Non-Markovian full counting statistics in quantum dot molecules

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Full counting statistics of electron transport is a powerful diagnostic tool for probing the nature of quantum transport beyond what is obtainable from the average current or conductance measurement alone. In particular, the non-Markovian dynamics of quantum dot molecule plays an important role in the nonequilibrium electron tunneling processes. It is thus necessary to understand the non-Markovian full counting statistics in a quantum dot molecule. Here we study the non-Markovian full counting statistics in two typical quantum dot molecules, namely, serially coupled and side-coupled double quantum dots with high quantum coherence in a certain parameter regime. We demonstrate that the non-Markovian effect manifests itself through the quantum coherence of the quantum dot molecule system, and has a significant impact on the full counting statistics in the high quantum-coherent quantum dot molecule system, which depends on the coupling of the quantum dot molecule system with the source and drain electrodes. The results indicated that the influence of the non-Markovian effect on the full counting statistics of electron transport, which should be considered in a high quantum-coherent quantum dot molecule system, can provide a better understanding of electron transport through quantum dot molecules.

Full counting statistics (FCS) of electron transport through mesoscopic system has attracted considerable attention both experimentally and theoretically because it can provide a deeper insight into the nature of electron transport mechanisms, which cannot be obtained from the average current. For instance, the shot noise measurements can be used to probe the dynamical in an open double quantum dots (QDs), the coherent coupling between serially coupled QDs, the evolution of the Kondo effect in a QD, and the conduction channels of quantum conductors. In particular, shot noise characteristics can provide information about the feature of the pseudospin Kondo effect in a laterally coupled double QDs, the spin accumulations in a electron reservoir, and the charge fractionalization in the \( v = 2 \) quantum Hall edge. In addition, the degree of entanglement of two electrons in the double QDs, the dephasing rate in a closed QD, the internal level structure of single molecule magnet, can be characterized by the super-Poissonian shot noise.

On the other hand, the quantum coherence in coupled QD system, which is characterized by the off-diagonal elements of the reduced density matrix of the QD system within the framework of the density matrix theory, plays an important role in the electron tunneling processes and has a significant influence on electron transport. In particular, theoretical studies have demonstrated that the high-order cumulants, e.g., the shot noise, the skewness, are more sensitive to the quantum coherence than the average current in the different types of QD systems, and the quantum coherence information in a side-coupled double QD system can be extracted from the high-order current cumulants. In fact, the non-Markovian dynamics of the QD system also plays an important role in the nonequilibrium electron tunneling processes. However, the above studies on current noise or FCS were mainly based on the different types of Markovian master equations. Although the influence of non-Markovian effect on the long-time limit of the FCS in the QD systems has received some attention, how the non-Markovian effect affects the FCS is still an open issue, especially the influence of the interplay between the quantum coherence and non-Markovian effect on the long-time limit of the FCS has not yet been revealed.

The aim of this report is thus to derive a non-Markovian FCS formalism based on the exact time-convolution-less (TCL) master equation and study the influences of the quantum coherence and non-Markovian effect on the FCS in QD molecule systems. It is demonstrated that the non-Markovian effect manifests itself through the quantum coherence of the considered QD molecule system, and has a significant impact on the FCS in the high quantum-coherent QD molecule system, which depends on the coupling of the considered QD molecule system with the incident and outgoing electrodes. Consequently, it is necessary to consider the influence of the
non-Markovian effect on the full counting statistics of electron transport in a high quantum-coherent single-molecule system.

**Results**

We now study the influences of the quantum coherence and non-Markovian effect on the FCS of electronic transport through the QD molecule system. In order to facilitate discussions effectively, we consider three typical QD systems, namely, single QD without quantum coherence, serially coupled double QDs and side-coupled double QDs with high quantum coherence in a certain parameter regime (see Fig. 1). In addition, we assume the bias voltage \( \mu_L = -\mu_R = V_B/2 \) is symmetrically entirely dropped at the QD-electrode tunnel junctions, which implies that the levels of the QDs are independent of the applied bias voltage even if the couplings are not symmetric, and choose meV as the unit of energy which corresponds to a typical experimental situation.

Single quantum dot without quantum coherence. In this subsection, we consider a single QD weakly coupled to two ferromagnetic electrodes. The Hamiltonian of the considered system is described by the Hamiltonian \( H_{total} = H_{dot} + H_{leads} + H_T \). The QD Hamiltonian \( H_{dot} \) is given by

\[
H_{dot,1} = \sum_{s} \varepsilon_s d_s^\dagger d_s + U d_1^\dagger d_1 d_1^\dagger d_1^\dagger,
\]

where \( d_s^\dagger (d_s) \) creates (annihilates) an electron with spin \( s \) and on-site energy \( \varepsilon_s \) which can be tuned by a gate voltage \( V_B \) in this QD system. \( U \) is the intradot Coulomb interaction between two electrons in the QD system.

The relaxation in the two ferromagnetic electrodes is assumed to be sufficiently fast, so that their electron distributions can be described by equilibrium Fermi functions. The two electrodes are thus modeled as non-interacting Fermi gases and the corresponding Hamiltonians can be expressed as

\[
H_{leads,1} = \sum_{s k \delta \bar{\delta}} \varepsilon_{s k \delta \bar{\delta}} a_{s k \delta \bar{\delta}}^\dagger a_{s k \delta \bar{\delta}},
\]

where \( a_{s k \delta \bar{\delta}}^\dagger (a_{s k \delta \bar{\delta}}) \) creates (annihilates) an electron with energy \( \varepsilon_{s k} \), spin \( s \) and momentum \( k \) in \( \delta \) (\( \delta \) = \( L \), \( R \)) electrode, and \( s = + (-) \) denotes the majority (minority) spin states with the density of states \( g_{s,\delta} \). The polarization vectors \( p_L \) (left lead) and \( p_R \) (right lead) parallel to each other, and their magnitudes are characterized by \( p_\delta = |p_\delta| = (g_{s,\delta} - g_{s,\bar{\delta}})/(g_{s,\delta} + g_{s,\bar{\delta}}) \). The tunneling between the QD and the electrodes is described by

\[
H_{T,1} = t_{L1} a_{L1}^\dagger d_1 + t_{R1} a_{R1}^\dagger d_1^\dagger + t_{L1} a_{L1}^\dagger d_1 + t_{R1} a_{R1}^\dagger d_1^\dagger + H.c.,
\]

where spin-up \( \uparrow \) and spin-down \( \downarrow \) are defined to be the majority spin and minority spin of the ferromagnet, respectively.

