Supplemental Material

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"Quantum interference and electron correlation in charge transport through triangular quantum dot molecules"

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EQUATION OF MOTION

In this section, we describe the equation of motion (EOM) method for the non-equilibrium Green's function[1]. The formalism provides a tool to calculate the transport properties of strongly-correlated multiple quantum dots (MQD) system in the Coulomb blockade regime. [2–7] The electron degrees of freedom within the MQD is treated exactly by the full many-body EOM, while the tunnelling effect to the metallic electrodes is included as the electron self-energy.

We consider an N quantum dot (QD) system (one level for each QD) with electron hopping and Coulomb interactions. This MQD system is weakly coupled to some metallic electrodes. The model Hamiltonian is

$$H = H_{QD} + H_T + H_{electrodes} \tag{1}$$

$$H_{QD} = \sum_{ij=1}^{N} t_{ij} d_i^\dagger d_j + \sum_{i<j=1}^{N} U_{ij} n_i n_j \tag{2}$$

$$H_T = \sum_{ik} (V_{ki} c_k^\dagger d_i + h.c.) \tag{3}$$

$$H_{electrodes} = \sum_k \epsilon_k c_k^\dagger c_k \ , \tag{4}$$

where t_{ij} denotes the electron hopping, and U_{ij} denotes the electron Coulomb interaction (which is real symmetric with zero diagonal elements), $d_j^\dagger$ and d_j are the creation and the annihilation operators for the electron at QD j, and $n_j = d_j^\dagger d_j$. The indices i, j labels all the discrete quantum numbers for the electrons in the MQD, including the spin. $c_k^\dagger$ and c_k are the creation and the annihilation operators for the electrons in the electrodes. The variable k labels all the quantum numbers for the electrons in the reservoirs including the momentum, the spin and the label for the reservoirs. We can derive the equation of motion which describes the full many-body degrees of freedom of electrons within the quantum dots. The reservoirs are taken into account by including the self-energy term. This approach is valid above the Kondo temperature.[2–7] For the contour-ordered double-time n-particle Green's function[1, 8]

$$G^{(n)}_{i_1 i_2 \ldots i_{2n}}(t,t') = -i\langle T[d^\dagger_{i_1}(t)\ldots d^\dagger_{i_{n-1}}(t) d_{i_n}(t)\ldots d_{i_{2n-1}}(t) d^\dagger_{i_{2n}}(t')]\rangle, \tag{5}$$

the EOM is

$$\begin{aligned} i\partial_t G^{(n)}_{i_1\ldots i_{2n}}(t,t') &= \delta(t-t') \sum_{\mu=n}^{2n-1} (-1)^{(\mu+1)} \delta_{i_\mu, i_{2n}} \\ &\times \langle d^\dagger_{i_1}\ldots d^\dagger_{i_{n-1}} d_{i_n}\ldots d_{i_{\mu-1}} d_{i_{\mu+1}}\ldots d_{i_{2n-1}}\rangle \\ &+ (-\sum_{\mu=1}^{n-1}\sum_j \bar{t}_{i_\mu j} + \sum_{\mu=n}^{2n-1}\sum_j t_{i_\mu j}) G^{(n)}_{i_1\ldots i_{\mu-1} j i_{\mu+1}\ldots i_{2n-1} i_{2n}}(t,t') \\ &+ (-\sum_{\mu=1}^{n-1}\sum_{\nu=\mu+1}^{n-1} U_{i_\mu i_\nu} + \sum_{\mu=n}^{2n-1}\sum_{\nu=n}^{\mu-1} U_{i_\mu i_\nu}) G^{(n)}_{i_1\ldots i_{2n}}(t,t') \\ &+ (-\sum_{\mu=1}^{n-1} + \sum_{\mu=n}^{2n-1}) \sum_j U_{i_\mu j} G^{(n+1)}_{i_1\ldots i_{n-1} j j i_n \ldots i_{2n-1} i_{2n}}(t,t') \\ &+ \int d\tau [-\sum_{\substack{\mu=1 \\ i_\mu \in I'}}^{n-1} \bar{\Sigma}_{i_\mu i_\mu}(t,\tau) + \sum_{\substack{\mu=n \\ i_\mu \in I}}^{2n-1} \Sigma_{i_\mu i_\mu}(t,\tau)] G^{(n)}_{i_1\ldots i_{2n}}(\tau,t'), \end{aligned} \tag{6}$$

