Enhancement of shot noise due to the fluctuation of Coulomb interaction

Duo Li, Lei Zhang, Fuming Xu and Jian Wang

Department of Physics and the center of theoretical and computational physics, The University of Hong Kong, Hong Kong, China

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We have developed a theoretical formalism to investigate the contribution of fluctuation of Coulomb interaction to the shot noise based on Keldysh non-equilibrium Green’s function method. We have applied our theory to study the behavior of dc shot noise of atomic junctions using the method of nonequilibrium Green’s function combined with the density functional theory (NEGF-DFT). In particular, for atomic carbon wire consisting 4 carbon atoms in contact with two Al(100) electrodes, first principles calculation within NEGF-DFT formalism shows a negative differential resistance (NDR) region in I-V curve at finite bias due to the effective band bottom of the Al lead. We have calculated the shot noise spectrum using the conventional gauge invariant transport theory with Coulomb interaction considered explicitly on the Hartree level along with exchange and correlation effect. Although the Fano factor is enhanced from 0.6 to 0.8 in the NDR region, the expected super-Poissonian behavior in the NDR region is not observed. When the fluctuation of Coulomb interaction is included in the shot noise, our numerical results show that the Fano factor is greater than one in the NDR region indicating a super-Poissonian behavior.

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I. INTRODUCTION

Quantum effects have become remarkably significant in nanoscale semiconductors and the traditional Boltzmann equation is no longer sufficient to describe transport phenomena. As proposed by Schottky in his seminar work, partition noise, or shot noise, is resulted from the quantization of charge. Accordingly, when electrons are uncorrelated, the classical value of Fano factor, which describes the magnitude of the electric fluctuation, should be one. When a Fano factor deviates from one, it shows a signature of interactions between current flow in different probes. It is known that shot noise is influenced by two crucial factors, namely, Pauli principle and Coulomb interaction, which coexist in electronic systems. Specifically, Pauli interaction can only suppress the Fano factor below one which corresponds to a sub-Poissonian case and has been confirmed convincingly by experiments. The Coulomb interaction, however, could either reduce the shot noise, or enhance it so that the Fano factor shows a super-Poissonian value, depending on details of mesoscopic devices. Hence, quantum enhancement of shot noise from the classical value has been the subject of growing interest in recent years and is explored intensively.

In mesoscopic systems shot noise is very important since it provides abundant information about transport properties of conductors, such as kinetics of electrons, distributions of energy, and correlations of electronic wave functions. In addition, people observed experimentally a shot noise enhancement in the negative differential resistance (NDR) region with or without the magnetic field in the tunneling structure. Various mechanisms have been proposed to generate a NDR, including enhancement of tunnel barriers, strong intramolecular correlations, band-gap inducement, and so on. One of early experiments by Li et al suggested that as the NDR region was approaching, the suppressed value of the Fano factor would increase. Further exploration by Panaccione et al showed that in the NDR region, the shot noise would go through a transition from a sub-Poissonian value to a super-Poissonian value in a nonlinear fashion. Nevertheless, a NDR was not a sufficient condition to generate the enhancement. As Song et al showed that there was no noise escalation in the super-lattice tunnel diode even though its I-V curve also exhibited a NDR region. This led to a conclusion that charge accumulation, which was related to the internal Coulomb potential, was ultimately responsible for the super-Poissonian shot noise. Given the good agreement between numerical calculations from semi-classical theory and those experiments, the Coulomb interaction was thought to be the reason for the noise enhancement. Since quantum effects dominate the transport behavior in mesoscopic systems, a quantum theory of shot noise capable describing the enhancement in the NDR region is clearly needed. In 1999, Blanter and Buttiker have studied the shot noise of resonant tunneling quantum well theoretically using scattering matrix method. In the nonlinear regime, Coulomb interaction (Hartree level) leads to hysteretic behavior in I-V curve. By including the fluctuation of Coulomb interaction, they identified an important energy scale, termed interaction energy, in the Fano factor. They found that in the NDR region where the interaction energy is very large, a super-poisonian behavior occurs due to the fluctuation of Coulomb interaction.