The QD-electrode coupling is assumed to be sufficiently weak, thus, the sequential tunneling is dominant and can be well described by the quantum master equation of reduced density matrix spanned by the eigenstates of the QD. The particle-number-resolved TCL quantum master equation for the reduced density matrix of the considered single QD is given by

\[
\dot{\rho}^{(a)}(t)_{dot,1} = -i\mathcal{L}\rho^{(a)} - \sum_{a \sigma} \left[ a_{L1}^\dagger a_{L1} \rho^{(a)} - a_{R1}^\dagger a_{R1} \rho^{(a)} - a_{L1}^\dagger a_{L1} \rho^{(a)} + a_{R1}^\dagger a_{R1} \rho^{(a)} \right] + H.c.,
\]

For more details, see Methods section. Here, the complete basis \( \{ |0,0\rangle, |1,0\rangle, |0,1\rangle, |1,1\rangle \) is chosen to describe the electronic states of this single QD system, and the single QD system parameters are chosen as \( \varepsilon_1 = \varepsilon_1 = 1 \), \( U = 5 \), \( p = 0.9 \) and \( k_B T = 0.04 \).

Figure 1 shows the first four current cumulants as a function of the bias voltage \( V_B \). Here, the two QD molecules possess high quantum coherence in the case of \( \Delta \ll k_B T \) (\( \Delta \) being the singly-occupied eigenenergy separation, \( k_B \) the Boltzmann constant, \( T \) the temperature of the QDs system). The hopping coupling between the two QDs, and the strength of coupling between the QDs system and the electrode \( \sigma \), are characterized by \( f \) and \( \Gamma_\sigma \), respectively.
Figure 2 | The average current ($\langle I \rangle$), shot noise ($C_S/C_V$), skewness ($C_A/C_V$) and kurtosis ($C_D/C_V$) versus bias voltage for the Markovian and the non-Markovian case at different coupling of the single QD with two ferromagnetic electrodes, respectively. Here, $C_V$ is the zero-frequency k-order cumulant of current fluctuations. The non-Markovian effect has no influence on the first four current cumulants of the considered single QD. The single QD system parameters: $\varepsilon_1 = \varepsilon_2 = 1$, $U = 5$, $p = 0.9$ and $k_B T = 0.04$.

$$\langle 0|\hat{\rho}_{2S}(t)|0\rangle$$

$$= - \left[ \Gamma_{L}\hat{f}_{L} + (\varepsilon_{1}) + \Gamma_{R}\hat{f}_{R} + (\varepsilon_{1}) \right] \langle 0|\rho_{S}(t)|0\rangle$$

$$+ \left[ \Gamma_{L}\hat{f}_{L} - (\varepsilon_{1}) + \Gamma_{R}\hat{f}_{R} - (\varepsilon_{1}) \right] \langle 0|\rho_{S}(t)|1\rangle$$

$$+ \left[ \Gamma_{L}\hat{f}_{L} - (\varepsilon_{1} - \varepsilon_{1}) + \Gamma_{R}\hat{f}_{R} - (\varepsilon_{1} - \varepsilon_{1}) \right] \langle 0|\rho_{S}(t)|\uparrow\downarrow\rangle$$

$$+ \left[ \Gamma_{L}\hat{f}_{L} - (\varepsilon_{1} - \varepsilon_{1}) + \Gamma_{R}\hat{f}_{R} - (\varepsilon_{1} - \varepsilon_{1}) \right] \langle 1|\rho_{S}(t)|\uparrow\downarrow\rangle,$$

$$\langle \downarrow|\hat{\rho}_{2S}(t)|\uparrow\downarrow\rangle$$

$$= \left[ \Gamma_{L}\hat{f}_{L} + (\varepsilon_{1}) + \Gamma_{R}\hat{f}_{R} + (\varepsilon_{1}) \right] \langle 0|\rho_{S}(t)|0\rangle$$

$$- \left[ \Gamma_{L}\hat{f}_{L} - (\varepsilon_{1}) + \Gamma_{R}\hat{f}_{R} - (\varepsilon_{1}) \right] \langle 0|\rho_{S}(t)|1\rangle$$

$$- \left[ \Gamma_{L}\hat{f}_{L} - (\varepsilon_{1} - \varepsilon_{1}) + \Gamma_{R}\hat{f}_{R} - (\varepsilon_{1} - \varepsilon_{1}) \right] \langle 0|\rho_{S}(t)|\uparrow\downarrow\rangle$$

$$+ \left[ \Gamma_{L}\hat{f}_{L} - (\varepsilon_{1} - \varepsilon_{1}) + \Gamma_{R}\hat{f}_{R} - (\varepsilon_{1} - \varepsilon_{1}) \right] \langle 1|\rho_{S}(t)|\uparrow\downarrow\rangle.$$

Here, $f_{x+}$ is the Fermi function of the electrode $x$, and $f_{x-} = 1 - f_{x+}$. The detailed procedure for calculation of the equation of motion of a reduced density matrix, see Methods section. Within the framework of the density matrix theory, the off-diagonal elements of the reduced density matrix characterize the quantum coherence of the considered QD system. Thus, the influence of the non-Markovian effect on the FCS may be associated with the quantum coherence of the considered QD system. In order to confirm this conclusion, we take serially coupled and side-coupled double QDs for illustration in the following two subsection.

