\hat{U}_+(t,\tau)\mathcal{P}+\hat{y}_-(t)\hat{U}_+(t,\tau)\mathcal{P},\end{aligned} \tag{3.11}$$

Eq. (3.6) is rewritten as

$$\begin{aligned}\hat{y}_-(t)=&\mathcal{Q}\hat{u}_-(t,t_0)+\{\hat{x}_-(t)+\hat{y}_-(t)\}\\&\times\int_{t_0}^{t}d\tau\hat{U}_+(t,\tau)\mathcal{P}i\hat{\mathcal{L}}_1(\tau)\mathcal{Q}\hat{u}_-(t,\tau).\end{aligned} \tag{3.12}$$

We solve Eq. (3.12) for $\hat{y}_-(t)$:

$$\hat{y}_-(t)=[\mathcal{Q}\hat{u}_-(t,t_0)-\hat{x}_-(t)(\hat{\Theta}_-(t)^{-1}-1)]\hat{\Theta}_-(t), \tag{3.13}$$

with

$$\hat{\Theta}_-(t)=\left(1-\int_{t_0}^{t}d\tau\hat{U}_+(t,\tau)\mathcal{P}i\hat{\mathcal{L}}_1(\tau)\mathcal{Q}\hat{u}_-(t,\tau)\right)^{-1}. \tag{3.14}$$

Thus, from Eqs. (3.2) and (3.13), we have a time-convolutionless type of decomposition of the form

$$\begin{aligned}\frac{d}{dt}\hat{x}_-(t)=&\hat{x}_-(t)i\hat{\mathcal{L}}_1(t)\mathcal{P}-\hat{x}_-(t)\{1-\hat{\Theta}_-(t)\}i\hat{\mathcal{L}}_1(t)\mathcal{P}\\&+\mathcal{Q}\hat{u}_-(t,t_0)\hat{\Theta}_-(t)i\hat{\mathcal{L}}_1(t)\mathcal{P}.\end{aligned} \tag{3.15}$$

Then the condition $\mathcal{P}A=A$ gives

$$\begin{aligned}\frac{d}{dt}\hat{A}(t)=&\hat{x}_-(t)i\hat{\mathcal{L}}_1(t)A-\hat{x}_-(t)\{1-\hat{\Theta}_-(t)\}i\hat{\mathcal{L}}_1(t)A\\&+\mathcal{Q}\hat{u}_-(t,t_0)\hat{\Theta}_-(t)i\hat{\mathcal{L}}_1(t)A\end{aligned} \tag{3.16}$$

for a dynamic variable $\hat{A}(t)$.

IV. TRANSCRIPTION RULES

Comparing basic formulas for the HP derived in Sec. II with the corresponding ones for the SP in Appendix A, we can deduce the following rules: In converting from the HP to the SP or vice versa, we are only to apply the following rules to the basic equations.

Rule 1. The constituent operators are replaced by

$$\left.\begin{matrix}\text{HP}\\ i\hat{\mathcal{L}}_1(t)\\ \mathcal{P}\end{matrix}\right\}\leftrightarrow\left\{\begin{matrix}\text{SP}\\ -i\hat{\mathcal{L}}_1(t).\\ \tilde{\mathcal{P}}\end{matrix}\right. \tag{4.1}$$

Rule 2. Order of the constituent operators is reversed as follows:

$$\left.\begin{matrix}\text{HP}\\ i\hat{\mathcal{L}}_1(t)\mathcal{Q}\\ T_-[e^{\int_\tau^t d\tau' i\hat{\mathcal{L}}_1(\tau')\mathcal{Q}}]\end{matrix}\right\}\leftrightarrow\left\{\begin{matrix}\text{SP}\\ -\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t)\\ T_+\left[\exp\left(\int_\tau^t d\tau'\,\tilde{\mathcal{Q}}(-i\tilde{\mathcal{L}}_1(\tau'))\right)\right].\end{matrix}\right. \tag{4.2}$$

These also yield the following replacements:

$$\left.\begin{array}{l}\text{HP}\\ \hat{x}_-(t)\\ \hat{u}_-(t,t_0)\end{array}\right\}\leftrightarrow\left\{\begin{array}{l}\text{SP}\\ \hat{x}_+(t)\\ \hat{u}_+(t,t_0)\end{array}\right. . \tag{4.3}$$

A simple example may serve for illustration: According to these rules, the second term in the right hand side of Eq. (3.8),

$$\int_{t_0}^{t} d\tau\, \hat{x}_-(\tau) i\hat{\mathcal{L}}_1(\tau)\mathcal{Q}\hat{u}_-(t,\tau) i\hat{\mathcal{L}}_1(t)\mathcal{P}, \tag{4.4}$$

is transformed into

$$\int_{t_0}^{t} d\tau\, \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{u}_+(t,\tau)\tilde{\mathcal{Q}}(-i\tilde{\mathcal{L}}_1(\tau))\hat{x}_+(\tau). \tag{4.5}$$

This is nothing but the second term in the right hand side of Eq. (A8).

V. EXPANSION FORMULAS

In order to apply the above method to actual problems, we derive a new type of cumulants by expanding the TC and TCL type decompositions.

(i) TC decomposition. For the TC type decomposition (3.10),

$$\frac{d}{dt}\hat{A}(t)=\hat{U}_-(t,t_0)\mathcal{P}i\hat{\mathcal{L}}_1(t)A+\int_{t_0}^{t} d\tau\, \hat{K}_-(t,\tau)A+\hat{\mathcal{J}}_-(t), \tag{5.1}$$

with

$$\hat{K}_-(t,\tau)\equiv\hat{U}_-(\tau,t_0)\mathcal{P}i\hat{\mathcal{L}}_1(\tau)\mathcal{Q}\hat{u}_-(t,\tau)i\hat{\mathcal{L}}_1(t) \tag{5.2}$$

and

$$\hat{\mathcal{J}}_-(t)\equiv\mathcal{Q}\hat{u}_-(t,t_0)i\hat{\mathcal{L}}_1(t)A, \tag{5.3}$$

we expand $\hat{u}_-(t,\tau)$ in Eqs. (5.2) and (5.3) as follows:

$$\begin{aligned}\int_{t_0}^{t} d\tau\, \hat{K}_-(t,\tau)=&\int_{t_0}^{t} dt_1\, \hat{U}_-(t_1,t_0)\mathcal{P}\,\hat{\Phi}_{-,2}(t,t_1)\\ &+\sum_{n=3}^{\infty}\int_{t_0}^{t} dt_1\int_{t_0}^{t_1} dt_2\cdots\int_{t_0}^{t_{n-2}} dt_{n-1}\\ &\times\hat{U}_-(t_{n-1},t_0)\mathcal{P}\,\hat{\Phi}_{-,n}(t,t_1,\ldots,t_{n-2},t_{n-1}),\end{aligned} \tag{5.4}$$

where

$$\hat{\Phi}_{-,2}(t,t_1)=i\hat{\mathcal{L}}_1(t_1)\mathcal{Q}i\hat{\mathcal{L}}_1(t), \tag{5.5}$$

$$\begin{aligned}&\hat{\Phi}_{-,n}(t,t_1,\ldots,t_{n-2},t_{n-1})\\ &\quad=i\hat{\mathcal{L}}_1(t_{n-1})\mathcal{Q}i\hat{\mathcal{L}}_1(t_{n-2})\cdots\mathcal{Q}i\hat{\mathcal{L}}_1(t_1)\mathcal{Q}i\hat{\mathcal{L}}_1(t)\quad(n\geqslant 3)\end{aligned} \tag{5.6}$$

and

$$\begin{aligned}\hat{\mathcal{J}}_-(t)=&\Bigg(\mathcal{Q}i\hat{\mathcal{L}}_1(t)+\int_{t_0}^{t} dt_1\,\mathcal{Q}\hat{\Phi}_{-,2}(t,t_1)\\ &+\sum_{n=3}^{\infty}\int_{t_0}^{t} dt_1\int_{t_0}^{t_1} dt_2\cdots\int_{t_0}^{t_{n-2}} dt_{n-1}\\ &\times\mathcal{Q}\hat{\Phi}_{-,n}(t,t_1,\ldots,t_{n-2},t_{n-1})\Bigg)A.\end{aligned} \tag{5.7}$$

In Eqs. (5.4) and (5.7), we find almost the same structure for $\hat{\Phi}_{-,n}(t)$ as the one for the SP except the chronological order [see Eqs. (B4) and (B7)]. When we represent the projection operator $\mathcal{P}$ as an appropriate averaging operation, namely, $\mathcal{P}\cdot\equiv\langle\langle\cdot\rangle\rangle$, we naturally obtain new cumulants and call them "antipartial cumulants" (APC), denoting them as

$$\begin{aligned}&\mathcal{P}\,\hat{\Phi}_{-,n}(t,t_1,\ldots,t_{n-2},t_{n-1})\\ &\quad\equiv\langle i\hat{\mathcal{L}}_1(t_{n-1})i\hat{\mathcal{L}}_1(t_{n-2})\cdots i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\quad(n\geqslant 2).\end{aligned} \tag{5.8}$$

