Non-Markovian full counting statistics of cotunneling assisted sequential tunneling in open quantum systems

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Abstract

We develop a non-Markovian full counting statistics formalism taking into account both the sequential tunneling and cotunneling based on the exact particle number resolved time-convolutionless master equation and the Rayleigh-Schrödinger perturbation theory. Then, in the sequential tunneling regime, we study the influences of the quantum coherence and the cotunneling processes on the non-Markovian full counting statistics of electron tunneling through an open quantum system, which consists of a side-coupled double quantum-dot system weakly coupled to two electron reservoirs. We demonstrate that, for the strong quantum-coherent side-coupled double quantum-dot system, the competition or interplay between the quantum coherence and the cotunneling processes, in the sequential tunneling regime, determines whether the super-Poissonian distributions of the shot noise, the skewness and the kurtosis take place (i.e., the Fano factor is larger than one), and whether the sign transitions of the values of the skewness and the kurtosis occur. These results suggest that, in the sequential tunneling regime, it is necessary to consider the influences of the quantum coherence and the cotunneling processes on the full counting statistics in the open strong quantum-coherent quantum systems, which provide a deeper insight into understanding of electron tunneling through open quantum systems.

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I. INTRODUCTION

In an open quantum system, for an intermediate coupling strength between the open quantum system and electron reservoir, the high-order tunneling processes, i.e., the cotunneling processes can influence the electron tunneling processes and bring out novel physical properties. Therefore, the electron cotunneling in open quantum systems, especially quantum dot (QD) systems, which is artificial molecules made from coupled QDs, and single molecules have been extensively studied both experimentally [1–6] and theoretically [7–12]. Particularly, the shot noise [13–28] and the full counting statistics (FCS) [26, 29–31] of cotunneling in QD systems have attracted considerable attention due to they can allow one to identify the intrinsic properties of the QD systems and access the information of electron correlation that cannot be obtained through the average current measurements. For example, in the Coulomb blockade regime, in which the transfer of electrons is dominated by cotunneling processes, it has been demonstrated experimentally [13–18] and theoretically [19–28] that the transport current displays super-Poissonian shot noise, which indicates that the super-Poissonian distribution of transferred-electron number and has a width being broader than its mean.

On the other hand, the quantum coherence, characterized by the off-diagonal elements of reduced density matrix of the considered system, also plays an important role in the electron tunneling through the strong quantum-coherent systems [28, 32–38]. In particular, theoretical studies have demonstrated that the non-Markovian effect of a strong quantum-coherent system plays an important role in the non-equilibrium electron tunneling processes [39], and manifests itself through the quantum coherence [38]. Consequently, in the intermediate coupling strength case, the electron tunneling through an open quantum system are mainly governed by the competitions or interplays between the cotunneling, sequential tunneling and quantum coherence. In the Coulomb blockade regime, it has been demonstrated that the electron cotunneling processes play a crucial role; whereas that, in the sequential tunneling regime where the transfer of electrons being dominated by sequential tunneling, has a slightly influence on the conduction and shot noise [3, 27, 39, 40]. Outside the Coulomb blockade regime, including the transition region from Coulomb blockade to sequential tunneling and the sequential tunneling regime, theoretical studies have demonstrated that the cotunneling assisted sequential tunneling processes have an important influence on the con-
duction \cite{8, 22} and shot noise \cite{21, 25}. However, in the sequential tunneling regime, the influences of quantum coherence and the cotunneling assisted sequential tunneling processes on the non-Markovian FCS has not yet been revealed.

In this work, we derive a non-Markovian FCS formalism taking into account both the sequential tunneling and cotunneling based on the exact particle number resolved time-convolutionless (TCL) master equation and the Rayleigh-Schrödinger perturbation theory developed in references \cite{41–43}. Then, in the sequential tunneling regime, we study the influences of the quantum coherence and the cotunneling processes on the non-Markovian FCS of electron tunneling through open quantum systems. For the sake of discussion, the considered open quantum system consists of a side-coupled double quantum-dot system weakly coupled to two electron electrodes (reservoirs). Here, the corresponding quantum coherence can be tuned by modulating the hopping strength between the two QDs relative to the coupling of this QD molecule with the source and drain electrodes. It is numerically demonstrated that, for the strong quantum-coherent side-coupled double QD system, the competition or interplay between the cotunneling processes and the quantum coherence, in the sequential tunneling regime, can take place in a range of the QD-electrode coupling strength and has a remarkable influence on the FCS. These characteristics depend on the temperature and the left-right asymmetry of the QD-electrode coupling. Therefore, in the open strong quantum-coherent quantum systems, it should be considered the effects of the quantum coherence and the cotunneling processes on the FCS, even through the electron tunneling is mainly dominated by the sequential tunneling processes.

II. MODEL AND FORMALISM

A. Hamiltonian of open quantum system and TCL master equations

We consider an open quantum system (OQS) weakly coupled to the two electrodes (reservoirs), which is described by the following Hamiltonian

\[
H = H_{\text{electrodes}} + H_{\text{OQS}} + H_{\text{hyb}}.
\]  

(1)

Here, the first term \( H_{\text{electrodes}} = \sum_{\alpha, k, \sigma} \varepsilon_{\alpha k} a_{\alpha k \sigma}^\dagger a_{\alpha k \sigma} \), characterized by the two noninteracting reservoirs, stands for the Hamiltonian of the two electrodes, with \( \varepsilon_{\alpha k} \) being the energy
dispersion, and \( a_{\alpha k\sigma}^\dagger \) (\( a_{\alpha k\sigma} \)) the creation (annihilation) operators in the \( \alpha \) electrode. The second term \( H_{\text{OQS}} = H_S (d_\mu^\dagger, d_\mu) \), which may contain vibrational or spin degrees of freedom and different types of many-body interactions, represents the OQS Hamiltonian, where \( d_\mu^\dagger \) (\( d_\mu \)) is the creation (annihilation) operator of electron in a quantum state denoted by \( \mu \). The third term \( H_{\text{hyb}} = \sum_{\alpha,\mu,k} (t_{\alpha\mu k} d_\mu^\dagger a_{\alpha k} + t^*_{\alpha\mu k} a_{\alpha k}^\dagger d_\mu) \), which is assumed to be a sum of bilinear terms that each create an electron in the OQS and annihilate one in the electrodes or vice versa, describes the tunneling coupling between the OQS and the two electrodes.

Due to the OQS-electrode coupling being sufficiently weak, thus, \( H_{\text{hyb}} \) can be treated perturbatively. In the interaction representation, the equation of motion for the total density matrix reads

\[
\frac{\partial}{\partial t} \rho_I (t) = -i [H_{\text{hyb}}^I (t), \rho_I (t)] \equiv \mathcal{L} (t) \rho_I (t), \tag{2}
\]

with

\[
H_{\text{hyb}}^I (t) = \sum_{\alpha,\mu} [f_{\alpha\mu}^\dagger (t) d_\mu (t) + f_{\alpha\mu} (t) d_\mu^\dagger (t)]
\]

where

\[
f_{\alpha\mu}^\dagger (t) = \sum_k t_{\alpha\mu k} \exp (i H_{\text{electrodes}} t) a_{\alpha k}^\dagger \exp (-i H_{\text{electrodes}} t) \tag{3}
\]

\[
d_\mu (t) = \exp (i H_{\text{OQS}} t) d_\mu \exp (-i H_{\text{OQS}} t) \tag{4}
\]

To derive an exact equation of motion for the reduced density matrix \( \rho_S \) of the OQS, it is convenient to define a super-operator \( \mathcal{P} \) according to

\[
\mathcal{P} \rho = \text{tr}_B [\rho] \otimes \rho_B = \rho_S \otimes \rho_B, \tag{5}
\]

where \( \rho_B \) is some fixed states of the two electrodes. Accordingly, a complementary super-operator \( \mathcal{Q} \),

\[
\mathcal{Q} \rho = \rho - \mathcal{P} \rho. \tag{6}
\]