**Serially coupled double quantum dots with high quantum coherence.**

We now consider two serially coupled double QDs weakly connected to two metallic electrodes, see Fig. 1(a). For the sake of simplicity, the spin degree of freedom has not been considered. The double-QD is described by a spinless Hamiltonian

$$H_{\text{dot},2} = \varepsilon_{1} d_{1}^{\dagger} d_{1} + \varepsilon_{2} d_{2}^{\dagger} d_{2} + U n_{1} n_{2} - J \left( d_{1}^{\dagger} d_{2} + d_{2}^{\dagger} d_{1} \right),$$

where $d_{i}^{\dagger}$ ($d_{i}$) creates (annihilates) an electron with energy $\varepsilon_{i}$ (which can be tuned by a gate voltage $V_g$) in ith QD. $U$ is the interdot Coulomb repulsion between two electrons in the double QD system, where we consider the intradot Coulomb interaction $U \rightarrow \infty$, so that the double-electron occupation in the same QD is prohibited. The last term of $H_{\text{dot}}$ describes the hopping coupling between the two dots with $J$ being the hopping parameter. The two metallic electrodes are modeled as non-interacting Fermi gases and the corresponding Hamiltonians are given by

$$H_{\text{leads},2} = \sum_{k} \varepsilon_{k} a_{k}^{\dagger} a_{k},$$

where $a_{k}^{\dagger}$ ($a_{k}$) creates (annihilates) an electron with energy $\varepsilon_{k}$ and momentum $k$ in $x (x = L, R)$ electrode. The tunneling between the double QDs and the two electrodes is described by
For the case of the weak QD-electrode coupling, the particle-number-resolved TCL quantum master equation for the reduced density matrix of the considered serially double-QD system reads

\[
\rho^{(n)}(t)_{\text{dot,2}} = -i[H_{\text{dot,2}}^{(n)}, \rho^{(n)}(t)] - \left[ d^+_2 A_R^{(-)} \rho^{(n)}(t) + \rho^{(n)}(t) A_L^{(+)} d^+_1 \right] \rho^{(n)}(t) + \rho^{(n)}(t) A_L^{(+)} d^+_1 - d^+_1 \rho^{(n)}(t) A_L^{(+)} - A_L^{(-)} \rho^{(n-1)}(t) d^+_1 - d^+_1 \rho^{(n)}(t) A_L^{(+)} + H.c. \]
\]

Here, we can diagonalize the serially coupled double QDs Hamiltonian \( H_{\text{dot,2}} \) in the basis represented by the electron occupation numbers in the QD-1 and QD-2 denoted respectively by \( N_L \) and \( N_R \), namely, \([0,0], [1,0], [0,1], [1,1]\), and obtain the corresponding four eigenstates of the considered serially coupled double QDs system:

\[
H_{\text{dot,2}}[0] = 0, [1] = 0, [1] = a_{\pm}[1,0] + b_{\pm}[0,1], \quad H_{\text{dot,2}}[2] = \epsilon_{\pm}[1,1] = (\epsilon_1 + \epsilon_2 + U)[2,2], [2] = [1,1],
\]

with

\[
\epsilon_{\pm} = \frac{(\epsilon_1 + \epsilon_2) \pm \sqrt{(\epsilon_1 - \epsilon_2)^2 + 4J^2}}{2},
\]

and

\[
a_{\pm} = \frac{\mp J}{\sqrt{(\epsilon_{\pm} - \epsilon_1)^2 + J^2}}, \quad b_{\pm} = \frac{\pm (\epsilon_{\pm} - \epsilon_1)}{\sqrt{(\epsilon_{\pm} - \epsilon_1)^2 + J^2}}.
\]

Here, we focus on the regime \((\epsilon_+ - \epsilon_-) \ll k_B T\), where the hopping coupling 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.

In the following numerical calculations, thus, we assume the parameters of the serially coupled double QDs system are chosen as \( \epsilon_1 = \epsilon_2 = 1 \), \( J = 0.001 \), \( U = 4 \) and \( k_B T = 0.05 \).

When the coupling of the QD-2 with the right (drain) electrode is stronger than that of the QD-1 with the left (source) electrode, namely, \( \Gamma_L/\Gamma_R < 1 \), we plot the first four current cumulants as a function of the bias voltage for different values of the QD-2-electrode coupling \( \Gamma_R \) at \( \Gamma_L/\Gamma_R = 0.1 \) in Figs. 3(a)–3(d). We found that the non-Markovian effect has a very weak influence on the FCS. Interestingly, the high-order current cumulants the skewness and the kurtosis can show the tiny differences, see Figs. 3(c) and 3(d).

Whereas for the \( \Gamma_L/\Gamma_R \gg 1 \) case, the non-Markovian effect has a significant impact on the FCS, see Fig. 4. Especially, for a relatively large value of the ratio \( \Gamma_L/\Gamma_R = 10 \) and the coupling of the QD-1 with the left electrode being stronger than the hoping.
coupling, namely, $\Gamma_L/\Gamma_R > 1$, the non-Markovian effect can induce a strong negative differential conductance (NDC) and super-Poissonian noise, see Figs. 4(e) and 4(f). In addition, in the case of $\Gamma_L/\Gamma_R \approx 1$ and $\Gamma_L/\Gamma_R > 1$, the transitions of the skewness and the kurtosis from positive (negative) to negative (positive) values are observed, see the dotted line in Fig. 4(c), the dotted and dash-dot-dotted lines in Fig. 4(d), and the dash-dot-dotted line in Fig. 4(h). It is well known that the skewness and the kurtosis (both its magnitude and sign) characterize, respectively, the asymmetry of and the peakedness of the distribution around the average transferred-electron number $n$ during a time interval $t$, thus that provides further information for the counting statistics beyond the shot noise. To discuss the underlying mechanisms of the current noise clearly, for the system parameters considered here, the two singly-occupied eigenstates and eigenvalues can be expressed as

$$|1\rangle^\pm = \mp \frac{\sqrt{2}}{2} |1,0\rangle + \frac{\sqrt{2}}{2} |0,1\rangle,$$

