Correlated two-electron transport: a principle for a novel charge pump

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Abstract

By considering a correlated two-electron transport process (TET) and using a diagrammatic analysis within the Keldysh nonequilibrium Green’s function formalism, we discuss a novel charge pump by which carriers are pumped from a contact with low chemical potential to another contact with a higher potential. The TET process involves two correlated incident electrons scattering and exchanging energy with each other. The process can significantly affect charge current density and it involves high empty states and/or low filled states of the Fermi liquid of the leads.

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In the past two decades, quantum electron transport through meso- and nanoscopic structures have received considerable attention [1]. Charge transport theories for nanostructures typically consider the situation of an incident electron from a lead that is scattered in the device scattering region and transmitted to other leads or reflected to where it came from. The scattering region can be rather complicated and may involve electron-electron, electron-phonon, electron-impurity, and other interactions and scattering mechanisms. The scattering region itself can be a semiconductor nanostructure such as a quantum dot (QD), a carbon nanotube, or a single molecule. The scattering processes may involve exchange of energy or spin. While situations and physics can vary in a wide range of ways, charge transport in nanostructures has largely, so far, been considered as involving a single electron incident from a non-interacting lead of the device, it traverses through a complicated scattering region in which interactions occur, and finally exits the device through the non-interacting leads. We will refer them as single electron tunneling or single electron transport (SET) processes.

In this paper, we go one step further by investigating correlated two-electron tunneling and transport (TET) processes which, to the best of our knowledge, has not been studied before. In this process, two incident electrons with energies $\epsilon_1$ and $\epsilon_2$ from the leads tunnel into the scattering region of a device, they scatter with each other and exchange energy (they could also scatter and exchange energy with other particles in the scattering region). Following the scattering event, they transmit to the outside world through the contacts, but with energies $\epsilon'_1$ and $\epsilon'_2$ which are different from the initial values $\epsilon_1$, $\epsilon_2$ due to energy exchange. If $\epsilon_1 + \epsilon_2 = \epsilon'_1 + \epsilon'_2$, i.e. the two incident electrons do not exchange energy with other particles, the transport process as a whole can be considered as elastic. In the TET process, the variation of electron number in the scattering region is always two: a situation that is different from SET. We show that such TET processes can indeed occur, and it affects measurable physical quantities such as charge current. In particular, TET induces a transport vertical flow—meaning carriers incident with energy $\epsilon_1$ exit at a different energy $\epsilon'_1$, resulting to a non-conserved current density, i.e. $\sum_n j_n(\epsilon) \neq 0$. Using the characteristics
of the correlated two-electron transport process, we design an interesting charge pump so that incident carriers can be pumped from a contact with low chemical potential to another contact having a higher potential.

We consider a QD coupled to two or three leads, described by the following Hamiltonian:

\[ H = \sum_\alpha \epsilon_\alpha d_\alpha^{\dagger} d_\alpha + U d_\uparrow^{\dagger} d_\uparrow d_\downarrow^{\dagger} d_\downarrow + \sum_{n,k,\alpha} \epsilon_{nka} d_{nka}^{\dagger} a_{nka} + \sum_{n,k,\alpha} [t_{nka} a_{nka}^{\dagger} d_\alpha + H.c.], \]

where \( a_{nka}^{\dagger} (a_{nka}) \) and \( d_\alpha^{\dagger} (d_\alpha) \) are creation (annihilation) operators in lead \( n \) and in QD, respectively. The QD includes two states and has an intradot Coulomb interaction \( U \). Subscript \( \alpha \) is the spin index, it may also indicate other quantum numbers. To account for a magnetic field, we let \( \epsilon_\uparrow \neq \epsilon_\downarrow \).

In the following we focus on investigating the elastic TET by analyzing the behavior of current density \( j_n(\epsilon) \) from lead \( n \) to the QD. \( j_n(\epsilon) \) relates to the current \( I_n \) through

\[ I_n = \int j_n(\epsilon) d\epsilon. \]

We also define the electron occupation number density operator \( \hat{N}_n(\epsilon, \tau) = \sum_k \int dt e^{i \epsilon t} a_{nka}^{\dagger}(\tau) a_{nka}(\tau + t) \).