For a factorizing initial condition \( \rho (t_0) = \rho_S (t_0) \otimes \rho_B \), \( \mathcal{P} \rho (t_0) = \rho (t_0) \), and then \( \mathcal{Q} \rho (t_0) = 0 \). Using the above TCL projection operator method, one can obtain the second-order and the fourth-order TCL master equations [44]

\[
\frac{\partial}{\partial t} \mathcal{P} \rho (t) \bigg|_{\text{second-order}} = \mathcal{K}_2 (t) \mathcal{P} \rho (t) = \int_{-\infty}^t dt_1 \mathcal{P} \mathcal{L} (t) \mathcal{L} (t_1) \mathcal{P} \rho (t) \tag{7}
\]
\[ \frac{\partial}{\partial t} \rho \left( t \right) \bigg|_{\text{fourth-order}} = K_4 \left( t \right) \rho \left( t \right) \]

\[ = \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \]

\[ \times \left[ P \mathcal{L} \left( t \right) \mathcal{L} \left( t_1 \right) \mathcal{L} \left( t_2 \right) \mathcal{L} \left( t_3 \right) \mathcal{P} - P \mathcal{L} \left( t \right) \mathcal{L} \left( t_1 \right) \mathcal{L} \left( t_2 \right) \mathcal{L} \left( t_3 \right) \mathcal{P} \right. \]

\[ \left. - P \mathcal{L} \left( t \right) \mathcal{L} \left( t_2 \right) \mathcal{P} \mathcal{L} \left( t_1 \right) \mathcal{L} \left( t_3 \right) \mathcal{P} - P \mathcal{L} \left( t \right) \mathcal{L} \left( t_3 \right) \mathcal{P} \mathcal{L} \left( t_1 \right) \mathcal{L} \left( t_2 \right) \mathcal{P} \right] \mathcal{P} \rho \left( t \right) \]

(8)

Both Eqs. (7) and (8) are the starting point of deriving the particle number resolved quantum master equation.

**B. The second-order particle number resolved TCL master equation**

In this subsection, we derive the second-order particle number resolved quantum master equation based on Eq. (7). Using both Eqs. (2) and (5), Eq. (7) can be rewritten as

\[ \frac{\partial}{\partial t} \rho_{I,S} \left( t \right) \bigg|_{\text{second-order}} = - \sum_{\alpha \beta} \int_{-\infty}^{t} dt_1 \text{tr}_B \left[ \rho_{I,S} \left( t \right) \otimes \rho_B \tilde{f}_{\alpha j}^\dagger \left( t_1 \right) d_j \left( t_1 \right) \tilde{d}_i \left( t \right) f_{\alpha i} \left( t \right) \right] \]

\[ - \sum_{\alpha \beta} \int_{-\infty}^{t} dt_1 \text{tr}_B \left[ \tilde{d}_i \left( t \right) f_{\alpha i} \left( t \right) \tilde{f}_{\alpha j}^\dagger \left( t_1 \right) d_j \left( t_1 \right) \rho_{I,S} \left( t \right) \otimes \rho_B \right] \]

\[ + \sum_{\alpha \beta} \int_{-\infty}^{t} dt_1 \text{tr}_B \left[ f_{\alpha i} \left( t \right) d_i \left( t \right) \rho_{I,S} \left( t \right) \otimes \rho_B \tilde{d}_j^\dagger \left( t_1 \right) f_{\alpha j} \left( t_1 \right) \right] \]

\[ + \sum_{\alpha \beta} \int_{-\infty}^{t} dt_1 \text{tr}_B \left[ \tilde{d}_i \left( t \right) f_{\alpha i} \left( t \right) \rho_{I,S} \left( t \right) \otimes \rho_B \tilde{f}_{\alpha j}^\dagger \left( t_1 \right) d_j \left( t_1 \right) \right] + \text{H.c.} \]

(9)

To fully describe the electron transport processes, the electron numbers, which emitted from the source electrode and passed through the OQS and arrived at the drain electrode, should be recorded. Following Refs. [45, 46], the Hilbert subspace \( B^{(n)} \) \((n = 1, 2, \ldots)\), which corresponds to \( n \) electrons arriving at the drain electrode and spanned by the product of all many-particle states of the two electrodes, is introduced and formally denoted as \( B^{(n)} \equiv \text{span} \left\{ \left| \Psi_L \right>^{(n)} \otimes \left| \Psi_R \right>^{(n)} \right\} \). Consequently, the entire Hilbert space of the two electrodes can be expressed as \( B = \bigoplus_n B^{(n)} \). With this classification of the states of the two electrodes, the average over states in the entire Hilbert space \( B \) in Eq. (9) should be replaced with the states in the subspace \( B^{(n)} \). Then, Eq. (9) can be expressed as a conditional TCL master
\[
\begin{align*}
\frac{\partial}{\partial t}\rho_{I,S}^{(n)}(t) \bigg|_{\text{second-order}} &= -\sum_{\alpha ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ \rho_{I,S}(t) \otimes \rho_B f_{ij}^\dagger(t_1) d_{ij}(t_1) d_{ij}^\dagger(t) \right] \\
&\quad - \sum_{\alpha ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ d_{ij}^\dagger(t) f_{\alpha ij}(t) f_{ij}^\dagger(t_1) d_{ij}(t_1) \rho_{I,S}(t) \otimes \rho_B \right] \\
&\quad + \sum_{\alpha ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ f_{ij}^\dagger(t_1) d_{ij}(t_1) \rho_{I,S}(t) \otimes \rho_B d_{ij}^\dagger(t) f_{\alpha ij}(t) \right] \\
&\quad + \sum_{\alpha ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ d_{ij}^\dagger(t) f_{\alpha ij}(t) \rho_{I,S}(t) \otimes \rho_B f_{ij}^{(n)}(t_1) d_{ij}(t_1) \right] + \text{H.c.} \quad (10)
\end{align*}
\]

Before proceeding, two physical considerations are implemented. (i) Instead of the conventional Born approximation for the entire density matrix \( \rho_T(t) \simeq \rho(t) \otimes \rho_B \), the ansatz \( \rho(t) \simeq \rho_B^{(n)}(t) \otimes \rho_B^{(n)} \) is proposed, where \( \rho_B^{(n)} \) being the density operator of two electrodes associated with \( n \) electrons arriving at the drain electrode. With this ansatz for the entire density operator (i.e., tracing over the subspace \( B^{(n)} \)), Eq. (10) can be reexpressed as

\[
\begin{align*}
\frac{\partial}{\partial t}\rho_{I,S}^{(n)}(t) \bigg|_{\text{second-order}} &= -\sum_{\alpha ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ f_{\alpha ij}^\dagger(t_1) f_{\alpha ij}(t) \rho_B \right] \rho_{I,S}^{(n)}(t) d_{ij}(t_1) d_{ij}^\dagger(t) \\
&\quad - \sum_{\alpha ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ f_{\alpha ij}(t) f_{ij}^\dagger(t_1) \rho_B \right] d_{ij}^\dagger(t) d_{ij}(t_1) \rho_{I,S}^{(n)}(t) \\
&\quad + \sum_{ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ f_{ij}(t) f_{ij}^\dagger(t_1) \rho_B \right] d_{ij}(t_1) \rho_{I,S}^{(n)}(t) d_{ij}^\dagger(t) \\
&\quad + \sum_{ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ f_{ij}^\dagger(t_1) f_{ij}(t) \rho_B \right] d_{ij}^\dagger(t) \rho_{I,S}^{(n)}(t) d_{ij}(t_1) \\
&\quad + \sum_{ij} \int_{-\infty}^{t} dt_1 tr_{B^{(n)}} \left[ f_{ij}^\dagger(t_1) f_{ij}(t) \rho_B \right] d_{ij}^\dagger(t) \rho_{I,S}^{(n)}(t) d_{ij}(t_1) + \text{H.c.} \quad (11)
\end{align*}
\]

