Non-Markovian correlation functions for open quantum systems

Jinshuang Jin\textsuperscript{1,2,3}, Christian Karlewski\textsuperscript{2,3} and Michael Marthaler\textsuperscript{3}

\textsuperscript{1} Department of Physics, Hangzhou Normal University, Hangzhou 310036, People’s Republic of China
\textsuperscript{2} Institute of Nanotechnology, Karlsruhe Institute of Technology (KIT), D-76344 Karlsruhe, Germany
\textsuperscript{3} Institut für Theoretische Festkörperphysik, Karlsruhe Institute of Technology, D-76131 Karlsruhe, Germany

E-mail: jsjin@hznu.edu.cn

Keywords: open quantum systems, correlation functions, non-Markovian, quantum dots

Abstract
Beyond the conventional quantum regression theorem, a general formula for non-Markovian correlation functions of arbitrary system operators both in the time- and frequency-domain is given. We approach the problem by transforming the conventional time-non-local master equation into dispersed time-local equations-of-motion. The validity of our approximations is discussed and we find that the non-Markovian terms have to be included for short times. While calculations of the density matrix at short times suffer from the initial value problem, a correlation function has a well defined initial state. The resulting formula for the non-Markovian correlation function has a simple structure and is as convenient in its application as the conventional quantum regression theorem for the Markovian case. For illustrations, we apply our method to investigate the spectrum of the current fluctuations of interacting quantum dots contacted with two electrodes. The corresponding non-Markovian characteristics are demonstrated.

1. Introduction
Open quantum systems, which are of great importance in many fields of physics, refer to a quantum system of primary interest coupled to an environment often called reservoir or bath [1–5]. The composite Hamiltonian ($H_{\text{tot}}$), in general, includes the system ($H_S$), the bath ($H_B$), and the coupling between the system and the bath ($H'$), i.e., $H_{\text{tot}} = H_S + H_B + H'$. It is well-known that the system is described by the reduced density operator, \( \rho(t) \equiv \text{tr}_B[\rho_{\text{tot}}(t)] \), i.e. the partial trace of the total density operator $\rho_{\text{tot}}$ over the bath space. The corresponding dynamics is determined by the master equation

\[
\dot{\rho}(t) = -i[H_S, \rho(t)] - \int_0^t dt \Sigma(t - \tau) \rho(\tau),
\]

where the effect of the bath is described by the second term with the self-energy $\Sigma(t - \tau)$. It contains the memory effect in principle even for weak system-reservoir coupling. Equation (1) is thus the so-called time-non-local (TNL) master equation describing non-Markovian dynamics.

As long as one knows the reduced density operator $\rho(t)$, a single-time expectation value of an arbitrary physical observable of the system, e.g., $\hat{O}$, is simply obtained via $\langle \hat{O}(t) \rangle = \text{Tr}[\hat{O}\rho(t)]$. However, it is not as easy to calculate two- or multiple-time correlation functions. Except for a small number of exactly solvable systems to use the exact procedure [6–10], convenient calculation of the correlation function is possible using the well-known quantum regression theorem (QRT) which is valid in the Born–Markovian approximation [1–5, 11, 12]. It is not valid any more for non-Markovian cases [5, 8, 9, 13, 14], due to memory effects which break the time translation invariance of the non-Markovian propagator.

The calculation of non-Markovian correlation functions for arbitrary system operators is a challenge and long-standing problem. Stimulated not only by fundamental interest but also by great demand because of the rapid progress in experiments which are able to access non-Markovian effects [15–17], there are many efforts to address this issue. For example, more than ten years ago, Shao and Markri [18] presented an iterative path integral algorithm for the numerically exact calculation of multitime correlation functions on the canonical ensemble. By using the stochastic Schrödinger equation approach and the Heisenberg equation, rather than
master equation, the method derived by Alonso and de Vega [19–21] is valid for zero-temperature environments and/or Hermitian system-environment coupling operators. Based on a generalized Born–Markov approximation, Budini [22] derived a quantum regression theorem which is applicable for non-Markovian Lindblad equations. Recently, Goan et al [23, 24] developed a scheme for the calculation of two-time correlation functions of the system operators with memory effects in terms of the ‘time-convolutionless’ master equation. Additionally some specific systems, such as the voltage fluctuation spectrum of the electron transport through a single electron transistor [25, 26] and the noise spectrum of the electron current through quantum point contact measuring the charge qubit [27], have been analyzed based on a real-time diagrammatic Keldysh technique [29, 30].

In this work, we aim to derive a general formula for non-Markovian correlation functions of arbitrary system operators in terms of the TNL-ME equation (1). We will consider weak system-reservoir coupling but short time-scales where non-Markovian effects should dominate. Later we will also analyze the relevant time scales in more detail. By using the fluctuation dissipation theorem of the bath correlator and introducing the auxiliary density operator in the frequency domain denoted by \( \phi^+(\omega, t) \), it is easy to transform the TNL-ME equation (1) into an equivalent set of coupled time-local equations of motion (for short TL-EOMs, expressed in equations (12) and (13)) [28], i.e.

\[
\dot{\rho}(t) = \mathbf{L}(\rho(t)), \quad \text{in the enlarged vector space with}
\]

\[
\dot{\rho}(t) \equiv \left\{ \rho(t), \phi^+(\omega, t), \phi(\omega, t) \right\}^T.
\]

The corresponding propagator in this enlarged vector space satisfies time-translation invariance and accordingly the correlation function can be treated similar to the Markovian case. The resulting equation takes a form very similar to the quantum regression theorem in a larger space

\[
\langle \hat{A}(t)\hat{B}(0) \rangle = \text{Tr} \left[ \hat{A} \left( \hat{P}(t, 0)\hat{\rho}(0) \right) \right].
\]  

The most important feature of this approach is the initial condition \( \hat{P}(0) = B\hat{\rho}(0) \), where \( \hat{\rho}(0) \) is the density matrix which has been time evolved from an initial time \( t_0 \to -\infty \). We assumed that system and reservoir decouple at the initial time \( t_0 \to -\infty \) which is the standard assumption for the TNL-ME. In the following we will derive equation (2) and we will discuss in detail the range of validity using the diagrammatic expansion on Keldysh contour [29]. This full non-Markovian description is applicable for both, fermionic and bosonic systems. As an example, we will discuss non-Markovian effects of the electronic reservoir on the current-fluctuation spectrum in quantum dots (QDs).

The paper is organized as follows. In section 2, we first introduce the conventional TNL-ME for weak system-reservoir coupling and then outline the equivalent TL-EOMs. Based on TL-EOMs, we get the formulas for the two-time non-Markovian correlation functions both in the time-domain and in the frequency-domain in section 3. We then implement the proposed scheme to study the current-fluctuation spectra of the electron tunneling through QDs in section 4. Finally, we conclude in section 5.

2. Non-markovian master equation: TNL-ME versus TL-EOMs

2.1. Time non-local master equation

The reservoir with infinite degrees of freedom is described by the non-interacting Hamiltonian \( H_B = \sum \epsilon_i \hat{c}_i \hat{c}_i^\dagger \) with the creation (annihilation) operator \( \hat{c}_i \) (\( \hat{c}_i^\dagger \)). The coupling Hamiltonian between the system and the bath, in general, is given by

\[
H' = Q^+ F^- + F^+ Q^+,
\]

where \( (Q^+) \equiv Q^- \) and \( (F^+) \equiv F^- \), with the operator of the central system \( Q^\pm \) and the operator of the bath \( F^\pm \). The coupling operator of the bath is defined as \( F^- = \sum t_i \hat{c}_i \) and contains the coupling coefficients \( t_i \). The result will be generalized for the case of coupling to multiple reservoirs in the appendix B. Note that the following derivations could be easily extended to the interaction Hamiltonian of the form \( H' = QX \) with setting \( Q^\dagger = Q \) and \( X = F^+ + F^- \) as the final formalism of correlation function given in section 3.2. The Hamiltonian of the small system is composed of the corresponding creation \( (\hat{a}_\mu^\dagger) \) and annihilation \( (\hat{a}_\mu) \) operators, i.e.

\[
H_S \equiv H_S(\hat{a}_\mu, \hat{a}_\mu^\dagger) \quad \text{which could include many-body interaction terms.}
\]