where the symbol "$\bar{A}$" denotes "taking the complex conjugate" of A. For $n = 1$ the contact term should be defined as $\delta_{i_1 i_2}\delta(t-t')$. I and I' are some extra selection rules we may impose according to the phenomenology. The detail of these selection rules is discussed in the last section. The hierarchy of equations terminates at $n = N$ since $G^{(N+1)} = 0$. Taking the Fourier transform and analytic continuation[1] leads to a set of linear equations for the retarded and the lessor Green's functions $G^{(n)r}_{i_1 i_2\ldots i_{2n}}$ and $G^{(n)<}_{i_1 i_2\ldots i_{2n}}$ in the frequency domain. These equations can be viewed as a generalization of the Dyson equation and the Keldysh equation[1] to the n-particle Green's function. They can be solved by any standard linear solver. In equilibrium, it can be shown that the equation for the lessor Green's function reduces to the fluctuation-dissipation theorem

$$G^{(n)<}_{i_1\ldots i_{2n}}(\epsilon) = -2if(\epsilon)\Im[G^{(n)r}_{i_1\ldots i_{2n}}(\epsilon)] \ , \tag{7}$$

where $\Im$ stands for taking the imaginary part. The self-consistent calculation is closed by the integral relation

$$\langle d^\dagger_{i_1}\ldots d^\dagger_{i_n} d_{i_{n+1}}\ldots d_{i_{2n}}\rangle = \int \frac{d\epsilon}{2\pi i} G^{(n)<}_{i_2\ldots i_{2n} i_1}(\epsilon) \ . \tag{8}$$

We are interested in the solution at the wide-band

limit,[9]

$$\Sigma^r_{jj}(\epsilon) = -i\sum_\alpha \frac{\Gamma^\alpha_j}{2} \tag{9}$$

$$\Sigma^<_{jj}(\epsilon) = i\sum_\alpha \Gamma^\alpha_j f_\alpha(\epsilon)\ , \tag{10}$$

where α is the label for the reservoirs, Γ^α_j is the tunnelling rate between the reservoir α and the level j, and f_α is the Fermi function for the reservoir α. After solving for the Green's function, we can obtain the non-equilibrium steady-state current flowing from the reservoir α into the QD system by the Meir-Wingreen formula [10]

$$J_\alpha \tag{11}$$
$$= \frac{ie}{h}\int d\epsilon \sum_j \Gamma^\alpha_j [G^{(1)<}_{jj}(\epsilon) + f_\alpha(\epsilon)(G^{(1)r}_{jj}(\epsilon) - G^{(1)a}_{jj}(\epsilon))].$$

DOUBLE QUANTUM DOTS

To examine our full numerical solution, we consider the double quantum dot (DQD) case with $E_L = E_R = E_F + 30\Gamma_0 - \Delta_{go}$ ($\Delta_{go} > 0$). The strong coupling condition with $t_{LR} = 16\Gamma_0$ and $U_{LR} = 25\Gamma_0$ are adopted in order to make comparison with Ref. [3]. Figure S1(a) shows the electrical conductance(G_e) as a function of gate voltage (Δ_{go}) applied to both QDs at $k_BT = 0$ and $k_BT = 1\Gamma_0$.