For instance, the lower order cumulants are explicitly given by

$$\begin{aligned}\langle i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}=&\mathcal{P}i\hat{\mathcal{L}}_1(t_1)\mathcal{Q}i\hat{\mathcal{L}}_1(t)\\ =&\langle\langle i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle\rangle\\ &-\langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\end{aligned} \tag{5.9}$$

and

$$\begin{aligned}&\langle i\hat{\mathcal{L}}_1(t_2)i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\\ &\quad=\mathcal{P}i\hat{\mathcal{L}}_1(t_2)\mathcal{Q}i\hat{\mathcal{L}}_1(t_1)\mathcal{Q}i\hat{\mathcal{L}}_1(t)\\ &\quad=\langle\langle i\hat{\mathcal{L}}_1(t_2)i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle\rangle-\langle\langle i\hat{\mathcal{L}}_1(t_2)i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\\ &\qquad\times\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle-\langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle\rangle\\ &\qquad+\langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle.\end{aligned} \tag{5.10}$$

(ii) TCL decomposition. Next, we treat Eq. (3.16), i.e.,

$$\frac{d}{dt}\hat{A}(t)=\hat{\Psi}_-(t)A+\hat{J}_-(t), \tag{5.11}$$

where

$$\hat{\Psi}_-(t)=\hat{U}_-(t,t_0)\mathcal{P}\,\hat{\Theta}_-(t)i\hat{\mathcal{L}}_1(t) \tag{5.12}$$

and

$$\hat{J}_-(t)=\mathcal{Q}\hat{u}_-(t,t_0)\hat{\Theta}_-(t)i\hat{\mathcal{L}}_1(t)A. \tag{5.13}$$

We find the first term of Eq. (5.11) in the form

$$\begin{aligned}\hat{\Psi}_-(t)=&\hat{U}_-(t,t_0)\sum_{n=0}^{\infty}\mathcal{P}(\hat{\sigma}_-(t))^n i\hat{\mathcal{L}}_1(t)\\ \equiv&\hat{U}_-(t,t_0)\sum_{n=1}^{\infty}\hat{\Psi}_{-,n}(t),\end{aligned} \tag{5.14}$$

where we defined $\hat{\sigma}_-(t)$ by

$$\hat{\sigma}_-(t) \equiv \int_{t_0}^{t} d\tau\, \hat{U}_+(t,\tau)\mathcal{P}i\hat{\mathcal{L}}_1(\tau)\mathcal{Q}\hat{u}_-(t,\tau). \quad (5.15)$$

Then, the lower order terms of the expansion are given by

$$\hat{\Psi}_{-,1}(t) = \mathcal{P}i\hat{\mathcal{L}}_1(t), \quad (5.16)$$

$$\begin{aligned}\hat{\Psi}_{-,2}(t) &= \int_{t_0}^{t} dt_1\, \mathcal{P}i\hat{\mathcal{L}}_1(t_1)\mathcal{Q}i\hat{\mathcal{L}}_1(t)\\ &= \int_{t_0}^{t} dt_1 \{\langle\langle i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle\rangle - \langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\\ &\quad \times \langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\}, \end{aligned} \quad (5.17)$$

$$\begin{aligned}\hat{\Psi}_{-,3}(t) &= \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \{\mathcal{P}i\hat{\mathcal{L}}_1(t_2)\mathcal{Q}i\hat{\mathcal{L}}_1(t_1)\mathcal{Q}i\hat{\mathcal{L}}_1(t)\\ &\quad - \mathcal{P}i\hat{\mathcal{L}}_1(t_1)\mathcal{P}i\hat{\mathcal{L}}_1(t_2)\mathcal{Q}i\hat{\mathcal{L}}_1(t)\}\\ &= \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \{\langle\langle i\hat{\mathcal{L}}_1(t_2) i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle\rangle\\ &\quad - \langle\langle i\hat{\mathcal{L}}_1(t_2) i\hat{\mathcal{L}}_1(t_1)\rangle\rangle \langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\\ &\quad - \langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle \langle\langle i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle\rangle\\ &\quad - \langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle \langle\langle i\hat{\mathcal{L}}_1(t_2) i\hat{\mathcal{L}}_1(t)\rangle\rangle\\ &\quad + \langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle \langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle \langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\\ &\quad + \langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle \langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle \langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\}. \end{aligned} \quad (5.18)$$

Comparing these lower order terms with the ones for SP, we find reversed chronological order terms in contrast with the ''ordered cumulant'' in the SP [see Eqs. (B17)–(B19)]. Thus we introduce new cumulants called ''antiordered cumulants'' (AOC), denoted by

$$\hat{\Psi}_{-,1}(t) \equiv \langle i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{AOC}}, \quad (5.19)$$

$$\hat{\Psi}_{-,2}(t) \equiv \int_{t_0}^{t} dt_1 \langle i\hat{\mathcal{L}}_1(t) i\hat{\mathcal{L}}_1(t_1)\rangle_{\mathrm{AOC}}, \quad (5.20)$$

$$\begin{aligned}\hat{\Psi}_{-,n}(t) &\equiv \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \cdots \int_{t_0}^{t_{n-2}} dt_{n-1}\\ &\quad \times \langle i\hat{\mathcal{L}}_1(t_{n-1}) \cdots i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{AOC}} \quad (n \geqslant 3). \end{aligned} \quad (5.21)$$

VI. RANDOM FREQUENCY MODULATION

In this section, we apply the above method to a model of random frequency modulation (the so-called Kubo-Anderson model) [34–37].

Let us consider a system whose time evolution is determined by the Hamiltonian

$$\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}_1(t) = \hbar[\omega_0 + \omega_1(t)]a^\dagger a, \quad (6.1)$$

where $a^\dagger$ (a) is a boson creation (annihilation) operator. We assume that ω_0 in Eq. (6.1) is constant and a random (angular) frequency $\omega_1(t)$ is governed by a stationary stochastic Gaussian-Markoffian process characterized by

$$\langle \omega_1(t)\rangle_B = 0, \quad (6.2)$$

and

$$\langle \omega_1(t)\omega_1(t_1)\rangle_B = \Delta_0^2 e^{-|t-t_1|/\tau_c}, \quad (6.3)$$

where the amplitude of modulation is denoted by Δ_0, τ_c being the characteristic time of the process. In Eqs. (6.2) and (6.3), we used a symbol $\langle \dots \rangle_B$ as an averaging procedure over the stochastic process of $\omega_1(t)$. This model describes time evolution of a boson field (for instance, a single mode of quantized electromagnetic field) under a random perturbation from its environment. When we replace $a^\dagger a$ by a spin operator S_z, this model describes a spin relaxation process under an adiabatic perturbation.

In order to analyze time evolution of the system, we want to obtain a differential equation of an average of annihilation operator $\hat{a}(t)$ in terms of the TC and TCL decompositions.

(i) TC decomposition. With the use of Eqs. (5.1) and (6.1), we find the following equation:

$$\frac{d}{dt}\hat{a}(t) = -i\langle \omega_1(t)\rangle_B \hat{a}(t) + \sum_{n=2}^{\infty} \hat{\Xi}_{-,n}(t) + \sum_{n=1}^{\infty} \hat{\mathcal{J}}_{-,n}(t). \quad (6.4)$$

The lower order terms of $\hat{\Xi}_{-,n}$ and $\hat{\mathcal{J}}_{-,n}$ are explicitly given by

$$\begin{aligned}\hat{\Xi}_{-,2}(t) &= \int_{t_0}^{t} dt_1 \hat{U}_-(t_1,t_0)\langle i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\, a\\ &= (-i)^2 \int_{t_0}^{t} dt_1 \{\langle \omega_1(t)\omega_1(t_1)\rangle_B\\ &\quad - \langle \omega_1(t)\rangle_B \langle \omega_1(t_1)\rangle_B\}\hat{a}(t_1), \end{aligned} \quad (6.5)$$

$$\begin{aligned}\hat{\hat{\Xi}}_{-,3}(t)=&\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\,\hat{U}_-(t_2,t_0)\\&\times\langle i\hat{\mathcal{L}}_1(t_2)i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\,a\\=&(-i)^3\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\{\langle\omega_1(t)\omega_1(t_1)\omega_1(t_2)\rangle_B\\&-\langle\omega_1(t)\rangle_B\langle\omega_1(t_1)\omega_1(t_2)\rangle_B\\&-\langle\omega_1(t)\omega_1(t_1)\rangle_B\langle\omega_1(t_2)\rangle_B\\&+\langle\omega_1(t)\rangle_B\langle\omega_1(t_1)\rangle_B\langle\omega_1(t_2)\rangle_B\}\hat{a}(t_2),\end{aligned}\tag{6.6}$$

$$\hat{\mathcal{J}}_{-,1}(t)=\mathcal{Q}i\hat{\mathcal{L}}_1(t)a=(-i)[\omega_1(t)-\langle\omega_1(t)\rangle_B]a,\tag{6.7}$$

$$\begin{aligned}\hat{\mathcal{J}}_{-,2}(t)=&\int_{t_0}^{t}dt_1\mathcal{Q}\hat{\Phi}_{-,2}(t_1,t)a=(-i)^2\int_{t_0}^{t}dt_1\{\omega_1(t)\omega_1(t_1)\\&-\langle\omega_1(t)\omega_1(t_1)\rangle_B-\langle\omega_1(t)\rangle_B\omega_1(t_1)\\&+\langle\omega_1(t)\rangle_B\langle\omega_1(t_1)\rangle_B\}a.\end{aligned}\tag{6.8}$$