Understanding electronic transport properties of atomic wire based structures is very important from the scientific viewpoint and due to its potential applications in molecular electronics. For example, combining the Lippmann-Schwinger equation and density functional theory (DFT), a NDR in the tunneling regime of atomic carbon wires was predicted by Lang. The shot noise of silicon atomic wires has also been studied using the same approach. In this paper, we develop a general theory for dc shot noise by including the fluctuation of Coulomb interaction. Our theory is based on non-equilibrium Green’s function (NEGF) method which can be coupled with DFT to study transport properties of nano-devices from first principles. As an application of our theory, we investigate the shot noise of an atomic carbon wire structure with four carbon atoms in the scattering region.
Its I-V characteristic and transport properties have been well understood. It was found that a band gap induced NDR occurs at high bias due to a shift of conduction channels in the central region. We have used the traditional formula to calculate the shot noise for $Al - C_4 - Al$ structure. The first term describes leads that dc voltages are applied on, and $\epsilon_{ka} = \epsilon_{ka}^{(0)} + qv_n$, which $\epsilon_{ka}^{(0)}$ is energy levels in the lead $\alpha$ and $v_n$ stands for an external voltage. The second term is for the isolated central region, where the self-consistent internal Coulomb potential under the Hartree approximation is defined as

$$U_n = \sum_m V_{nm} <d_m^\dagger d_m>$$

where $V_{nm}$ is a matrix element of the Coulomb potential. In the real space $V(x, x') = 1/|x - x'|$, $q$ is the electron charge.

The last term corresponds to a coupling between the central region and leads described by a coupling constant $t_{kon}$. The current operator of the lead $\alpha$ is defined as ($\hbar = 1$):

$$\hat{I}_{\alpha}(t) = \frac{q}{\pi} \int dE \{ f_{\alpha}(1 - f_{\beta}) + f_{\beta}(1 - f_{\alpha}) \} \text{Tr} \{ \hat{T} \}$$

(1)

to calculate the shot noise for $Al - C_4 - Al$ structure. Our results show that shot noise is sub-Poissonian. When the fluctuation of Coulomb interaction is included, large shot noise was found in NDR region showing super-Poissonian behavior.

Our paper is organized as follows. In section II, we derive a general theory for dc shot noise when the fluctuation of Coulomb interaction is included in the first order. The detailed derivation is given in Appendix. In section III, we describe some technical details and show the numerical results in the atomic carbon chain system along with an analysis and discussion of the result. Finally, the summary is given in section IV.

II. THEORETICAL FORMALISM

In this section, a NEGF theory is developed to calculate dc shot noise in the regime of NDR, which involves the Coulomb interaction between electrons. A key ingredient of the new theory is that to account for large shot noise in the NDR region both self-consistent Coulomb potential and its fluctuation have to be considered.

A. General Expression

We start from a quantum coherent two-lead conductor defined by the Hamiltonian

$$\hat{H}_0 = \sum_{ka} \epsilon_{ka} \hat{C}_{ka}^\dagger \hat{C}_{ka} + \sum_n (\epsilon_n + qU_n) d_n^\dagger d_n$$

$$+ \sum_{kam} [t_{kan} \hat{C}_{ka}^\dagger \hat{d}_n + \text{c.c.}]$$

(2)

where $\hat{C}_{ka}$ ($\hat{C}_{ka}$), $d_n^\dagger$ ($d_n$) are the creation (annihilation) operators of electrons in leads and the scattering region, respectively. The first term describes leads that dc voltages are applied on, and $\epsilon_{ka} = \epsilon_{ka}^{(0)} + qv_n$, which $\epsilon_{ka}^{(0)}$ is energy levels in the lead $\alpha$ and $v_n$ stands for an external voltage. The second term is for the isolated central region, where the self-consistent internal Coulomb potential under the Hartree approximation is defined as

$$U_n = \sum_m V_{nm} <d_m^\dagger d_m>$$

(3)

where $V_{nm}$ is a matrix element of the Coulomb potential. In the real space $V(x, x') = 1/|x - x'|$, $q$ is the electron charge.

The current operator of the lead $\alpha$ is defined as ($\hbar = 1$):

$$\hat{I}_{\alpha}(t) = \frac{q}{\pi} \int dE \{ f_{\alpha}(1 - f_{\beta}) + f_{\beta}(1 - f_{\alpha}) \} \text{Tr} \{ \hat{T} \}$$

(4)

where $\hat{N}_{\alpha} = \sum_k \hat{C}_{ka}^\dagger \hat{C}_{ka}$ is the number operator for electrons in the lead $\alpha$.