(16)

Here we have utilized the equations $\epsilon_1 = \epsilon_2 = \epsilon$ and $\epsilon \gg J$. In this situation, the equations of motion of the six elements of the reduced density matrix are given by

$$
\begin{align*}
\langle 0,0 | \rho_S(t) | 0,0 \rangle \\
= & - \frac{1}{2} \left[ \Gamma_{\ell R} + (e + \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle + \\
& \frac{1}{2} \left[ \Gamma_{\ell L} - (e - \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle + \\
& - \frac{1}{2} \left[ \Gamma_{\ell R} - (e - \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle - \\
& \frac{1}{2} \left[ \Gamma_{\ell L} - (e + \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle + \\
& + \frac{1}{2} \left[ \Gamma_{\ell L} + (e - \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle - \\
& + \frac{1}{2} \left[ \Gamma_{\ell R} + (e + \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle
\end{align*}
$$

(17)

$$
\begin{align*}
\langle 1 | \pm \rho_S^n(t) | 1 \rangle^\pm \\
= & \frac{1}{2} \left[ \Gamma_{\ell L} + (e + \Gamma_R) \left\{ \rho_S^n(t) - \rho_S^n(t) \right\} \rangle + \langle 0,0 | \rho_S(t) | 0,0 \rangle \\
& - \frac{1}{2} \left[ \Gamma_{\ell R} + (e - \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle - \\
& - \frac{1}{2} \left[ \Gamma_{\ell L} + (e - \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle - \\
& - \frac{1}{2} \left[ \Gamma_{\ell R} + (e + \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle + \\
& + \frac{1}{2} \left[ \Gamma_{\ell L} + (e + \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle + \\
& + \frac{1}{2} \left[ \Gamma_{\ell R} + (e - \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle - \\
& + \frac{1}{2} \left[ \Gamma_{\ell L} + (e - \Gamma_R) \right] \langle 1 | \rho_S(t) | 1 \rangle
\end{align*}
$$

(18)

Figure 4 | The average current ($\langle I \rangle$), shot noise ($C_2/C_1$), skewness ($C_3/C_1$) and kurtosis ($C_4/C_1$) versus bias voltage for the Markovian and the non-Markovian case at different values of the QD-2-electrode coupling $\Gamma_R$. (a)–(d) for $\Gamma_L/\Gamma_R = 1$,(e)–(h) for $\Gamma_L/\Gamma_R = 10$. Here, $C_i$ is the zero-frequency $i$-th order cumulant of current fluctuations. The non-Markovian effect in the $\Gamma_L/\Gamma_R \approx 1$ case has a significant impact on the first four cumulants of transport current. The serially coupled double QDs system parameters: $\epsilon_1 = \epsilon_2 = 1$, $J = 0.001$, $U = 4$ and $k_B T = 0.05$. 

5
\[
\langle 1,1|\hat{\rho}_{5}(t)|1,1\rangle = \frac{1}{2} \left[ \Gamma I_{L}^{1} + (e + U) + \Gamma R I_{R}^{1} + (e + U) e^{-i\phi}\right] \langle 1| \hat{\rho}_{5}(t)|1\rangle^{+} \\
+ \frac{1}{2} \left[ \Gamma I_{L}^{1} + (e + U) - \Gamma R I_{R}^{1} + (e + U) e^{i\phi}\right] \langle 1| \hat{\rho}_{5}(t)|1\rangle^{-} \\
+ \frac{1}{2} \left[ \Gamma I_{L}^{1} + (e + U) - \Gamma R I_{R}^{1} + (e + U) e^{i\phi}\right] \langle 1| \hat{\rho}_{5}(t)|1\rangle^{-} \\
- \left[ \Gamma I_{L}^{1} - (e + U) + \Gamma R I_{R}^{1} - (e + U) e^{-i\phi}\right] \langle 1,1|\hat{\rho}_{5}(t)|1,1\rangle,
\]

where \(\Phi_{x} = \Phi_{x}(\epsilon + U) - \Phi_{x}(\epsilon), \Phi_{y} = \Phi_{y}(\epsilon + U) - \Phi_{y}(\epsilon).\) Compared with the Markovian case, it is obvious that the non-Markovian effect manifests itself through the off-diagonal elements of the reduced density matrix, namely, the quantum coherence of the considered QDs system. In Fig. 5(a), we plot the functions \(\Phi_{1} = -0.1\Phi_{R}(\Gamma_{R} = 0.1\Gamma_{L}), \Phi_{2} = \Phi_{R}(\Gamma_{R} = 0.1\Gamma_{L})\) and \(0.1\Phi_{1} - \Phi_{R}(\Gamma_{R} = 0.1\Gamma_{R})\) as a function of bias voltage. It is clearly evident that the values of the functions \(\Phi_{1} = -0.1\Phi_{R}\) and \(\Phi_{2} - \Phi_{R}\) show significant variations with increasing bias voltage, especially in the vicinity of the bias voltages \(V_{b} = 2\) and \(V_{b} = 10\) because the new transport channels begin to participate in quantum transport; while \(0.1\Phi_{1} - \Phi_{R}\) has a gentle variation. Consequently, the non-Markovian effects in the \(\Gamma_{I}/\Gamma_{R} \geq 1\) case have a remarkable impact on the FCS, see Fig. 4. Moreover, for \(\Gamma_{I}/\Gamma_{R} < 10\) case, the non-Markovian effect has a more significant impact on the FCS than the Markovian case, it is obvious that the non-Markovian effect on the FCS depends on the quantum coherence of the considered QD system. To prove whether this conclusion is universal or not, we take side-coupled double QDs for further illustration in the following subsection.