Here, we have used the orthogonality between the states in different subspaces. (ii) The extra electrons arriving at the drain electrode will flow back into the source electrode via
the external closed transport circuit. Additionally, the rapid relaxation processes in the electrodes will bring the electrodes to the local thermal equilibrium states quickly, which are determined by the chemical potentials. After the procedure done in Eq. (11), the density matrices of two electrodes $\rho_B^{(n)}$ and $\rho_B^{(n+1)}$ should be replaced by $\rho_B^{(0)}$. In the Schrödinger representation, Eq. (11) can be written as

$$\frac{\partial}{\partial t} \rho_S^{(n)}(t) \bigg|_{\text{second-order}} = -i \left[ H_S, \rho_S^{(n)}(t) \right]$$

$$- \sum_{\alpha ij} \int_{-\infty}^{t} dt_1 C_{\alpha ij}^{(+)}(t_1 - t) \rho_S^{(n)}(t) e^{-iH_S(t-t_1)} d_j e^{iH_S(t-t_1)} d_i^\dagger$$

$$- \sum_{\alpha ij} \int_{-\infty}^{t} dt_1 C_{\alpha ij}^{(-)}(t - t_1) d_i^\dagger e^{-iH_S(t-t_1)} d_j e^{iH_S(t-t_1)} \rho_S^{(n)}(t)$$

$$+ \sum_{ij} \int_{-\infty}^{t} dt_1 C_{ij}^{(-)}(t - t_1) e^{-iH_S(t-t_1)} d_j e^{iH_S(t-t_1)} \rho_S^{(n)}(t) d_i^\dagger$$

$$+ \sum_{ij} \int_{-\infty}^{t} dt_1 C_{ij}^{(+)}(t_1 - t) d_i^\dagger \rho_S^{(n)}(t) e^{-iH_S(t-t_1)} d_j e^{iH_S(t-t_1)}$$

$$+ \sum_{ij} \int_{-\infty}^{t} dt_1 C_{ij}^{(+)}(t_1 - t) d_i^\dagger \rho_S^{(n+1)}(t) e^{-iH_S(t-t_1)} d_j e^{iH_S(t-t_1)} + \text{H.c.}$$

$$= (12)$$

where the correlation functions are defined as

$$C_{\alpha ij}^{(+)}(t - t_1) = \text{tr}_R \left[ f_{\alpha i}^\dagger(t) f_{\alpha j}(t_1) \rho_B \right] = \left\langle f_{\alpha i}^\dagger(t) f_{\alpha j}(t_1) \right\rangle,$$  \hspace{1cm} (13)

$$C_{\alpha ij}^{(-)}(t - t_1) = \text{tr}_R \left[ f_{\alpha i}(t) f_{\alpha j}^\dagger(t_1) \rho_B \right] = \left\langle f_{\alpha i}(t) f_{\alpha j}^\dagger(t_1) \right\rangle.$$  \hspace{1cm} (14)

Introducing the following super-operators

$$A_{\alpha i}^{(+)}(t) = \sum_j \int_{-\infty}^{t} dt_1 C_{\alpha ij}^{(+)}(t_1 - t) e^{-iH_S(t-t_1)} d_j e^{iH_S(t-t_1)},$$

$$\hspace{1cm} (15)$$

$$A_{\alpha i}^{(-)}(t) = \sum_j \int_{-\infty}^{t} dt_1 C_{\alpha ij}^{(-)}(t_1 - t) e^{-iH_S(t-t_1)} d_j e^{iH_S(t-t_1)},$$

$$\hspace{1cm} (16)$$
then, Eq. (12) can be rewritten as a compact form

\[
\frac{\partial}{\partial t} \rho_S^{(n)}(t) \bigg|_{\text{second-order}} = -i \left[ H_S, \rho_S^{(n)}(t) \right] - \sum_i \left\{ \rho_S^{(n)}(t) A_i^{(+)}(t) d_i^+ + d_i^+ A_i^{(-)}(t) \rho_S^{(n)}(t) - A_L^{(-)}(t) \rho_S^{(n)}(t) d_i^+ \right. \\
\left. - A_R^{(-)}(t) \rho_S^{(n-1)}(t) d_i^+ - d_i^+ \rho_S^{(n)}(t) A_L^{(+)}(t) - d_i^+ \rho_S^{(n+1)}(t) A_R^{(+)}(t) + \text{H.c.} \right\} .
\]

(17)

where \( A_i^{(\pm)}(t) = \sum_a A_i^{a(\pm)}(t) \). The equation (17) is the starting point of the non-Markovian FCS calculation taking into account the sequential tunneling processes only.

C. The fourth-order particle number resolved TCL master equation

In this subsection, we derive the fourth-order particle number resolved quantum master equation based on Eq. (8). Using both Eqs. (2) and (5), Eq. (8) can be expressed as

\[
\frac{\partial}{\partial t} \rho_{I,S}^{(n)}(t) \bigg|_{\text{fourth-order}} = \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \sum_{ijkl} \times \left\{ \text{tr}_B \left[ H_I(t), \left[ H_I(t_1), \left[ H_I(t_2), \left[ H_I(t_3), \rho_S \otimes \rho_B \right] \otimes \rho_B \right] \otimes \rho_B \right] \right] \\
- \text{tr}_B \left[ H_I(t), \left[ H_I(t_1), \text{tr}_B \left[ H_I(t_2), \left[ H_I(t_3), \rho_S \otimes \rho_B \right] \otimes \rho_B \right] \otimes \rho_B \right] \right] \\
- \text{tr}_B \left[ H_I(t), \left[ H_I(t_2), \text{tr}_B \left[ H_I(t_1), \left[ H_I(t_3), \rho_S \otimes \rho_B \right] \otimes \rho_B \right] \otimes \rho_B \right] \right] \\
- \text{tr}_B \left[ H_I(t), \left[ H_I(t_3), \text{tr}_B \left[ H_I(t_1), \left[ H_I(t_2), \rho_S \otimes \rho_B \right] \otimes \rho_B \right] \otimes \rho_B \right] \otimes \rho_B \right] \right\} 
\]

(18)

To further facilitate this derivation, the tunneling coupling between the OQS and the two electrodes \( H_{\text{hyb}}^I(t) \) is rewritten as the following equation

\[
H_{\text{hyb}}^I(t) = \sum_{\alpha,\mu} F_{\alpha \mu}(t) D_{\mu}(t) ,
\]