From the Heisenberg equation of motion,

$$\frac{d\hat{N}_{\alpha}}{dt} = -i [\hat{N}_{\alpha}, \hat{H}_0]$$

(5)

we have

$$\frac{d\hat{N}_{\alpha}}{dt} = -i \sum_{kn} [t_{kan} \hat{C}_{ka}^\dagger \hat{d}_n + \text{h.c.}]$$

(6)

where $\hat{H}$ is the system Hamiltonian with constant Coulomb potential and h.c. denotes the Hermitian conjugate.

Hence the current operator becomes

$$\hat{I}_{\alpha}(t) = -i q \sum_{kn} [t_{kan} \hat{C}_{ka}^\dagger (t) \hat{d}_n (t) + \text{h.c.}]$$

(7)

On the mean field level, the current is a functional of Coulomb interaction, i.e., $\langle \hat{I} = \hat{I} [\hat{U}] \rangle$. Here we have treated the operator of Coulomb interaction $\hat{U}$ as a C number meaning that the fluctuation of Coulomb interaction is assumed not important. However, this is not always true. For instance, in order to reflect the Coulomb interaction between electrons in the NDR region, we have to consider the fluctuation of the Coulomb potential. Fig.1 shows the physical picture of the NDR. For simplicity, we assume that the scattering region has one resonant level $E_0$ with a width characterizing the lifetime of the resonant level. We also assume that there is an effective band bottom for the lead which is crucial for the phenomenon of NDR. As shown in Fig.1, when the bias voltage is increased the current increases because the resonant level is brought down by the external bias. As the bias is increased further such that the resonant level falls below the band bottom of the lead the current starts to decrease giving rise to the NDR. The above physical picture is static where the Coulomb potential is included on the mean field level and the correlation effect of Coulomb interaction has been neglected. For the current correlation in the NDR region, the correlation effect of Coulomb interaction has to be considered. In this picture, when the resonant energy level $E_0$ is about to fall below the band bottom of the lead, the internal potential of the scattering region due to the Coulomb interaction of injected electron will push it up, leading to a positive correlation between incoming electron flows. This positive correlation is a dynamic process and can not be described by a Hartree field. In another word, the fluctuation of Coulomb interaction has to be considered for the positive correlation in the NDR region. As demonstrated by Larade and confirmed by our calculation, for the atomic
wire with even number carbon atoms like $C_4$ and $C_6$, there is an effective band bottom responsible for the NDR. Therefore, the fluctuation of Coulomb potential $\hat{U}$ should be important. For odd number wires such as $C_5$ and $C_7$, however, there is no apparent NDR effect. Hence there no effective band bottom and the fluctuation of $\hat{U}$ can be neglected.

To treat the fluctuation of Coulomb interaction, we follow the idea of Ref. [12] and expand the current in terms of Coulomb potential operator about its equilibrium value up to linear order. After the expansion, the total current in the real space could be expressed as

$$\hat{I}_\alpha(t) \simeq \hat{I}_{\alpha 0}(t) + \sum_i \frac{\delta \hat{I}_{\alpha 0}(t)}{\delta U_i(t)} |_{U_i(t)=U_i} (\hat{U}_i(t) - U_i)$$

$$\simeq \hat{I}_{\alpha 0}(t) + \sum_i \frac{\delta I_{\alpha 0}}{\delta U_i} (\hat{U}_i(t) - U_i)$$

$$= \hat{I}_{\alpha 0}(t) + \sum \lambda_{\alpha i} \delta \hat{U}_i(t)$$

where $I_{\alpha 0} = \langle \hat{I}_{\alpha 0} \rangle$, $U_i = \langle \hat{U}_i \rangle$, and $\delta \hat{U}_i(t) = \hat{U}_i(t) - U_i$. We have also introduced a quantity $\lambda_{\alpha i} = \delta I_{\alpha i} / \delta U_i$.

