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Master-equation approach to macroscopic quantum tunneling of charge in ultrasmall single-electron-tunneling double junctions

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It is known that classical Coulomb blockade effects of single-electron-tunneling (SET) devices are disturbed by higher-order tunneling effects denoted as macroscopic quantum tunneling of charge (q-MQT) or cotunneling. Starting from the von Neumann equation, a master-equation approximation of cotunneling is given for ultrasmall SET double junctions in the limit $T=0$. The influence of an external electromagnetic environment modeled by an additional impedance in the circuit is studied both in the low-impedance and high-impedance limits. General tunneling channels described by quasiparticle current amplitudes $\mathrm{Im}I_q(\omega)$ are considered just as the Ohmic approximation which leads to some known results. The rates of cotunneling show in the high-impedance case the effect of a "quantum" Coulomb blockade. By regularizing the logarithmic singularities in the quantum rates using linewidth effects, the master-equation description (Markov property) is complete in the sense of the tunneling theory up to fourth order.

I. INTRODUCTION

The understanding of tunneling of single electrons in ultrasmall normal conducting double junctions is of essential importance for the control of more complicated single-electron tunneling (SET) devices. The reason not to consider simpler single junctions is that for a voltage-driven single junction without any external environmental impedance there is no Coulomb blockade which is responsible for the interesting SET effects.[1,2] But in the case of a double junction one tunnel junction influences the other and vice versa producing in this way the Coulomb blockade. We take into account the presence of an electromagnetic environment modeled by an additional impedance $Z(\omega)$ in the circuit (cf. Fig. 1); this environment generates an additional effect and one junction is influenced by the other one and the external environment. Hence, the values of Coulomb blockade voltages are modified and consequently the physical effects, too. The model is characterized in Sec. III.

One can expect that the SET effects are relevant for low temperatures satisfying the relation

$$E_C=\frac{e^2}{2C}\gg k_B T\ .$$

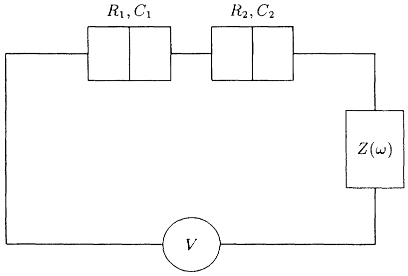

FIG. 1. Scheme of the considered circuit.

Typical SET junctions are characterized by capacities C of about $10^{-16}\ldots10^{-17}F$. To realize charge effects experimentally the quantum fluctuations should be significantly smaller than the typical energy E_C. This can be realized if the tunnel resistances of the junctions $R_{1/2}$ satisfy the relation

$$R_{1/2}\gg R_Q=\frac{h}{e^2}=25.8\ \mathrm{k}\Omega\ .$$

Typical tunnel resistances are at least of the order of 100 kΩ.

The influence of the environment on SET effects has frequently been discussed.[1,3-8] It is assumed that the charge distribution after a tunneling event relaxes in the electromagnetic environment in the circuit. For small $Z(\omega)$, i.e., in the low-impedance case

$$\mathrm{Re}Z(\omega)\ll R_Q\ ,$$

the relaxation is very fast. The contrary case is that of high impedance characterized by

$$\mathrm{Re}Z(\omega)\gg R_Q\ .$$

The stationary mean tunneling current in a double junction has been calculated in first-order perturbation theory with and without environment, using as well the "golden rule" arguments as the more fundamental master-equation model.[1,9,10] This last method is based on the general theory of the density matrix (cf. Sec. II). The master-equation approach of this process, denoted as ordinary tunneling, is sketched in Sec. IV.

Beyond this ordinary tunneling there is a tunneling effect of higher order denoted as macroscopic quantum tunneling of charge (q-MQT), or cotunneling. It has been investigated in the low-impedance case.[4,11-14] Here, one considers the tunneling process via the double junction as one act. This process is also possible for voltages below the Coulomb blockade because the energy difference for the whole process is negative even though the energy differences for the tunneling events at both junctions are positive. In other words, tunneling from the left to the right electrode or vice versa happens via virtual electron states on the central electrode. This process can be viewed as quantum tunneling through a barrier built by the two junctions and the central electrode which is macroscopic. Therefore, this process is called macroscopic quantum tunneling of charge (q-MQT). From another point of view this process looks like a simultaneous tunneling of two electrons through the two junctions which suggests the term cotunneling. Simultaneous means that the time difference obeys the Heisenberg uncertainty principle with the energy differences mentioned above.