(19)

with

\[
F_{\alpha \mu}(t) = \sum_k e^{iH_{\text{electrodes}} t} F_{\alpha \mu k} e^{-iH_{\text{electrodes}} t},
\]

\[
D_{\mu}(t) = e^{iH_{\text{dot}} t} D_{\mu} e^{-iH_{\text{dot}} t},
\]
where $F_{\alpha \mu k} = t_{\alpha \mu k} a_{\alpha \mu k} + t_{\alpha \mu k}^* a_{\alpha \mu k}^*$, $D_\mu = d_\mu + d_\mu^*$. Inserting Eq. (19) into Eq. (18), one can obtain

$$
\frac{\partial}{\partial t} \rho_{I,S} (t) \bigg|_{\text{fourth-order}} = \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \sum_{ijkl} \left\{ C_{02} C_{13} \left[ 0, \left[ \hat{1}, \hat{2} \right] 3 \rho_S \right] - C_{02} C_{31} \left[ 0, \left[ \hat{1}, \hat{2} \right] 3 \rho_S \right] + C_{03} C_{12} \left[ 0, \left[ \hat{1}, \hat{2} \right] 3 \rho_S \right] - C_{03} C_{12} \left[ 0, \left[ \hat{1}, \hat{2} \right] 3 \rho_S \right] \right\} + \text{H.c.,}
$$

with

$$
C_{02} = \sum_{\alpha \mu k} \text{tr}_B \left[ F_{\alpha i} (t) F_{\alpha k} (t_2) \right], \quad C_{03} = \sum_{\alpha \mu l} \text{tr}_B \left[ F_{\alpha i} (t) F_{\alpha l} (t_3) \right],
$$

$$
C_{12} = \sum_{\alpha \mu j} \text{tr}_B \left[ F_{\alpha j} (t_1) F_{\alpha k} (t_2) \right], \quad C_{21} = \sum_{\alpha \mu j} \text{tr}_B \left[ F_{\alpha k} (t_2) F_{\alpha j} (t_1) \right],
$$

$$
C_{13} = \sum_{\alpha \mu j} \text{tr}_B \left[ F_{\alpha j} (t_1) F_{\alpha l} (t_3) \right], \quad C_{31} = \sum_{\alpha \mu j} \text{tr}_B \left[ F_{\alpha l} (t_3) F_{\alpha j} (t_1) \right],
$$

$$
\hat{0} = D_i (t), \quad \hat{1} = D_j (t_1), \quad \hat{2} = D_k (t_2), \quad \hat{3} = D_l (t_3).
$$

In the Schrödinger representation, Eq. (20) can be expressed as

$$
\frac{\partial \rho_S (t)}{\partial t} \bigg|_{\text{fourth-order}} = -i \left[ H_S, \rho_S (t) \right] + \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \sum_{ijkl} \left[ I_1 + I_2 + II_1 + II_2 + II_3 + \text{H.c.} \right],
$$

with

$$
I_1 = C_{02} C_{13} D_i e^{-i \xi_S (t_1)} D_j e^{-i \xi_S (t_2)} D_k e^{-i \xi_S (t_3)} D_l \rho_S (t)
$$

$$
+ C_{02} C_{13} e^{-i \xi_S (t_2)} D_k e^{-i \xi_S (t_1)} D_j e^{-i \xi_S (t_3)} D_l \rho_S (t) D_i
$$

$$
- C_{02} C_{13} D_i e^{-i \xi_S (t_2)} D_k e^{-i \xi_S (t_1)} D_j e^{-i \xi_S (t_3)} D_l \rho_S (t)
$$

$$
- C_{02} C_{13} e^{-i \xi_S (t_3)} D_k e^{-i \xi_S (t_1)} D_j e^{-i \xi_S (t_2)} D_l \rho_S (t) D_i,
$$

(22)
Here, we define the super-operator $L$ as $e^{-i L t} O e^{i L t} \equiv e^{-i L t} O$. Now, we derive the fourth-order particle number resolved quantum master equation based on Eq. (21). Without loss of generality, we consider the case of Eq. (22). Considering the Hamiltonian $H_{\text{hyb}}^t = \sum_{\alpha, \mu} [f_{\alpha \mu}(t) d_\mu(t) + f_{\alpha \mu}(t) d_\mu^\dagger(t)]$, the $C_{02}, C_{13}$ and $D_{i,j,k,l}$ have the following forms

$$C_{02}^{(t)} = \sum_{\alpha i k} \text{tr}_B \left[ f_{\alpha i}(t) f_{\alpha k}(t) \right],$$

$$I_2 = C_{02} C_{31} e^{-i L S(t-t_1)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ e^{-i L S(t-t_3)} D_l D_i$$

$$+ C_{02} C_{31} D_i e^{-i L S(t-t_2)} D_k e^{-i L S(t-t_1)} D_j \rho_S(t) \ e^{-i L S(t-t_3)} D_l D_i$$

$$- C_{02} C_{31} e^{-i L S(t-t_1)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ e^{-i L S(t-t_3)} D_l D_i$$

$$- C_{02} C_{31} D_i e^{-i L S(t-t_2)} D_k e^{-i L S(t-t_1)} D_j \rho_S(t) \ e^{-i L S(t-t_3)} D_l D_i, \ (23)$$

$$H_1 = C_{03} C_{12} D_i e^{-i L S(t-t_1)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ e^{-i L S(t-t_3)} D_l D_i$$

$$+ C_{03} C_{12} e^{-i L S(t-t_3)} D_i e^{-i L S(t-t_1)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ D_i$$

$$- C_{03} C_{12} D_i e^{-i L S(t-t_3)} D_j e^{-i L S(t-t_1)} D_k \rho_S(t) \ e^{-i L S(t-t_2)} D_i D_j$$

$$- C_{03} C_{12} e^{-i L S(t-t_1)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ e^{-i L S(t-t_3)} D_i D_j, \ (24)$$

$$H_2 = C_{03} C_{12} D_i e^{-i L S(t-t_3)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ e^{-i L S(t-t_1)} D_l D_i$$

$$+ C_{03} C_{12} e^{-i L S(t-t_2)} D_i e^{-i L S(t-t_3)} D_j \rho_S(t) \ e^{-i L S(t-t_1)} D_l D_i$$

$$- C_{03} C_{12} D_i e^{-i L S(t-t_2)} D_j e^{-i L S(t-t_3)} D_k \rho_S(t) \ e^{-i L S(t-t_1)} D_l D_i$$

$$- C_{03} C_{12} e^{-i L S(t-t_3)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ e^{-i L S(t-t_1)} D_l D_i, \ (25)$$

$$H_3 = C_{03} C_{21} D_i e^{-i L S(t-t_3)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ e^{-i L S(t-t_1)} D_l D_i$$

$$+ C_{03} C_{21} e^{-i L S(t-t_2)} D_i e^{-i L S(t-t_3)} D_j e^{-i L S(t-t_1)} D_k D_i$$

$$- C_{03} C_{21} D_i e^{-i L S(t-t_2)} D_j e^{-i L S(t-t_3)} D_k \rho_S(t) \ e^{-i L S(t-t_1)} D_k D_i$$

$$- C_{03} C_{21} e^{-i L S(t-t_3)} D_j e^{-i L S(t-t_2)} D_k \rho_S(t) \ e^{-i L S(t-t_1)} D_k D_i, \ (26)$$

Here, $D_i, D_j, D_l, D_k$ are the density matrices of the subsystems.
\[ C_{02}^{(-)} = \sum_{\alpha ik} \text{tr}_B \left[ f_{\alpha i}(t) f^\dagger_{\alpha k}(t_2) \right], D_i = d_i^\dagger, D_k = d_k, \quad (28) \]

\[ C_{13}^{(+)} = \sum_{\alpha jl} \text{tr}_B \left[ f^\dagger_{\alpha j}(t_1) f_{\alpha l}(t_3) \right], D_j = d_j, D_l = d_l^\dagger, \quad (29) \]

\[ C_{13}^{(-)} = \sum_{\alpha jl} \text{tr}_B \left[ f_{\alpha j}(t_1) f^\dagger_{\alpha l}(t_3) \right], D_j = d_j^\dagger, D_l = d_l, \quad (30) \]

respectively. Therefore, the particle number resolved formation of Eq. (22) can be expressed as follows

\[ I_1 = I_{1,n-1} + I_{1,n} + I_{1,n+1}, \quad (31) \]

with

\[ I_{1,n-1} = \]

\[ + C_{R,0.2}^{(-)} C_{L,1,3}^{(-)} A_k A_j^\dagger A_i^\dagger \rho_S^{(n-1)}(t) d_i^\dagger + C_{R,0.2}^{(-)} C_{R,1,3}^{(-)} A_k A_i^\dagger A_j^\dagger \rho_S^{(n-1)}(t) d_i^\dagger \]