It is easy to see that this new current operator gives the same current but different shot noise. With the new current operator, the current correlation is obtained up to the linear order in $\delta U$,

$$S_{\alpha \beta}^{(1)} = \langle \Delta \hat{I}_\alpha(t) \Delta \hat{I}_\beta(t') \rangle = \langle \hat{I}_\alpha(t) \hat{I}_\beta(t') \rangle - I_{\alpha} I_{\beta}$$

$$\simeq \langle \hat{I}_{\alpha 0}(t) \hat{I}_{\beta 0}(t') \rangle - I_{\alpha} I_{\beta} + \Delta_{\alpha \beta}^{(1)}$$

where $S_{\alpha \beta} = (1/2) [S_{\alpha \beta}^{(1)} + S_{\alpha \beta}^{(2)}]$. The first two terms in Eq. (9) corresponds to the current correlation $S_{\alpha \beta}^{(1)}$ in the absence of Coulomb potential fluctuation. They have been calculated before and is given by Eq. (1). The last term in Eq. (9) is defined as

$$\Delta_{\alpha \beta}^{(1)} = \sum_i [\lambda_{\beta i} < \hat{I}_{\alpha 0}(t) \hat{U}_i(t') > + \lambda_{\alpha i} < \hat{U}_i(t) \hat{I}_{\beta 0}(t') >$$

$$- (\lambda_{\alpha i} U_i \alpha + \lambda_{\beta i} U_i \beta)]$$

Using the NEGF method, we have derived the expression of $\Delta_{\alpha \beta}^{(1)}$ (see the appendix). Finally, the shot noise in the presence of Coulomb potential fluctuation is written as

$$S_{\alpha \beta} = S_{\alpha \beta 0} + \Delta_{\alpha \beta}$$

where

$$\Delta_{\alpha \beta} = -\frac{q^2}{2\pi} \sum_{ij} [\lambda_{\beta i} V_{ij} \text{Im}(\Xi_{\alpha j j}) + \lambda_{\alpha i} V_{ij} \text{Im}(\Xi_{\beta j j})]$$

with

$$\lambda_{\alpha i} = -\frac{q}{2\pi} \int dE \sum_{\beta} (f_{\alpha} - f_{\beta}) (G^r \Gamma_{\beta} G^a \Gamma_{\alpha} G^f)_{ij} + h.c.$$
In addition, from Eq. (13), we have

$$\lambda_L = \frac{-q}{2\pi} \int_{E_F}^{E} dE(G' \Gamma_R G^* \Gamma_L G') + h.c.$$  

$$\lambda_R = \frac{q}{2\pi} \int_{E_q}^{E} dE(G' \Gamma_L G^* \Gamma_R G') + h.c. \quad (19)$$

With Eqs. (17), (18), and (19), the two-probe shot noise can be calculated.

### III. Numerical Results

In this section we use state-of-the-art first-principle quantum transport package MATDCAL to investigate the general transport properties of atomic carbon-chain systems coupled with Al leads. In the package DFT is carried out within the formalism of the Keldysh nonequilibrium Green’s function. Numerically, the effective Kohn-Sham (KS) equations is solved by a linear combination of the atomic orbitals (LCAO) basis set. We define the atomic core by a nonlocal norm conserving pseudopotential and treat the exchange-correlation at the LDA level. DFT determines the atomic structure and the Hamiltonian while NEGF contributes to the nonequilibrium transport properties. Under an external bias the transport boundary conditions are treated by the real space numerical techniques. For further references, the theoretical background and practical execution of this formalism can be found in Ref[24]. A numerical error tolerance is set to be $10^{-4}$ to confirm self-consistency.

Generally speaking, we have carried out our calculation on the atomic chain structure with four carbon atoms $Al - C_4 - Al$. The carbon chain lies in the central simulation box in contact with electron reservoirs through two semi-infinite Al electrodes. The schematic structure is shown in Fig.2 where there are 18 Al atoms in the unit cell of the semi-infinite electrodes with a cross section along (100) direction.

![FIG. 2: The schematic plot of Al – C₄ – Al system. The atomic wire with four carbon atoms is linked by two semi-infinite Al electrodes. The (100) direction Al electrodes extend to ±∞ where electron reservoirs are located.](image)

The contact distance between the Al electrode and the carbon chain is fixed at 0.378 a.u. while the distance between the nearby carbon atoms is equal to 2.50 a.u. In our calculation, we have set temperature to be zero.