When we average Eq. (6.4) over the whole system and use the relations (6.2) and (6.3), all terms of $\hat{\mathcal{J}}_{-,n}$ and the odd terms of $\hat{\hat{\Xi}}_{-,n}$ disappear. Thus only the even terms of $\hat{\hat{\Xi}}_{-,n}$ contribute to time evolution of an average of the annihilation operator $\langle\hat{a}(t)\rangle$:

$$\begin{aligned}\frac{d}{dt}\langle\hat{a}(t)\rangle=&(-i)^2\Delta_0^2\int_{t_0}^{t}dt_1\,\xi_1(t-t_1)\langle\hat{a}(t_1)\rangle+(-i)^4\Delta_0^4\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_2}dt_3\,2\xi_1(t-t_1)\xi_2(t_1-t_2)\xi_1(t_2-t_3)\langle\hat{a}(t_3)\rangle\\&+(-i)^6\Delta_0^6\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_2}dt_3\int_{t_0}^{t_3}dt_4\int_{t_0}^{t_4}dt_5\{6\xi_1(t-t_1)\xi_2(t_1-t_2)\xi_3(t_2-t_3)\xi_2(t_3-t_4)\xi_1(t_4-t_5)\\&+4\xi_1(t-t_1)\xi_2(t_1-t_2)\xi_1(t_2-t_3)\xi_2(t_3-t_4)\xi_1(t_4-t_5)\}\langle\hat{a}(t_5)\rangle+\cdots,\end{aligned}\tag{6.9}$$

where we defined $\xi_n(t)$ by

$$\xi_n(t)\equiv e^{-nt/\tau_c}.\tag{6.10}$$

Since we find a convolution type of integrals, Laplace transform of Eq. (6.9) gives us a series of successive algebraic equations. That is, defining the Laplace transform of a function $f(t)$ by

$$f[s]=\int_0^{\infty}dt\,f(t)e^{-st},\tag{6.11}$$

we can solve Eq. (6.9) as follows:

$$\langle\hat{a}[s]\rangle=\frac{\langle\hat{a}(0)\rangle}{s-\Sigma_1[s]},\tag{6.12}$$

where we set $t_0=0$ and $\Sigma_1[s]$ is written successively by the relation

$$\Sigma_n[s]\equiv\frac{n(-i\Delta_0)^2}{\xi_n^{-1}[s]-\Sigma_{n+1}[s]},\tag{6.13}$$

with

$$\xi_n[s]\equiv\frac{1}{s-n/\tau_c}.\tag{6.14}$$

Namely, $\langle\hat{a}[s]\rangle$ is solved in terms of the form of a continued fraction, which agrees with the known result [37].

(ii) TCL decomposition. When we apply the expansion formula (5.11) of TCL decomposition to the model of random frequency modulation, we have

$$\frac{d}{dt}\hat{a}(t)=-i\langle\omega_1(t)\rangle_B\,\hat{a}(t)+\sum_{n=2}^{\infty}\hat{\Gamma}_{-,n}(t)+\sum_{n=1}^{\infty}\hat{J}_{-,n}(t).\tag{6.15}$$

The lower order terms of $\hat{\Gamma}_{-,n}$ and $\hat{J}_{-,n}$ are explicitly given by

$$\begin{aligned}\hat{\Gamma}_{-,2}(t)=&\hat{U}_-(t,t_0)\int_{t_0}^{t}dt_1\langle i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{AOC}}\,a\\=&(-i)^2\int_{t_0}^{t}dt_1\{\langle\omega_1(t)\omega_1(t_1)\rangle_B\\&-\langle\omega_1(t)\rangle_B\langle\omega_1(t_1)\rangle_B\}\hat{a}(t),\end{aligned}\tag{6.16}$$

$$\begin{aligned}\hat{\Gamma}_{-,3}(t)=&\hat{U}_-(t,t_0)\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\\&\times\langle i\hat{\mathcal{L}}_1(t_2)i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{AOC}}\,a\\=&(-i)^3\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\{\langle\omega_1(t)\omega_1(t_1)\omega_1(t_2)\rangle_B\\&-\langle\omega_1(t)\rangle_B\langle\omega_1(t_1)\omega_1(t_2)\rangle_B\\&-\langle\omega_1(t)\omega_1(t_1)\rangle_B\langle\omega_1(t_2)\rangle_B-\langle\omega_1(t)\omega_1(t_2)\rangle\\&\times\langle\omega_1(t_1)\rangle+\langle\omega_1(t)\rangle_B\langle\omega_1(t_1)\rangle_B\langle\omega_1(t_2)\rangle_B\\&+\langle\omega_1(t)\rangle_B\langle\omega_1(t_2)\rangle_B\langle\omega_1(t_1)\rangle_B\}\hat{a}(t),\end{aligned}\tag{6.17}$$

$$\hat{J}_{-,1}(t)=\mathcal{Q}i\hat{\mathcal{L}}_1(t)a=(-i)[\omega_1(t)-\langle\omega_1(t)\rangle_B]a,\tag{6.18}$$

$$\hat{J}_{-,2}(t) = \int_{t_0}^{t} dt_1 \mathcal{Q}\hat{\Phi}_{-,2}(t_1,t)a$$
$$= (-i)^2 \int_{t_0}^{t} dt_1 \{\omega_1(t)\omega_1(t_1) - \langle \omega_1(t)\omega_1(t_1)\rangle_B$$
$$-\langle \omega_1(t)\rangle_B \omega_1(t_1) + \langle \omega_1(t)\rangle_B \langle \omega_1(t_1)\rangle_B\} a. \quad (6.19)$$

Averaging Eq. (6.15) over the whole system, and using Eqs. (6.2) and (6.3), we find

$$\frac{d}{dt}\langle \hat{a}(t)\rangle = -\int_{t_0}^{t} dt_1 \langle \omega_1(t)\omega_1(t_1)\rangle_B \langle \hat{a}(t)\rangle, \quad (6.20)$$

since the relevant cumulants higher than the third order and $\hat{J}_-$ disappear [24]. Thus Eq. (6.20) is solved to give ($t_0 = 0$)

$$\langle \hat{a}(t)\rangle = e^{-\Delta_0^2 \int_0^t dt_1 \int_0^{t_1} dt_2\, \xi_1(t_1 - t_2)} \langle \hat{a}\rangle = e^{-\alpha^2 (t/\tau_c - 1 + e^{-t/\tau_c})} \langle \hat{a}\rangle, \quad (6.21)$$

which is again an exact result [37]. In Eq. (6.21), we defined α by

$$\alpha = \Delta_0 \tau_c . \quad (6.22)$$

In this model, the TC and TCL decomposition mutually play a complementary role. Namely, the TC decomposition gives the solution in the frequency domain for $s \equiv i\omega$, while the TCL decomposition gives the solution in time domain. We can flexibly choose the desired solution depending on our purpose.

VII. LOW FIELD RESONANCE IN QUANTUM ENVIRONMENT

Next, we apply our formalism to a spin relaxation phenomenon caused by a nonadiabatic interaction with a quantum environment. Our whole system is specified by the following Hamiltonian:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 , \quad (7.1)$$

where

$$\mathcal{H}_0 = \hbar \omega_0 S_z + \mathcal{H}_B , \quad (7.2)$$

$$\mathcal{H}_1 = \hbar(\omega_- S_+ + \omega_+ S_-). \quad (7.3)$$

In Eqs. (7.2) and (7.3), $\mathcal{H}_B$ and $\omega_\pm$ denote the Hamiltonian and the reservoir operators, respectively. We assume that the reservoir is composed of a collection of harmonic oscillators where $\mathcal{H}_B$ and $\omega_\pm$ are of the form

$$\mathcal{H}_B = \sum_j \hbar \omega_j b_j^\dagger b_j , \quad (7.4)$$

$$\omega_- \equiv \sum_j \kappa_j b_j \equiv (\omega_+)^\dagger . \quad (7.5)$$

In these expressions, b_j ($b_j^\dagger$) is an annihilation (creation) operator of the jth oscillator and κ_j the coupling constant between the relevant spin and the jth oscillator.

Now, we proceed to determine time evolution of a transverse component of the spin, $S_+ \equiv S_x + iS_y$, in order to analyze the relaxation process. From Eqs. (2.10) and (2.16), we have

$$S_+(t) = U_-(t,t_0)S_+ = \hat{U}_-(t,t_0)e^{i\mathcal{L}_0(t-t_0)}S_+$$
$$= e^{i\omega_0(t-t_0)}\hat{U}_-(t,t_0)S_+ \equiv e^{i\omega_0(t-t_0)}\hat{S}_+(t). \quad (7.6)$$

Then we obtain time evolution of $\hat{S}_+(t)$ by using the TC decomposition in the HP, Eq. (5.1):

$$\frac{d}{dt}\hat{S}_+(t) = \hat{U}_-(t,t_0)\mathcal{P}i\hat{\mathcal{L}}_1(t)S_+ + \int_{t_0}^{t} d\tau \hat{K}_-(t,\tau)S_+ + \hat{\mathcal{J}}_-(t). \quad (7.7)$$

According to Eq. (2.8), the operator $\hat{\mathcal{L}}_1(t)$ in Eq. (7.7) is explicitly given by ($t_0 \equiv 0$)

$$\hat{\mathcal{L}}_1(t)\cdot = e^{i\mathcal{L}_0 t}\mathcal{L}_1(t)e^{-i\mathcal{L}_0 t}\cdot = \frac{1}{\hbar}[e^{(i/\hbar)\mathcal{H}_0 t}\mathcal{H}_1 e^{-(i/\hbar)\mathcal{H}_0 t},\cdot]$$
$$\equiv [\tilde{\omega}_-(t)S_+ + \tilde{\omega}_+(t)S_-,\cdot], \quad (7.8)$$

where

$$\tilde{\omega}_+(t) = \sum_j e^{-i(\omega_0 - \omega_j)t}\kappa_j^* b_j^\dagger = \tilde{\omega}_-(t)^\dagger . \quad (7.9)$$

From Eq. (C9) in Appendix C, the projection operator $\mathcal{P}$ is given by

$$\mathcal{P}\cdot = \mathrm{tr}_B\, \rho_B \cdot = \frac{1}{Z_B}\mathrm{tr}_B\, e^{-\beta\mathcal{H}_B}\cdot \equiv \langle \cdot \rangle_B , \quad (7.10)$$

with a partition function Z_B and

$$\beta \equiv \frac{1}{k_B T}, \quad (7.11)$$

where k_B is the Boltzmann constant and T is the temperature of the reservoir.