Current density \( j_n(\epsilon) \) can be calculated from the time evolution of \( \hat{N}_n(\epsilon, \tau) \):

\[ j_n(\epsilon) = -e \text{Im} \sum_\alpha \frac{\Gamma_n(\epsilon)}{2\pi} [2 f_n(\epsilon) G^r_\alpha(\epsilon) + G^K_\alpha(\epsilon)] \tag{1} \]

where linewidth function \( \Gamma_n(\epsilon) \equiv 2\pi \sum_k |t_{nk}|^2 \delta(\epsilon - \epsilon_{nk}) \); \( f_n(\epsilon) \) is the Fermi distribution function for lead \( n \); \( G^r_{\alpha, <}(\epsilon) \) are the retarded and Keldysh Green’s functions of the QD. Using the standard equation of motion technique, \( G^r_{\alpha}(\epsilon) \) has already been solved in previous work:

\[ G^r_{\alpha}(\epsilon) = \frac{1 + U A_{\alpha} n_\alpha}{\epsilon - \epsilon_\alpha - \Sigma^{(0)}(\epsilon) + U A_{\alpha} (\Sigma^a_\alpha + \Sigma^b_\alpha)} \tag{2} \]

where \( A_{\alpha}(\epsilon) = [\epsilon - \epsilon_\alpha - U - \Sigma^{(0)}(\epsilon) - \Sigma^{(1)}_\alpha(\epsilon) - \Sigma^{(2)}_\alpha(\epsilon)]^{-1}; \Sigma^{(0)}_\alpha \) is the lowest-order self-energy, \( \Sigma^{(1), (2), a, b}_\alpha \) are the higher-order self-energies; and \( n_\alpha = \text{Im} \int \frac{d\epsilon}{2\pi} G^K_\alpha(\epsilon) \) is the intradot electron occupation number of state \( \alpha \). It is worth to mention that if temperature \( T \) is lower than the Kondo temperature \( T_K \), the solution of Eq.(2) has a Kondo resonance at the Fermi level which was the subject of many previous studies.
We investigate TET processes at a temperature higher than $T_K$. To this end we need to solve the the Keldysh Green’s function $G^<_\alpha(\epsilon)$. Note that if one applies the commonly used ansatz for interacting lesser and greater self-energies \[5\], or using the large-$U$ limit non-crossing approximation \[4,6\] to solve $G^<_\alpha(\epsilon)$, the two-electron scattering will be lost in these approximations. Therefore a more precise analysis is needed in our problem and we proceed as follows. Introducing the intradot electron occupation number density operator

$$\hat{N}_\alpha(\epsilon, \tau) = \int e^{i\epsilon t} d^\dagger_\alpha(\tau) d_\alpha(\tau + t) \frac{dt}{2\pi}$$

and due to the steady state condition $\langle \frac{d}{d\tau} \hat{N}_\alpha(\epsilon, \tau) \rangle = 0$, we have:

$$-e \sum_n i\frac{\Gamma_n}{2\pi} [G^<_\alpha + f_n(G^r_\alpha - G^a_\alpha)] = i_\alpha(\epsilon)$$

(3)

where $i_\alpha(\epsilon)$ gives the intradot vertical flow: $i_\alpha(\epsilon) \equiv ieU \int e^{i\epsilon t} \{ < d^\dagger_\alpha(0)d_\alpha(0)d_\alpha(t)d^\dagger_\alpha(t) > - < d^\dagger_\alpha(0)d^\dagger_\alpha(t)d_\alpha(t)d_\alpha(0) > \} \frac{dt}{2\pi}$. The quantity $i_\alpha(\epsilon)$ can be viewed as an intradot “vertical” current density \[1\] at energy $\epsilon$ that is contributed by carriers with other energies due to the e-e scattering process. Summing over the index $\alpha$, Eq.(3) reduces to $\sum_\alpha j_n(\epsilon) + \sum_\alpha i_\alpha(\epsilon) = 0$, which is exactly the steady state current conservation equation so that the total current density—including the vertical flow, through the intradot energy level $\epsilon$ is zero. If $G^r_\alpha(\epsilon)$ and $i_\alpha(\epsilon)$ have been solved, from Eq.(3) $G^<_\alpha(\epsilon)$ can be obtained immediately.