Assuming weak system-bath coupling and performing Born but without Markovian approximation, the self-energy for the expansion up to second-order of the coupling Hamiltonian is expressed as

\[
\Sigma(t - \tau) \equiv \langle [H'(t), e^{-i\hat{H}(t-\tau)}] \rangle_{H_S} \text{in the } H_B\text{-interaction picture. The corresponding diagram is schematically shown in figure 1(b). Here, } \langle \cdots \rangle_{H_B} \text{ stands for the statistical average over the bath in thermal equilibrium. The explicit formalism for the self-energy in equation (1) thus reads}
\]

\[
\Sigma(t - \tau) \equiv [Q^\dagger, \Pi(t, \tau)C^{(\pm)}(t - \tau)]^\dagger,
\]

where we introduced the free propagator \( \Pi(t, \tau) \equiv e^{-i\hat{H}_0(t-\tau)} \) with \( \hat{L} \equiv [H_0, \cdot] \), and

\[
C^{(\pm)}(t) \equiv C^{(\dagger)}(t)Q^\pm - \cdot C^{(\pm)}Q\pm,
\]

\[
C^{(\dagger)}(t) = \mathcal{F}(-i\hat{A}(t)) = \mathcal{F}(-i\rho(t)) = \mathcal{F}(-i\mathbf{L}(\rho(t))).
\]
with the correlator of the bath
\[ C^{(\pm)}(t - \tau) = \langle F^\pm(t) F^\mp(\tau) \rangle_B. \]  
(6)

Here \( \langle \cdots \rangle_B \) stands for the statistical average over the bath which is assumed to be in thermal equilibrium. Consequently, the TNL-ME equation (1) is explicitly given by \( \langle \hat{b} = 1 \rangle \) \cite{31--33},
\[ \dot{\rho}(t) = -i\mathcal{L}_S \rho(t) - \sum_{\pm} \int_0^t d\tau [Q^{\pm}, \Pi_0(t, \tau) C^{(\pm)}_Q(t - \tau) \rho(\tau)]. \]  
(7)

Note that equation (7) is derived assuming initial decoupling at \( t_0 \rightarrow -\infty \), \( \rho_{\text{tot}}(t_0) = \rho(t_0) \rho_B \) and \( \rho_B \) is the equilibrium state of the bath. Obviously, the non-Markovian dynamics described by equation (7) is determined by the superoperator \( C^{(\pm)}_Q(t - \tau) \) which contains the memory effect in the bath correlator \( C^{(\pm)}(t - \tau) \) defined by equation (6). If the fluctuation of the bath is considered white noise corresponding to the delta function of the correlator, e.g., \( C^{(\pm)}(t - \tau) \propto \gamma^{(\pm)}(t - \tau) \) with the constantly dissipative rate \( \gamma^{(\pm)} \), the TNL-ME of equation (7) immediately recast to Markovian master equation. It is time-local with the time-independent dissipative term and the QRT is valid very well. However, the practical bath has more complicated fluctuations and the bath correlator in general is not a simple delta or single exponential function but the expression of sum over many exponential terms as formally given by equation (31) containing the finite memory time. Therefore, the non-Markovian treatment is necessary.

Let us define the corresponding propagator \( \Pi(t, t_0) \) for equation (7) (equation (1))
\[ \rho(t) = \Pi(t, t_0) \rho(t_0), \]  
(8)
which satisfies the Dyson equation \cite{29} as shown in figure 1 (a). This propagator does not satisfy time-translation invariance, i.e. \( \Pi(t, t_0) = \Pi(t, t_0) \Pi(t_0, t_0) \). The conventional quantum regression theorem is thus broken.

2.2. Time-local equations-of-motion

The key to the calculation of the non-Markovian correlation function, e.g., \( \langle \hat{A}(t) \hat{B}(\tau) \rangle \), is to expand the master equation to an extend space \( \rho \rightarrow \hat{\rho} \) which preserves again time translation symmetry. This will allow us to case the correlator into the form of the regression theorem equation (2).

We adopt the multi-frequency-dispersed scheme developed in \cite{28} and define the bath correlator equation (6) in the frequency-domain \( (C^{(\pm)}(\omega)) \) as
\[ C^{(\pm)}(\omega) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{i\omega t} C^{(\pm)}(\omega), \]  
(9)
where \( C^{(\pm)}(\omega) \) is directly related to the spectral density of the bath depending on the specific operator \( (F^\pm) \). Correspondingly, the Liouville operator of equation (5) in the frequency-domain is (see equation (A.5))
\[ C^{(\pm)}_Q(\omega) \equiv C^{(\pm)}(\omega) Q^{\pm} - 1 \cdot C^{(\mp)}(\omega) Q^{\mp}. \]  
(10)

Furthermore, we introduce the auxiliary density operators in the frequency-domain defined by
\[ \phi^{(\pm)}(\omega, t) = -i \int_{t_0}^t d\tau e^{-i(\omega+\omega')\tau-1} C^{(\pm)}_Q(\omega) \rho(\tau), \]  
(11)
which means \( \phi^{(\pm)}(\omega, t_0) = 0 \) is applicable for the initially decoupled system-reservoir with \( t_0 \rightarrow -\infty \) as we mentioned above. Taking the time derivatives of the auxiliary density operators, it is easy to recast TNL-ME of equation (7) (equation (1)) in the form
\[ \dot{\rho}(t) = -i\mathcal{L}_S \rho(t) - i \sum_{\pm} \int \frac{d\omega}{2\pi} [Q^{\mp}, \phi^{(\pm)}(\omega, t)], \]  
(12)
\[ \dot{\phi}^\pm(\omega, t) = -i(L_S \mp \omega)\phi^\pm(\omega, t) - iC_Q^{(+)}(\omega)\rho(t), \]  
(13)

which are the so-called TL-EOMs due to the involved time-independent dissipative coefficients. TL-EOMs of equations (12) and (13) is the lowest-tier truncation of hierarchical equation of motion [28] which has the linearity of the hierarchical Liouville space as demonstrated in [34].

Following the arrangement in [34] we introduce a vector composed of the reduced density operator and auxiliary density operators, i.e.

\[ \tilde{\rho}(t) \equiv [\rho(t), \phi^+(\omega, t), \phi^-(\omega, t)]^T. \]  
(14)

The TL-EOMs equations (12) and (13) can then be further compacted with

\[ \hat{\rho}(t) = \Lambda \tilde{\rho}(t). \]  
(15)

Here according to equations (12) and (13), \( \Lambda \) can be formally written as

\[ \Lambda = \begin{pmatrix} -iL_S & -i \int \frac{d\omega}{2\pi} Q^+ & -i \int \frac{d\omega}{2\pi} Q^- \\ -iC_Q^{(+)}(\omega) & -i(L_S - \omega) & 0 \\ -iC_Q^{(-)}(\omega) & 0 & -i(L_S + \omega) \end{pmatrix}, \]  
(16)

where we introduced \( Q^{\pm} = [Q^+, Q^-]. \)

Apparently, equation (15) leads to \( \hat{\rho}(t) = \tilde{\Pi}(t, t_0)\tilde{\rho}(t_0) \) with \( \tilde{\Pi}(t, t_0) = e^{\Lambda(t-t_0)}. \) In this vector space, the propagator satisfies the time-translation invariance, i.e. \( \tilde{\Pi}(t, t_0) = \tilde{\Pi}(t, \tau)\tilde{\Pi}(\tau, t_0) \) and the correlation function in equation (2) can be calculated straightforwardly in a form similar to the Markovian case based on the quantum regression theorem.

### 3. Non-markovian correlation function

#### 3.1. Two-time correlation function

Using the vector of \( \tilde{\rho}(t) \) defined in equation (14), a single-time expectation value of the system operator \( \hat{A} \) can be expressed as

\[ \langle \hat{A}(t) \rangle = \text{Tr}[\hat{A}\rho(t)] = \text{Tr}[\hat{A}\tilde{\Pi}(t, t_0)\tilde{\rho}(t_0)], \]  
(17)

with the initial condition being that system and reservoir decouple \( (\phi^+(\omega, t_0 \to -\infty) = 0) \), i.e.

\[ \tilde{\rho}(t_0) = \{ \rho(t_0), 0, 0 \}. \]

Since time-translation invariance of the propagator has been restored, we can now follow the derivation of the Markovian correlation function based on the quantum regression theorem [4]. Therefore the non-Markovian two-time correlation function in the vector space is given by

\[ \langle \hat{A}(t)\hat{B}(0) \rangle = \text{Tr}[\hat{A}\tilde{\Pi}(t, \tau)\tilde{B}\tilde{\rho}(0)], \]  
(18)

where the components of the density operators in the vector are now

\[ \hat{\rho}(t) \equiv [\hat{\rho}(t), \hat{\phi}^+(\omega, t), \hat{\phi}^-(\omega, t)]^T. \]  
(19)

The initial state at \( t = 0 \) is given by

\[ \hat{B}\tilde{\rho}(0) = \{ \hat{B}\rho(0), \hat{B}\phi^+(\omega, 0), \hat{B}\phi^-(\omega, 0) \}, \]  
(20)

where \( \tilde{\rho}(0) \) is the density matrix which has been time evolved from a initial time \( t_0 \to -\infty \).