The curve at zero temperature is shown only for comparison purpose, although it is below the Kondo temperature.[11, 12] There are twelve distinct peaks in the G_e spectrum, which can be identified by finding the enigenvalues of the Hubbard Hamiltonian H_{QDs} in configurations with various occupation numbers.[13] The positions of peaks are related to the difference of eigenvalues of the (n+1)-particle and n-particle states in various configurations. The peaks with appreciable strength occur at:
$\epsilon_1 = E_L - t_{LR}$,
$\epsilon_2 = E_L - t_{LR} + \frac{U_0+U_{LR}}{2} - \frac{1}{2}\sqrt{(U_0-U_{LR})^2 + 16t^2_{LR}}$,
$\epsilon_3 = E_L + U_{LR} - t_{LR}$, $\epsilon_4 = E_L + t_{LR}$,
$\epsilon_5 = E_L + t_{LR} + \frac{U_0+U_{LR}}{2} - \frac{1}{2}\sqrt{(U_0-U_{LR})^2 + 16t^2_{LR}}$,
$\epsilon_6 = E_L + U_{LR} + t_{LR}$, $\epsilon_7 = E_L + U_0 + U_{LR} - t_{LR}$,
$\epsilon_8 = E_L - t_{LR} + \frac{U_0+3U_{LR}}{2} + \frac{1}{2}\sqrt{(U_0-U_{LR})^2 + 16t^2_{LR}}$,
$\epsilon_9 = E_L + U_0 + 2U_{LR} - t_{LR}$, $\epsilon_{10} = E_L + U_0 + U_{LR} + t_{LR}$,
$\epsilon_{11} = E_L + t_{LR} + \frac{U_0+3U_{LR}}{2} + \frac{1}{2}\sqrt{(U_0-U_{LR})^2 + 16t^2_{LR}}$,
$\epsilon_{12} = E_L + U_0 + 2U_{LR} + t_{LR}$.
The magnitude of G_e is smaller than the quantum conductance $2e^2/h$ for $t_{LR}/\Gamma \gg 1$ as a result of electron Coulomb interactions.[3] The mechanism for understanding the unusual G_e behavior in nanostructure junction systems is a subject of high interest.[14] Due to electron Coulomb interactions, the magnitudes of peaks are related to the probability weights of quantum paths, which are related to single-particle occupation numbers and many-particle correlation functions.[6] At $k_BT = 1\Gamma_0$, the peaks in G_e are suppressed and broadened. [See dashed curve in Fig. S1(a)]

Figure S1(b) shows the one-particle occupation number $N_{\ell,\sigma} \equiv \langle n_{\ell,\sigma}\rangle$, intradot two-particle correlation function $\langle n_{\ell,\bar\sigma} n_{\ell,\sigma}\rangle$, interdot correlation function $\langle n_{\ell,\bar\sigma} n_{j,\sigma}\rangle$, and three-particle correlation function $\langle n_{\ell,\bar\sigma} n_{j,\bar\sigma} n_{j,\sigma}\rangle$ as functions of Δ_{go} at $k_BT = 0$. The one-particle occupation number (black solid line) exhibits four plateaus with values 1/4, 1/2, 3/4, and 1, corresponding to one-, two-, three- and four-particle configurations. At zero temperature and $\Gamma \to 0$, N_ℓ will be a stair-case function, which becomes broadened as a result of the coupling between dots and electrodes. The first peak height can be evaluated approximately by the probability weight $1 - N_{L,\bar\sigma} - N_{R,\bar\sigma} - N_{R,\sigma} + \langle n_{L,\bar\sigma} n_{R,\bar\sigma}\rangle + \langle n_{L,\bar\sigma} n_{R,\sigma}\rangle + \langle n_{R,\bar\sigma} n_{R,\sigma}\rangle - \langle n_{L,\bar\sigma} n_{R,\bar\sigma} n_{R,\sigma}\rangle$. Due to symmetry of the system, we have $N_{L,\bar\sigma} = N_{R,\bar\sigma}$, $\langle n_{L,\bar\sigma} n_{L,\sigma}\rangle = \langle n_{R,\bar\sigma} n_{R,\sigma}\rangle$, and so on. At $\Delta_{go} = \epsilon_1$, we have $N_L = N_R = 0.125$ and $G_e \approx 0.625$ in units of quantum conductance $2e^2/h$, which is close to the anomalous value 0.7 reported in Ref. [14]. The magnitudes of other G_e peaks can also be analyzed by their probability weights. For example, the peak labeled by ϵ_{12} is approximately $\langle n_{L\bar\sigma} n_{R\bar\sigma} n_{R\sigma}\rangle$. Comparing Fig. S1(a) with Fig. S1(b), we see that the four main peaks labeled by ϵ_1, ϵ_5, ϵ_8 and ϵ_{12} occur at Δ_{go} where N_ℓ makes a jump. The ϵ_5 peak corresponds to the transition form one-particle state to two-particle state in a spin-singlet configuration, which is evidenced by the quick increase of the correlation function $\langle n_{L,\sigma} n_{R,\bar\sigma}\rangle$, while ϵ_6 peak is related to a significant decrease in $\langle n_{L,\sigma} n_{R,\bar\sigma}\rangle$, indicating a change of the two-particle state form spin singlet to a partial mixture of spin triplet. We noticed that the G_e spectrum for $\Delta_g > 115\Gamma_0$ is a mirror image of that for $\Delta_g < 115\Gamma_0$ due to the electron-hole symmetry. Thus, physical mechanisms for the ϵ_7-ϵ_{12} peaks can be interpreted in terms of mechanisms for the ϵ_1-ϵ_6 peaks by using the hole picture. It is worth noting that the results shown in Fig. S1 are identical to those of Ref [3], which confirms the validity of the procedures of our full numerical calculation.