Next, we solve the vertical flow quantity $i_\alpha(\epsilon)$. We introduce two particle contour-ordered Green’s functions $B_\alpha(t, 0)$ and $B_c(t, 0)$, $B_\alpha(t, 0) \equiv - < T_C[d_\alpha(t)d_\alpha(0^+)d^\dagger_\alpha(0)] >$ and $B_c(t, 0) \equiv - < T_C[d_\alpha(t)d^\dagger_\alpha(0)d^\dagger_\alpha(t)] >$. Although there are four operators in the definition $B_{\alpha/c}(t, 0)$, only two time indices at $(t, 0)$ appear. We can therefore write the contour ordered quantities as $B^{+-}_{\alpha/c}(t, 0)$, $B^{+\pm}_{\alpha/c}$, $B^{-\pm}_{\alpha/c}$, and $B^{--}_{\alpha/c}$. Their Fourier transformations can be defined as $B(\epsilon) \equiv \int e^{i\epsilon t} B(t, 0) dt$. Using the two-particle Green’s function, the vertical flow $i_\alpha(\epsilon)$ is reduced to $i_\alpha(\epsilon) = -\frac{eU}{\pi} Im B^{+-}_\alpha(\epsilon)$.

The contour Green’s function $B_{\alpha/c}$ is solved by means of a Feynman diagram expansion using the Wick’s theorem. We take the interacting part of the Hamiltonian as $H_I = Ud^\dagger \uparrow d_\uparrow d^\dagger \downarrow d_\downarrow$, and the remaining part $H - H_I$ as the non-interacting Hamiltonian $H_0$. 


The first-order irreducible self-energy which we consider is shown in Fig.1. It is clear that the first-order graph describes a two-particle propagation involving the exchange of an interacting energy $U$. Two important points should be mentioned here. (i) The vertex we calculate sums up all reducible diagrams constructed by the irreducible self-energy of Fig.1 (also see Eq.(4)). This level of approximation is equivalent to that of a typical random phase approximation, \textit{i.e.} we compute the irreducible self-energy up to order $U^{-1}$. It is reasonable to neglect other higher order irreducible diagrams which are of orders $U^{-2}$ and higher, because the interaction energy $U$ is large. (ii) Single solid lines in Fig.1 stand for the intradot contour-ordered Green’s functions $G_\alpha$ of the Hamiltonian $H$ (not $H_0$). This means that we have summed over all terms in the Feynman diagram expansion of $G_\alpha$.

The corresponding equation for Fig.1 is

$$B_{c/\alpha}(t,0) = -G_\alpha(t,0)G_\bar{\alpha}(t/0^+,0) + iU \int_{t_1} dt G_\alpha(t,t_1)G_\bar{\alpha}(t/0^+,t_1)B_c(t_1,0).$$

Finally, we get $B^+_{c/\alpha}(t,0)$, $B^-_{c/\alpha}(t,0)$, and $B^{+-}_{c/\alpha}(t,0)$ from Eq.(4), and upon taking a Fourier transformation, $B^{+-}_{\alpha}(\epsilon)$ can be expressed in terms of $G^{++}_{\alpha}(\epsilon)$, $G^{+-}_{\alpha}(\epsilon)$, $G^{-+}_{\alpha}(\epsilon)$, and $G^{-+}_{\alpha}(\epsilon)$—these four Green’s functions are directly related to $G^r_\alpha$ and $G^<_\alpha$. This completes the analytical derivations.