A similar equation has been derived using linear response theory in [34]. In this case all high-order contributions in the self-energy have been considered and one should keep all hierarchical EOMs for the numerical calculation of the correlation function [28, 34].

In this paper we consider the lowest-order contribution for weak system-reservoir coupling. In terms of equations (18)–(20) together with the TL-EOMs equations (12) and (13), (see appendix A for the detail), we finally get

\[ \langle \hat{A}(t)\hat{B}(0) \rangle = \text{Tr}[\hat{A}\Pi(t, 0)\hat{B}\tilde{\rho}(0)] = \sum_{t_2} \int_0^t dt_1 \int_0^{t_1} dt_2 \text{Tr}\{\hat{A}\Pi(t, t_2)\times [Q^+, \Pi_0(t_2, 0)\hat{B}\Pi_0(0, t_1)C_Q^{(+)}(t_2 - t_1)\rho(t_1)]\}, \]  
(21)

with the steady-state \( \rho(0) = \Pi(0, t_0)\rho(t_0) = \tilde{\rho} \) and \( \rho(t_1) = \Pi(t_1, t_0)\rho(t_0) = \tilde{\rho} \) for \( t_0 \to -\infty \). Compared to the Markovian correlation function, the second modification in equation (21) arises from the memory effect and it is the vertex corrections which will be further illustrated in the coming subsection based on a diagrammatic representation.
Similarly, it is easy to get the NMK-CF of \( \langle \hat{A}(0) \hat{B}(t) \rangle \) as expressed in equation (A.9). In this vector space, Non-Markovian multiple-time correlation functions can be calculated by \( \langle \hat{A}(t) \hat{C}(\tau) \cdots \hat{B}(0) \rangle = \text{Tr} \{ \hat{G}(t, \tau) \hat{G}(\tau, \tau) \cdots \hat{G}(t, 0) \} \). 

3.2. Spectrum of the two-time correlation function

Since the widely measured quantity in experiments is the spectrum of the correlation function (in Fourier space, i.e. \( \mathcal{F} \{ f(t) \} = \int_{-\infty}^{\infty} dt \ e^{i\omega t} f(t) \)), we will now calculate the spectrum for the stationary two-time correlation function

\[
S_{AB}(\omega) \equiv \mathcal{F} \{ \langle A(t) B(0) \rangle \} = 2 \text{Re} \{ \mathcal{L} \{ \langle A(t) B(0) \rangle \} \},
\]

where the last identity is assumed to be \( \hat{A}^\dagger = \hat{B} \), and \( \mathcal{L} \{ \langle A(t) B(0) \rangle \} \) is the Laplace transformation defined by \( \mathcal{L} \{ f(t) \} = \int_{0}^{\infty} dt \ e^{-\omega t} f(t) \). Based on either equation (21) directly or equation (18), we finally obtain (for the detail derivation see the appendix A)

\[
\mathcal{L} \{ \langle A(t) B(0) \rangle \} = \text{Tr} \{ \hat{A} \hat{G}(\omega) \hat{B} \hat{\rho} \} - \frac{i}{\omega} \text{Tr} \{ \hat{A} \hat{G}(\omega) \hat{B} \hat{\rho} \}
\times \sum_{+,-} \{ Q^\pm, \hat{B} (\hat{C}_{Q}^{(\pm)}(L_\omega, 0) - \hat{C}_{Q}^{(\pm)}(L_\omega, \omega)) \hat{\rho} \},
\]

where \( \hat{G}(\omega) \) and \( \hat{C}_{Q}^{(\pm)}(L_\omega, \omega) \) are the counterparts in the frequency domain by Laplace transformation of \( \hat{G}(t) \) and \( e^{-i \hat{C}_{Q}^{(\pm)} (t)} \hat{b} \) (see equation (8)), respectively. Explicitly, they are given by

\[
\hat{G}(\omega) = [i(\hat{L_\omega} - \omega) - \hat{\Sigma}(\omega)]^{-1},
\]

and

\[
\hat{C}_{Q}^{(\pm)}(L_\omega, \omega) \cdot = [\hat{C}_{Q}^{(\pm)} (\omega - L_\omega) Q^\pm \cdot - \cdot \hat{C}_{Q}^{(\pm)} (\omega - L_\omega) Q^\pm],
\]

with the frequency-domain of the self-energy (equation (4))

\[
\hat{\Sigma}(\omega) \cdot = \sum_{+,-} \{ Q^\pm, \hat{C}_{Q}^{(\pm)}(L_\omega, \omega) \cdot \},
\]

and the frequency-domain of the bath correlation (see equations (6) and (9))

\[
\hat{C}_{Q}^{(\pm)}(\omega) = \int_{-\infty}^{\infty} d\omega' \frac{i}{2\pi} \omega \pm \omega' + i0^+ \hat{C}_{Q}^{(\pm)}(\omega').
\]

It can be further give by

\[
\hat{C}_{Q}^{(\pm)}(\omega) = \frac{1}{2} [\hat{C}_{Q}^{(\pm)}(\mp \omega) + i\hat{N}^{\pm}(\omega)],
\]

where the real part \( \hat{C}_{Q}^{(\pm)}(\omega) \) is described by equation (9) and the imaginary part is

\[
\hat{N}^{\pm}(\omega) \equiv \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{1}{\omega + \omega'} \hat{C}_{Q}^{(\pm)}(\omega'),
\]

with \( \mathcal{P} \) denoting the principle value of the integral. 

Similarly, the spectrum of the correlation function of \( \langle \hat{A}(0) \hat{B}(t) \rangle \), is given by equation (A.10). The formulas equations (21) and (23) are the main contributions of the present work for the calculation of the non-Markovian two-time correlation function of the system operators (denoted by \( \hat{A} \) and \( \hat{B} \)) in time-domain and the frequency-domain, respectively.

Note that the above for often relevant Hermitian coupling operator, i.e. \( H' = QX \) (see equation (3)) with \( Q^\dagger = Q \) and \( X = F^+ + F^- \), equations (21) and (23) is simplified to

\[
\langle \hat{A}(t) \hat{B}(0) \rangle = \text{Tr} \{ \hat{A} \Pi(t, t_0) \hat{B} \hat{\rho}(0) \} = \sum_{+, -} \int_{t_0}^{t_0} dt_1 \int_{t_0}^{t_0} dt_2 \text{Tr} \{ \hat{A} \Pi(t, t_2) \}
\]

\[
\times \{ Q, \Pi_0(t_2, 0) \hat{B} \Pi_0(t, t_0) \hat{C}_{Q}(t_2 - t_0) \hat{\rho}(t_0) \},
\]

and

\[
\mathcal{L} \{ \langle A(t) B(0) \rangle \} = \text{Tr} \{ \hat{A} \hat{G}(\omega) \hat{B} \hat{\rho} \} - \frac{i}{\omega} \text{Tr} \{ \hat{A} \hat{G}(\omega) \hat{B} \hat{\rho} \}
\times \{ Q, \hat{B} (\hat{C}_{Q}(L_\omega, 0) - \hat{C}_{Q}(L_\omega, \omega)) \hat{\rho} \},
\]

respectively, where we used \( Q^- = Q^+ = Q \) and \( \hat{C}_{Q} = \hat{C}_{Q}^- + \hat{C}_{Q}^+ \), \( \hat{C}_{Q} = \hat{C}_{Q}^+ + \hat{C}_{Q}^- \).
3.3. Discussion and comments

Now we are in the position to discuss the applicability and the range of validity of the present NMK-CF formula equation (21) (or equation (23)). For convenience, we consider the coupling Hamiltonian $H' = QX$, with the operator of the bath $X$ containing the coupling coefficient $g$, as an example. The resulting formula for non-Markovian correlation function is given by equations (29) and (30). The bath correlator in equation (6) is recast to

$$C(t - \tau) = \langle X^\dagger(t)X(\tau) \rangle_B = \sum_{m=0}^{\infty} \eta_{m} e^{-\lambda_{m}t},$$

where the second identity is written as a parametric decomposition such as Matsubara expansion [35, 36] or pade approximation [37], with $\eta_{m} \propto g^{2}$ and $1/\lambda_{m}$ representing the correlation time of the bath or the so-called the memory time. For the present considered weak system-reservoir coupling, the self-energy expressed in equation (4) contains the lowest-order contribution (or the first-order contraction by Wick theorem), e.g., $\Sigma(t) \propto C(t)$. Let the index $f$ represent the number of contractions. The lowest-order contribution considered in the self-energy means $f = 1$.

