Cotunneling at resonance for the single-electron transistor

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We study electron transport through a small metallic island in the perturbative regime. Using a diagrammatic real-time technique, we calculate the occupation of the island as well as the conductance through the transistor in forth order in the tunneling matrix elements, a process referred to as cotunneling. Our formulation does not require the introduction of a cut-off. At resonance we find significant modifications of previous theories and quantitative agreement with recent experiments.

Electron transport through small metallic islands is strongly influenced by the charging energy associated with low capacitance of the junctions [1–3]. A variety of single-electron effects, including Coulomb blockade phenomena and gate-voltage dependent oscillations of the conductance, have been observed. If the conductance of the barriers is low

$$\alpha_0 \equiv h/(4\pi^2 e^2 R_{\rm T}) \ll 1 \qquad (1)$$

they can be described within the "orthodox theory" [1] which treats tunneling in lowest order perturbation theory (golden rule). This corresponds to the classical picture of incoherent, sequential tunneling processes. On the other hand, there is experimental and theoretical evidence that in several regimes higher-order tunneling processes have to be taken into account.

First, in the Coulomb blockade regime, sequential tunneling is exponentially suppressed. The leading contribution to the current is a second-order process in α_0 where electrons tunnel via a virtual state of the island. Averin and Nazarov [4] evaluated the transition rate of this "inelastic cotunneling" process at zero temperature. A divergence which arises at finite temperature requires a regularization, which they treated by an approximation which is valid far away from the resonances. In this regime, their results were confirmed by experiments [5].

Second, even at resonance where sequential tunneling occurs, higher-order processes can have a significant effect on the gate-voltage dependent linear and nonlinear conductance [6,7]. Similar effects were discussed for the equilibrium properties of the single-electron box [8–12]. A diagrammatic real-time technique developed for metallic islands [6,7] as well as for quantum dots [13,14] allows a systematic description of the nonequilibrium tunneling processes. The effects from quantum fluctuations become observable either for strong tunneling $\alpha_0 \sim 1$ or at low temperatures $\alpha_0 \ln E_{\rm C}/T \sim 1$, where $E_{\rm C}$ denotes the charging energy (see below). The theory has been evaluated in the limit where only two adjacent charge states are included (even virtually). Therefore, it was necessary to introduce a band-width cut-off $\sim E_{\rm C}$. The predicted broadening of the conductance peak as well as the reduction of its height was confirmed qualitatively in recent experiments on a single-electron transistor in the strong tunneling regime by Joyez et al. [15]. However, a quantitative fit between theory and experiment requires using a renormalized value for the charging energy. This value has been determined independently in the experiments.

In this Letter, we use the real-time diagrammatic technique to obtain the current in second order in α_0 including *all* relevant states. In this case no cut-off is required; all terms are regularized in a natural way. This analysis allows an unambiguous comparison with experiments where only bare system parameters enter. At resonance we obtain new contributions compared to the earlier theory of electron cotunneling. They emerge from a change of the occupation probabilities and a renormalization of the charge excitation energy. For realistic parameters $T/E_{\rm C} \sim 0.05$ and $\alpha_0^{\rm L} = \alpha_0^{\rm R} \sim 0.02$ the corrections are of order 20%. We compare with recent experiments [15] and find excellent agreement without any fitting parameter.

The system is modeled by the Hamiltonian

$$H = H_{\rm L} + H_{\rm R} + H_{\rm I} + H_{\rm ch} + H_{\rm T} = H_0 + H_{\rm T}. \qquad (2)$$

Here $H_{\rm r} = \sum_{kn} \epsilon^{\rm r}_{kn} a^\dagger_{{\rm r}kn} a_{{\rm r}kn}$ and $H_{\rm I} = \sum_{qn} \epsilon_{qn} c^\dagger_{qn} c_{qn}$ describe the noninteracting electrons in the two leads r=L,R and on the island. The wave vectors k and q numerate the states of the electrons, while $n = 1, \ldots N$ is the transverse channel index which includes the spin. In the following, we consider "wide" metallic junctions with $N \gg 1$. The Coulomb interaction of the electrons on the island is described by the the capacitance model $H_{\rm ch} = E_{\rm C}(\hat{n} - n_{\rm x})^2$, where $E_{\rm C} = e^2/(2C)$ with $C = C_{\rm L} + C_{\rm R} + C_{\rm g}$. The excess particle number operator on the island is given by $\hat{n}$. Furthermore, the 'external charge' $en_{\rm x} = C_{\rm L}V_{\rm L} + C_{\rm R}V_{\rm R} + C_{\rm g}V_{\rm g}$, accounts for the applied gate and transport voltages. The charge transfer processes are described by the tunneling Hamiltonian

$$H_{\rm T} = \sum_{{\rm r=L,R}} \sum_{kqn} T^{{\rm r}n}_{kq} a^\dagger_{{\rm r}kn} c_{qn} {\rm e}^{-i\hat{\varphi}} + {\rm h.c.}\,. \qquad (3)$$