**Side-coupled double quantum dots with high quantum coherence.** We consider here a side-coupled double QDs system. In this case, the QD-1 is only weakly coupled to the two electrodes, see Fig. 1(b). The QD-electrode tunneling is thus described by

\[
\hat{H}_{T,3} = \sum_{\alpha,k} \left( t_{\alpha k} a_{\alpha k}^{\dagger} d_{1} + t_{\alpha k}^{*} d_{1}^{\dagger} a_{\alpha k} \right).
\]

In the case of the QD-electrode weak coupling, the particle-number-resolved TCL quantum master equation for the side-coupled double QDs can be expressed as

\[
\dot{\rho}(t) = -i\mathcal{L}_{3}\rho(t) - \left[ d_{1}^{\dagger} A_{11}^{(-)} \rho(t) + d_{1} A_{11}^{(-)} \rho(t) \right] \\
+ \rho(t) A_{11}^{(+)} d_{1}^{\dagger} + \rho(t) A_{11}^{(+)} d_{1}^{\dagger} A_{11}^{(-)} A_{11}^{(-)} d_{1}^{\dagger} \\
- A_{11}^{(-)} \rho(t) A_{11}^{(+)} d_{1}^{\dagger} - d_{1}^{\dagger} \rho(t) A_{11}^{(+)} A_{11}^{(-)} d_{1}^{\dagger} \\
- d_{1}^{\dagger} \rho(t) A_{11}^{(+)} + H.c.
\]

Here, the eigenstates and eigenvalues of the side-coupled double QDs system are the same as the serially coupled double QDs system. In the following numerical calculations, the parameters of the side-coupled QDs system are chosen as \(\epsilon_{1} = \epsilon_{2} = 1, J = 0.001, U = 5\) and \(k_{B}T = 0.1.\)

**Figure 5** | (a) The functions \(\Phi_{x} = \Phi_{x}(\epsilon + U) - \Phi_{x}(\epsilon), \Phi_{y} = \Phi_{y}(\epsilon + U) - \Phi_{y}(\epsilon)\) as a function of bias voltage with \(U = 4\) and \(k_{B}T = 0.05.\) (b) The functions \(\Phi_{x} = 0.1\Phi_{R}(\Gamma_{R} = 0.1\Gamma_{L}), \Phi_{y} = \Phi_{R}(\Gamma_{R} = 0.1\Gamma_{L})\) and \(0.1\Phi_{x} - \Phi_{R}(\Gamma_{R} = 0.1\Gamma_{R})\) as a function of bias voltage with \(U = 5\) and \(k_{B}T = 0.1.\)
For the present side-coupled QDs system with high quantum coherence, we find that for \( \Gamma_j/\Gamma_R \approx 1 \) case the non-Markovian effect has a more remarkable impact on the FCS than that in the serially coupled double QDs system, but the NDC does not appear, see Figs. 4 and 6. For instance, in the case of \( \Gamma_j/\Gamma_R > 1 \) and \( \Gamma_j/\Gamma_R = 1 \), the non-Markovian effect can further enhance the super-Poissonian shot noise, see the dotted and dash-dot-dotted lines in Fig. 6(b); and the transitions of the skewness and the kurtosis from a relatively small positive to a large negative values take place, especially for a relatively large value \( \Gamma_j/\Gamma_R \) the kurtosis can be further decreased to a very large negative value, see the dotted and dash-dot-dotted lines in Figs. 6(c) and 6(d). While for the \( \Gamma_j/\Gamma_R > 1 \) and \( \Gamma_j/\Gamma_R = 10 \) case the non-Markovian effect can enhance the shot noise to a super-Poissonian value, see the dotted and dash-dot-dotted lines in Fig. 6(f), and the transition of the kurtosis from small positive to large negative values only takes place, see the dotted and dash-dot-dotted lines in Fig. 6(h).

For the system parameters considered here, namely, in the limit of \( \epsilon_1 = \epsilon_2 = \epsilon = J \), the equations of motion of the six elements of the reduced density matrix read

\[
\rho(t) = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \rho_1(t) & 0 & 0 & 0 \\
0 & 0 & 0 & \rho_2(t) & 0 & 0 \\
0 & 0 & 0 & 0 & \rho_3(t) & 0 \\
0 & 0 & 0 & 0 & 0 & \rho_4(t)
\end{pmatrix}
\]

\[
\begin{align*}
\rho_1(t) &= \frac{1}{2} \left[ \Gamma f_{L_+} + (e) + \Gamma f_{R_+} - (e) \right] \rho_1(t) \\
\rho_2(t) &= \frac{1}{2} \left[ \Gamma f_{L_-} - (e) + \Gamma f_{R_-} - (e) \right] \rho_2(t) \\
\rho_3(t) &= \frac{1}{2} \left[ \Gamma f_{L_+} - (e) + \Gamma f_{R_+} - (e) \right] \rho_3(t) \\
\rho_4(t) &= \frac{1}{2} \left[ \Gamma f_{L_-} + (e) + \Gamma f_{R_-} + (e) \right] \rho_4(t)
\end{align*}
\]