Because there is no "classical" Coulomb blockade in this case, the second-order contributions to the tunneling current, even though they are much smaller than first-order contributions (by the factor $R_Q/R_{1/2}$), could disturb experiments based on the blockade effect which has really been observed.[15–18]

In a former paper[19] it has been shown that in the presence of a high-impedance environment, there is only a partial breakdown of the Coulomb blockade by cotunneling. A new "quantum" Coulomb blockade is generated which is approximately half the "classical" Coulomb blockade. The aim of this paper is to present the more fundamental master-equation approach of cotunneling in the zero-temperature limit. The master equation is derived in Sec. V.

How special tunnel channels are made available is described by general quasiparticle current amplitudes $\mathrm{Im}I_q(\omega)$. By using the Ohmic approximation, one gets the known results. The rates of the master equation of cotunneling show in the high-impedance case the effect of the "quantum" Coulomb blockade. These and other properties are specified in Sec. VI.

Averin and Nazarov[13,14] have shown that the q-MQT current consists of two parts. The first one which dominates depends on the product of the squared absolute values of the tunneling amplitudes $T^{(i)}$ and is, therefore, called incoherent. The second part is smaller than the first part by a factor Δ/E_C where Δ^{-1} is the electron density of states in the central electrode. This part depends on a mixture of the tunneling amplitudes of both junctions. In this way it describes the interference properties of q-MQT and is called coherent. This term depends on the internal structure of the central electrode and vanishes if Δ^{-1} becomes large.

Since the condition (30) (cf. Sec. V) was used this master-equation approach yields only the incoherent part of the mean cotunneling current.

The calculated cotunneling current has a logarithmic singularity at the "classical" Coulomb blockade.[4,11,12,19] This singularity is an artifact of the perturbation theory and has to be regularized. This can be done by using usual linewidth arguments of quantum theory (cf. Sec. VII). Simultaneously, this regularization guarantees the Markov property of the tunneling process which makes a master-equation description possible.

II. DENSITY OPERATOR THEORY

The dynamics of a system characterized by the Hamiltonian H_0 coupled to a reservoir labeled H_R will be described by the statistical operator ρ satisfying the von Neumann equation

$$\dot{\rho}(t)=\frac{1}{i\hbar}[H_0+H_R+H_T,\rho(t)] \ . \qquad (1)$$

The sum in brackets on the right-hand side corresponds to the total Hamiltonian. H_T denotes the interaction part which is given here by tunneling. It is assumed that the interaction is switched on adiabatically for $t\to-\infty$ (see Sec. IV). The operator ρ can be expressed using a quasiseparation[20] in the following way:

$$\rho=\sigma f_R+\Delta\rho \ . \qquad (2)$$

The density operator of the reservoir f_R is given by the canonical expression($\beta=1/k_BT$)

$$f_R=\frac{e^{-\beta H_R}}{\mathrm{Tr}_R\{e^{-\beta H_R}\}} \ . \qquad (3)$$

Tr_R means the trace with respect to the reservoir states. Note that there are the equations

$$\mathrm{Tr}_R\{\Delta\rho\}=0 \ , \quad \sigma=\mathrm{Tr}_R\{\rho\} \ .$$

Using Eq. (1), the density operator σ of the dynamical system obeys the equation

$$\dot{\sigma}(t)=\frac{1}{i\hbar}[H_0,\sigma(t)]+\frac{1}{i\hbar}\mathrm{Tr}_R[H_T,\Delta\rho(t)] \ . \qquad (4)$$

The unknown interaction term $\Delta\rho$ in this equation can be determined by successive approximation

$$\Delta\rho(t)^{(n)}=\Delta\rho(t)^{(1)}+\frac{1}{i\hbar}\int_{-\infty}^{t}dt'U(t,t')\times\{(1-f_R\,\mathrm{Tr}_R)[H_T,\Delta\rho(t')^{(n-1)}]\}\times U^{-1}(t,t') \qquad (5)$$

where

$$\Delta\rho^{(1)}(t)=\frac{1}{i\hbar}\int_{-\infty}^{t}dt'U(t,t')\times[H_T,\sigma,(t')f_R]U^{-1}(t,t')$$