The matrix elements $T^{{\rm r}n}_{kq}$ are considered independent of the states k and q. They are related to the tunneling resistances $R_{\rm T,r}$ of the left and right junction by $\frac{1}{R_{\rm T,r}} = N\frac{2\pi e^2}{\hbar} N_{\rm r}(0) N_I(0) |T^{{\rm r}n}|^2$, where $N_{\rm I/r}(0)$ are the density of states of the island/leads. The operator ${\rm e}^{\pm i\hat{\varphi}}$ changes the charge on the island by $\pm e$.

We proceed using the diagrammatic technique developed in Ref. [6,7]. The nonequilibrium time evolution of the charge degrees of freedom on the island is described by a density matrix, which we expand in $H_{\rm T}$. The reservoirs are assumed to remain in thermal equilibrium (with electrochemical potential $\mu_{\rm r}$) and are traced out using Wick's theorem, such that the Fermion operators are contracted in pairs. For a large number of channels N, the "simple loop" configurations dominate where the two operators in $a^\dagger_{{\rm r}kn}c_{qn}$ from one term $H_{\rm T}$ are contracted with the corresponding two operators $c^\dagger_{qn}a_{{\rm r}kn}$ from another term $H_{\rm T}$, while the contribution of more complicated configurations are small.

The time evolution of the reduced density matrix in a basis of charge states is visualized in Fig. (1). The forward and backward propagator (on the Keldysh contour) are coupled by "tunneling lines", representing tunneling in junction r. In Fourier space they are given by

$$\alpha^\pm_{\rm r}(\omega) = \pm\alpha^{\rm r}_0 \frac{\omega-\mu_{\rm r}}{\exp[\pm\beta(\omega-\mu_{\rm r})]-1}\,. \qquad (4)$$

if the line is directed backward/forward with respect to the closed time path. Furthermore, we associate with each tunneling vertex at time t a factor $\exp(\pm i\Delta_n t)$ depending on the energy difference of the adjacent charge states $\Delta_n = E_{\rm ch}(n+1) - E_{\rm ch}(n)$. If the vertex lies on the backward path it acquires a factor -1. We define $\alpha_0 = \sum_{\rm r}\alpha^{\rm r}_0$ and $\alpha(\omega) = \sum_{\rm r}\alpha_{\rm r}(\omega)$.

The time evolution of the density matrix leads to a formally exact master equation [6,7]

$$\dot{p}_n = \sum_{n'\neq n}\left[p_{n'}\Sigma_{n',n} - p_n\Sigma_{n,n'}\right] \qquad (5)$$

for the probability p_n of charge state n and the transition rates $\Sigma_{n,n'}$ between n and n'. In the perturbative regime we write $p_n = p^{(0)}_n + p^{(1)}_n + p^{(2)}_n + \ldots$ and $\Sigma_{n,n'} = \Sigma^{(1)}_{n,n'} + \Sigma^{(2)}_{n,n'} + \Sigma^{(3)}_{n,n'} + \ldots$ where $p^{(k)}_n$ and $\Sigma^{(k)}_{n,n'}$ denotes the term $\sim \alpha_0^k$ of the expansion. The master equation must hold in each order. In lowest order (sequential tunneling) in the stationary state it reads $p^{(0)}_n\alpha^+(\Delta_n) - p^{(0)}_{n+1}\alpha^-(\Delta_n) = 0$. At low temperature at most two charge states ($n = 0, 1$) are important, all other states are suppressed exponentially.

Due to higher-order processes the occupation is modified and also the probability for the other charge states can be nonzero (they are algebraically suppressed, but not exponentially). The expansion of the master equation up to α_0^2 gives a relation between the rates in second order $\Sigma^{(2)}_{n,n'}$ (diagrams with two lines) and the occupation in first order $p^{(1)}_n$ which lead to a correction in the average occupation $\langle n\rangle = \sum_n np_n = \langle n\rangle^{(0)} + \langle n\rangle^{(1)} + \ldots$.