In the following discussion we will not limit us to $f = 1$, but consider arbitrary number of the contractions such that the self-energy is exact. Based on the diagrammatic technique [29], we can obtain (see appendix C.1 for details)

$$\langle \hat{A}(t)\hat{B}(0) \rangle = \text{Tr}[\hat{A}\Pi(t, 0)\hat{B}\rho(0)] + \int_{0}^{t} dt_{2} \int_{t_{0}}^{t} dt_{1} \text{Tr}[\hat{A}\Pi(t_{2}, t_{1})\Sigma_{B}\Pi(t_{1}, t_{0})\rho(t_{0})],$$

which is formally exact. The relevant diagrams are shown schematically in figure 2. Here, $\Sigma_B$ is the self-energy of vertex corrections including the operator $\hat{B}$ and contain all the unseparable diagrams as well as the self-energy $\Sigma$ in the propagator $\Pi$. It is worth noting that equation (32) has the same form as equation (29) (or equation (21)) where $\Sigma_B$ can be extracted and $\Sigma$ is expressed explicitly in equation (4) which has the same order of the magnitude to $\Sigma_B$.

We will now use this formalism to discuss the range of validity of the non-Markovian correlation function. As has been shown in [38], in general non-Markovian effects are of the same order has higher order contractions in the self-energy. However, for the correlation function we have a well defined time scale and for short times scales, the combination of non-Markovian master equation and lowest order self-energy can be valid.

Let us make the Taylor expansion of the time-derivative of the correlation function, i.e. $G^{f}(t) \equiv \frac{d\langle\hat{A}(t)\hat{B}(0)\rangle}{dt}$ in $H_{B}^{f}$—interaction, for small $t = 0^{+}$,

$$G^{f}(t) = G^{f}(0) + \left. \frac{dG^{f}(t)}{dt} \right|_{t=0} \cdot t + \frac{1}{2} \left. \frac{d^{2}G^{f}(t)}{dt^{2}} \right|_{t=0} \cdot t^{2} + \cdots.$$  

(33)

Following the estimation of the order of magnitude of the self-energy in [38], we roughly get (see appendix C.2 for the detail)

$$G^{f}(t) \sim \text{Tr}[\hat{A}f(Q, \hat{B}, \rho)] \sum_{l} \frac{g^{2l}}{\gamma^{2l-1}} t^{2l} + \text{Tr}[\hat{A}f(Q)\hat{B}\rho + f(Q, \hat{B}, \rho)] \sum_{l} \frac{g^{2l}}{\gamma^{2l-2}} t^{2l} + \frac{1}{2} \text{Tr}[\hat{A}f(Q)\hat{B}\rho + f(Q, \hat{B}, \rho)] \sum_{l} \frac{g^{2l}}{\gamma^{2l-3}} t^{2l} + \cdots,$$

(34)

where $\gamma$ is the smallest decay rate $\gamma_{m}$ of $C(t)$ in equation (31), $f(Q, \hat{B}, \rho)$ and $f(Q)$ are just the formal expressions arising from $\Sigma_B$ and $\Sigma$, respectively. Note that the magnitudes of $f(Q)\hat{B}\rho$ and $f(Q, \hat{B}, \rho)$ are nearly...
equal. We conclude that the expansion is valid for time $t \lesssim 1/\gamma$, even if only the lowest order contributions $l = 1$ are considered.

4. Current fluctuations of the electron transport in QDs

For the demonstration of characteristic non-Markovian effects, we consider electron transport through QDs contacted with two electrodes (left $L$ and right $R$). This is a typical fermionic open quantum system where we will consider the non-Markovian effects in the spectrum in high-frequency regime [26, 31, 39].

The coupling Hamiltonian of equation (3) is specified by

$$H' = \sum_{\mu} (a_{\mu}^d F_{\mu} + F_{\mu}^a a_{\mu}),$$

with the system operator $Q_{\mu}^s$ being $a_{\mu}^d (a_{\mu})$ the creation (annihilation) operator of an electron of the $\mu$th-level of the dot, and the operator of the reservoirs $F_{\mu}^s = \sum_{\alpha = \text{L,R}} F_{\mu}^{\alpha s}$ with $F_{\mu}^{\alpha s} = \sum_{k} t_{\alpha \mu} c_{\alpha \mu}$. The correlator of lead $\alpha$ in the frequency-domain of equation (9) is thus $C_{n\mu}^{\alpha}(\omega) = \Gamma_{n\mu}(\omega)f^{\alpha}_{n\mu}(\omega)$ with the spectral density of the reservoir $\Gamma_{n\mu}(\omega) = 2\pi \sum_{k} t_{n\mu} t_{\alpha k}^* \delta(\omega - \epsilon_k)$, the Fermi function of $f^{\alpha}_{n\mu}(\omega) = f^{\alpha}_{n\mu}(\omega) = \frac{1}{1 + \exp(\beta(\omega - \mu_{\alpha})/T)}$, $f^{\alpha}_{n\mu}(\omega) = 1 - f^{\alpha}_{n\mu}(\omega)$, and $\beta = 1/k_B T$ the inverse of the temperature. For the studied model, we consider $C^{(\pm\omega)}_{n\mu}(t) = C^{(\pm\omega)}_{n\mu}(t)\delta_{n\mu}$ and symmetrical bias voltage $\mu_L = -\mu_R = eV/2$.

Assuming a Lorentzian spectrum centered around the Fermi energy of the lead [30, 31, 40, 42–44],

$$\Gamma_{n\mu}(\omega) = \frac{\Gamma_{\mu R}^{\omega\omega}}{(\omega - \mu_{\mu R})^2 + \omega^2},$$

with high cut-off frequency $\omega = 100\Gamma$ with $\Gamma = \sum_{n\mu} \Gamma_{n\mu}$, it leads to equation (27) (equation (B.5)) [30, 44] $C^{(\pm\omega)}_{n\mu}(\omega) = \Gamma_{n\mu}(\omega)f^{\alpha}_{n\mu}(\omega) \approx \Gamma_{n\mu} f^{\alpha}_{n\mu}(\omega)$ and

$$A^{(\pm\omega)}_{n\mu}(\omega) = \frac{\Gamma_{n\mu}}{\pi} \left\{ \operatorname{Re} \left[ \Psi \left( \frac{1}{2} + \frac{\beta(\omega - \mu_{\mu})}{2\pi} \right) \right] - \Psi \left( \frac{1}{2} + \frac{\beta \omega_{\mu R}}{2\pi} \right) \pm \frac{\omega - \mu_{\mu}}{\omega_{\mu R}} \right\},$$

with $\Psi(\chi)$ the Digamma function.

Here we focus on the investigation of the non-Markovian current fluctuations in the central dots, i.e. $(\dot{Q}(t)Q(0))$ with $Q = e\dot{N}(t)$ and $\dot{N} = \sum_{\alpha} a_{\alpha}^d a_{\alpha}$. Since it satisfies the charge conservation of $\dot{Q}(t) = -[H_{\mu\mu} + Ig(t)]$, the corresponding spectrum is expected to be closely related to the noise spectrum of the transport current $I(t)$ in the reservoir $\mu$. Actually, the current-fluctuation spectrum in the central dot can be easily obtained in terms of the corresponding charge fluctuation defined by $S_{\mu\mu}(\omega) = \mathcal{F}[\langle \dot{N}(t)N(0)\rangle]$, via the relation of $S_{\mu\mu}(\omega) = \mathcal{F}[\langle \dot{Q}(t)Q(0)\rangle]$ calculated by equation (23), the result agrees completely with that given in [26] based on diagrammatic technique.

4.1. Single QD

Let us first study the simplest model of electron transport through a spin-less one-level QD in the sequential tunneling regime, $\mu_L > \varepsilon > \mu_R$, described by the Hamiltonian $H_{\mu} = e\dot{a}^d a$. Two states are considered in the dot, the empty state $|0\rangle$ and the single-electron occupied state $|1\rangle$, which leads to $a = |0\rangle \langle 1|$. For the spectrum of the charge fluctuation $S_{\mu\mu}(\omega) = \mathcal{F}[\langle \dot{N}(t)N(0)\rangle]$ calculated by equation (23), the result agrees completely with that given in [26] based on diagrammatic technique.