\[ - C_{R,0.2}^{(-)} C_{L,1,3}^{(-)} A_j^\dagger A_k A_i^\dagger \rho_S^{(n-1)}(t) d_i^\dagger - C_{R,0.2}^{(-)} C_{R,1,3}^{(-)} A_j^\dagger A_k A_i^\dagger \rho_S^{(n-1)}(t) d_i^\dagger \]

\[ + C_{R,0.2}^{(+)} C_{L,1,3}^{(-)} A_k A_j A_i^\dagger \rho_S^{(n-1)}(t) d_i^\dagger + C_{R,0.2}^{(+)} C_{R,1,3}^{(-)} A_k A_j A_i^\dagger \rho_S^{(n-1)}(t) d_i^\dagger \]

\[ - C_{R,0.2}^{(+)} C_{L,1,3}^{(+)} A_j A_k A_i^\dagger \rho_S^{(n-1)}(t) d_i^\dagger - C_{R,0.2}^{(+)} C_{R,1,3}^{(+)} A_j A_k A_i^\dagger \rho_S^{(n-1)}(t) d_i^\dagger \quad (32) \]
\[ I_{1,n} = \\
+ C_{0,2}^{(-)} C_{1,3}^{(-)} d_i^\dagger A_k^\dagger A_l \rho_{S}^{(n)} (t) + C_{0,2}^{(-)} C_{1,3}^{(+)} d_i^\dagger A_k^\dagger A_l \rho_{S}^{(n)} (t) \\
- C_{0,2}^{(-)} C_{1,3}^{(-)} d_i^\dagger A_k^\dagger A_l \rho_{S}^{(n)} (t) - C_{0,2}^{(-)} C_{1,3}^{(+)} d_i^\dagger A_k^\dagger A_l \rho_{S}^{(n)} (t) \\
+ C_{0,2}^{(+)} C_{1,3}^{(-)} d_i A_k^\dagger A_l \rho_{S}^{(n)} (t) + C_{0,2}^{(+)} C_{1,3}^{(+)} d_i A_k^\dagger A_l \rho_{S}^{(n)} (t) \\
- C_{0,2}^{(+)} C_{1,3}^{(-)} d_i A_k^\dagger A_l \rho_{S}^{(n)} (t) - C_{0,2}^{(+)} C_{1,3}^{(+)} d_i A_k^\dagger A_l \rho_{S}^{(n)} (t) \\
+ C_{L,0,2}^{(-)} C_{L,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i^\dagger + C_{L,0,2}^{(-)} C_{R,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i^\dagger \\
- C_{L,0,2}^{(-)} C_{L,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i^\dagger - C_{L,0,2}^{(-)} C_{R,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i^\dagger \\
+ C_{L,0,2}^{(+)} C_{L,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i^\dagger + C_{L,0,2}^{(+)} C_{R,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i^\dagger \\
- C_{L,0,2}^{(+)} C_{L,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i^\dagger - C_{L,0,2}^{(+)} C_{R,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i^\dagger \\
+ C_{L,0,2}^{(+)} C_{L,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i + C_{L,0,2}^{(+)} C_{R,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i \\
- C_{L,0,2}^{(+)} C_{L,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i - C_{L,0,2}^{(+)} C_{R,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i \\
+ C_{L,0,2}^{(+)} C_{L,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i + C_{L,0,2}^{(+)} C_{R,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i \\
- C_{L,0,2}^{(+)} C_{L,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i - C_{L,0,2}^{(+)} C_{R,1,3}^{(+)} A_k^\dagger A_l \rho_{S}^{(n)} (t) d_i \tag{33} \\
\]

\[ I_{1,n+1} = \\
+ C_{R,0,2}^{(+)} C_{L,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n+1)} (t) d_i + C_{R,0,2}^{(+)} C_{R,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n+1)} (t) d_i \\
- C_{R,0,2}^{(+)} C_{L,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n+1)} (t) d_i - C_{R,0,2}^{(+)} C_{R,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n+1)} (t) d_i \\
+ C_{R,0,2}^{(+)} C_{L,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n+1)} (t) d_i + C_{R,0,2}^{(+)} C_{R,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n+1)} (t) d_i \\
- C_{R,0,2}^{(+)} C_{L,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n+1)} (t) d_i - C_{R,0,2}^{(+)} C_{R,1,3}^{(-)} A_k^\dagger A_l \rho_{S}^{(n+1)} (t) d_i \tag{34} \\
\]

where
\[ A_j = e^{-i \mathcal{L}_S (t-t_1)} d_j, A_j^\dagger = e^{-i \mathcal{L}_S (t-t_1)} d_j^\dagger, \tag{35} \]
\[ A_k = e^{-i \mathcal{L}_S (t-t_2)} d_k, A_k^\dagger = e^{-i \mathcal{L}_S (t-t_2)} d_k^\dagger, \tag{36} \]
\[ A_l = e^{-i \mathcal{L}_S (t-t_3)} d_l, A_l^\dagger = e^{-i \mathcal{L}_S (t-t_3)} d_l^\dagger. \tag{37} \]

According to the procedure described above, one can obtain the particle-number-resolved density matrices corresponding to Eq. (21), which is the starting point of the non-Markovian FCS calculation taking the cotunneling processes into account. Therefore, the particle num-
ber resolved quantum master equation taking into account both the sequential tunneling and cotunneling can be written as

$$\frac{\partial \rho_S^{(n)}(t)}{\partial t} = -i [H_S, \rho_S(t)] + \left. \frac{\partial \rho_S^{(n)}(t)}{\partial t} \right|_{\text{second-order}} + \left. \frac{\partial \rho_S^{(n)}(t)}{\partial t} \right|_{\text{fourth-order}}$$ (38)

D. FULL COUNTING STATISTICS

The FCS formalism based on Eq. (38) can be obtained from the cumulant generating function (CGF) $F(\chi)$ \cite{47}

$$e^{-F(\chi)} = \sum_n P(n, t) e^{in\chi},$$ (39)

where $\chi$ is the counting field, and $P(n, t) = \text{Tr} \left[ \rho_S^{(n)}(t) \right]$. Thus, one has $e^{-F(\chi)} = \text{Tr} [S(\chi, t)]$ by defining $S(\chi, t) = \sum_n \rho_S^{(n)}(t) e^{in\chi}$, where the trace is over the eigenstates of the OQS. Since Eq. (38) has the following form

$$\dot{\rho}_S^{(n)} = A\rho_S^{(n)} + C_1\rho_S^{(n+1)} + D_1\rho_S^{(n-1)} + C_2\rho_S^{(n+2)} + D_2\rho_S^{(n-2)},$$ (40)

then $S(\chi, t)$ satisfies

$$\dot{S} = AS + e^{-i\chi}C_1S + e^{i\chi}D_1S + e^{-2i\chi}C_2S + e^{2i\chi}D_2S \equiv L_\chi S,$$ (41)

where $S$ is a column matrix, and $A$, $C_1$, $D_1$, $C_2$ and $D_2$ are five square matrices. Here, for the second-order case $C_2 = D_2 = 0$, and the specific form of $L_\chi$ can be obtained by performing a discrete Fourier transformation to the matrix elements of Eq. (38).