and

$$U(t,t')=\exp\left\{\frac{1}{i\hbar}(H_0+H_R)(t-t')\right\} .$$

Using Eq. (5) up to third order and the splitting of the unitary transformation

$$U(t,t')=R(t,t')S(t,t')$$

with

$$R(t,t')=\exp\left[\frac{1}{i\hbar}H_R(t-t')\right] ,$$

$$S(t,t')=\exp\left[\frac{1}{i\hbar}H_0(t-t')\right] ,$$

Eq. (4) can be written as

$$\dot{\sigma}(t)=F_T^{(1)}[\sigma(t)]+F_T^{(2)}[t;\sigma(t)]+F_T^{(3)}[t;\sigma(t)]+F_T^{(4)}[t;\sigma(t)]+\dots . \quad (6)$$

The terms on the right-hand side are given by

$$F_T^{(1)}=\frac{1}{i\hbar}[H_0,\sigma(t)] , \quad (7)$$

$$F_T^{(2)}=\frac{1}{(i\hbar)^2}\int_{-\infty}^{t}dt'\,\mathrm{Tr}_R\{[H_T,S(t,t')[H_T(t'-t),\sigma(t')f_R]S^\dagger(t,t')]\} , \quad (8)$$

$$F_T^{(3)}=\frac{1}{(i\hbar)^3}\int_{-\infty}^{t}dt'\int_{-\infty}^{t'}dt''\,\mathrm{Tr}_R\{[H_T,S(t,t')[H_T(t'-t),S(t',t'')[H_T(t''-t),\sigma(t'')f_R]S^\dagger(t',t'')]S^\dagger(t,t')]\} , \quad (9)$$

$$F_T^{(4)}=\frac{1}{(i\hbar)^4}\int_{-\infty}^{t}dt'\int_{-\infty}^{t'}dt''\int_{-\infty}^{t''}dt'''\,\mathrm{Tr}_R\{[H_T,S(t,t')[H_T(t'-t),S(t',t'')\times((1-f_R\,\mathrm{Tr}_R)[H_T(t''-t),S(t'',t''')[H_T(t'''-t),\sigma(t''')f_R]\times S^\dagger(t'',t''')]S^\dagger(t',t'')]S^\dagger(t,t')]\} . \quad (10)$$

$$F_T^{(n)}[t;\sigma(t)]\propto\frac{1}{(i\hbar)^n} .$$

It makes no sense to add in Eq. (6) terms of still higher than fourth order in $1/(i\hbar)$. The reason is that the last term already corresponds to the usual perturbation-theory treatment with respect to one single junction. Higher-order terms would destroy consistency with the model of the tunneling Hamiltonian which is as an approximation limited to small transmittancy. Because there is no explicit time dependence of H_T the time argument in $H_T(\tau)$ corresponds to the unitary transformation

$$H_T(\tau)=R^\dagger(\tau)H_TR(\tau) .$$

III. MODEL

This theory will be applied to tunneling in a double junction with the electromagnetic environment of Fig. 1. The dynamical part of the total Hamiltonian is given by an operator expression of the Coulomb energy. The operators are not labeled in a special way to distinguish them from ordinary c numbers. Using the notations $C_\Sigma=C_1+C_2$, $\kappa_i=C_j/C_\Sigma$, $i,j=1,2$, and $i\neq j$, one writes

$$H_0=\frac{(Q_1-Q_2)^2}{2C_\Sigma}-V(\kappa_1Q_1+\kappa_2Q_2) . \quad (11)$$

$Q_{1,2}=en_{1,2}$ corresponds to the charges which have already tunneled through the respective junction . It is convenient to introduce operators of the difference charge q and of the total charge Q as follows:

$$q=Q_1-Q_2 , \quad Q=\kappa_1Q_1+\kappa_2Q_2 . \quad (12)$$

Now, H_0 can be written as

$$H_0=\frac{q^2}{2C_\Sigma}-VQ . \quad (13)$$

Corresponding to the principles of quantum theory, one defines as canonical conjugates of the charge operators the flux operators $\Phi_{1,2}$ and $\{\phi,\Psi\}$, respectively, with

$$\Phi=\Phi_1+\Phi_2 , \quad \Psi=\kappa_2\Phi_1-\kappa_1\Phi_2$$

which obey the commutation relations

$$[\Phi_i,Q_j]=i\hbar\delta_{ij} , \quad [\Psi,q]=i\hbar , \quad [\Phi,Q]=i\hbar . \quad (14)$$