Technically, the correction term in Eq.(12) has to be solved in real space due to the Coulomb like interaction involving $V_{ij}$ which reads $V(x, x') = 1/|x - x'|$ in real space. Since quantities $\lambda$ and $\Xi$ play the role of charge, we can define the potential induced by $\Xi$,

$$\Omega_{\alpha x} = \sum_{x'} V(x, x') \text{Im}(\Xi(x', x')) \quad (20)$$

Since $\nabla^2 V(x, x') = -4\pi \delta(x-x')$, $\Omega_{\alpha}$ satisfies the Poisson like equation,

$$\nabla^2 \Omega_{\alpha}(x) = -4\pi \text{Im}(\Xi(x, x)) \quad (21)$$

We solve this equation for the leads to obtain the boundary condition for the scattering region. It turns out $\Omega_{\alpha}(x)$ in the lead is very small so that the boundary condition of $\Omega_{\alpha}(x)$ can be safely set to zero. Once $\Omega_{\alpha}(x)$ is obtained the correction term can be easily calculated from Eq.(12)

$$\Delta_{\alpha \beta} = -\frac{q^2}{2\pi} \int [\lambda_{\beta}(x) \Omega_{\alpha}(x) + \lambda_{\alpha}(x) \Omega_{\beta}(x)] dx \quad (22)$$

The I-V characteristics is shown in Fig.3, where the inset plots the transmission coefficient $T$ versus the energy $E$ at zero bias. The shot noise and the corresponding Fano factor are shown in Fig 4. In Fig 3 and 4, the Coulomb interaction is included on the Hartree level and the Coulomb potential fluctuation is neglected. Following observations are in order. (1) The I-V curve is similar to the result obtained by Larade et al.[21] (2) At zero bias the resonant energy is higher than Fermi energy of the system (chosen to be zero). As we apply a voltage to the right electrode, the resonant energy level should drop and move closer to Fermi energy of the left electrode. With the increasing of voltage, the effective band bottom of

![FIG. 3: The I-V curve of Al – C₄ – Al structure. A NDR region begins to show up around 0.65 V. Inset: the transmission coefficient for this system at zero bias.](image)
the emitter and the main resonant level would be aligned, and this gives the maximum current around 0.65 V. When the voltage increases further, a significant decrease of current occurs.

3) The shot noise in the absence of Coulomb potential fluctuation has a similar behavior as that of I-V except that the maximum is at 0.7V instead of 0.65V. (4) The Fano factor is nearly a constant of order of 0.6 in positive differential resistance (PDR) region when bias is smaller than 0.6V. It starts to increase sharply upon entering the NDR region and the Fano factor eventually shoots up to 0.8. We conclude that although the Fano factor calculated on the Hartree level shows enhancement in the NDR region, it is still sub-Poissonian which does not agree with experimental result.

In general, when electrons tunnel through the left barrier to occupy empty energy levels, Pauli principle inhibits other tunneling electrons to reach the same energy level but higher ones. As a consequence Pauli exclusion principle gives the negative effect for current correlation. Coulomb interaction, however, can give either positive or negative correlation effect to shot noise. This can be understood as follows. It is known that the maximum current corresponds to the situation that the energy of incoming electron is in line with the resonant level. Hence in both PDR and NDR regions, most of electrons are off resonance. Due to the Coulomb interaction the incoming electron can push up the resonant level so that the next electron will be further away from the resonance in the PDR region or closer to the resonance in the NDR region, giving rise negative or positive correlations. Our numerical results indeed confirm this physical picture. In Fig. 5 and 6 we present the result of shot noise and Fano factor in the presence of fluctuation of Coulomb interaction. We have also included the shot noise and Fano factor in the absence of Coulomb potential fluctuation for comparison. In Fig. 5, we see that the correction term solved via the Poisson like equation is very small at low bias when $V < 0.5$ and becomes negative until around 1.0V where shot noise increases sharply. In Fig. 6, we see that a large Fano factor great than 3 occurs near $V = 1.0$. This result is in qualitatively agreement with others’ work.

IV. CONCLUSION

The traditional formalism can only describe the suppression of shot noise, which corresponds to the PDR region. In order to treat enhancement of shot noise correctly in the NDR region, we have to include the fluctuation of Coulomb interaction. In this paper we have developed such a general dc theory for calculating the shot-noise in the NDR region. The theoretical framework is based on the combination of NEGF-DFT formalism with the self-consistent Coulomb potential and its fluctuation included. Our theory (Eq. (11)) can also be applied to mesoscopic conductors. We have applied our theory to molecular devices. Specifically, we have calculated the shot noise of $Al – C_4 – Al$ structure which is an ideal system since its I-V curve exhibits a NDR region. We found a large Fano factor in the NDR region exhibiting super-Poissonian behav-
V. APPENDIX

In this appendix, we will derive the expression of $\lambda_{\alpha,\beta}$ and $\Delta_{\alpha,\beta}$ using the theory of NEGF.