The numerical result is shown in figure 3. It depicts non-Markovian features (solid-line) compared to the Markovian results (dashed-line) for charge fluctuation and (b) for current fluctuation, respectively. For low frequency regime at $\omega < \min \left\{ |\mu_L - \varepsilon| \right\}$ corresponding to the long time limit, the results based on both non-Markovian and Markovian treatments are consistent due to the disappearance of non-Markovian effect. With increasing the frequency higher than the energy-resonance, i.e. $\omega > \omega_{\mu R}$, it enters the non-Markovian regime where the non-Markovian characteristic occurs in the spectra, showing steps at $\omega \approx \omega_{\mu R}$ in the current fluctuation spectrum (see figure 3 (b)). This is consistent with the noise spectrum of the transport current through the reservoirs studied in [10, 31, 39].

The non-Markovian feature showing steps at $\omega_{\mu R}$ in $S_{\mu\mu}(\omega)$ provides the information of the energy structure in the central dot. The heights of the steps contain the information of the tunneling rate as demonstrated in the following. For the studied single-level dot in the regime of $\mu_L > \varepsilon > \mu_R$, the stationary population of the empty and single-electron occupied states are, $p_{00} = \frac{\Gamma_{10}}{\Gamma}$ and $p_{11} = \frac{\Gamma_{11}}{\Gamma}$, respectively. Considering the spectrum in the positive frequency regime ($\omega > 0$), it corresponds to the energy absorption processes. Accordingly, when the dot is in the empty state $|0\rangle$ with probability $p_{00}$ the electrons in the right reservoir absorb the energy, i.e. $\mu_R + \omega = \varepsilon$ and tunnel to the dot, which leads to the step at $\omega_{\mu R} = |\varepsilon - \mu_R|$ with the height of...
The typical non-Markovian feature shows step at $\omega_{\text{em}} = |\epsilon - \mu_L|$ in the spectra. The other parameters are (in unit of $\Gamma = \Gamma_L + \Gamma_R$): $\epsilon = 1$, $k_B T = 0.1$, and $\mu_L = -\mu_R = eV/2 = 6$.

Similarly, in the reversed regime ($\omega < 0$) corresponding to the energy emission processes, we get

$$h_L' \approx \tilde{\rho}_{00} \Gamma_L = \frac{\Gamma_L}{T},$$

and tunnel to the left reservoir, which leads to the step at $\omega_{\text{em}} = |\mu_L - \epsilon|$ with the height of $h_L \approx \rho_{11} \Gamma_L = \frac{\Gamma_L}{T}$. The ratio of the heights in the positive frequency regime thus is

$$h_L / h_R \approx \frac{\Gamma_L^2}{\Gamma_R^2}, \quad (\omega > 0).$$

Similarly, the energy, i.e., $\epsilon + \omega = \mu_L$ and tunnel to the left reservoir, which leads to the step at $\omega_{\text{em}} = |\mu_L - \epsilon|$ with the height of $h_L \approx \rho_{11} \Gamma_L = \frac{\Gamma_L}{T}$. The ratio of the heights in the positive frequency regime thus is

$$h_L / h_R \approx \frac{\Gamma_L^2}{\Gamma_R^2}, \quad (\omega < 0),$$

which is insensitive to the tunneling rate. These analytical solutions of equations (37) and (38) are consistent with the numerical results shown in figure 3(b) (dashed–dotted–line) for asymmetrical coupling $\Gamma_L = 2 \Gamma_R$ as an example.

We further consider the single level in the dot with spin–dependence as described by the Hamiltonian,

$$H_S = \sum_{\mu=\uparrow, \downarrow} \epsilon_{\mu} a_{\mu}^\dagger a_{\mu} + U h \tilde{\eta}_k,$$

where $\tilde{\eta}_k = a_k^\dagger a_k$ and $N = \sum_{\mu=\uparrow, \downarrow} \tilde{\eta}_k$. The involved states in the dot are $|0\rangle$, $| \uparrow \rangle$, $| \downarrow \rangle$, and $|2 \rangle \equiv | \uparrow \downarrow \rangle$, denoting the empty, two single-occupation spin states, and the double-occupation spin–pair state, respectively. In this state–basis, we have $a_\uparrow = |0\rangle \langle 1 \uparrow | + | 1 \uparrow \rangle \langle 2 \uparrow |$ and $a_\downarrow = |0\rangle \langle 1 \downarrow | + | 1 \downarrow \rangle \langle 2 \downarrow |$. The corresponding spectrum of the current fluctuations with different Coulomb interaction is shown in figure 4.

Besides the steps at the energy-resonance $\omega_{\text{em}}$, we also find the steps induced by Coulomb interaction at $\omega_{\text{RU}} \equiv |\epsilon + U - \mu_R|$. The different Coulomb interaction modifies the positions and the heights of the steps in the spectrum. In the positive high frequency limit at $\omega > \max \{\omega_{\text{RU}}, \omega_{\text{RU}}\}$, the current fluctuation spectra nearly approach the same value due to the absorption of enough energy to open all the tunneling channels.

We identify the regimes as (i) weak $U$ with $\mu_L > \epsilon$, $\epsilon + U > \mu_R$ and (ii) strong $U$ with $\epsilon + U > \mu_L > \epsilon > \mu_R$. In figure 4, for $U = 0$, the ratios of the heights of the steps both in the positive and negative parts are the same as spinless single level. However, compared to figure 3, the magnitude of the spectrum is doubled due to the two–energy levels (spin–up | $\uparrow \rangle$ and spin–down | $\downarrow \rangle$) involved in the transport. After the similar derivation in spinless single-level dot as demonstrated above, the ratios of the heights of the steps for the positive (denoted by $h^+$) and negative (denoted by $h^-$) frequencies in the spectrum are given by, respectively

$$h_{\text{RU}}: h_R: h_{\text{LU}} = \Gamma_L \Gamma_R' : \Gamma_R^2 : \Gamma_L \Gamma_R', \quad (39)$$

and

$$h_{\text{RU}}: h_R: h_{\text{LU}} = \Gamma_L \Gamma_R : \Gamma_R : \Gamma_L, \quad (40)$$

for (i) weak $U$ (the short–dashed–line in figure 4), and

$$h_{\text{RU}}: h_R: h_{\text{LU}} = \Gamma_L \Gamma_R : \Gamma_R^2 : \Gamma_L^2, \quad (41)$$

for (ii) strong $U$.
for (ii) strong $U$ (the dotted–dashed line in figure 4). Since the stationary double occupation is not allowed for strong $U$, there are no Coulomb-induced steps in the negative part of the spectrum.

### 4.2. Coupled double QDs

Now let us consider the electron transport through the system of two coupled QDs described by the Hamiltonian $H_b = \sum_{\mu=L,R} \hat{a}_{\mu}^\dagger \hat{a}_{\mu} + \frac{U}{2} \hat{n}_L \hat{n}_R + \Omega (\hat{a}_L^\dagger \hat{a}_R + \hat{a}_R^\dagger \hat{a}_L)$, where $U$ is the interdot Coulomb interaction, $\hat{n}_\mu = \hat{a}_{\mu}^\dagger \hat{a}_{\mu}$ and $\hat{N} = \sum_{\mu=L,R} \hat{n}_\mu$. The involved states of the double dot are $|0\rangle$ for the empty double dot, $|L\rangle$ for the left dot occupied, $|R\rangle$ for the right dot occupied, and $|LR\rangle$ for the two dots occupied. Here, we assume at most one electron in each dot. In this space, we have $a_L = |0\rangle \langle L| + |R\rangle \langle 2|$ and $a_R = |0\rangle \langle R| - |L\rangle \langle 2|$. The description of the involved states in this double dots is quite similar to that in the single dot with spin-dependence studied above. However, the essential difference is that the states of $|L\rangle$ and $|R\rangle$ are not the eigenstates of the system Hamiltonian $H_b$, which have the intrinsic coherent Rabi oscillation demonstrated by the coherent coupling strength $\Omega$. The corresponding Rabi frequency denoted by $\Delta$ is the energy difference between the eigenstates ($\varepsilon_{\pm}$), e.g., $\Delta = \varepsilon_+ - \varepsilon_- = 2\Omega$ for the degenerate double-dots system considered here.

The current–fluctuation spectrum for the coupled double dots is numerically displayed in figure 5. Similar to the single QD, the spectrum has the feature of a energy-resonance step at not only $\omega_{\alpha 0}$ (denoted by the dashed arrows), but also $\omega_{\alpha U} = |\varepsilon_+ + U - \mu_\alpha|$ induced by Coulomb interaction. The other parameters are the same as in figure 3.

$$h^\prime_b : h^\prime_r = 1 : 1,$$

(42)
\( \varepsilon_{L,R} \). Besides the same step behavior as in the single QD, of particular interest is the signal of the coherent Rabi oscillation of the coupled double dots in the current-fluctuation spectrum.