The stationary current $I_{\rm r} = -ie\sum_{n,n'}p_n\Sigma^{\rm r+}_{n,n'}$ through reservoir r uses the rates $\Sigma^{\rm r+}_{n,n'}$ where the rightmost tunneling line corresponds to reservoir r and is an outgoing (incoming) one if the rightmost vertex lies on the upper (lower) propagator (and vice versa for $\Sigma^{\rm r-}_{n,n'}$). There are two types of diagrams contributing to the second-order correction of the current $I^{(2)}$: those of the form $p^{(0)}\Sigma^{(2)}$ and others like $p^{(1)}\Sigma^{(1)}$. The first ones correspond to the cotunneling processes derived by Averin and Nazarov [4]. The second ones are due to changes in the occupation probabilities in higher orders. They have not been considered in previous theories, but is equally important as the first one.

In lowest order the average occupation $\langle n\rangle^{(0)} = \alpha^+(\Delta_0)/\alpha(\Delta_0)$ is smeared only by temperature. Quantum fluctuations yield in next order

$$\langle n\rangle^{(1)} = \frac{1}{2E_{\rm C}}\frac{\partial}{\partial n_{\rm x}}\left[p^{(0)}_0\left(\phi_0-\phi_{-1}\right) + p^{(0)}_1\left(\phi_1-\phi_0\right)\right] \qquad (6)$$

where $\phi_n = \sum_{\rm r}\alpha^{\rm r}_0(\Delta_n-\mu_{\rm r})\,{\rm Re}\,\Psi\left(i\frac{\beta}{2\pi}(\Delta_n-\mu_{\rm r})\right)$ and Ψ denotes the digamma function.

In equilibrium, i.e. at $V = 0$, the transistor is equivalent to the single-electron box. A systematic perturbative expansion of the partition function (up to order α_0^2) was performed by Grabert [12]. The result Eq. (6) is identical to his result in order α_0, which at $T = 0$ reads $\langle n\rangle^{(1)} = \alpha_0\ln[(1+2n_{\rm x})/(1-2n_{\rm x})]$. Eq. (6) generalizes the analysis to nonequilibrium situation, i.e. $V \neq 0$.

The current $I = I_{\rm L} = -I_{\rm R}$ is in lowest order given by

$$I^{(1)}(\Delta_0) = \frac{4\pi^2 e}{h}\frac{\alpha_{\rm L}(\Delta_0)\alpha_{\rm R}(\Delta_0)}{\alpha(\Delta_0)}\left[f_{\rm R}(\Delta_0) - f_{\rm L}(\Delta_0)\right]. \qquad (7)$$

The total second order, "cotunneling" contribution can be divided into three parts $I^{(2)}(\Delta_0) = \sum_{i=1}^3 I^{(2)}_i(\Delta_0)$

$$I^{(2)}_1(\Delta_0) = \int d\omega\, I^{(1)}(\omega)\alpha(\omega)\,{\rm Re}\left[p^{(0)}_0 R_-(\omega)^2 + p^{(0)}_1 R_+(\omega)^2\right], \qquad (8)$$

$$I^{(2)}_2(\Delta_0) = -I^{(1)}(\Delta_0)\int d\omega\,{\rm Re}\sum_{\sigma=\pm}\alpha^\sigma(\omega)R_\sigma(\omega)^2\,, \qquad (9)$$

$$I^{(2)}_3(\Delta_0) = -\frac{\partial I^{(1)}(\Delta_0)}{\partial\Delta_0}\int d\omega\,{\rm Re}\sum_{\sigma=\pm}\alpha^\sigma(\omega)R_\sigma(\omega)\,, \qquad (10)$$

where we used the definition $R_\pm(\omega) = \frac{1}{\omega-\Delta_0+i0^+} - \frac{1}{\omega-\Delta_{\pm1}+i0^+}$. The poles at $\omega = \Delta$ are regularized in a natural way (it comes out of our theory and is *not* added by hand) as Cauchy's principal values ${\rm Re}\frac{1}{x+i0^+} = P\frac{1}{x}$ and their derivative ${\rm Re}\frac{1}{(x+i0^+)^2} = -\frac{d}{dx}P\frac{1}{x}$.