Expression of $\lambda_{\alpha,\beta}$

Using the NEGF, the current is given by

$$I_\alpha = -\frac{q}{2\pi} \int dE \sum_\beta [Tr(\Gamma_\alpha G^* \Gamma_\beta G^\alpha)](f_\alpha - f_\beta)$$

where $f_{\alpha,\beta}$ are Fermi distribution functions in corresponding leads. $G^{r,a}$ are the respective retarded and advanced Green functions of the scattering region and $\Gamma_{\alpha,\beta}$ are the linewidth functions related to the coupling of leads and the scattering region.

To obtain $\lambda_{\alpha}$, we first calculate the following quantity,

$$\lambda_{\alpha}(E) = -\frac{q}{2\pi} \sum_\beta Tr \frac{\partial}{\partial U_i}[\Gamma_\alpha G^\alpha \Gamma_\beta G^\alpha]$$

$$= -\frac{q}{2\pi} \sum_\beta Tr[\Gamma_\alpha \frac{\partial G^r}{\partial U_i}]G^a + \Gamma_\alpha G^r \Gamma_\beta (\frac{\partial G^a}{\partial U_i})]$$

$$= -\frac{q}{2\pi} \sum_\beta Tr[\Gamma_\alpha \frac{\partial G^r}{\partial U_i}]G^a] + h.c. \tag{24}$$

The quantity $\frac{\partial}{\partial U_i} G^r$ can be calculated from the Dyson equation:

$$G^r = G^r_0 + G^r_0 U G^r$$

where $G^r_0$ is the retarded Green’s function in the absence of Coulomb interaction. Taking the derivative with respect to $U_i$, we have

$$\frac{\partial}{\partial U_i} G^r = \frac{1}{1 - G^r_0 U} G^r_0 D_i G^r + G^r_0 U \frac{\partial G^r}{\partial U_i} \tag{26}$$

where $D_i$ is a diagonal matrix with the matrix element $(D_i)_{jk} = \delta_{ji}\delta_{kj}$, i.e., there is only one nonzero matrix element. From Eq. (26), we find

$$\frac{\partial}{\partial U_i} G^r = \frac{1}{1 - G^r_0 U} G^r_0 D_i G^r + G^r_0 U \frac{\partial G^r}{\partial U_i} \tag{27}$$

where we have used the following Dyson equation again,

$$G^r = \frac{1}{1 - G^r_0 U} G^r_0$$

Substituting Eq. (27) into Eq. (24), we can get

$$\lambda_{\alpha}(E) = -\frac{q}{2\pi} \sum_\beta Tr(\Gamma_\alpha G^r D_i G^\alpha \Gamma_\beta G^\alpha) + h.c. \tag{28}$$

which is equivalent to Eq. (13).

Expression of $\Delta_{\alpha,\beta}$

For simplicity, we only deal with the first term of $\Delta_{\alpha,\beta}$ explicitly, i.e., $<\hat{I}_{\alpha}(t)\hat{U}_i(t')>$. The second term can be calculated similarly. Using the current and Coulomb potential operators in Eqs. (7) and (8), we obtain

$$< -iq^2 \sum_{kn} [t_{kan} \hat{C}_{\alpha}^t(t) \hat{d}_{an}(t) - h.c.] \sum_m V_{im} \hat{d}_{rn}^\dagger(t') \hat{d}_m(t') >$$

$$= -iq^2 \sum_{knm} V_{im} [t_{kan} < \hat{C}_{\alpha}^t(t) \hat{d}_{an}(t) \hat{d}_m(t') \hat{d}_m(t') >$$

$$- t_{kan}^* < \hat{d}_{rn}^\dagger(t') \hat{d}_m(t') \hat{d}_m(t') >]$$

$$= -iq^2 \sum_{knm} V_{im} [t_{kan} < \hat{C}_{\alpha}^t(t) \hat{d}_{an}(t) \hat{d}_m(t') >$$

$$- t_{kan}^* < \hat{d}_{rn}^\dagger(t') \hat{d}_m(t') \hat{d}_m(t') >] + I_\alpha U_i \tag{29}$$