From Eqs. (2,3,4), the intradot occupation number $n_\alpha$ and the vertical flow $i_\alpha(\epsilon)$ are determined self-consistently. Let’s consider a two-probe ($n = L, R$) device and the wideband approximation in which $\Gamma_n(\epsilon)$ is independent of energy $\epsilon$. Fig.2 plots the vertical flow $i(\epsilon) = \sum_\alpha i_\alpha(\epsilon)$ (solid line) as well as the current density $j_{L/R}(\epsilon)$ at a high bias. Clearly, the vertical flow $i(\epsilon)$ is non-zero due to the TET processes, and its value $|i(\epsilon)|$ has four peaks at energies $\epsilon_\uparrow$, $\epsilon_\downarrow$, $\epsilon_\uparrow + U$, and $\epsilon_\downarrow + U$, respectively. This means that the incident electron indeed can vary its energy in QD by e-e scattering. In contrast, in a typical SET process the electron keeps its energy and does not induce any vertical flow $i(\epsilon)$.

The physics of the TET process that induces the vertical flow is shown by inset (a) of Fig.2. To start, two incident electrons from left lead having energies $\epsilon_\downarrow$ and $\epsilon_\uparrow + U$ tunnel
into the QD. They scatter with each other inside the QD and exchange energy to final states \( \epsilon_\uparrow \) and \( \epsilon_\downarrow + U \). Afterwards they tunnel out of the QD. In the vertical flow curve of Fig.2, two peaks at \( \epsilon_\downarrow \) and \( \epsilon_\uparrow + U \) are negative (dips), and the other two peaks at \( \epsilon_\uparrow \) and \( \epsilon_\downarrow + U \) are positive: precisely indicating the transfer of states from the initial ones at \( \epsilon_\downarrow \) and \( \epsilon_\uparrow + U \) to the final ones at \( \epsilon_\uparrow \) and \( \epsilon_\downarrow + U \). It is also worth mentioning that besides the new TET process, the usual SET processes also exist in charge transport through the QD in the present case.

So far we have demonstrated that TET processes can exist. In the following we analyze several important questions concerning TET. What is its consequence? (i) TET makes current density a non-conserved quantity, i.e. \( \sum_n j_n(\epsilon) \neq 0 \). Of course, the total current is still conserved, i.e. \( \sum_n I_n = 0 \). This can be easily proved from the definition of the vertical flow \( i(\epsilon) \), namely \( i(\epsilon) \) has the property \( \int i(\epsilon) d\epsilon = 0 \). (ii) TET can involve high energy empty states, namely states which are higher by about \( U \) than the highest chemical potential \( \max(\mu_L, \mu_R) \) (see inset (a) of Fig.2); it may also involve electrons deep inside the Fermi sea, namely states which are lower by about \( U \) than the lowest chemical potential \( \min(\mu_L, \mu_R) \) (see TET process shown in inset (b) of Fig.2). (iii) TET may induce a current density that is flowing out from the high voltage terminal, i.e. the left lead (indicated by the negative peak at \( \epsilon_\downarrow + U \) in \( j_L(\epsilon) \) curve, and by the arrow A in inset (a) of Fig.2). Similarly, TET may also induce a current density that is flowing in from the low voltage terminal, i.e. the right lead (see arrow A in inset (b) of Fig.2). These characteristics are rather different from the typical elastic SET processes.

Under what conditions does TET or the vertical flow \( i(\epsilon) \) exist? (i) We found that an increase (decrease) of temperature \( T \) or linewidth \( \Gamma \) will widen (narrow) the peaks of vertical flow, but does not affect peak positions and heights significantly. (ii) If \( U = 0 \), the vertical flow \( i(\epsilon) = 0 \) identically: TET crucially depends on this parameter. (iii) If \( U \to \infty \), \( i(\epsilon) \) tends to zero. This is because at large \( U \), the intradot two-electron occupation is prohibited therefore TET is blockaded. In this case only SET processes occur. (iv) When bias potential \( eV = \mu_L - \mu_R \) is less than \( U \), \( i(\epsilon) \) decreases drastically. In the limit of \( eV = 0 \), \( i(\epsilon) = j_n(\epsilon) = 0 \).
Are observable quantities of charge transport affected by the TET process? (i) Clearly the current density $j_{L/R}(\epsilon)$ is affected significantly as already discussed above. (ii) In general, the current, conductance, and $n_\alpha$ will be affected significantly by TET (see below). The current noise, which reflects the e-e time correlation, will increase due to TET processes. However, if one uses the wideband approximation, the TET dependence in charge current will be lost.