Our next task is to calculate an expansion series of Eq. (7.7). With the use of Eq. (5.4),

$$\int_0^t d\tau \hat{K}_-(t,\tau)S_+$$
$$= \int_0^t dt_1 \hat{U}_-(t_1,t_0)\langle i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\, S_+$$
$$+ \sum_{n=3}^{\infty} \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{n-1}} dt_n\, \hat{U}_-(t_{n-1},t_0)$$
$$\times \langle i\hat{\mathcal{L}}_1(t_n)i\hat{\mathcal{L}}_1(t_{n-1})\cdots i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\, S_+ , \quad (7.12)$$

each expansion term is obtained by evaluating the ''antipartial cumulants.''

Explicitly, we find the lower order terms as follows:

$$
\begin{aligned}
&\langle i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\, S_+ \\
&\quad = [\langle i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle_B - \langle i\hat{\mathcal{L}}_1(t_1)\rangle_B \langle i\hat{\mathcal{L}}_1(t)\rangle_B] S_+ \\
&\quad = -[\langle \tilde{\omega}_+(t)\tilde{\omega}_-(t_1)\rangle_B + \langle \tilde{\omega}_-(t_1)\tilde{\omega}_+(t)\rangle_B \\
&\qquad -2\langle \tilde{\omega}_+(t)\rangle_B \langle \tilde{\omega}_-(t_1)\rangle_B] S_+ + [\langle \tilde{\omega}_+(t)\tilde{\omega}_+(t_1)\rangle_B \\
&\qquad + \langle \tilde{\omega}_+(t_1)\tilde{\omega}_+(t)\rangle_B - 2\langle \tilde{\omega}_+(t)\rangle_B \langle \tilde{\omega}_+(t_1)\rangle_B] S_- \\
&\quad = -[\langle \tilde{\omega}_+(t)\tilde{\omega}_-(t_1)\rangle_B + \langle \tilde{\omega}_-(t_1)\tilde{\omega}_+(t)\rangle_B] S_+ . \qquad (7.13)
\end{aligned}
$$

In the last line of Eq. (7.13), we used Eqs. (D2) and (D10).

As is seen from Appendix D, the odd-order moments disappear for the harmonic oscillator bath. Therefore, the odd-order "antipartial cumulants" also disappear:

$$
\langle i\hat{\mathcal{L}}_1(t_{2n})\cdots i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\, S_+ = 0 \quad (n \geqslant 1). \qquad (7.14)
$$

Then, the next order cumulant contributing to the time evolution of S_+ is of the form

$$
\begin{aligned}
\langle i\hat{\mathcal{L}}_1(t_3) i\hat{\mathcal{L}}_1(t_2) i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{APC}}\, S_+ &= [\langle i\hat{\mathcal{L}}_1(t_3) i\hat{\mathcal{L}}_1(t_2) i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle - \langle i\hat{\mathcal{L}}_1(t_3) i\hat{\mathcal{L}}_1(t_2)\rangle \langle i\hat{\mathcal{L}}_1(t_1) i\hat{\mathcal{L}}_1(t)\rangle] S_+ \\
&= 2\{\langle \tilde{\omega}_-(t_3)\tilde{\omega}_+(t_1)\rangle_B \langle \tilde{\omega}_+(t)\tilde{\omega}_-(t_2)\rangle_B + \langle \tilde{\omega}_-(t_2)\tilde{\omega}_+(t)\rangle_B \langle \tilde{\omega}_+(t_1)\tilde{\omega}_-(t_3)\rangle_B \\
&\quad + \langle \tilde{\omega}_-(t_2)\tilde{\omega}_+(t_1)\rangle_B \langle \tilde{\omega}_+(t)\tilde{\omega}_-(t_3)\rangle_B + \langle \tilde{\omega}_-(t_1)\tilde{\omega}_+(t_2)\rangle_B \langle \tilde{\omega}_+(t)\tilde{\omega}_-(t_3)\rangle_B \\
&\quad + \langle \tilde{\omega}_-(t_3)\tilde{\omega}_+(t)\rangle_B \langle \tilde{\omega}_+(t_1)\tilde{\omega}_-(t_2)\rangle_B + \langle \tilde{\omega}_-(t_3)\tilde{\omega}_+(t)\rangle_B \langle \tilde{\omega}_+(t_2)\tilde{\omega}_-(t_1)\rangle_B\} S_+ .
\end{aligned} \tag{7.15}
$$

Thus, with the use of Eqs. (D19) and (D20), time evolution of $\langle \hat{S}_+(t)\rangle$ is given by

$$
\begin{aligned}
\frac{d}{dt}\langle \hat{S}_+(t)\rangle =& -\Delta^2 \int_0^t dt_1\, e^{-\gamma(t-t_1)-i(t-t_1)(\omega_o-\omega_b)}[1+2n(\omega_b)]\langle \hat{S}_+(t_1)\rangle + \Delta^4 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3\, e^{-\gamma(t+t_1-t_2-t_3)} \\
&\times (8e^{-i(t+t_1-t_2-t_3)(\omega_o-\omega_b)} + 4e^{-i(t-t_1+t_2-t_3)(\omega_o-\omega_b)}) n(\omega_b)[1+n(\omega_b)]\langle \hat{S}_+(t_3)\rangle \\
&-\Delta^6 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 \int_0^{t_4} dt_5 \{e^{-\gamma(t+t_1-t_2+t_3-t_4-t_5)}(16e^{-i(t+t_1-t_2+t_3-t_4-t_5)(\omega_o-\omega_b)} \\
&+8e^{-i(t-t_1+t_2+t_3-t_4-t_5)(\omega_o-\omega_b)} + 8e^{-i(t+t_1-t_2-t_3+t_4-t_5)(\omega_o-\omega_b)} + 4e^{-i(t-t_1+t_2-t_3+t_4-t_5)(\omega_o-\omega_b)}) \\
&+e^{-\gamma(t+t_1+t_2-t_3-t_4-t_5)}(8e^{-i(t+t_1-t_2+t_3-t_4-t_5)(\omega_o-\omega_b)} + 8e^{-i(t-t_1+t_2+t_3-t_4-t_5)\omega_o-\omega_b}) \\
&+8e^{-i(t+t_1-t_2-t_3+t_4-t_5)(\omega_o-\omega_b)} + 8e^{-i(t-t_1+t_2-t_3+t_4-t_5)(\omega_o-\omega_b)}) n(\omega_b)[1+n(\omega_b)][1+2n(\omega_b)]\langle \hat{S}_+(t_5)\rangle + \cdots .
\end{aligned} \tag{7.16}
$$

In the same way as in Sec. VI, Eq. (7.16) is written in the form of the convolution type of integrals:

$$
\begin{aligned}
\frac{d}{dt}\langle \hat{S}_+(t)\rangle =& -\Delta^2 \int_0^t dt_1\, \xi_1(t-t_1)\, \eta_1(t-t_1)[1+2n(\omega_b)]\langle \hat{S}_+(t_1)\rangle + \Delta^4 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3\, \xi_1(t-t_1)\xi_2(t_1-t_2)\xi_1(t_2-t_3) \\
&\times [8\,\eta_1(t-t_1)\,\eta_2(t_1-t_2)\,\eta_1(t_2-t_3) + 4\,\eta_1(t-t_1)\,\eta_1(t_2-t_3)] n(\omega_b)[1+n(\omega_b)]\langle \hat{S}_+(t_3)\rangle \\
&-\Delta^6 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 \int_0^{t_4} dt_5 \{\xi_1(t-t_1)\xi_2(t_1-t_2)\xi_1(t_2-t_3)\xi_2(t_3-t_4)\xi_1(t_4-t_5) \\
&\times [16\,\eta_1(t-t_1)\,\eta_2(t_1-t_2)\,\eta_1(t_2-t_3)\,\eta_2(t_3-t_4)\,\eta_1(t_4-t_5) + 8\,\eta_1(t-t_1)\,\eta_1(t_2-t_3)\,\eta_2(t_3-t_4)\,\eta_1(t_4-t_5) \\
&+8\,\eta_1(t-t_1)\,\eta_2(t_1-t_2)\,\eta_1(t_2-t_3)\,\eta_1(t_4-t_5) + 4\,\eta_1(t-t_1)\,\eta_1(t_2-t_3)\,\eta_1(t_4-t_5)] + \xi_1(t-t_1)\xi_2(t_1-t_2) \\
&\times \xi_3(t_2-t_3)\xi_2(t_3-t_4)\xi_1(t_4-t_5)[8\,\eta_1(t-t_1)\,\eta_2(t_1-t_2)\,\eta_1(t_2-t_3)\,\eta_2(t_3-t_4)\,\eta_1(t_4-t_5) + 8\,\eta_1(t-t_1) \\
&\times \eta_1(t_2-t_3)\,\eta_2(t_3-t_4)\,\eta_1(t_4-t_5) + 8\,\eta_1(t-t_1)\,\eta_2(t_2-t_3)\,\eta_1(t_3-t_4)\,\eta_1(t_4-t_5) + 8\,\eta_1(t-t_1)\,\eta_1(t_2-t_3) \\
&\times \eta_1(t_4-t_5)]\} n(\omega_b)[1+n(\omega_b)][1+2n(\omega_b)]\langle \hat{S}_+(t_5)\rangle + \cdots ,
\end{aligned} \tag{7.17}
$$