Interestingly, the emergence of the coherent signal of the Rabi oscillation is nearly determined by the strength of the Coulomb interaction \( U \). For weak Coulomb interaction in the regime of \( \mu_a > \varepsilon_{\pm}, \varepsilon_{\pm} + U > \mu_R \), such as \( U = 0 \) (solid-line) and \( U = 2\Gamma \) (dashed line) in figure 5(a), no signal at the Rabi frequency \( \omega = \Delta \) occurs in the spectrum. While in the double-dot Coulomb blockade regime, either \( \varepsilon_{\pm} + U > \mu_a > \varepsilon_{\pm}, \varepsilon_{\pm} + U > \mu_R \) or \( \varepsilon_{\pm} + U > \mu_a > \mu_R \), the current-fluctuation spectrum always shows a peak at the Rabi frequency \( \omega = \Delta \) as shown in figure 5(a) with \( U = 6\Gamma \) (short-dashed–dotted-line) and \( U = 15\Gamma \) (dashed–dotted-line). Although the coherent signal peak appears for asymmetrical coupling with \( \Gamma_a = 4\Gamma_R \) shown in the inset of figure 5(a), it is quite weak compared to that induced by Coulomb interaction. This means the coherent Rabi oscillation signal in the current-fluctuation spectrum is sensitive to the dynamical blockade channel. This characteristic of Rabi coherence signal is also consistent with Markovian treatment studied in [45], where the symmetrical current fluctuation spectrum was considered.

Furthermore, by increasing the coherent coupling strength \( \Omega \), we find that the coherent signal of the Rabi oscillation is moved to the high-frequency regime with strong non-Markovian effect as shown in figure 5(b). Simultaneously, the peak of the coherent signal in the spectrum gradually increases monotonically and sharply increases at the resonance regime where the Rabi frequency approaches the bias voltage, i.e. \( \Delta = 2\Omega = eV \), as shown in figure 5(c). This arises from the interplay between the Rabi resonance and the lead-dot tunneling resonance, i.e. \( \varepsilon_{\pm} = \pm eV / 2 \), combined with strong non-Markovian effect. It may suggest that this resonant regime is good for the observation of the coherent signal in the current-fluctuation spectrum experimentally. Beyond the resonance regime, the system will enter into the cotunneling regime which is beyond the present approach and we have to recur to more advanced approaches such as the hierarchical equations of motion [28, 34, 41, 46] and the real-time diagrammatic technique [29, 30] for the consideration of higher-order contributions in the self-energy.

### 5. Summary

In summary, using the frequency-dispersed technique by transforming the typical TNL master equation into equivalent TL-EOMs, we established an efficient formula for the two-time non-Markovian correlation function of arbitrary system operators in open quantum systems. The key to the calculation of the non-Markovian correlation function is how to restore an effective time-translation symmetry to the propagator. We find that this corresponds to the vertex corrections as further demonstrated by the real-time diagrammatic technique. The final result has an elegant structure and is as convenient to apply as the widely used quantum regression theorem for the Markovian case.

We applied the present method to study the current-fluctuation spectra in the interacting single QD and coupled double dots, respectively, contacted by two electrodes. The typical non-Markovian effect have been demonstrated. We found that the non-Markovian step behavior in the current-fluctuation spectrum of the single QD is consistent with that in the noise spectrum of the transport current through the leads. The sharp peak of the coherent Rabi signal in the double dots occurs at the resonance regime where the eigenenergy levels are comparable to the chemical potential in the leads under the applied bias voltage. From this current-fluctuation spectrum covering the full-frequency regime, the information of the energy structure of the QDs, the tunneling rate as well as the Coulomb interaction and even the coherent Rabi signal can be extracted directly.

Experimentally, the measurement of current-fluctuation spectrum can be realized via the detector such as a quantum point contact or single electron transistor coupled to the dots for the realization of the real-time single-electron tunneling through the dots [47, 48]. It is worth noting that the previous studies have widely focused on the low frequency limit which can be accessible in the current experiments [45, 49]. Although more difficult, experimental progress has also been made toward measuring the noise spectrum of electron transport in the high-frequency regime [30–53]. We expect the present results to be tested in the near future.

### Acknowledgments

We acknowledge helpful discussions with G Schön, X Q Li, and Y J Yan. JSJ acknowledges support from a fellowship of the KIT, as well as support from the Program of HNUEYT, the NNSF of China (Grant No. 11274085), NSF of Zhejiang Province (Grant No. LZ13A040002), and Hangzhou Innovation Funds for Quantum Information and Quantum Optics.
Appendix A. The detail derivation of the two-time NMK-CF expressed in equations (21) and (23)

In this appendix, we give a detailed derivation of the non-Markovian two-time correlation function given by the equation (21). For the calculation of the steady state as the initial condition of correlation function, the stationary solution of auxiliary density operators \( \hat{\phi}^\pm (\omega) \equiv \phi^\pm (\omega, 0) \) are given by equation (11)

\[
\hat{\phi}^\pm (\omega) = -i \int_0^\infty dt_e e^{i (\xi + \omega_t) t} C_Q^{\pm}(\omega, \rho (t_e)). \tag{A.1}
\]

Updated with the initial condition of \( \hat{\rho} (0) = \hat{B} \rho (0) = \{ \hat{B} \rho, \hat{B} \phi^+(\omega), \hat{B} \phi^- (\omega) \} \) for equation (15), we get the formula of the auxiliary density operators

\[
\hat{\phi}^\pm (\omega, t) = e^{-i (\xi + \omega_t t) t} \hat{B} \phi^\pm (\omega) - i \int_0^t dt_e e^{-i (\xi + \omega_t(t-t_e))} C_Q^{\pm}(\omega, \rho (t_e)). \tag{A.2}
\]

Inserting it into equation (12) yields

\[
\hat{\rho} (t) = -i \mathcal{L}_s \hat{\rho} (t) + \int_0^t d\tau \Sigma (t-\tau) \hat{\rho} (\tau) = -i \sum_{\pm} \int_0^\infty d\omega \frac{1}{2\pi} \left[ Q^\mp, e^{-i (\xi + \omega_t) t} \hat{B} \phi^\pm (\omega) \right], \tag{A.3}
\]

which has the solution

\[
\hat{\rho} (t) = \Pi (t, 0) \hat{B} \rho - i \sum_{\pm} \int_0^t dt_e \Pi (t, t_e) \int_0^\infty d\omega \frac{1}{2\pi} \left[ Q^\mp, e^{-i (\xi + \omega_t) t} \hat{B} \phi^\pm (\omega) \right]. \tag{A.4}
\]

Here, we used the initial condition \( \hat{\rho} (0) = \hat{B} \rho (0) = \hat{B} \rho \). With the use of equation (A.1) and the relation (see equations 9 and 10)

\[
\int_{\pm} d\omega \frac{e^{i \omega t} C_Q^{\pm}(\omega)}{2\pi} = \delta_Q (t), \tag{A.5}
\]

based on equation (18), we finally get the NMK-CF expressed by equation (21). We could directly get the spectrum of the correlation function in the frequency domain by a Laplace transformation based on equation (21). An alternative way is that we first obtain the stationary solution of auxiliary density operators in equation (A.1) which reads

\[
\hat{\phi}^\pm (\omega) = \frac{C_Q^{\pm}(\omega, \rho)}{\pm \omega - \xi + i \omega^+}. \tag{A.6}
\]

Then, we transform equation (A.3) into the frequency-domain with the relation

\[
\int_{\pm} d\omega \frac{\hat{\phi}^\pm (\omega)}{2\pi - \xi + i \omega^+} = -i \omega \left( C_Q^{\pm}(\mathcal{L}, 0) - C_Q^{\pm}(\mathcal{L}, \omega) \right) \hat{\rho}, \tag{A.7}
\]

The result is

\[
\left[ \hat{\Pi} (\omega) \right]^{-1} \hat{\rho} (\omega) = \hat{B} \rho - \sum_{\pm} \left[ Q^\mp, \hat{B} \left( C_Q^{\pm}(\mathcal{L}, 0) - C_Q^{\pm}(\mathcal{L}, \omega) \right) \right] \hat{\rho}. \tag{A.8}
\]

Finally, according to the first identity of equation (18) which suggests \( L \{ (A (t) B (0)) \} = \text{Tr} [ \hat{A} \hat{B} (\omega) ] \), we get the formula of the NMK-CF in the frequency-domain as expressed in equation (23).