Figure 6 | The average current (\( \langle I \rangle \)), shot noise (\( C_S/C_V \)), skewness (\( C_3/C_V \)) and kurtosis (\( C_4/C_V \)) versus bias voltage for the Markovian and the non-Markovian case at different values of the QD-1-electrode coupling \( \Gamma_R \) (a)–(d) for \( \Gamma_j/\Gamma_R = 1 \), (e)–(h) for \( \Gamma_j/\Gamma_R = 10 \). Here, \( C_i \) is the zero-frequency \( k \)-order cumulant of current fluctuations. The non-Markovian effect in the \( \Gamma_j/\Gamma_R \approx 1 \) case has a more remarkable impact on the first four cumulants of transport current than that in the serially coupled double QDs system, but the NDC does not appear. The side-coupled double QDs system parameters: \( \epsilon_1 = \epsilon_2 = J = 0.001 \), \( U = 5 \) and \( k_B T = 0.1 \).
\[
\langle 1,1|\hat{\rho}_S(t)|1,1 \rangle \\
= \frac{1}{2} \left[ \Gamma_L f_L + (e + U) + \Gamma_R f_R + (e + U)e^{-i\omega t} \right] \langle 1|^{+} \rho_S(t)|1 \rangle^{+} \\
+ \frac{1}{2} \left[ \Gamma_L f_L + (e + U) + \Gamma_R f_R + (e + U)e^{-i\omega t} \right] \langle 1|^{-} \rho_S(t)|1 \rangle^{-} \\
+ \frac{1}{2} \left[ \Gamma_L f_L + (e + U) + \Gamma_R f_R + (e + U)e^{-i\omega t} \right] \langle 1|^{+} \rho_S(t)|1 \rangle^{+} \\
+ \frac{1}{2} \left[ \Gamma_L f_L + (e + U) + \Gamma_R f_R + (e + U)e^{-i\omega t} \right] \langle 1|^{-} \rho_S(t)|1 \rangle^{-} \\
- \langle \Gamma_L f_L - (e + U) + \Gamma_R f_R - (e + U) \rangle\langle 1|\rho_S(t)|1 \rangle \\
\text{(26)}
\]

From the above four equations, we find that these characteristics also originate from the quantum coherence of the side-coupled double QDs, and can also be understood in terms of the functions \( \Phi_L + 0.1\Phi_R + \Phi_L + \Phi_R \), which have considerable variations in the vicinity of the bias voltages \( V_S = 2 \) and \( V_D = 12 \) because the new transport channels begin to enter the bias voltage window, see the solid and dashed lines in Fig. 5(b). As for the \( \Gamma_L/\Gamma_R < 1 \) case the non-Markovian effect has a slightly influence on the FCS because the function \( 0.1\Phi_L + \Phi_R \) has a gentle variation with bias voltage, see the dotted line in Fig. 5(b), which is the same as the serially coupled double QDs system, see Figs. 3(a)–3(d) and 7.

In addition, it should be pointed out that for \( \Gamma_L/\Gamma_R = 1 \) the non-Markovian effect has a stronger impact on the FCS than that for \( \Gamma_L/\Gamma_R > 1 \) case, which is contrary to the case of the serially coupled double QDs system. For the the side-coupled double QDs system, the quantum coherence originates from the quantum interference between the direct electron tunneling process, namely, the conduction-electron tunneling into the QD-1 and then directly tunneling out of the QD-1 onto the drain electrode, and the indirect tunneling process, namely, the conduction-electron from the source electrode first tunneling from the QD-1 to the QD-2, then tunneling back into the QD-1, and at last tunneling out of the QD-1. Thus, the fast direct tunneling process in the \( \Gamma_L = 10\Gamma_R \) case can be suppressed compared with the \( \Gamma_L = \Gamma_R \) case, which leads to the non-Markovian effect has a relatively strong impact on the FCS in the \( \Gamma_L/\Gamma_R = 1 \) case.

**Discussion**

We have developed a non-Markovian FCS formalism based on the exact TCL master equation, and studied the influence of the interplay between the quantum coherence and non-Markovian effect on the long-time limit of the FCS in three QD systems, namely, single QD, serially coupled double QDs and side-coupled double QDs. It is demonstrated that the non-Markovian effect manifests itself through the quantum coherence of the considered QD molecule system, and especially has a significant impact on the FCS in the high quantum-coherent QD molecule system, which depends on the coupling of the considered QD molecule system with the source and drain electrodes. For the single QD system without quantum coherence, the non-Markovian effect has no influence on the current noise properties; whereas for the serially coupled and side-coupled double QDs systems with high quantum coherence, that has a remarkable impact on the FCS when the coupling of the considered QD molecule with the incident electrode is equal to or stronger than that with the outgoing electrode. For instance, for the high quantum-coherent serially coupled double QDs system, the non-Markovian effect can induce a strong NDC and change the shot noise from the sub-Poissonian to super-Poissonian distribution in the case of \( \Gamma_L/\Gamma_R \gg 1 \) and \( \Gamma_L > \Gamma_R \) while for the high quantum-coherent side-coupled double QDs system, that can remarkably enhance the super-Poissonian noise or the sub-Poissonian noise for the \( \Gamma_L/\Gamma_R \simeq 1 \) case. Moreover, the non-Markovian effect can also lead to the occurrences of the skewness and kurtosis from small positive to large negative values. These results indicated that the influence of the non-Markovian effect on

**Figure 7** The average current (\( \langle I \rangle \)), shot noise (\( C_2/C_1 \)), skewness (\( C_3/C_1 \)) and kurtosis (\( C_4/C_1 \)) versus bias voltage for the Markovian and the non-Markovian case at different values of the QD-1-electrode coupling \( \Gamma_R \) with \( \Gamma_L/\Gamma_R = 0.1 \). Here, \( C_k \) is the zero-frequency \( k \)-order cumulant of current fluctuations. The non-Markovian effect in the \( \Gamma_L/\Gamma_R = 0.1 \) case has a slightly influence on the first four current cumulants. The other system parameters are the same as in Fig. 6.
the long-time limit of the FCS should be considered in a highly quantum-coherent single-molecule system.