where we have defined

$$\xi_n(t) \equiv e^{-n\gamma t}, \tag{7.18}$$

and

$$\eta_n(t) \equiv e^{-ni(\omega_o-\omega_b)t}. \tag{7.19}$$

In this way, we can successively determine the time-dependent coefficients of $\langle \hat{S}_+(t)\rangle$. With the Laplace transform of Eq. (7.17), we found the absorption spectrum in a form similar to that of the continued fraction shown in the preceding section. We plan to publish the details soon. We are satisfied here only with the result (7.17) as an example of actual calculations showing the manipulating procedure for quantum systems.

VIII. DISCUSSION AND CONCLUDING REMARKS

In this paper, we formulated a unified and general method of projection operator for the dynamical variables. Without specifying the projection operator, a systematic formalism is established in order to obtain basic equations such as TC and TCL decompositions. The formalism enables us to have flexible treatment of problems including a time-dependent Liouville operator. Expansion formulas for both decompositions are also obtained. These characteristic features of the formalism are parallel to the one for the Liouville–von Neumann equation. Moreover, we applied the formulas to the problems of random frequency modulation and low field resonance.

Now, we examine existing theories [27–30] from our viewpoint. When the Liouville operator $\mathcal{L}$ is time independent, time evolution of an operator A is determined by $e^{i\mathcal{L}(t-t_0)}$. We have thus

$$\frac{\partial}{\partial t} e^{i\mathcal{L}(t-t_0)} = e^{i\mathcal{L}(t-t_0)} i\mathcal{L}, \tag{8.1}$$

which corresponds to Eq. (3.1). Therefore, the main results of Sec. III and Appendix A are transformed as follows:

$$\left.\begin{array}{r} T_+[e^{-\int_{t_0}^t dt' i\hat{\mathcal{L}}_1(t')}] \\ T_-[e^{\int_{t_0}^t dt' i\hat{\mathcal{L}}_1(t')}] \\ T_+[e^{-\int_{t_0}^t dt' \overline{\mathcal{Q}} i\hat{\mathcal{L}}_1(t')}] \\ T_-[e^{\int_{t_0}^t dt' i\hat{\mathcal{L}}_1(t')\mathcal{Q}}] \end{array}\right\} \rightarrow \left\{\begin{array}{l} e^{-i\mathcal{L}(t-t_0)} \\ e^{i\mathcal{L}(t-t_0)} \\ e^{-\mathcal{Q}i\mathcal{L}(t-t_0)} \cdot \\ e^{i\mathcal{L}\mathcal{Q}(t-t_0)} \end{array}\right. \tag{8.2}$$

Further, if we specify the projection operator $\mathcal{P}$ as

$$\mathcal{P}X = (X,A^\dagger)(A,A^\dagger)^{-1}A, \tag{8.3}$$

in Eq. (3.10), we obtain the TC equation in [27]. In Eq. (8.3), the symbol (X,Y) is often defined by a canonical average:

$$(X,Y) \equiv \frac{1}{\beta}\int_0^\beta d\lambda \langle e^{\lambda\mathcal{H}} X e^{-\lambda\mathcal{H}} Y\rangle. \tag{8.4}$$

As a result, an expansion formula for Mori's TC equation is also obtained by specifying the projection operator $\mathcal{P}$ as Eq. (8.3) in Eq. (5.1) with the replacement (8.2). Moreover, when the application of the replacement (8.2) to Eq. (3.16) is done, we have the TCL type equation derived by operator manipulation in [28].

There is another work where the TC and/or TCL equation for a dynamical variable $A(t)$ is obtained by defining projection operators,

$$\mathcal{P}\cdot = \sum_{k,l} A_k g_{kl}\, \mathrm{Tr}\{B_l \cdot\} \tag{8.5}$$

and

$$\tilde{\mathcal{P}}\cdot = \sum_{k,l} B_k \tilde{g}_{kl}\, \mathrm{Tr}\{A_l \cdot\}, \tag{8.6}$$

for the HP and the SP, respectively [30,38]. In Eqs. (8.5) and (8.6), A and B are arbitrary operators. Keeping in mind that both the basic equations for the HP and the SP should give the same mean value of an operator A, they inferred the basic equations for the HP from the TC and TCL equations in the SP with the use of the ''dual'' relations

$$\begin{aligned} \mathrm{Tr}\{X(\mathcal{P}Y)\} &= \mathrm{Tr}\{(\tilde{\mathcal{P}}X)Y\}, \\ \mathrm{Tr}\{X(\tilde{\mathcal{P}}Y)\} &= \mathrm{Tr}\{(\mathcal{P}X)Y\}. \end{aligned} \tag{8.7}$$

Especially in [30], from our point of view, a sort of transcription rule was suggested for the system of a time-independent Hamiltonian. Such a transcription rule coincides with ours when the replacements (8.2) are made to the rules in Sec. IV.

Another type of TCL equation was also proposed [39,40]:

$$\begin{aligned} \tilde{\mathcal{P}}\dot{W}(t) = &-\tilde{\mathcal{P}}i\mathcal{L}\tilde{\mathcal{P}}W(t) - \tilde{\mathcal{P}}i\mathcal{L}\{e^{-i\mathcal{L}t}n_+(t)^{-1}-1\}\tilde{\mathcal{P}}W(t) \\ &-\tilde{\mathcal{P}}i\mathcal{L}e^{-i\mathcal{L}t}n_+(t)^{-1}\tilde{\mathcal{Q}}W(t_0), \end{aligned} \tag{8.8}$$

where

$$n_+(t) = \tilde{\mathcal{Q}} + \tilde{\mathcal{P}}e^{-i\mathcal{L}t} \tag{8.9}$$

$$= 1 + \tilde{\mathcal{P}}(e^{-i\mathcal{L}t}-1). \tag{8.10}$$

The TCL equation for a time-independent Liouville operator, which is obtained by applying the replacement (8.2) to Eq. (A16), is proved to be equivalent to Eq. (8.8) in [40].

With the use of the rule of replacement (8.2) and rules in Sec. IV, we can extend the TCL equation (8.8).

(1) Application (in the reversed direction) of Eq. (8.2) to Eq. (8.8) enables us to find an equation for the time-dependent Liouville operator:

$$\begin{aligned} \tilde{\mathcal{P}}\dot{W}(t) = &-\tilde{\mathcal{P}}i\mathcal{L}(t)\tilde{\mathcal{P}}W(t) \\ &-\tilde{\mathcal{P}}i\mathcal{L}(t)\{U_+(t,t_0)N_+(t)^{-1}-1\}\tilde{\mathcal{P}}W(t) \\ &-\tilde{\mathcal{P}}i\mathcal{L}(t)U_+(t,t_0)N_+(t)^{-1}\tilde{\mathcal{Q}}W(t_0), \end{aligned} \tag{8.11}$$

where

$$N_+(t) = \tilde{\mathcal{Q}} + \tilde{\mathcal{P}}U_+(t,t_0) \tag{8.12}$$

$$=1+\tilde{\mathcal{P}}(U_+(t,t_0)-1). \tag{8.13}$$

(2) With the aid of the rules in Sec. IV and replacement (8.2), we immediately obtain an equation for a dynamical variable $A(t)$:

$$\dot{A}(t)=e^{i\mathcal{L}t}\mathcal{P}i\mathcal{L}A+e^{i\mathcal{L}t}\mathcal{P}\{n_-(t)^{-1}e^{i\mathcal{L}t}-1\}i\mathcal{L}A+\mathcal{Q}n_-(t)^{-1}e^{i\mathcal{L}t}i\mathcal{L}A, \tag{8.14}$$

where

$$n_-(t)=\mathcal{Q}+e^{i\mathcal{L}t}\mathcal{P} \tag{8.15}$$

$$=1+(e^{i\mathcal{L}t}-1)\mathcal{P}. \tag{8.16}$$

(3) Further use of the rules in Sec. IV gives the following equation for $A(t)$:

$$\dot{A}(t)=U_-(t,t_0)\mathcal{P}i\mathcal{L}(t)A+U_-(t,t_0)\mathcal{P}\times\{N_-(t)^{-1}U_-(t,t_0)-1\}i\mathcal{L}(t)A+\mathcal{Q}N_-(t)^{-1}U_-(t,t_0)i\mathcal{L}(t)A, \tag{8.17}$$

where

$$N_-(t)=\mathcal{Q}+U_-(t,t_0)\mathcal{P} \tag{8.18}$$

$$=1+[U_-(t,t_0)-1]\mathcal{P}. \tag{8.19}$$

Thus the transcription rules are quite powerful in obtaining other types of equations when a single equation is known.