In the low frequency limit, the low order cumulants of transferred-electron number $C_k$ can be calculated based on Eq. (41) and the Rayleigh–Schrödinger perturbation theory developed in Refs. \cite{38,41,43,46,49}. Here, the first four cumulants are directly related to the peak position (i.e., the average current $\langle I \rangle = eC_1/t$), the peak-width (i.e., shot noise characterized by Fano factor $C_2/C_1$), the skewness ($C_3/C_1$) and the kurtosis ($C_4/C_1$) of the distribution of transferred-electron number. In general, the shot noise, skewness and kurtosis are represented by the Fano factors $F_2 = C_2/C_1$, $F_3 = C_3/C_1$ and $F_4 = C_4/C_1$, respectively.
III. TRANSPORT THROUGH SIDE-COUPLED DOUBLE QD SYSTEM

A. Hamiltonian of the side-coupled double QD system

In order to facilitate discussions effectively, we consider a side-coupled double QD system weakly connected to two metallic electrodes, see Fig. 1. For the sake of simplicity, we neglect electron-spin. The Hamiltonian of the side-coupled double-QD system is described by

\[
H_{\text{dot}} = \varepsilon_1 d_1^\dagger d_1 + \varepsilon_2 d_2^\dagger d_2 + U_{12} \hat{n}_1 \hat{n}_2 - J \left( d_1^\dagger d_2 + d_2^\dagger d_1 \right),
\]  

where \( d_i^\dagger \) (\( d_i \)) is the creation (annihilation) operator of an electron with energy \( \varepsilon_i \) in \( i \)th QD, and \( U_{12} \) the interdot Coulomb repulsion between two electrons in different QDs. Here, we assume that the intradot Coulomb interaction \( U \to \infty \), thus, the double-electron occupation in different QDs is permitted only. The last term of \( H_{\text{dot}} \) describes the hopping between the two QDs with \( J \) being the hopping parameter. To facilitate the following calculation, the eigenstates of \( H_{\text{dot}} \) are used to describe the electronic states of the side-coupled double QD system. Here, The Hamiltonian \( H_{\text{dot}} \) can be diagonalized in the basis represented by the electron occupation numbers of the QD-1 and the QD-2, i.e., \( |0\rangle_1 |0\rangle_2, |1\rangle_1 |0\rangle_2, |0\rangle_1 |1\rangle_2, |1\rangle_1 |1\rangle_2 \). Consequently, the four eigenvalues of and the corresponding four eigenstates of the side-coupled double QD system are given by

\[
H_{\text{dot}} |0\rangle = 0, |0\rangle = |0\rangle_1 |0\rangle_2, 
\]

\[
H_{\text{dot}} |1\rangle^\pm = \varepsilon_\pm |1\rangle^\pm, |1\rangle^\pm = a_\pm |1\rangle_1 |0\rangle_2 + b_\pm |0\rangle_1 |1\rangle_2, 
\]

\[
H_{\text{dot}} |2\rangle = \varepsilon_{1,1} |2\rangle, |2\rangle = |1\rangle_1 |1\rangle_2, 
\]

with

\[
\varepsilon_\pm = \frac{(\varepsilon_1 + \varepsilon_2) \pm \sqrt{(\varepsilon_1 - \varepsilon_2)^2 + 4J^2}}{2}, 
\]

\[
\varepsilon_{1,1} = \varepsilon_1 + \varepsilon_2 + U_{12} 
\]

and

\[
a_\pm = \frac{\mp J}{\sqrt{(\varepsilon_\pm - \varepsilon_1)^2 + J^2}} 
\]

14
\[ b_\pm = \pm \frac{(\varepsilon_\pm - \varepsilon_1)}{\sqrt{2 + J^2}} \]  

(49)

The electron distributions of the two metallic electrodes, in which the relaxation is assumed to be sufficiently fast, are described by the equilibrium Fermi functions and the corresponding Hamiltonian reads

\[ H_{\text{electrodes}} = \sum_{\alpha k} \varepsilon_{\alpha k} a_{\alpha k}^{\dagger} a_{\alpha k} \]  

(50)

where \( a_{\alpha k}^{\dagger} (a_{\alpha k}) \) is the \( \alpha \)-electrode electron creation (annihilation) operator with energy \( \varepsilon_{\alpha k} \) and momentum \( k \).

The tunneling between the QD-1 and the two electrodes is described by

\[ H_{\text{hyb}} = \sum_{\alpha k} \left( t_{\alpha k} a_{\alpha k}^{\dagger} d_1 + t_{\alpha k}^{\ast} d_1^{\dagger} a_{\alpha k} \right), \]  

(51)

where the tunneling amplitudes \( t_{\alpha} \) and the density of states \( g_\alpha \) are assumed to be independent of wave vector and energy, thus, the electronic tunneling rate can be characterized by \( \Gamma_\alpha = 2\pi |t_{\alpha}|^2 g_\alpha \).

In the side-coupled double QD system, the quantum coherence can be tuned by modulating the magnitude of the hopping parameter \( J \) relative to the tunneling coupling strength between the QD-1 and the two electrodes. In the case of \( J \ll \Gamma (\Gamma = \Gamma_L + \Gamma_R) \), the hopping strength between the two QDs strongly modifies the internal dynamics, and the off-diagonal elements of the reduced density matrix play an essential role in the electron tunneling processes \[35, 38, 50\]; while in the regime \( J \gg \Gamma \), the off-diagonal elements of the reduced density matrix have very little influence on the electron tunneling processes \[35\]. In the following calculation, the parameters of the side-coupled double QD system are taken as \( \varepsilon_1 = \varepsilon_2 = 2.35 \), \( U_{12} = 4 \) and \( k_B T = 0.1 \) (if not explicitly stated otherwise), where the unit of energy is chosen as meV \[51\].

**B. The side-coupled double QD system with strong quantum coherence**

We first study the influences of the off-diagonal elements of the reduced density matrix, namely, the quantum coherence, and the electron cotunneling processes on the FCS of elec-
tron transport through this QD system with strong quantum coherence. For the side-coupled double QD system, the quantum coherence has an important influence on the electron sequential tunneling processes in the bias voltage range in which the transitions between the singly-occupied and empty-occupied eigenstates take place. Consequently, the following discussions focus on this bias voltage region, the applied bias voltage is here chosen as $V_b = 4.5$ based on the parameters of the QD system. To determine the dependence of the FCS on the quantum coherence and the electron cotunneling processes, we consider the average current, shot noise, skewness and kurtosis as a function of the tunneling rate $\Gamma_\alpha$ for the four different cases, (1) considering the diagonal elements of the reduced density matrix in the sequential tunneling processes only, (2) considering the diagonal and off-diagonal elements of the reduced density matrix in the sequential tunneling processes, (3) considering the diagonal elements of the reduced density matrix in the cotunneling assisted sequential tunneling processes only, (4) considering the diagonal and off-diagonal elements of the reduced density matrix in the cotunneling assisted sequential tunneling processes.

Figures 2, 3 and 4 show the influence of the temperature of the QD system on the first four current cumulants with the different values of the left-right asymmetry of the QD-electrode coupling $\Gamma_L/\Gamma_R$. In the case of the coupling of the QD-1 with the source-electrode is stronger than that of the QD-1 with the drain-electrode, i.e., $\Gamma_L/\Gamma_R > 1$, we in Fig. 2 plot the first four current cumulants as a function of the tunneling rate $\Gamma_L$ with different temperatures $k_B T$ at $\Gamma_L/\Gamma_R = 10$. We found that, in the $\Gamma/J < 1$ case, the electron cotunneling processes play a essential role in determining the values of the shot noise and high-order current cumulants; whereas in the $\Gamma/J \gg 1$ case the quantum coherence plays a crucial role in determining whether the Fano factors of the shot noise, the skewness and the kurtosis are larger than one or not, see Fig. 2. In the case of the intermediate value of $\Gamma/J$, the competition between the electron cotunneling processes and the quantum coherence takes place. This leads to the formation of a crossover region, but the range of which depends on the temperature $k_B T$, see Fig. 2.