**Methods**

Particle-number-resolved time-convolutionless quantum master equation. We consider a general transport setup consisting of a single-level QD molecule weakly coupled to the two electrodes, see Fig. 1, which is described by the following Hamiltonian

$$H = H_{\text{electrodes}} + H_{\text{rud}} + H_{\text{hyb}}.$$  

(27)

Here, the first term \(H_{\text{electrodes}} = \sum_{\alpha \in \{e, h\}} E_{\alpha} d_{\alpha}^\dagger d_{\alpha}\) stands for the Hamiltonians of the two electrodes, with \(E_{\alpha}\) being the energy dispersion, and \(d_{\alpha}^\dagger\) (\(d_{\alpha}\)) the annihilation (creation) operator of electrons in the \(\alpha\) electrode. The second term \(H_{\text{rud}} = H_{\text{rud}}(d_{\alpha}^\dagger d_{\beta})\), which may contain vibrational or spin degrees of freedom and different types of many-body interaction, represents the QD molecule Hamiltonian, where \(d_{\alpha}^\dagger\) (\(d_{\alpha}\)) is the creation (annihilation) operator of electrons in a quantum state denoted by \(\mu\). The third term \(H_{\text{hyb}} = \sum_{\alpha \in \{e, h\}} \sum_{\nu} \left( r_{\alpha \nu} d_{\nu}^\dagger d_{\alpha} + i s_{\alpha \nu} d_{\alpha}^\dagger d_{\nu} \right)\) describes the tunneling coupling between the QD molecule and the two electrodes, which is assumed to be a sum of bilinear terms that each create an electron in the QD molecule and annihilate one in the electrodes or vice versa.

The QD-electrode coupling is assumed to be sufficiently weak, so that \(H_{\text{hyb}}\) can be treated perturbatively. In the interaction representation, the equation of motion for the total density matrix reads

$$\frac{\partial}{\partial t} \rho(t) = -i[H_{\text{hyb}}(t), \rho(t)] \equiv L(t)\rho(t),$$  

(28)

with

$$H_{\text{hyb}}(t) = -\sum_{\alpha \in \{e, h\}} \sum_{\nu} \left( r_{\alpha \nu} d_{\nu}^\dagger d_{\alpha} + i s_{\alpha \nu} d_{\alpha}^\dagger d_{\nu} \right)$$

where \(f_{\alpha \nu}(t) = \sum_{\mu} c_{\alpha \mu}^* e^{i(\omega_{\mu\nu} - \omega_{\alpha\mu})t} d_{\mu}^\dagger + d_{\mu}, \quad d_{\nu}(t) = e^{i\omega_{\mu\nu} t} d_{\nu} e^{-i\omega_{\mu\nu} t}\). In order to derive an exact equation of motion for the reduced density matrix \(\rho_{\text{QD}}\) of the QD molecule system, it is convenient to define a super-operator \(P\) according to

$$P\rho = \rho - \mathbb{Q}\rho,$$

(29)

with \(\mathbb{Q}\rho\) being some fixed state of the electron electrode. Accordingly, a complementary super-operator \(Q\) reads

$$Q\rho = \rho - P\rho,$$  

(30)

For a factorizing initial condition \(\rho(t_0) = \rho_0(t_0) \otimes \rho_{\text{rud}} \rho(t_0) = \rho_0(t_0)\), and \(Q\rho(t_0) = 0\). Using the TCL projection operator method\(^{11}\), one can obtain the second-order TCL master equation

$$\frac{\partial}{\partial t} \rho^{(n)}(t) = -\sum_{\alpha \in \{e, h\}} \int_{-\infty}^t dt'^* \mathbb{P}(t') \mathcal{L}(t') P\rho^{(n)}(t).$$  

(31)

The Eq. (31) is the starting point of deriving the particle-number-resolved quantum master equation. Using Eqs. (28) and (29), after some algebraic calculations we can rewrite Eq. (31) as

$$\frac{\partial}{\partial t} \rho_{\text{QD}}(t) = -\sum_{\alpha \in \{e, h\}} \int_{-\infty}^t dt \mathbf{Tr}_{\text{rud}} [\rho_{\text{QD}}(t) \otimes \rho_{\text{rud}}(t)] (d_{\alpha}^\dagger d_{\alpha}) + H.c.$$  

(32)

In order to fully describe the electron transport problem, we should record the number of electrons arriving at the drain electrode, which emitted from the source electrode and passing through the QD molecule. We follow Li and co-authors\(^{10,11}\) and introduce the Hilbert subspace \(B^{(n)} = \text{span}(\{|\Psi_{\text{rud}}(n)\rangle\otimes|\Psi_{\text{rud}}(n)\rangle\})\) corresponding to \(n\) electrons arriving at the drain electrode, which is spanned by the product of all many-particle states of the two isolated electrodes, and formally denoted as \(B^{(n)} = \text{span}(\{|\Psi_{\text{rud}}(n)\rangle\otimes|\Psi_{\text{rud}}(n)\rangle\})\). Then, the entire Hilbert space of the two electrodes can be expressed as \(B = \bigotimes_{n} B^{(n)}\).