Moreover, the formalism developed in this paper should not be restricted within the rigid framework of the Liouville–von Neumann equation and the Heisenberg equation of motion. In other words, we may use the derived basic TC and TCL decompositions flexibly. For instance, let us consider an operator defined by

$$g(t)=\delta(a-A(t))=e^{i\mathcal{L}t}\delta(a-A), \tag{8.20}$$

where a is one of the realization values of a system operator A. The time evolution operator of $g(t)$ obeys Eq. (8.1) and therefore we find

$$\dot{g}(t)=e^{i\mathcal{L}t}\mathcal{P}i\mathcal{L}\delta(a-A)+\int_{t_0}^{t}d\tau\, e^{i\mathcal{L}\tau}\mathcal{P}i\mathcal{L}\mathcal{Q}e^{i\mathcal{L}\mathcal{Q}(t-\tau)}i\mathcal{L}\times\delta(a-A)+\mathcal{F}(t), \tag{8.21}$$

where

$$\mathcal{F}(t)=\mathcal{Q}e^{i\mathcal{L}\mathcal{Q}t}i\mathcal{L}\delta(a-A). \tag{8.22}$$

The quantity $\mathcal{F}(t)$ is regarded as a ''fluctuating force'' for the probability distribution operator $g(t)$. Equation (8.21) is a microscopic version of the Boltzmann-Langevin equation [41,42].

Even for the Schrödinger equation

$$|\dot{\psi}(t)\rangle=-\frac{i}{\hbar}\mathcal{H}|\psi(t)\rangle, \tag{8.23}$$

we can use the method of projection operator for the HP: The formal solution of Eq. (8.23) is obtained as

$$|\psi(t)\rangle=e^{-(i/\hbar)\mathcal{H}(t-t_0)}|\psi(t_0)\rangle \tag{8.24}$$

to give the relation

$$\frac{\partial}{\partial t}e^{-(i/\hbar)\mathcal{H}(t-t_0)}=e^{-(i/\hbar)\mathcal{H}(t-t_0)}\left(-\frac{i}{\hbar}\mathcal{H}\right), \tag{8.25}$$

which is analogous to Eq. (8.1). When we define the projection operator by

$$\mathcal{P}=\sum_{\lambda}|\lambda\rangle\langle\lambda|, \tag{8.26}$$

we immediately find the following wave equation with a ''fluctuating force'' ($t_0\equiv 0$):

$$|\dot{\psi}_\lambda(t)\rangle=-\frac{i}{\hbar}\sum_{\lambda'}|\psi_{\lambda'}(t)\rangle\mathcal{H}_{\lambda',\lambda}-\frac{1}{\hbar^2}\sum_{\lambda'}\int_0^t d\tau|\psi_{\lambda'}(\tau)\rangle\langle\mathcal{Q}f_{\lambda'}(0)|\mathcal{Q}f_\lambda(t-\tau)\rangle+\frac{1}{\hbar}|\mathcal{Q}f_\lambda(t)\rangle, \tag{8.27}$$

where

$$|\mathcal{Q}f_\lambda(t)\rangle=-ie^{-i\mathcal{Q}\mathcal{H}\mathcal{Q}t/\hbar}\mathcal{Q}\mathcal{H}|\lambda\rangle. \tag{8.28}$$

This is the equation derived in [43]. In obtaining Eq. (8.27), we used the relation of the form

$$\mathcal{Q}e^{-i\mathcal{H}\mathcal{Q}t/\hbar}=e^{-i\mathcal{Q}\mathcal{H}t/\hbar}\mathcal{Q} \tag{8.29}$$

$$=\mathcal{Q}e^{-i\mathcal{Q}\mathcal{H}\mathcal{Q}t/\hbar}\mathcal{Q}. \tag{8.30}$$

Thus, we need not restrict ourselves to strict usage of terminologies for the Schrödinger picture and the Heisenberg picture.

Next, we discuss the expansion formulas deduced from the basic equations. When we define the projection operator appropriately, the expansion formulas are written in terms of certain kinds of cumulants. Especially, the cumulants for the TCL equation in the SP (see Appendix A) coincide with the ones for stochastic equations [44–47]. They are called the ''ordered cumulants'' [45,46,23]. In this paper, using the projection operator and expansion formulas flexibly, we have given a general and unified formalism in which dynamical evolution of the observable itself is expressed by the cumulant functions. The newly obtained formulas would be of practical use in treating actual problems.

Finally, we briefly comment on the low field resonance model of Sec. VII where a quantum mechanical environment is used. In the model, a spin of magnitude 1/2 interacts nonadiabatically with the environment. When we rotate an axis of quantization in our model Hamiltonian, our system reduces to the so-called Caldeira-Leggett model [48,49]. Then, the basic equation in Sec. VII with higher order terms essentially determines time evolution of the Caldeira-Leggett model which has attracted considerable interest so far, though it is quite difficult to find a solution of the model. We plan to give details of the calculations in the future.

We hope the formalism developed in this paper will be used to solve various actual problems in related fields as well as in different fields of physics.

APPENDIX A: BASIC EQUATIONS IN THE SCHRÖDINGER PICTURE

In this appendix, we give a brief derivation of basic equations in the SP in order to find a correspondence with the ones in the HP in Sec. II. With the use of Eqs. (2.1), (2.4), and (2.6), time evolution of the operator $\hat{U}_+(t,t_0)$ is found to be

$$\frac{\partial}{\partial t}\hat{U}_+(t,t_0) = -i\hat{\mathcal{L}}_1(t)\hat{U}_+(t,t_0). \tag{A1}$$

Instead of Eqs. (3.4) and (3.5), we introduce

$$\hat{x}_+(t) = \tilde{\mathcal{P}}\hat{U}_+(t,t_0) \tag{A2}$$

and

$$\hat{y}_+(t) = \tilde{\mathcal{Q}}\hat{U}_+(t,t_0). \tag{A3}$$

Time evolution of these quantities is governed by the following equations:

$$\frac{d}{dt}\hat{x}_+(t) = \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{x}_+(t) + \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{y}_+(t) \tag{A4}$$

and

$$\frac{d}{dt}\hat{y}_+(t) = \tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(t))\hat{x}_+(t) + \tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(t))\hat{y}_+(t). \tag{A5}$$

These are derived by operating $\tilde{\mathcal{P}}$ and $\tilde{\mathcal{Q}}$ on Eq. (A1) from the left.

(i) Time-convolution formula. Equation (A5) is solved to give

$$\hat{y}_+(t) = \hat{u}_+(t,t_0)\tilde{\mathcal{Q}} + \int_{t_0}^{t} d\tau\, \hat{u}_+(t,\tau)\tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(\tau))\hat{x}_+(\tau), \tag{A6}$$

where

$$\hat{u}_+(t,\tau) = T_+\left[\exp\left(\int_{\tau}^{t} d\tau'\, \tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(\tau'))\right)\right]. \tag{A7}$$

Substitution of Eq. (A6) into Eq. (A4) gives a time-convolution type of equation

$$\begin{aligned}\frac{d}{dt}\hat{x}_+(t) &= \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{x}_+(t) \\ &\quad + \int_{t_0}^{t} d\tau\, \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{u}_+(t,\tau)\tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(\tau)) \\ &\quad \times \hat{x}_+(\tau) + \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{u}_+(t,t_0)\tilde{\mathcal{Q}}.\end{aligned} \tag{A8}$$

With the use of Eqs. (A2) and (A8), we obtain

$$\begin{aligned}\frac{d}{dt}\tilde{\mathcal{P}}\hat{W}(t) &= \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\tilde{\mathcal{P}}\hat{W}(t) \\ &\quad + \int_{t_0}^{t} d\tau\, \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{u}_+(t,\tau)\tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(\tau)) \\ &\quad \times \tilde{\mathcal{P}}\hat{W}(\tau) + \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{u}_+(t,t_0)\tilde{\mathcal{Q}}W(t_0)\end{aligned} \tag{A9}$$

for a density operator

$$\hat{W}(t) \equiv \hat{U}_+(t,t_0)\hat{W}(t_0). \tag{A10}$$

(ii) Time-convolutionless formula. Using the relation

$$\hat{x}_+(\tau) = \tilde{\mathcal{P}}\hat{U}_+(\tau,t_0) = \tilde{\mathcal{P}}\hat{U}_-(t,\tau)(\tilde{\mathcal{P}} + \tilde{\mathcal{Q}})\hat{U}_+(t,t_0), \tag{A11}$$

we have from Eq. (A6)

$$\begin{aligned}\hat{y}_+(t) &= \hat{u}_+(t,t_0)\tilde{\mathcal{Q}} + \int_{t_0}^{t} d\tau\, \hat{u}_+(t,\tau)\tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(\tau))\tilde{\mathcal{P}}\hat{U}_-(t,\tau) \\ &\quad \times \{\hat{x}_+(t) + \hat{y}_+(t)\}.\end{aligned} \tag{A12}$$

Thus we obtain a solution for $\hat{y}_+(t)$ as

$$\hat{y}_+(t) = \Theta_+(t)\{\hat{u}_+(t,t_0)\tilde{\mathcal{Q}} - [\hat{\Theta}_+(t)^{-1} - 1]\hat{x}_+(t)\}, \tag{A13}$$

where

$$\hat{\Theta}_+(t) = \left(1 - \int_{t_0}^{t} d\tau\, \hat{u}_+(t,\tau)\tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(\tau))\tilde{\mathcal{P}}\hat{U}_-(t,\tau)\right)^{-1}. \tag{A14}$$

From Eqs. (A4) and (A13), we have a time-convolutionless type of equation of the form

$$\begin{aligned}\frac{d}{dt}\hat{x}_+(t) &= \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{x}_+(t) \\ &\quad - \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\{1 - \hat{\Theta}_+(t)\}\hat{x}_+(t) \\ &\quad + \tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{\Theta}_+(t)\hat{u}_+(t,t_0)\tilde{\mathcal{Q}}.\end{aligned} \tag{A15}$$

In the same way we obtained Eq. (A9), we have

$$\frac{d}{dt}\tilde{\mathcal{P}}\hat{W}(t)=\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\tilde{\mathcal{P}}\hat{W}(t)-\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\{1-\hat{\Theta}_+(t)\}\tilde{\mathcal{P}}\tilde{W}(t)+\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{\Theta}_+(t)\hat{u}_+(t,t_0)\tilde{\mathcal{Q}}W(t_0) \tag{A16}$$

for the density operator $\hat{W}(t)$.