Similarly, the correlation function of \( \hat{A} (0) \hat{B} (t) \) is given by

\[
\left\langle \hat{A} (0) \hat{B} (t) \right\rangle = \text{Tr} \left\{ \hat{B} \Pi (t, 0) (\hat{A} \hat{B}) \right\} = \sum_{\pm} \int_0^t dt_2 \int_{-\infty}^0 dt_1 \times \text{Tr} \left\{ \hat{B} \Pi (t, t_2) \left[ Q^\mp, \Pi_{\pm} (t_2, 0) (\Pi_{\pm} (0, t_2) C_Q^{\pm} (t_2 - t_1) \hat{A}) \right] \right\}, \tag{A.9}
\]

in the time-domain and

\[
L \{ (A (t) B (0)) \} = \text{Tr} \left\{ \hat{B} \left[ \hat{A} \right] (\omega) \right\} = \frac{1}{\omega} \text{Tr} \left\{ \hat{B} \hat{\Pi} (\omega) \right\} \times \sum_{\pm} \left[ Q^\mp, C_Q^{\pm}(\mathcal{L}, 0) - C_Q^{\pm}(\mathcal{L}, \omega) \right] \hat{A}, \tag{A.10}
\]

in the frequency-domain.
Appendix B. The two-time correlation function for multiple coupling

For simplicity so far, we showed the derivation of the NMK-CF for a single-operator coupling formalism as expressed in equation (3). Realistically, the coupling Hamiltonian is given by the multiple-operator coupling formalism, i.e. \( H' = \sum_{\mu} Q^{(\mu)}_C F^{(\mu)}_C + \text{h.c.} \) as we illustrated in section 4. The final formulas of equations (21) and (23) can be generalized by simply replacing the operators \( Q^{(\mu)}_C \) and \( C^{(\mu)}_Q \) with \( \tilde{Q}^{(\mu)}_C \) and \( \tilde{C}^{(\mu)}_Q \), respectively, and further adding the sum of \( \mu \). Thus, the formalism presented in sections 2 and 3 as well as appendix A are the same but adding the symbol \( \mu \) with the summation. Especially we pay attention on the final formula of NMK-CF in the time-domain of equation (21), where \( C^{(\mu)}_Q \) expressed in equation (5) should be replaced by

\[
C^{(\mu)}_Q(t - \tau) \cdot = \sum_{\nu} \left[ C^{(+)}_{\mu\nu}(t - \tau) Q^{(\nu)}_C - \right. \cdot \left. C^{(-)}_{\mu\nu}(t - \tau) Q^{(\nu)}_C \right], \tag{B.1}
\]

with \( C^{(\mu)}_Q(t - \tau) = \langle \tilde{F}^{(\mu)}_C(t) \tilde{F}^{(\mu)}_C(\tau) \rangle \), and the counterpart in the frequency-domain expressed in equation (10) should be replaced by

\[
\tilde{C}^{(\mu)}_Q(\omega, \omega) \cdot = \sum_{\nu} \left[ \tilde{C}^{(+)}_{\mu\nu}(\omega) \tilde{Q}^{(\nu)}_C - \tilde{C}^{(-)}_{\mu\nu}(\omega) \tilde{Q}^{(\nu)}_C \right], \tag{B.2}
\]

with (see equation (27))

\[
\tilde{C}^{(\mu)}_{\nu}(\omega) = \int_{-\infty}^{\infty} \frac{dw'}{2\pi} \omega + \omega' + i0^+ C^{(\mu)}_{\nu} (\omega'), \tag{B.3}
\]

For instance, the NMK-CF of equation (23) is generalized to

\[
L \left[ \langle A(t) B(0) \rangle \right] = \text{Tr} \left[ \tilde{A} \tilde{H}(\omega) \rho \right] - \omega \text{Tr} \left( \tilde{A} \tilde{H}(\omega) \right) \times \sum_{\mu, \nu, -} \left[ Q^{(+)}_C, \tilde{B} \left( \tilde{C}^{(\nu)}_Q(\omega, 0) - \tilde{C}^{(\nu)}_Q(\omega, \omega) \right) \rho \right] \tag{B.4}
\]

Appendix C. Diagram description

C.1. The derivation of formal exact two-time correlation function in equation (32)

The two-time correlator of two operators \( \hat{A} \) and \( \hat{B} \) acting in the Hilbert space of the system of interest is given by

\[
\langle \hat{A}(t) \hat{B}(0) \rangle = \text{Tr} \left[ \rho_T(t_0) U(t_0, t) \hat{A} U(t, 0) \hat{B} U(0, t_0) \right]. \tag{C.1}
\]

The functions \( U(t', t) \) are the unitary time evolutions \( U(t', t) = T \{ \exp \left[ \int_{t'}^{t} dt'' \hat{H}_s(t'') \right] \} \) and \( \rho_T(t) \) is the density matrix of the total system. The operator \( T \) is the time ordering operator. We assume that we can write this density matrix for \( t_0 \) as a direct product of the reduced density matrices of the system and the bath, which is valid for the limit \( t_0 \to -\infty \),

\[
\rho_T(t_0) = \rho_B(t_0) \otimes \sum_{s'} \rho_{s'}(t_0) | s' \rangle \langle s' |, \tag{C.2}
\]

\[
\langle \hat{A}(t) \hat{B}(0) \rangle = \text{Tr} \left[ \rho_B(t_0) \otimes \sum_{s'} \rho_{s'}(t_0) | s' \rangle \langle s' | U(t_0, t) \hat{A} U(t, 0) \hat{B} U(0, t_0) \right], \tag{C.3}
\]

\[
= \sum_{s', s} \rho_{s'}(t_0) | s' \rangle \text{Tr} \left[ \rho_B(t_0) U(t_0, t) \hat{A} U(t, 0) \hat{B} U(0, t_0) | s \rangle \right] \tag{C.4}
\]

where \{ | s \rangle \} is a basis set for the central system. By changing to the interaction picture (operators and states marked by tilde) the equation can be further simplified. Therefore, we introduce the Hamiltonian \( H_0 = H_s + H_b \) and the unitary time evolution \( U_0(t', t) = T \{ \exp \left[ \int_{t'}^{t} dt'' \hat{H}_s(t'') \right] \} \). The time evolution in the interaction picture is \( \tilde{U}(t', t) = U_0(t', t) U(t', t) = T \{ \exp \left[ \int_{t'}^{t} dt'' \hat{H}(t'') \right] \} \). An operator in the interaction picture is given by \( \tilde{A}(t) = U_0^\dagger(t, t_0) \hat{A} U_0(t, t_0) \) and an expansion of the exponential functions \( \tilde{U}(t, t') \) yields an expansion in the couplings \( \hat{H}' \)

\[
\tilde{U}(t_0, t) \hat{A}(t) \tilde{U}(t, 0) \hat{B}(0) \tilde{U}(0, t_0) = \langle \hat{A}(t) \hat{B}(0) + \int_0^t dt_0 \int_0^{t_0} dt_2 \hat{H}'(t_2) \hat{A}(t) \hat{H}'(t_2) \hat{B}(0) \nonumber
\]

\[
- \int_0^t dt_0 \int_0^{t_0} dt_2 \hat{H}'(t_2) \hat{H}'(t_0) \hat{A}(t) \hat{B}(0) + \cdots \tag{C.5}
\]
\[ \langle A(t)B(0) \rangle^I = \sum_{s,s'} \rho_{ss'}(t_0) \prod_{s,s'} (t_{0s} \hat{A}(t), \hat{B}(0)). \]  

which leads to

\[ \langle \hat{A}(t)\hat{B}(0) \rangle^I = \sum_{s,s'} \rho_{ss'}(t_0) \prod_{s,s'} (t_{0s} \hat{A}(t), \hat{B}(0)). \]

Each dot in a diagram denotes a coupling Hamiltonian \( \hat{H} \). The full superoperator is the full time evolution of the density matrix including the two operators \( \hat{A} \) and \( \hat{B} \). By using Wicks theorem the trace over the bath decays into two point functions represented by a contraction between the dots. So we can write

\[ \prod_{s,s'} \hat{A}(t) = \prod_{s,s'} \hat{A}(t) + \prod_{s,s'} \hat{B}(t) \]

Here, \( \Sigma_{ab} \) which is the self-energy for vertex corrections contains all the unseparable diagrams and equation (C.8) is still exact. We can rewrite this equation in algebraic form as

\[ \langle \hat{A}(t)\hat{B}(0) \rangle^I = \text{Tr} \{ \hat{A}(t)\hat{B}(0) \} \hat{B}(0) \hat{B}(0) \hat{A}(t) \hat{B}(0) \]

Using the relation of \( \rho(0) = \Pi(0) \rho(t_0) \rho(t_0) \) and \( \rho(t_1) = \Pi(t_1) \rho(t_0) \rho(t_0) \), equation (C.9) immediately recast to equation (32) in the Schrödinger picture.