With this classification of the electrode states, the average over states in the entire Hilbert space \(B\) in Eq. (32) should be replaced with the states in the subspace \(B^{(n)}\), and leading to a conditional TCL master equation

$$\frac{\partial}{\partial t} \rho_{\text{QD}}^{(n)}(t) = -\sum_{\alpha \in \{e, h\}} \int_{-\infty}^t dt \mathbf{Tr}_{\text{rud}} [\rho_{\text{QD}}^{(n)}(t) \otimes \rho_{\text{rud}}(t)] (d_{\alpha}^\dagger d_{\alpha}) + H.c.$$  

(33)

To proceed, two physical considerations are further implemented. (i) Instead of the conventional Born approximation for the entire density matrix \(\rho(t) \approx \rho(t) \otimes \rho_{\text{rud}}\), the ansatz \(\rho(t) \approx \rho^{(n)}(t) \otimes \rho_{\text{rud}}^{(n)}(t)\) is proposed, where \(\rho_{\text{rud}}^{(n)}(t)\) being the electrode density operator associated with \(n\) electrons arriving at the drain electrode. With this ansatz for the entire density operator, tracing over the subspace \(B^{(n)}\), the Eq. (33) can be rewritten as

$$\frac{\partial}{\partial t} \rho_{\text{QD}}^{(n)}(t) = -\sum_{\alpha \in \{e, h\}} \int_{-\infty}^t dt \mathbf{Tr}_{\text{rud}} [\rho_{\text{QD}}^{(n)}(t) \otimes \rho_{\text{rud}}(t)] (d_{\alpha}^\dagger d_{\alpha}) + H.c.$$  

(34)

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. Moreover, 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. Consequently, after the procedure done in Eq. (34), the electrode density matrices \(\rho_{\text{rud}}^{(n)}(t)\) and \(\rho_{\text{rud}}^{(n+1)}(t)\) should be replaced by \(\rho_{\text{rud}}^{(0)}(t)\). In the Schrödinger representation, the Eq. (34) can be expressed as

$$\frac{\partial}{\partial t} \rho_{\text{QD}}^{(n)}(t) = -\sum_{\alpha \in \{e, h\}} \int_{-\infty}^t dt \mathbf{Tr}_{\text{rud}} [\rho_{\text{QD}}^{(n)}(t) \otimes \rho_{\text{rud}}(t)] (d_{\alpha}^\dagger d_{\alpha}) + H.c.$$  

(35)
where the correlation function are defined as
\[ C_{nn}^{(n)}(t_1 - t_2) = \text{tr} \left[ f_{cn}(t_1) f_{cn}(t_2) \right] = \left\langle f_{cn}(t_1) f_{cn}(t_2) \right\rangle, \]
(36)

Introducing the following super-operators
\[ A_{nn}^{(+)}(t) = \sum_{v} \int_{-\infty}^{\infty} dt C_{nn}^{(+)}(t_1 - t) e^{-iH(t-n)} d\psi e^{iH(t-n)}, \]
(37)
then, the Eq. (35) can be rewritten as a compact form
\[ \frac{\partial}{\partial t} \rho^{(n)}(t) = -i [H_{s}, \rho^{(n)}(t)] \]
\[ - \sum_{l} \left\{ \rho^{(0)}(0) A_{n0}^{(+)}(t) d_{l}^{+} + d_{l}^{+} A_{nl}^{(-)}(t) \rho^{(n)}(t) \right\} \]
\[ - A_{nl}^{(-)}(t) \rho_{nl}^{(n)}(t) d_{l}^{+} - A_{nl}^{(+)}(t) \rho_{nl}^{(-)}(t) d_{l}^{+} \]
\[ - d_{l}^{+} \rho^{(0)}(0) A_{nl}^{(-)}(t) d_{l}^{+} - d_{l}^{+} \rho_{nl}^{(+)}(t) A_{nl}^{(-)}(t) d_{l}^{+} + H C \}, \]
\[ A_{nn}^{(+)}(t) = \sum_{l} a_{l} A_{nl}^{(+)}(t). \]

From Eq. (40) we can identify the first four cumulant currents:
\[ C_{1}/t = \left\langle \left\langle [L_1(t)], 0 \right\rangle \right\rangle /i, \]
(41)
\[ C_{2}/t = \left\langle \left\langle [L_1(0)], -2 \left\langle \left\langle [L_1, RL_1(0)] \right\rangle \right\rangle \right\rangle /i^2, \]
(42)
\[ C_{3}/t = \left\langle \left\langle [L_1(0)], -3 \left\langle \left\langle [L_2, RL_1(0)], L_2 + L_1 \right\rangle \right\rangle \right\rangle /i^3, \]
(43)
\[ C_{4}/t = \left\langle \left\langle [L_1(0)], -4 \left\langle \left\langle [L_2, RL_1(0)], L_2 + L_1 \right\rangle \right\rangle \right\rangle /i^4. \]
(44)

Here, it is important to emphasize that the first four cumulant currents \( C_i \) are directly related to the transport characteristics. For example, the first-order cumulant (the peak position of the distribution of transferred-electron number) \( C_1 = 0 \) gives the average current \( \langle \dot{\theta} \rangle = eC_i/t \). The zero-frequency shot noise is related to the second-order cumulant (the peak-width of the distribution) \( S = 2eC_2/t = 2e^2 (\beta^2 - n^2) /\hbar \). The third-order cumulant \( C_3 = (n-n)^2 \) and fourth-order cumulant \( C_4 = (n-n)^4 \) characterize, respectively, the skewness and kurtosis of the distribution.

\[ \langle \left\langle [L_1(t)], 0 \right\rangle \rangle \]
\[ \langle \left\langle [L_1(0)], -2 \left\langle \left\langle [L_1, RL_1(0)] \right\rangle \right\rangle \right\rangle \]
\[ \langle \left\langle [L_1(0)], -3 \left\langle \left\langle [L_2, RL_1(0)], L_2 + L_1 \right\rangle \right\rangle \right\rangle \]
\[ \langle \left\langle [L_1(0)], -4 \left\langle \left\langle [L_2, RL_1(0)], L_2 + L_1 \right\rangle \right\rangle \right\rangle \]

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Author contributions
H.B.X. conceived the idea and designed the research and performed calculations. H.J.J., J.Q.L. and W.M.L. contributed to the analysis and interpretation of the results and prepared the manuscript.

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