APPENDIX B: EXPANSION FORMULAS IN THE SCHRÖDINGER PICTURE

The basic equations obtained in Appendix A are expanded to give some kinds of cumulants in this appendix.

(i) TC formula. Expanding the TC formula, namely,

$$\frac{d}{dt}\tilde{\mathcal{P}}\hat{W}(t)=\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\tilde{\mathcal{P}}\hat{W}(t)+\int_{t_0}^{t}d\tau\hat{K}_+(t,\tau)\hat{W}(t_0)+\hat{\mathcal{J}}_+(t), \tag{B1}$$

where

$$\hat{K}_+(t,\tau)\equiv\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{u}_+(t,\tau)\tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(\tau))\tilde{\mathcal{P}}\hat{U}_+(\tau,t_0), \tag{B2}$$

with

$$\hat{\mathcal{J}}_+(t)\equiv\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{u}_+(t,t_0)\tilde{\mathcal{Q}}W(t_0), \tag{B3}$$

we obtain

$$\int_{t_0}^{t}d\tau\hat{K}_+(t,\tau)=(-1)^2\int_{t_0}^{t}dt_1\,\tilde{\mathcal{P}}\hat{\Phi}_{+,2}(t,t_1)\tilde{\mathcal{P}}\hat{U}_+(t_1,t_0)+\sum_{n=3}^{\infty}(-1)^n\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\cdots\int_{t_0}^{t_{n-2}}dt_{n-1}\times\tilde{\mathcal{P}}\hat{\Phi}_{+,n}(t,t_1,\ldots,t_{n-2},t_{n-1})\times\tilde{\mathcal{P}}\hat{U}_+(t_{n-1},t_0), \tag{B4}$$

with

$$\hat{\Phi}_{+,2}(t,t_1)=i\hat{\mathcal{L}}_1(t)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_1), \tag{B5}$$

$$\hat{\Phi}_{+,n}(t,t_1,\ldots,t_{n-2},t_{n-1})=i\hat{\mathcal{L}}_1(t)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_1)\cdots\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_{n-2})\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_{n-1})\quad(n\geqslant 3) \tag{B6}$$

and

$$\hat{\mathcal{J}}_+(t)=\left(\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))+(-1)^2\int_{t_0}^{t}dt_1\,\tilde{\mathcal{P}}\hat{\Phi}_{+,2}(t,t_1)+\sum_{n=3}^{\infty}(-1)^n\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\cdots\int_{t_0}^{t_{n-2}}dt_{n-1}\times\tilde{\mathcal{P}}\hat{\Phi}_{+,n}(t,t_1,\ldots,t_{n-2},t_{n-1})\right)\tilde{\mathcal{Q}}\hat{W}(t_0). \tag{B7}$$

For the projection operator $\tilde{\mathcal{P}}$ defined by

$$\tilde{\mathcal{P}}\cdot=\langle\langle\cdot\rangle\rangle, \tag{B8}$$

where $\langle\langle\cdot\rangle\rangle$ is a symbol to take a certain average, we obtain a kind of cumulant represented by

$$\tilde{\mathcal{P}}\hat{\Phi}_{+,n}(t,t_1,\ldots,t_{n-2},t_{n-1})\equiv\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}(t_1)\cdots i\hat{\mathcal{L}}_1(t_{n-1})\rangle_{\mathrm{PC}}\quad(n\geqslant 2). \tag{B9}$$

In Eq. (B9), the subscript PC indicates the cumulants called ‘‘partial cumulants’’ [24]. In order to show the difference of the chronological ordering structure between the SP and the HP explicitly, we write down a few lower order cumulants:

$$\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)\rangle_{\mathrm{PC}}=\tilde{\mathcal{P}}i\hat{\mathcal{L}}_1(t)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_1)=\langle\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)\rangle\rangle-\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle, \tag{B10}$$

$$\begin{aligned}\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t_2)\rangle_{\mathrm{PC}}&=\tilde{\mathcal{P}}i\hat{\mathcal{L}}_1(t)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_1)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_2)\\&=\langle\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t_2)\rangle\rangle\\&\quad-\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t_2)\rangle\rangle\\&\quad-\langle\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle\\&\quad+\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle.\end{aligned} \tag{B11}$$

(ii) TCL formula. We also expand the TCL formula:

$$\frac{d}{dt}\tilde{\mathcal{P}}\hat{W}(t)=\hat{\Psi}_+(t)\hat{W}(t_0)+\hat{J}_+(t), \tag{B12}$$

where

$$\hat{\Psi}_+(t)=\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{\Theta}_+(t)\tilde{\mathcal{P}}\hat{U}_+(t,t_0), \tag{B13}$$

$$\hat{J}_+(t)=\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\hat{\Theta}_+(t)\hat{u}_+(t,t_0)\tilde{\mathcal{Q}}W(t_0). \tag{B14}$$

Equation (B13) is expanded as

$$\hat{\Psi}_+(t)=\tilde{\mathcal{P}}(-i\hat{\mathcal{L}}_1(t))\sum_{n=0}^{\infty}[\hat{\sigma}_+(t)]^n\tilde{\mathcal{P}}\hat{U}_+(t,t_0)\equiv\sum_{n=1}^{\infty}(-1)^n\hat{\Psi}_{+,n}(t)\tilde{\mathcal{P}}\hat{U}_+(t,t_0), \tag{B15}$$

where we introduced $\hat{\sigma}_+$ as

$$\hat{\sigma}_+(t)\equiv\int_{t_0}^{t}d\tau\,\hat{u}_+(t,\tau)\tilde{\mathcal{Q}}(-i\hat{\mathcal{L}}_1(\tau))\tilde{\mathcal{P}}\hat{U}_-(t,\tau). \tag{B16}$$

The lower order terms of the expansion are explicitly given by

$$\hat{\Psi}_{+,1}(t)=\tilde{\mathcal{P}}i\hat{\mathcal{L}}_1(t), \tag{B17}$$

$$\hat{\Psi}_{+,2}(t)=\int_{t_0}^{t}dt_1\,\tilde{\mathcal{P}}i\hat{\mathcal{L}}_1(t)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_1)=\int_{t_0}^{t}dt_1\{\langle\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)\rangle\rangle-\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\}, \tag{B18}$$

$$\begin{aligned}\hat{\Psi}_{+,3}(t)&=\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\{\tilde{\mathcal{P}}i\hat{\mathcal{L}}_1(t)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_1)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_2)-\tilde{\mathcal{P}}i\hat{\mathcal{L}}_1(t)\tilde{\mathcal{Q}}i\hat{\mathcal{L}}_1(t_2)\tilde{\mathcal{P}}i\hat{\mathcal{L}}_1(t_1)\}\\&=\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\{\langle\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t_2)\rangle\rangle-\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)i\hat{\mathcal{L}}_1(t_2)\rangle\rangle-\langle\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle\\&\quad-\langle\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_2)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle+\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle+\langle\langle i\hat{\mathcal{L}}_1(t)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_2)\rangle\rangle\langle\langle i\hat{\mathcal{L}}_1(t_1)\rangle\rangle\}.\end{aligned} \tag{B19}$$

These cumulants are called ''ordered cumulants'' (OC) [45,46,23,24] and are denoted by

$$\hat{\Psi}_{+,1}(t)\equiv\langle i\hat{\mathcal{L}}_1(t)\rangle_{\mathrm{OC}},$$

$$\hat{\Psi}_{+,2}(t)\equiv\int_{t_0}^{t}dt_1\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)\rangle_{\mathrm{OC}}, \tag{B20}$$

$$\hat{\Psi}_{+,n}(t)\equiv\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\cdots\int_{t_0}^{t_{n-2}}dt_{n-1}\times\langle i\hat{\mathcal{L}}_1(t)i\hat{\mathcal{L}}_1(t_1)\cdots i\hat{\mathcal{L}}_1(t_{n-1})\rangle_{\mathrm{OC}}\quad(n\geqslant 3) \tag{B21}$$