In the Coulomb blockade regime, we have $p^{(0)}_0 = 1$, $p^{(0)}_1 = 0$ and $I^{(1)}(\Delta_0) = 0$. Consequently, only the first term of $I^{(2)}_1$ contributes. At $T = 0$, the integrand is zero at the poles, and we can omit the term $+i0^+$. This gives the well-known result of inelastic cotunneling [4]. At finite temperature, however, the regularization scheme is

needed which is not provided by previous theories [16]. Our result is also well-defined for $T \neq 0$.

Furthermore, we are able to describe the system at resonance. In this regime, $I_2^{(2)}$ and $I_3^{(2)}$ become important. The origin of the second term may intuitively be interpreted as the reduction of the first order contribution $I^{(1)}(\Delta_0)$ since quantum fluctuations lead to an occupation of the adjacent charge states $n = -1$ and 2. Therefore, the probability of the system to be in state $n = 0$ or 1 is decreased. The third term may indicate the appearance of a renormalization of the excitation energy Δ_0 [6,7,9,11]. Due to this renormalization the system is effectively "closer" to the resonance as the original parameters would suggest. The current would then, in second order, be roughly given by the derivative of the first order term times the renormalization.

The behavior of the system at resonance (and its crossover to the Coulomb blockade regime) was also described in Ref. [6,7] within the resonant tunneling approximation for the two charge state model. Therefore, the expansion of the resonant tunneling formula up to α_0^2 yields Eqs. (8) - (10) if we omit all terms with Δ_1 and Δ_{-1}. The integrals, then, become divergent and a cut-off (of the order of the charging energy) has to be introduced. In this Letter, however, we took into account all processes, and, therefore, no cut-off is needed.

In Fig. 2 we show the second-order contribution to the linear differential conductance $G = \partial I/\partial V$ (in the following we choose $\alpha_0^{\rm L} = \alpha_0^{\rm R}$). In Figs. 3 and 4 a comparison of the first order, the sum of the first and second order, and the resonant tunneling approximation (where the cut-off is adjusted at $E_{\rm C}$) is displayed for the linear and nonlinear regime. The deviation from sequential tunneling is significant and of the order 20%. The agreement with the resonant tunneling approximation provides a clear criterium for the choice of the bandwidth cut-off. Furthermore, and most importantly, it shows the existence of a parameter regime where renormalizations of $E_{\rm C}$, α_0, and Δ_0 by higher-order charge states can be neglected although the current deviates significantly from the classical result. We have checked the significance of third order terms $\sim \alpha_0^3$ by using the resonant tunneling formula [6,7] and exact results for the average charge in third-order at zero temperature [12]. For the parameter sets used in the figures, the deviations to the sum of first and second order terms were smaller than about 2%. Therefore, at not too low temperatures, second-order perturbation theory is a good approximation even if the tunneling resistance approaches the quantum resistance.

In Fig. 5 we compare our results with recent experiments [15]. The temperature dependence of the Coulomb oscillations were measured for two samples with different conductances. For one with $\alpha_0 = 0.015$, our results in second-order perturbation theory agree perfectly in the whole temperature and gate voltage range. Also for the other sample with $\alpha_0 = 0.063$ the agreement is very good. For still stronger tunneling higher-order effects such as the inelastic resonant tunneling [7] would be relevant.

We emphasize, that only bare values for α_0 and $E_{\rm C}$ have been used here, as determined unambiguously in the experiment. In contrast the resonant tunneling approximation with the bare value of the charging energy would lead to a deviation from the experiment by about 10%. Thus, the inclusion of higher-order charge states within second-order perturbation theory, as presented in this Letter, is an important improvement of the theory.

The peak conductance shown in Fig. 5 depends logarithmically on temperature. From Eqs. (8) - (10) we find at low temperature $G_{\rm max}/G_{\rm as} = \frac{1}{2} - \alpha_0 \left(\gamma + \log\left(\frac{E_C}{\pi T}\right)\right) + O(\alpha_0^2)$ with γ being Euler's constant and $G_{\rm as} = 1/(R_L + R_R)$ the asymptotic high temperature limit. This result may be interpreted as a renormalization of $G_{\rm as}$ or α_0 [6,7,9,11]. It shows a typical logarithmic temperature dependence since, at least in the equilibrium situation, the low-energy behavior of the system is expected to be that of the multichannel Kondo model [9]. While at resonance the new terms are crucial, the Coulomb blockade regime is sufficiently described by Eq. (8) to find a very good agreement between theory and experiment.

In conclusion we have presented a consistent calculation of the current of the single-electron transistor up to second-order perturbation theory (cotunneling). The approach is free of any divergences and provides cut-off independent results. At resonance we find new terms which are significant for experimentally realistic parameters and are responsible for logarithmic behavior. A comparison with experiments shows good quantitative agreement without renormalization of system parameters.