In the rest of this paper, we apply the property of TET to design a device so that electrons can be pumped from a lead with lower chemical potential to another lead having a higher chemical potential. Consider a device with three leads ($n = 1, 2, 3$) and consider the non-wideband case. We use a model of quasi-square bands where the coupling $\Gamma_n(\epsilon) = \Gamma/\{\exp[(|\epsilon - c_n| - W)/0.05] + 1\}$, the width of the band is set by $2W = 1$ and its center at $c_n$ which is dependent on the terminal voltage $eV_n = \mu_n$ but $\mu_n - c_n$ is kept fixed. More specifically, let's assume lead 1 to be a p-type semiconductor with $\mu_1 - c_1 = 0.4$; lead 2 an n-type semiconductor with $\mu_2 - c_2 = -0.4$; and lead 3 a metal with $\mu_3 - c_3 = 0$ (see Fig.4). The energy diagram of the device is set by external voltages as that shown in Fig.4 so that $\mu_2 > \mu_1 > \mu_3$. The current density $j_n(\epsilon)$ in this case is shown in Fig.3. We note that $j_1(\epsilon)$ (dotted line) has two positive peaks at $\epsilon_\downarrow$ and $\epsilon_\uparrow + U$; $j_2(\epsilon)$ and $j_3(\epsilon)$ each has one negative peak at $\epsilon_\downarrow + U$ and $\epsilon_\uparrow$, respectively; and $i(\epsilon) = -\sum_n j_n(\epsilon)$ has two negative peaks at $\epsilon_\downarrow$ and $\epsilon_\uparrow + U$, and two positive peaks at $\epsilon_\downarrow + U$ and $\epsilon_\uparrow$. The current $I_n = \int j_n(\epsilon)d\epsilon$ is quite large. We emphasize two points for this pump. (i) In this device the SET process almost does not occur because bands of different leads do not overlap. Then, clearly, the large current $I_n$ originates from the TET process: $I_n \rightarrow 0$ if the vertical flow $i(\epsilon) \rightarrow 0$. This demonstrates that TET can significantly affect charge current in the general case of non-wideband coupling. (ii) The charge current in the terminal with the highest bias voltage, e.g. lead 2, is negative (solid line in the inset of Fig.3), which demonstrates the pump effect. The pumps works because when an electron tunnels from lead 1 to 3, it emits energy $U$ to pump another electron from lead 1 to lead 2, through the TET process.

More clearly, the working principle of the TET pump is summarized in Fig.4. (a) We
start from the situation where no charge is in the QD so that levels $\epsilon_\uparrow$ and $\epsilon_\downarrow$ are empty. In this situation, an electron in the Fermi sea of lead 1 having energy $\epsilon_\downarrow$ can easily tunnel into the QD (Fig.4a). (b) After this electron tunnels into the QD and occupies the QD level of $\epsilon_\downarrow$, the other intradot level $\epsilon_\uparrow$ is raised up to $\epsilon_\uparrow + U$, so that another electron with energy $\epsilon_\uparrow + U$ in lead 1 tunnels into the QD (Fig.4b). (c) When the second electron comes into QD, due to the e-e Coulomb interaction $U$, the level $\epsilon_\downarrow$ with its electron is raised up to $\epsilon_\downarrow + U$, leading to a negative peak at $\epsilon_\downarrow$ and a positive peak at $\epsilon_\downarrow + U$ in the vertical flow curve $i(\epsilon)$. Now the intradot two-electron system has total energy $\epsilon_\uparrow + \epsilon_\downarrow + U$ (Fig.4c). Afterwards the first electron in state $\epsilon_\downarrow + U$ easily tunnels to lead 2 and takes away energy $\epsilon_\downarrow + U$. The net effect is that the two electrons exchanged energy $U$, which is the TET process discussed above. When the first electron leaves the QD, the other electron at $\epsilon_\uparrow + U$ falls down to $\epsilon_\uparrow$, leading to a negative peak at $\epsilon_\uparrow + U$ and a positive peak at $\epsilon_\uparrow$ in the curve of $i(\epsilon)$. (d) Finally, the second electron tunnels to lead 3 (Fig.4d) and our device returns to its initial conformation of Fig.4a. This way an electron is pumped from lead 1 to lead 2, where $\mu_1 < \mu_2$, via the TET process. We emphasize that each tunneling event from Fig.4a-d is a first-order normal tunneling event in which tunneling occurs at two aligning states (not like higher-order virtual co-tunneling process) [7]. We therefore conclude that the TET process should have large probability to occur so that $i(\epsilon)$ can be near the unit value $e/h$ (see Fig.3).