APPENDIX C: A RELATION BETWEEN PROJECTION OPERATORS

Our formalism developed in this paper is free from a choice of projection operators. However, it is natural to impose a requirement that $\mathcal{P}$ and $\tilde{\mathcal{P}}$ give the same result for averaged quantities. That is, we require a certain relation between the projection operators $\mathcal{P}$ and $\tilde{\mathcal{P}}$ so as to yield the same averaged value. With the use of such a relation, we can determine the projection operator $\mathcal{P}$ consistent with $\tilde{\mathcal{P}}$ and vice versa keeping the idempotent relations

$$\mathcal{P}^2=\mathcal{P}, \tag{C1}$$

and

$$\tilde{\mathcal{P}}^2=\tilde{\mathcal{P}}. \tag{C2}$$

For a system operator A, an average is obtained in the *SP* and the *HP* by Eq. (2.9):

$$\langle A\rangle_t=\mathrm{Tr}\,AW(t)=\mathrm{Tr}\,A(t)W(t_0)\equiv\langle A(t)\rangle. \tag{C3}$$

When a total system consists of a relevant system (S) and a bath (B), Tr in Eq. (C3) is recognized as a trace operation for the total system, namely,

$$\mathrm{Tr}\cdot\equiv\mathrm{tr}_B\,\mathrm{tr}_S\cdot. \tag{C4}$$

The left hand side of Eq. (C3) is manipulated as follows:

$$\langle A\rangle_t=\mathrm{Tr}\,AW(t)=\mathrm{Tr}\,A\rho_B\,\mathrm{tr}_B\,W(t)\equiv\mathrm{Tr}\,A(\tilde{\mathcal{P}}W(t)), \tag{C5}$$

where ρ_B is the density operator for the B (bath) system alone, satisfying

$$\mathrm{tr}_B\,\rho_B=1, \tag{C6}$$

and we have defined $\tilde{\mathcal{P}}$ by

$$\tilde{\mathcal{P}}\cdot\equiv\rho_B\,\mathrm{tr}_B\cdot. \tag{C7}$$

In contrast, the right hand side of Eq. (C3) is rewritten as follows:

$$\begin{aligned}\langle A(t)\rangle&=\mathrm{Tr}\,A(t)W(t_0)=\mathrm{Tr}\,AW(t)\\&=\mathrm{Tr}(\mathrm{tr}_B\,\rho_BA)W(t)\\&\equiv\mathrm{Tr}(\mathcal{P}A)W(t),\end{aligned} \tag{C8}$$

where we have also defined $\mathcal{P}$ by

$$\mathcal{P}\cdot\equiv\mathrm{tr}_B\,\rho_B\,. \tag{C9}$$

Then, Eq. (C3) is equivalent to

$$\mathrm{Tr}\,A(\tilde{\mathcal{P}}W(t))=\mathrm{Tr}(\mathcal{P}A)W(t). \tag{C10}$$

This is the specialized version of the ''dual'' relation (8.7) [30,38] obtained by requiring the same averaged value of A both in the *HP* and the *SP*.

APPENDIX D: MOMENTS OF VARIABLES OF QUANTUM ENVIRONMENT

In order to analyze the spin relaxation process in Sec. VII, we have to evaluate moments of operators, $\tilde{\omega}_\mu(t)$ ($\mu=+$ or $-$).

With the definition (7.10),

$$\dot{\mathcal{P}}=\mathrm{tr}_B\,\rho_B\cdot=\frac{1}{Z_B}\mathrm{tr}_B\,e^{-\beta\mathcal{H}_B}\cdot\equiv\langle\cdot\rangle_B\,, \tag{D1}$$

the lower order moments of the reservoir variable b_j and $b_j^\dagger$ are found to be

$$\langle b_j\rangle_B=\langle b_j^\dagger\rangle_B=0, \tag{D2}$$

$$\langle b_j^\dagger b_j\rangle_B=\frac{1}{e^{\lambda_j}-1}\equiv n(\omega_j), \tag{D3}$$

$$\langle b_j b_j^\dagger\rangle_B=1+\langle b_j^\dagger b_j\rangle_B=\frac{1}{1-e^{-\lambda_j}}\equiv\overline{n}(\omega_j), \tag{D4}$$

$$\langle b_j b_j\rangle_B=\langle b_j^\dagger b_j^\dagger\rangle_B=0, \tag{D5}$$

with

$$\lambda_j\equiv\beta\hbar\omega_j. \tag{D6}$$

Thus, for the reservoir variables,

$$\tilde{\omega}_+(t)=\sum_j e^{-i(\omega_0-\omega_j)t}\kappa_j^* b_j^\dagger=\tilde{\omega}_-(t)^\dagger, \tag{D7}$$

the relations (D2)–(D5) yield

$$\begin{aligned}\langle\tilde{\omega}_+(t)\tilde{\omega}_-(t_1)\rangle_B&=\sum_j\sum_l \kappa_j^*\kappa_l e^{-i(\omega_0-\omega_j)t+i(\omega_0-\omega_l)t_1}\langle b_j^\dagger b_l\rangle_B\\&=\sum_j|\kappa_j|^2e^{-i(\omega_0-\omega_j)(t-t_1)}n(\omega_j),\end{aligned} \tag{D8}$$

$$\begin{aligned}\langle\tilde{\omega}_-(t)\tilde{\omega}_+(t_1)\rangle_B&=\sum_j\sum_l \kappa_j\kappa_l^* e^{i(\omega_0-\omega_j)t-i(\omega_0-\omega_1)t_1}\langle b_j b_l^\dagger\rangle_B\\&=\sum_j|\kappa_j|^2e^{i(\omega_0-\omega_j)(t-t_1)}\overline{n}(\omega_j)\end{aligned} \tag{D9}$$

and

$$\langle\tilde{\omega}_\pm(t)\tilde{\omega}_\pm(t_1)\rangle_B=0. \tag{D10}$$

For higher moments, we have

$$\begin{aligned}\langle\varphi_1\varphi_2\cdots\varphi_n\rangle_B&=\langle\varphi_1\varphi_2\rangle_B\langle\varphi_3\cdots\varphi_n\rangle_B+\langle\varphi_1\varphi_3\rangle_B\langle\varphi_2\cdots\varphi_n\rangle_B\\&\quad+\cdots+\langle\varphi_1\varphi_n\rangle_B\langle\varphi_2\varphi_3\cdots\varphi_{n-1}\rangle_B\quad(n\geqslant 3),\end{aligned} \tag{D11}$$

where

$$\varphi_n\equiv\tilde{\omega}_+(t_n)\ \text{ or }\ \tilde{\omega}_-(t_n). \tag{D12}$$

Repeated use of Eq. (D11) gives the theorem due to (Wick) Bloch and de Dominicis. When we also note Eq. (D2), we find that the moments of odd order disappear:

$$\langle\varphi_1\varphi_2\cdots\varphi_n\rangle_B=0\quad(\text{for odd } n). \tag{D13}$$

Now, we introduce a frequency distribution $\rho(\omega)$ of the coupling strength κ_j. That is, the distribution is defined by

$$\rho(\omega)\equiv\sum_j|\kappa_j|^2\delta(\omega-\omega_j). \tag{D14}$$

Next, we assume the distribution to be a Lorentzian with width γ, centered at ω_b:

$$\rho(\omega)\equiv\frac{\gamma}{\pi}\frac{\Delta^2}{(\omega-\omega_b)^2+\gamma^2}. \tag{D15}$$

Further, the average number $n(\omega_j)$ is assumed to be constant around the frequency range where $\rho(\omega)$ changes appreciably.

Thus we have

$$\begin{aligned}\int_{-\infty}^{\infty}e^{-i\omega t}\rho(\omega)d\omega&=\sum_j|\kappa_j|^2e^{-i\omega_j t}\\&=\frac{\gamma\Delta^2}{\pi}\int_{-\infty}^{\infty}e^{-i\omega t}\frac{1}{(\omega-\omega_b)^2+\gamma^2}d\omega\\&=\Delta^2e^{-i\omega_b t-\gamma|t|}.\end{aligned} \tag{D16}$$

From Eq. (D16), we find

$$\sum_j|\kappa_j|^2e^{-i\omega_j t}=\Delta^2e^{-i\omega_b t-\gamma|t|}, \tag{D17}$$

which gives a relation among characteristic parameters.

The correlation function for $\tilde{\omega}_\pm(t)$'s is calculated as follows: Since we have a relation,

$$\begin{aligned}\int_{-\infty}^{\infty}e^{-i\omega t}\rho(\omega)n(\omega)d\omega&=\sum_j|\kappa_j|^2e^{-i\omega_j t}n(\omega_j)\\&\cong\sum_j|\kappa_j|^2e^{-i\omega_j t}n(\omega_b)\\&=\Delta^2e^{-i\omega_b t-\gamma|t|}n(\omega_b),\end{aligned} \tag{D18}$$

the correlation functions of the reservoir variable are found to be

$$\begin{aligned}\langle\tilde{\omega}_+(t)\tilde{\omega}_-(t_1)\rangle_B&=\sum_j|\kappa_j|^2e^{-i(\omega_0-\omega_j)(t-t_1)}n(\omega_j)\\&\cong\Delta^2e^{-i(\omega_0-\omega_b)(t-t_1)-\gamma|t-t_1|}n(\omega_b)\end{aligned} \tag{D19}$$

and

$$\begin{aligned}\langle\tilde{\omega}_-(t)\tilde{\omega}_+(t_1)\rangle_B&=\sum_j|\kappa_j|^2e^{i(\omega_0-\omega_j)(t-t_1)}\overline{n}(\omega_j)\\&\cong\Delta^2e^{i(\omega_0-\omega_b)(t-t_1)-\gamma|t-t_1|}\overline{n}(\omega_b).\end{aligned} \tag{D20}$$

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