C.2. The derivation for equation (34)

We introduce \( \varrho_B(t) = \Pi(t, 0) \hat{B}(t) \hat{B}(0) \) with \( \varrho_B(0) = \hat{B} \hat{B} \) and the auxiliary density operator describing the vertex contribution

\[ \varrho_{\Sigma B}(t) = \int_0^t dt_1 \Pi(t, t_1) \Sigma_{ab}(t_1 - t_0) \hat{B}(t_0) \hat{B}(0) \]

with \( \varrho_{\Sigma B}(0) = 0 \) apparently. Since we are interested in the discussion of the order of the magnitude roughly, it is more convenient in the \( H_S \)-interaction picture. The corresponding time-derivation equations are

\[ \dot{\varrho}_B(t) = \int_0^t d\tau \Sigma(t - \tau) \hat{B}(t), \]

\[ \dot{\varrho}_{\Sigma B}(t) = \int_0^t d\tau \Sigma(t - \tau) \hat{B}(t) + \hat{f}(t), \]

with \( \hat{f}(t) = \int_0^\infty dt_1 \Sigma_{ab}(t_1) \hat{B}(t_0) \hat{B}(0) \). Then the time-derivation of the two-time correlation function (see equation (C.9)) reads

\[ G^{I}(t) \equiv \frac{d\langle A(t)\hat{B}(0) \rangle^I}{dt} = \text{Tr} \{ \hat{A} [ \hat{B}(t) + \hat{f}(t)] \}. \]

The Taylor expansion of the time-derivative of the correlation function for small \( t = 0^+ \) is

\[ G^{I}(t) = G^{I}(0) + \frac{dG^{I}(t)}{dt} \bigg|_{t=0} t + \frac{1}{2} \frac{d^2G^{I}(t)}{dt^2} \bigg|_{t=0} t^2 + \cdots. \]

With equations (C.11) and (C.12), we get

\[ G^{I}(t) = \text{Tr} \{ \hat{A} [ \hat{f}_B(0) ] \} + \text{Tr} \{ \hat{A} [ \hat{B}(0) + \hat{\Sigma}_B(0) ] \} \rho \}

\[ + \frac{1}{2} \text{Tr} \{ \hat{A} [ \hat{f}_B(0) ] \} \rho \}

\[ t^2 + \cdots. \]
Using the estimation of the order of magnitude presented in [38], we roughly get

\[
\tilde{f}_B(0) \sim \sum_l \frac{g^{2l}}{\gamma^{2l+1}} f(Q, \tilde{B}, \tilde{p}),
\]

\[
\Sigma_0(0) \tilde{p} \sim \sum_l \frac{g^{2l}}{\gamma^{2l+2}} f(Q, \tilde{B}, p),
\]

\[
\tilde{\Sigma}_0(0) \tilde{p} \sim \sum_l \frac{g^{2l}}{\gamma^{2l+2}} f(Q, \tilde{B}, \tilde{p}),
\]

\[
\Sigma(0) \sim \sum_l \frac{g^{2l+1}}{\gamma^{2l+2}} f(Q),
\]

\[
\tilde{\Sigma}(0) \sim \sum_l \frac{g^{2l}}{\gamma^{2l+2}} f(Q),
\]

where \(\gamma\) is the minimum decay rate \(\gamma\) of \(C(t)\) in equation (31), \(f(Q, \tilde{B}, \tilde{p})\) and \(f(Q)\) are just the formal expression arising from \(\Sigma_0\) and \(\Sigma\), respectively. Inserting equation (C.16) into (C.15), we finally get equation (34).

References

[1] Carmichael HJ 1993 An Open System Approach to Quantum Optics (Berlin: Springer)
[2] Scully M and Zubairy M S 1997 Quantum Optics (Cambridge: Cambridge University Press)
[3] Gardiner C W and Zoller P 2000 Quantum Noise (Berlin: Springer)
[4] Breuer H P and Petruccione F 2002 The Theory of Open Quantum Systems (Oxford: Oxford University Press)
[5] Weiss U 2008 Quantum Dissipative Systems 3rd edn (Singapore: World Scientific)
[6] Grabert H, Schramm P and Ingold G L 1988 Phys. Rep. 168 115
[7] Hu B L, Par J P and Zhang Y 1992 Phys. Rev. D 45 2843
[8] Ford G W and O’Connell R F 1996 Phys. Rev. Lett. 77 798
[9] Ford G W and O’Connell R F 2000 Opt. Commun. 179 451
[10] Yang P Y, Lin C Y and Zhang W M 2013 Phys. Rev. B 89 115411
[11] Lax M 1963 Phys. Rev. 129 2342
[12] Lax M 1967 Phys. Rev. 157 213
[13] Ford G and O’Connell R 1999 Annu. Phys., NY 276 144
[14] Lax M 2000 Opt. Commun. 179 463
[15] Berthelot A et al 2006 Nat. Phys. 2 759
[16] Winger M et al 2009 Phys. Rev. Lett. 103 207403
[17] Ulrich S M et al 2011 Phys. Rev. Lett. 106 247402
[18] Shao J and Makri N 2001 Chem. Phys. 268 1
[19] Alonso D and de Vega I 2005 Phys. Rev. Lett. 94 200403
[20] Alonso D and de Vega I 2007 Phys. Rev. A 75 052108
[21] de Vega I and Alonso D 2006 Phys. Rev. A 73 022110
[22] Budini A 2008 J. Stat. Phys. 131 51
[23] Chen P W, Jian C C and Goan H S 2011 Phys. Rev. B 83 115439
[24] Chen P W, Jian C C and Goan H S 2011 J. Chem. Phys. 134 124112
[25] Shnirman A, Mozyrsky D and Martin I 2002 LANL arXiv:cond-mat/0211618
[26] Kick A, Wendin G and Johansson G 2003 Phys. Rev. B 67 035301
[27] Johansson G, Kick A and Wendin G 2002 Phys. Rev. Lett. 88 046802
[28] Jin J S, Zheng X and Yan Y 2008 J. Chem. Phys. 128 234703
[29] Schoeller H and Schön G 1994 Phys. Rev. B 50 18436
[30] König J 1995 Diploma Thesis University Karlsruhe
[31] Jin J S, Li X Q, Luo M and Yan Y J 2011 J. Appl. Phys. 109 053704
[32] Yan Y J and Xu R X 2005 Annu. Rev. Phys. Chem. 56 187
[33] Makhlín Y, Schön G and Shnirman A 2001 Rev. Mod. Phys. 73 357
[34] Wang S K, Zheng X, Jin J S and Yan Y J 2013 Phys. Rev. B 88 035129
[35] Meier C and Tannor D J 1999 J. Chem. Phys. 111 3365
[36] Xu R X, Cui P, Li X Q, Mo Y and Yan Y 2005 J. Chem. Phys. 122 041103
[37] Hu J, Xu R X and Yan Y J 2010 J. Chem. Phys. 133 101106
[38] Karlewski C and Marthaler M 2014 Phys. Rev. B 90 104302
[39] Engel H A and Loss D 2004 Phys. Rev. Lett. 93 136602
[40] Wingreen N S and Meir Y 1994 Phys. Rev. B 49 11040
[41] Ishizaki A and Tanimura Y 2005 J. Phys. Soc. Japan 74 3131
[42] Maciejko J, Wang I and Guo H 2006 Phys. Rev. B 74 085324
[43] Zheng X, Jin J S and Yan Y J 2008 New J. Phys. 10 093016
[44] Jin J S, Li J, Li X Q and Yan Y J 2014 J. Chem. Phys. 140 244111
[45] Luo J Y, Li X Q and Yan Y J 2007 Phys. Rev. B 76 085325
[46] Jin J S, Wang S K, Zheng X and Yan Y J 2015 J. Chem. Phys. 142 234108
[47] Gustavsson S, Leturcq R, Simovic B, Schleser R, Ihn T, Studerus P, Ensslin K, Driscoll D C and Gossard A C 2006 Phys. Rev. Lett. 96 076605
[48] Fujisawa T, Hayashi T, Tomita R and Hirayama Y 2006 Science 312 1634
[49] Kielich G, Scholl E, Brandes T, Hohls F and Haug R J 2007 Phys. Rev. Lett. 99 206602
[50] Onac E, Balestro F, Willems van Beveren L H, Hartmann U, Nazarov Y V and Kouwenhoven L P 2006 Phys. Rev. Lett. 96 176601
[51] Zakka-Bajjani Ségalas E J, Portier F, Roche R and Glattli D C 2007 Phys. Rev. Lett. 99 236803
[52] Basset J, Bouchiat H and Deblock R 2010 Phys. Rev. Lett. 105 166801
[53] Ubbelohde N, Fricke C, Flindt C, Hohls F and Haug R J 2012 Nat. Commun. 3 612