We have also investigated the terminal voltage ($e.g.$ $V_3$) dependence of current $I_n$, shown in the inset of Fig.3. As $V_3$ is increased so that $eV_3 = \mu_3$ passes the lowest resonance state $\epsilon_\uparrow$, the tunneling event in Fig.4d can not occur and the TET process is blockaded, leading to a significant reduction of all currents $I_n$ (including the pumping current $I_2$) (see inset of Fig.3).

In summary, we have investigated the two-electron correlated scattering process in mesoscopic system. TET induces a vertical flow in the scattering region so that electrons enter and exit the device with different energies. TET is found to affect current density significantly, and the process can involve high empty states or/and low filled states of the leads. The properties of TET suggests an interesting working principle of an electron pump which
pumps charge carries from a lead with low chemical potential to another lead with a higher chemical potential. In fact, if the bands of lead 1 is full and bands of leads 2 and 3 are empty, these results are not affected. Our proposed pump is very different as compared to the electron-photon or parametric pumps [8,9]. It should be experimentally feasible even for devices fabricated in two-dimensional electron gas. In that case, the bands in our theory can be replaced with Landau levels.

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FIGURES

FIG. 1. Relevant Feynman diagrams considered in this work. The single solid lines, doubles solid lines, and the wave lines stand for the Green’s functions $G_\alpha$, $B_{\alpha/c}$, and interaction $U$, respectively.

FIG. 2. $j_{L/R}(\epsilon)$ and $i(\epsilon)$ versus energy $\epsilon$ in the wideband limit. The parameters are: $\epsilon^\uparrow = -0.5$, $\epsilon^\downarrow = 0.1$, $U = 1$, $T = \Gamma = 0.1$, and $\mu_L = -\mu_R = 0.7$. Insets (a) and (b) are schematic plots for two kinds of TET processes.

FIG. 3. The main plot shows $j_n(\epsilon)$ versus $\epsilon$ with $\mu_3 = -1.3$ and the inset shows $I_n$ versus $eV_3 = \mu_3$ for the non-wideband case. Other parameters are: $\epsilon^\uparrow = -1.1$, $\epsilon^\downarrow = -0.1$, $U = 1.2$, $T = \Gamma = 0.05$, $\mu_1 = 0.4$, and $\mu_2 = 0.9$. The dotted, solid, dashed curves in the main plot correspond to $j_1(\epsilon)$, $j_2(\epsilon)$, and $j_3(\epsilon)$, respectively; they correspond to $I_1, I_2, I_3$ in the inset.

FIG. 4. Schematic plots for the working principle of the TET charge pump.
Fig. 1
Fig. 2
Fig. 3

The figure shows the current density $j(\varepsilon)$ and the current $I(e\Gamma/h)$ as functions of energy $\varepsilon$. The inset highlights the behavior of the current at lower energies, labeled as $eV_3$. The x-axis represents the energy $\varepsilon$, while the y-axis represents the current density and current.
Fig. 4