COMPUTATIONAL COST

We compare the computational effort of our Green's function approach to the rate equation approach. In the rate equation, the quantity to be solved is the density matrix of the size $2^N \times 2^N = 4^N$, which grows exponentially. In our Green's function approach, the number of correlators to be solved is $\sum_{n=1}^N \binom{N}{n}\binom{N}{n} = \binom{2N}{N} - 1 \to \frac{4^N}{\sqrt{\pi N}}$, which grows sub-exponentially. However, to get these correlators, the number of Green's functions to be solved is $\sum_{n=1}^N \binom{N}{n-1}\binom{N}{n} N$, which grows super-exponentially.

For the triple QD system ($N = 6$), we need to solve for 4752 Green's functions to get 923 correlators, while the number of the density matrix elements is 4096.

SELF-ENERGY TERM

The non-equilibrium tunnelling is treated by the Bułka-Kostyrko ansatz.[3] The recipe provides a good lowest order approximation to the transport physics in the Coulomb blockade regime. [2–7] In this section, we give a derivation[15–17] which makes the Bułka-Kostyrko ansatz plausible. The higher order effect may be included by employing more complicated self-energy term. [16, 18–20]

For the Hamiltonian of Eq. (1), the EOM is $i\partial_t G^{(n)}_{i_1\ldots i_{2n}}$ =(equilibrium term) + (self-energy term), where the equilibrium term is generated by H_{QD} and the self-energy term is generated by $H_T + H_{electrodes}$. The equilibrium term can be derived straight forwardly, so we focus on the self-energy term here. The derivatives of the operators of the electrons in the reservoirs are

$$i\partial_t c_k(t) = \epsilon_k c_k(t) + \sum_j V_{kj} d_j(t) \tag{12}$$

$$i\partial_t c_k^\dagger(t) = -\epsilon_k c_k^\dagger(t) + \sum_j -\bar{V}_{kj} d_j^\dagger(t)\ . \tag{13}$$

Integrate these equations to get

$$c_k(t) = \sum_j V_{kj} \int d\tau g_k(t,\tau) d_j(\tau) \tag{14}$$

$$c_k^\dagger(t) = \sum_j \bar{V}_{kj} \int d\tau \bar{g}_k(t,\tau) d_j^\dagger(\tau)\ , \tag{15}$$

where $g_k(t,t') = (i\partial_t - \epsilon_k)^{-1}$ and $\bar{g}_k(t,t') = (-i\partial_t - \epsilon_k)^{-1}$.

Consider the derivatives of the operators of the electrons in the QDs, the contribution of $H_T + H_{electrodes}$ is

$$i\partial_t d_j(t) = \sum_k \bar{V}_{kj} c_k(t) \tag{16}$$

$$i\partial_t d_j^\dagger(t) = \sum_k -V_{kj} c_k^\dagger(t)\ . \tag{17}$$

Now we can eliminate the electrodes degrees of freedom,

$$i\partial_t d_j(t) = \int d\tau \sum_{kl} \bar{V}_{kj} V_{kl} g_k(t,\tau) d_l(\tau) \tag{18}$$

$$= \int d\tau \sum_l \Sigma_{jl}(t,\tau) d_l(\tau) \tag{19}$$

$$i\partial_t d_j^\dagger(t) = -\int d\tau \sum_{kj} V_{kj} \bar{V}_{kl} \bar{g}_k(t,\tau) d_l^\dagger(\tau) \tag{20}$$