The underlying physics of the cotunneling effect can be understood in terms of the cotunneling-induced redistribution of the occupation probabilities of the QD’s different eigenstates. In the $\Gamma_L/\Gamma_R = 10$ case, the occupation probabilities of the two singly-occupied eigenstates are much larger than that of empty-occupied eigenstate, leading to a relatively long dwell time of the conduction electron before tunneling out the QD system. When
Γ/J \ll 1$, the conduction electrons can tunnel back and forth between the two singly-occupied eigenstates very rapidly, and then enhance the cotunneling processes induced by the transitions between the doubly-occupied $|2\rangle$ and singly-occupied $|1\rangle^{\pm}$ eigenstates. Consequently, the cotunneling processes can dramatically decrease and increase the occupation probabilities of the singly-occupied and empty-occupied eigenstates with decreasing ratio of $\Gamma$ to $J$, respectively, see Figs. 5(a1)-5(a3). This indicates that the sequential-induced blocking of electron tunneling can be removed by the cotunneling processes, which leads to the shot noise being decreased, see Figs. 2(b1)-2(b3). Whereas in the $\Gamma/J \gg 1$ case the conduction electrons can tunnel back and forth between the two singly-occupied eigenstates very slowly, and then suppress the cotunneling processes. Thus, the quantum coherence has a very significant influence on the electron tunneling processes and the cotunneling-induced probability distributions for the singly-occupied and empty-occupied eigenstates have a slight variation, see Figs. 5(a1)-5(a3). In addition, the cotunneling-induced non-equilibrium electron distribution depends on the temperature $k_B T$, see Figs. 5(a1)-5(a3), which are responsible for the slight influence of the cotunneling effect on the FCS with decreasing the temperature $k_B T$.

Compared with the $\Gamma_L/\Gamma_R > 1$ case, in the case of $\Gamma_L/\Gamma_R \leq 1$, the range of the crossover region is very small, which also depends on the temperature $k_B T$, see Figs. 3 and 4. Particularly, in the cases of $\Gamma_L/\Gamma_R \leq 1$ and $\Gamma/J \gg 1$, the interplay between the electron cotunneling processes and the quantum coherence determine the FCS properties of transferred-electron number, such as, whether the super-Poissonian distributions of the shot noise, the skewness and the kurtosis ($F_i > 1$) occur or not, and whether the signs of the values of the skewness and the kurtosis become negative from a positive value or not, see Figs. 3 and 4. In particular, the magnitudes and signs of the skewness and kurtosis characterize the asymmetry of and the combined weight of the tails relative to the rest of the probability distribution of transferred-electron number, respectively. Thus, they can provide much more information for the counting statistics that the shot noise. Moreover, in the cases of $\Gamma_L/\Gamma_R = 1$ and $\Gamma/J \gg 1$, the behavior of the shot noise is mainly governed by the quantum coherence, see Figs. 4(b1), 4(b2) and 4(b3). However, these characteristics also depend on the temperature $k_B T$, i.e., the quantum coherence will play an essential role in the electron tunneling processes with decreasing temperature, see Figs. 3(a3)-3(d3) and 4(a3)-4(d3).

These properties of the $\Gamma_L/\Gamma_R \leq 1$ case can also be explained through the cotunneling-
induced redistribution of the occupation probabilities. In the $\Gamma_L/\Gamma_R = 0.1$ case, the occupation probabilities of the two singly-occupied eigenstates are much smaller than that of empty-occupied eigenstate, thus, the conduction electrons have a very short dwell time, which is contrary to the $\Gamma_L/\Gamma_R = 10$ case. In the $\Gamma/J \gg 1$ case, the two electron tunneling can occur through the cotunneling processes induced by the transitions between the doubly-occupied $|2\rangle$ and singly-occupied $|1\rangle^\pm$ eigenstates and the succeed sequential processes induced by the transitions between the singly-occupied $|1\rangle^\pm$ and empty-occupied $|0\rangle$ eigenstates. Thus, the cotunneling assisted sequential tunneling processes in the $\Gamma/J \gg 1$ case still play an important role in determining the FCS. Additionally, in the $\Gamma/J \ll 1$ case, the cotunneling processes can further increase the occupation probability of the empty-occupied eigenstate with decreasing ratio of $\Gamma$ to $J$, see Figs. 5(b1)-5(b3). This effect can further block the electron tunneling, then leading to the shot noise being relatively enhanced, see Figs. 3(b1)-3(b3).

Figures 6 and 7 show the influence of the left-right asymmetry of the QD-electrode coupling $\Gamma_L/\Gamma_R$ on the first four current cumulants for a given temperature $k_B T = 0.1$. In the cases of $\Gamma_L/\Gamma_R > 1$ and $\Gamma > J$ (the intermediate value), the range of the crossover region, in which the electron cotunneling processes decrease the values of Fano factors while the quantum coherence increase that of Fano factors, increases with increasing the ratio of $\Gamma_L$ to $\Gamma_R$, see Fig. 6. Whereas in the cases of $\Gamma_L/\Gamma_R < 1$ and $\Gamma/J \gg 1$, the interplay between the electron cotunneling processes and the quantum coherence has a relatively remarkable influence on the high-order current cumulants with decreasing the ratio of $\Gamma_L$ to $\Gamma_R$, and determines whether the super-Poissonian distributions of the shot noise and the kurtosis take place or not, and whether the transition of the skewness from positive to negative values occurs or not, see Fig. 7. These results can also be understood with the help of the redistribution of the occupation probability induced by the left-right asymmetry of the QD-electrode coupling, see Fig. 8.

C. The side-coupled double QD system with weak quantum coherence

We finally discuss the influences of the electron cotunneling processes on the first four current cumulants in the side-coupled double QD system with weak quantum coherence. Here, the hopping parameter is thus chosen as $J = 1$. According to the parameters of the
QD system, we choose the three fixed bias voltages, under which the different transitions between the QD eigenstates participate in the electron tunneling processes, namely, \( V_b = 2.5 \) corresponding to the transitions between the singly-occupied \( |1\rangle^- \) and empty-occupied eigenstates, \( V_b = 4.5 \) corresponding to the transitions between the singly-occupied \( |1\rangle^\pm \) and empty-occupied eigenstates, and \( V_b = 6.5 \) corresponding to the transitions between the singly-occupied \( |1\rangle^\pm \) and empty-occupied eigenstates and the transitions between the doubly-occupied \( |2\rangle \) and singly-occupied \( |1\rangle^+ \) eigenstates. In this situation, the properties of the first four current cumulants are well determined by the electron cotunneling processes because the quantum coherence indeed has a very small influence on the values of the first four current cumulants, see Figs. 9 and 10. In the case of \( \Gamma_L/\Gamma_R > 1 \), the electron cotunneling processes have a relatively obvious influence on the first four current cumulants, see Fig. 9; whereas in the case of \( \Gamma_L/\Gamma_R < 1 \) that have a slight influence on the first four current cumulants, see Fig. 10. It is important to note that the electron cotunneling processes do not change the intrinsic statistical properties of current cumulants, namely, whether the super-Poissonian distribution of the current cumulants take place or not, and whether the sign transitions of the values of the skewness and the kurtosis occur or not, see Figs. 9 and 10.

IV. CONCLUSIONS

We have developed an efficient non-Markovian FCS formalism taking into account both the sequential tunneling and cotunneling processes, and studied the influences of the quantum coherence and the cotunneling assisted sequential tunneling processes on the first four current cumulants in a side-coupled double QD system. In the strong quantum-coherent side-coupled double QD system, it is numerically demonstrated that, in the sequential tunneling regime, the competition or interplay between the cotunneling processes and the quantum coherence determines that whether the super-Poissonian distributions of the shot noise, the skewness and the kurtosis take place, and whether the sign transitions of the values of the skewness and the kurtosis occur. These characteristics depend on the temperature of the QD system, the left-right asymmetry of the QD-electrode coupling, and the magnitude of the coupling strengths. However, in the weak quantum-coherent side-coupled double QD system, the cotunneling processes has a relatively slight influence on the statistical properties
of current cumulants, which also depends on the left-right asymmetry of the QD-electrode coupling and the corresponding coupling strengths. Consequently, the dependence of the FCS on the quantum coherence and the cotunneling processes is necessary to be considered in the open strong quantum-coherent quantum systems, even through the electron tunneling is mainly dominated by the sequential tunneling processes.