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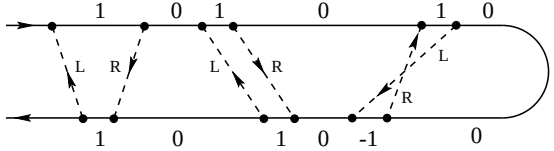

FIG. 1. A diagram showing contributions to sequential tunneling ($\Sigma^{\mathrm{L},-}_{0,1}$ and $\Sigma^{\mathrm{R},+}_{1,0}$) and cotunneling ($\Sigma^{\mathrm{R},+}_{0,0}$ and $\Sigma^{\mathrm{L},-}_{0,0}$).

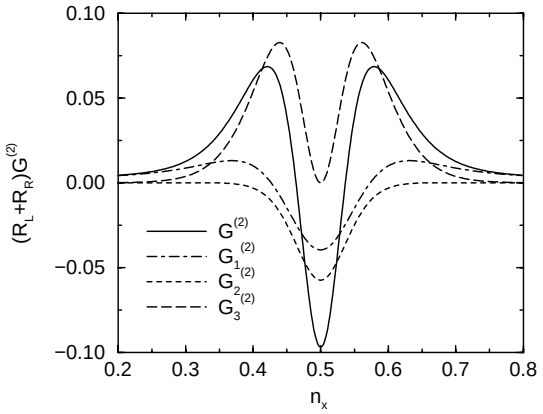

FIG. 2. The second-order contribution of the differential conductance $G^{(2)} = \sum_{i=1}^{3} G_i^{(2)}$ for $T/E_{\mathrm{C}} = 0.05$, $\alpha_0 = 0.04$ and $V = 0$.

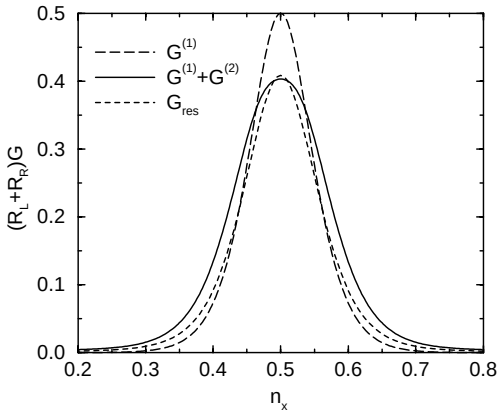

FIG. 3. The differential conductance for $T/E_{\mathrm{C}} = 0.05$, $\alpha_0 = 0.04$ and $V = 0$: sequential tunneling, sequential plus cotunneling, and resonant tunneling approximation.

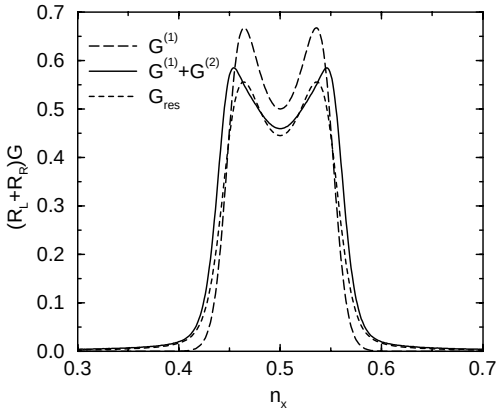

FIG. 4. The differential conductance for $T/E_C = 0.01$, $\alpha_0 = 0.02$ and $V/E_C = 0.2$: sequential tunneling, sequential plus cotunneling, and resonant tunneling approximation.

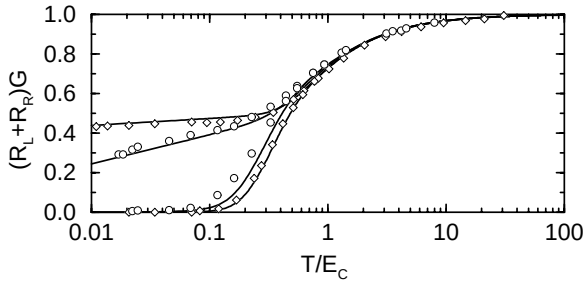

FIG. 5. Maximal and minimal linear conductance for $E_C = 1.47K$ and $\alpha_0 = 0.015$, and $E_C = 1K$ and $\alpha_0 = 0.063$. The dots are experimental data from Ref. [15].