In terms of Green’s function$^{27}$

$$G^{<}_{m,ka}(t,t') = i < \hat{C}_{\alpha}^t(t') \hat{d}_a(t) > \tag{30}$$

$$G^{>}_{nm}(t',t) = - i < \hat{d}_a(t') \hat{d}_m(t) > \tag{31}$$

$$G^{<}_{mn}(t,t') = i < \hat{d}_a(t) \hat{d}_m(t') > \tag{32}$$

$$G^{>}_{ka,m}(t',t) = - i < \hat{C}_{\alpha}(t') \hat{d}_a(t) > \tag{33}$$

we obtain,

$$< \hat{I}_{\alpha}(t)\hat{U}_i(t') >$$

$$= -iq^2 \sum_{knm} V_{im} [t_{kan} G^{<}_{m,ka}(t,t') G^{>}_{nm}(t',t)$$

$$- t_{kan}^* G^{<}_{mn}(t,t') G^{>}_{ka,m}(t',t)] + I_\alpha U_i \tag{34}$$

Applying the Langreth theorem of analytic continuation$^{28}$ and suppressing time indices, we have

$$G^{<}_{m,ka} = \sum_l (G^{l*}_{m,l} t_{kal} g^{a}_{ka} + G^{l*}_{m,l} t_{kal} g^{a}_{ka}) \tag{35}$$

$$G^{>}_{ka,m} = \sum_l (g^{<}_{ka,l} t_{kal} G^{a}_{lm} + g^{<}_{ka,l} t_{kal} G^{a}_{lm}) \tag{36}$$

where $g^{<,>}_{\alpha,r,a}$ are the corresponding Green’s functions in the lead $\alpha$.

For dc transport, the Green functions depend only on $t' - t$. 

ior.
After the Fourier transform from time to energy, it becomes

\[
< \hat{I}_{\alpha \beta} \hat{U}_i > - I_a U_i \\
= - \frac{i q^2}{2 \pi} \sum_{k n l m} V_{m n} \int dE [t_{k m} (G^{<}_{m l t_{k a l} G^{<}_{k n} \\
+ G^{<}_{m l t_{k a l} G^{<}_{k n} - t_{k a n} G^{<}_{m a n} (G^{<}_{k a l t_{k o a l} G^{<}_{l m}} \\
+ g_{k a l t_{k o a l} G^{<}_{l m}})] \\
= - \frac{i q^2}{2 \pi} \sum_{m} V_{m} \int dE [(G^{<}_{\Sigma^{<}_{\alpha} + G^{<}_{\Sigma^{<}_{\alpha}}}) G^{>}_{\alpha} - G^{<}_{\Sigma^{<}_{\alpha} G^{<}} \\
+ \Sigma^{<}_{\alpha} G^{>}])_{m m} \\
= - \frac{i q^2}{2 \pi} \sum_{m} V_{m} \hat{\Sigma}^{\alpha}_{m m} 
\]

where \(\Sigma^{<,>,r,a}_{\alpha}\) are the corresponding functions of self-energy due to the lead \(\alpha\). Using the Keldysh equation and properties of Green’s functions, we find

\[
\Xi^{\alpha}_{i} = i \int dE [G^{\dagger}_{\alpha \beta} G^{\dagger}_{\alpha \beta} f_{\alpha} - G^{\dagger}_{\alpha \beta} \Sigma^{<}_{\alpha} G^{>}_{\alpha} (f_{\alpha} - 1) \\
+ G^{\dagger}_{\alpha \beta} \Sigma^{<}_{\alpha} G^{>}_{\alpha} \Sigma^{<}_{\alpha} G^{>}_{\alpha} ] 
\]

(42)

Substituting this equation into Eqs. (37) and (10), we finally arrive at

\[
\Delta^{(1)}_{\alpha \beta} = - \frac{i q^2}{2 \pi} \sum_{i j} (\lambda_{\beta i} V_{ij} \Xi_{\alpha j} - \lambda_{\alpha i} V_{ij} \Xi^{*}_{\beta j}) 
\]

(43)

where we have used the fact that \(< \hat{I}_{\alpha \beta} (t) \hat{U}_i (t') > = < \hat{U}_i (t) \hat{I}_{\alpha \beta} (t') >\).

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