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FIG. 1: (Color online) The open quantum system consists of a quantum-coherent-tunable side-coupled single-level double quantum-dot (QD) system weakly coupled to two electron reservoirs (electrodes). Here, $J$ and $\Gamma_\alpha$ characterize the hopping between the two QDs and the tunneling coupling between the QD-1 and the electrode $\alpha$, respectively. The QD molecule possess strong quantum coherence in the case of $J \ll \Gamma$ ($\Gamma = \Gamma_L + \Gamma_R$), whereas in the case of $J \gg \Gamma$ that possess weak quantum coherence.
FIG. 2: (Color online) The average current $\langle I \rangle$, shot noise $C_2/C_1$, skewness $C_3/C_1$ and kurtosis $C_4/C_1$ as a function of the tunneling rate $\Gamma_L$ with different temperatures of the QD system $k_BT$ at $\Gamma_L/\Gamma_R = 10$, where $C_k$ is the zero-frequency $k$-order cumulant of transferred-electron number. Here, the four different cases are considered, namely, (1) considering the diagonal elements of the reduced density matrix in the sequential tunneling processes only, denoted by second-order diagonal, (2) considering the diagonal and off-diagonal elements of the reduced density matrix in the sequential tunneling processes, denoted by second-order off-diagonal, (3) considering the diagonal elements of the reduced density matrix in the cotunneling assisted sequential tunneling processes only, denoted by fourth-order diagonal, (4) considering the diagonal and off-diagonal elements of the reduced density matrix in the cotunneling assisted sequential tunneling processes, denoted by fourth-order off-diagonal. In the case of $\Gamma/J < 1$, the properties of current cumulants are mainly governed by the electron cotunneling processes; whereas in the case of $\Gamma/J \gg 1$ that are mainly governed by the quantum coherence. In the case of the intermediate value of $\Gamma/J$, the competition between the electron cotunneling processes and the quantum coherence takes place, which leads to the formation of a crossover region. However, the range of the crossover region depends on the temperature $k_BT$. The side-coupled double QD system parameters: $\epsilon_1 = \epsilon_2 = 2.35$, $J = 0.001$, $U_{12} = 4$ and $V_b = 4.5$, where meV is chosen as the unit of energy.
FIG. 3: (Color online) The average current $\langle I \rangle$, shot noise $C_2/C_1$, skewness $C_3/C_1$ and kurtosis $C_4/C_1$ as a function of the tunneling rate $\Gamma_R$ with different temperatures of the QD system $k_B T$ at $\Gamma_L/\Gamma_R = 0.1$. In the $\Gamma/J \gg 1$ case, the interplay between the electron cotunneling processes and the quantum coherence determines whether the super-Poissonian distributions of the shot noise and the kurtosis ($F_i > 1$) occur, and whether the signs of the values of the skewness become negative from positive values, which also depends on the temperature $k_B T$. The notations and the parameters of the QD system are the same as in Fig. 2.
FIG. 4: (Color online) The average current $\langle I \rangle$, shot noise $C_2/C_1$, skewness $C_3/C_1$ and kurtosis $C_4/C_1$ as a function of the tunneling rate $\Gamma_L$ with different temperatures of the QD system $k_BT$ at $\Gamma_L/\Gamma_R = 1$. In the $\Gamma/J \gg 1$ case, the quantum coherence plays an essential role in determining whether the super-Poissonian shot noise takes place; whereas the interplay between the electron cotunneling processes and the quantum coherence determines whether the super-Poissonian distributions of the skewness and the kurtosis occur, and whether the signs of the values of the kurtosis become a large negative from a small positive values, which depends on the temperature $k_BT$. The notations and the parameters of the QD system are the same as in Fig. 2.
FIG. 5: (Color online) The occupation probabilities of the QD’s eigenstates as a function of the tunneling rate $\Gamma_L$ ($\Gamma_R$) with different values of the ratio of $\Gamma_L$ to $\Gamma_R$ and the temperature $k_B T$. The parameters of the QD system of (a1)-(a3), (b1)-(b3) and (c1)-(c3) are the same as in Figs. 2, 3 and 4, respectively.
FIG. 6: (Color online) The average current ⟨I⟩, shot noise C₂/C₁, skewness C₃/C₁ and kurtosis C₄/C₁ as a function of the tunneling rate Γₐ with different values of the ratio of Γₐ to Γᵣ at Γₐ > Γᵣ and k_BT = 0.1. In the crossover region, the electron cotunneling processes decrease the values of Fano factors, while the quantum coherence increase that of Fano factors. However, the range of the crossover region increases with increasing the ratio of Γₐ to Γᵣ. The notations and the parameters of the QD system are the same as in Fig. 2.
FIG. 7: (Color online) The average current $\langle I \rangle$, shot noise $C_2/C_1$, skewness $C_3/C_1$ and kurtosis $C_4/C_1$ as a function of the tunneling rate $\Gamma_R$ with different values of the ratio of $\Gamma_L$ to $\Gamma_R$ at $\Gamma_L < \Gamma_R$ and $k_B T = 0.1$. In the $\Gamma/J \gg 1$ case, the interplay between the electron cotunneling processes and the quantum coherence determines whether the super-Poissonian distributions of the shot noise and the kurtosis take place, and whether the transition of the skewness from positive to negative values occurs. The notations and the parameters of the QD system are the same as in Fig. 2.
FIG. 8: (Color online) The occupation probabilities of the QD’s eigenstates as a function of the tunneling rate $\Gamma_L$ ($\Gamma_R$) with different values of the ratio of $\Gamma_L$ to $\Gamma_R$. The parameters of the QD system of (a1)-(a3) and (b1)-(b3) are the same as in Figs. 5 and 6, respectively.
FIG. 9: (Color online) The average current $\langle I \rangle$, shot noise $C_2/C_1$, skewness $C_3/C_1$ and kurtosis $C_4/C_1$ as a function of the tunneling rate $\Gamma_L$ with different bias voltages $V_b$ at $k_BT = 0.1$ and $\Gamma_R = 0.001$. Here, the three fixed bias voltages, under which the different transitions involved in the electron tunneling, are considered, namely, (1) $V_b = 2.5$ corresponding to the transitions between the singly-occupied $|1\rangle^-$ and empty-occupied eigenstates, (2) $V_b = 4.5$ corresponding to the transitions between the singly-occupied $|1\rangle^\pm$ and empty-occupied eigenstates, (3) $V_b = 6.5$ corresponding to the transitions between the singly-occupied $|1\rangle^\pm$ and empty-occupied eigenstates, and the transitions between the doubly-occupied $|2\rangle$ and singly-occupied $|1\rangle^+$ eigenstates. In the case of $\Gamma_L/\Gamma_R > 1$, the electron cotunneling processes have a relatively obvious influence on the first four order current cumulants. The other notations and the parameters of the QD system are the same as in Fig. 2.
FIG. 10: (Color online) The average current $\langle I \rangle$, shot noise $C_2/C_1$, skewness $C_3/C_1$ and kurtosis $C_4/C_1$ as a function of the tunneling rate $\Gamma_R$ with different bias voltages $V_b$ at $k_B T = 0.1$ and $\Gamma_L = 0.001$. In the case of $\Gamma_L/\Gamma_R < 1$, the electron cotunneling processes have a slight influence on the first four order current cumulants. The other notations and the parameters of the QD system are the same as in Fig. 2.