$$= -\int d\tau \sum_l \bar{\Sigma}_{jl}(t,\tau) d_l^\dagger(\tau)\ . \tag{21}$$

If we take the Markov approximation[17] and consider only the diagonal tunnelling matrix elements, then we have

$$\begin{aligned} &i\partial_t (d_{i_1}^\dagger \ldots d_{i_{n-1}}^\dagger d_{i_n} \ldots d_{i_{2n-1}}(t)) \\ &= \int d\tau \{[-\sum_{\mu=1}^{n-1} \bar{\Sigma}_{i_\mu i_\mu}(t,\tau) + \sum_{\mu=n}^{2n-1} \Sigma_{i_\mu i_\mu}(t,\tau)] \\ &\times [d_{i_1}^\dagger \ldots d_{i_{n-1}}^\dagger d_{i_n} \ldots d_{i_{2n-1}}(\tau)]\}, \end{aligned} \tag{22}$$

and hence we get the final result,

$$\begin{aligned} &\text{self-energy term} \\ &= \int d\tau [-\sum_{\mu=1}^{n-1} \bar{\Sigma}_{i_\mu i_\mu}(t,\tau) + \sum_{\mu=n}^{2n-1} \Sigma_{i_\mu i_\mu}(t,\tau)] G^{(n)}_{i_1\ldots i_{2n}}(\tau,t'). \end{aligned} \tag{23}$$

The self-energy term is quite general, but it contains many extra terms comparing to the previous works. [2–7] Numerical experiments also show that these terms break the particle-hole symmetry. This is due to the larger broadening appears in the higher particle Green's functions, which comes from the extra terms. This fact is easy to see from the expression.

The remedy is to employ some phenomenological arguments to drop certain self-energy terms. The selection rules are:

1. If one creation operator $d_i^\dagger(t)$ and one annihilation operator $d_i(t)$ of the same level i appear simultaneously, this pair is assumed to be stationary and the associated self-energy is dropped.

2. If the annihilation operator $d_i(t)$ is of the opposite spin with respect to the creation operator $d_j^\dagger(t')$, then the associated self-energy is dropped.

3. The remaining self-energy associated with the creation operator $d_i^\dagger(t)$ is dropped.

With these restrictions, the self-energy term above becomes the Bułka-Kostyrko ansatz:

$$\begin{aligned} &\text{self-energy term} \\ &= \int d\tau \sum_{i_\mu \in I_{BK}} \Sigma_{i_\mu i_\mu}(t,\tau) G^{(n)}_{i_1\ldots i_{2n}}(\tau,t'), \end{aligned} \tag{24}$$

where

$$\begin{aligned} I_{BK} = &\{i_\mu | n \le \mu \le 2n-1, \text{spin}(i_\mu) \ne \text{spin}(i_{2n})\} \\ &\backslash \{i_\mu | 1 \le \mu \le n-1\}. \end{aligned} \tag{25}$$

This is the formula used in practical simulations.

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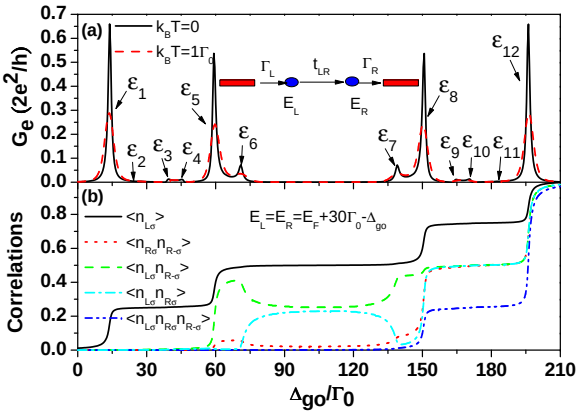

Figure S 1. (a) Electrical conductance and (b) Correlation functions of DQD as functions of gate voltage Δ_{go} with $E_L = E_R = E_F + 30\Gamma_0 - \Delta_{go}$, $t_{LR} = 16\Gamma_0$, $U_{LR} = 25\Gamma_0$, and $\Gamma = 1.6\Gamma_0$.

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