The reservoir Hamiltonian consists of the terms corresponding to the left (Λ), right (P), and central (M) electrodes as well as the environment (E) and reads as

$$H_R=H_\Lambda+H_P+H_M+H_E \quad (15)$$

with

$$H_\Lambda=\sum_\lambda\epsilon_\lambda a_\lambda^\dagger a_\lambda , \quad (16)$$

$$H_P=\sum_\rho\epsilon_\rho b_\rho^\dagger b_\rho , \quad (17)$$

$$H_M=\sum_\mu\epsilon_\mu d_\mu^\dagger d_\mu . \quad (18)$$

a_λ, b_ρ, and d_μ stand for quasi-electron annihilation operators satisfying the anticommutation relations whereas ϵ denotes the energies of the quasiparticles in the electrodes with respective wave vectors. The spin is neglected. Here, e.g., metallic systems with a high density of electron states are presumed. This provides the justification of the ansatz (11) with classical capacities. The environment is modeled by a chain of LC circuits labeled by the index q. It corresponds to a sum of harmonic oscillators described by the Bose operators c_q (Refs. 1, 5, 6, and 21) in the following way:

$$H_E=\sum_q\hbar\omega_qc_q^\dagger c_q . \quad (19)$$

In what follows it is necessary to take the continuum limit with respect to the index q.

Corresponding to Eq. (15) the reservoir density operator f_R can be split into a product of density operators belonging to the electrodes (e) and the environment (E),

$$f_R=f_e f_E \ . \tag{20}$$

The interaction part of the Hamiltonian describes the tunneling of an electron through the respective junction which is connected to an excitation in the electromagnetic environment. Following Grabert and co-workers[1,5] one has

$$H_T=H_{1^+}+H_{1^-}+H_{2^+}+H_{2^-} \ , \tag{21}$$

with

$$H_{1^+}=\sum_{\lambda,\mu} T^{(1)}_{\lambda\mu} d^\dagger_\mu a_\lambda e^{-i(e/\hbar)\Phi_1} \ , \quad H_{1^-}=H^\dagger_{1^+} \ , \tag{22}$$

$$H_{2^+}=\sum_{\mu,\rho} T^{(2)}_{\mu\rho} b^\dagger_\rho d_\mu e^{-i(e/\hbar)\Phi_2} \ , \quad H_{2^-}=H^\dagger_{2^+} \tag{23}$$

and tunneling matrix elements $T^{(1)}$ and $T^{(2)}$. The terms labeled "+" in Eq. (21) mean tunneling from the left to the right (cf. Fig. 1), in contrast to the Hermitian conjugates which describe the reverse process. The basic algebra underlying this approach is the following relation:[22,23]

$$H_{i\pm}F(Q_j)=F(Q_j\mp e\delta_{ij})H_{i\pm} \ , \tag{24}$$

where F is an arbitrary function of the junction charge operators Q_i, $i=1,2$.

IV. MASTER EQUATION OF ORDINARY TUNNELING

The evaluation of the density matrix according to Eq. (6) up to second order in $1/(i\hbar)$ is determined only by the term $F^{(2)}_T[t;\sigma(t)[$. This corresponds to the Averin-Likharev theory[22–25] of single-electron tunneling without cotunneling. Then, Eq. (6) leads for $R_Q \ll R_i$ to a classical master equation for the diagonal elements σ in the n representation ($n=n_1-n_2$). As has already been mentioned, this condition guarantees that quantum fluctuations can be neglected. The resulting master equation reads as

$$\begin{aligned}\dot\sigma(n,t)=&[r_1(n-1)+l_2(n-1)]\sigma(n-1,t)\\&+[l_1(n+1)+r_2(n+1)]\sigma(n+1,t)\\&-[r_1(n)+l_1(n)+r_2(n)+l_2(n)]\sigma(n,t) \ .\end{aligned} \tag{25}$$

The rate coefficients $r_{1,2}$ (to the right) and $l_{1,2}$ (to the left) are given by the energy differences connected with tunneling of single electrons at the respective junction depending only on the netto charge $q=ne$. Details can be found in Refs. 1, 2, 9, and 24–27.

The calculation of the mean current shows in the zero-temperature limit that there is an exact classical Coulomb blockade $V^{(\mathrm{cl})}_{\mathrm{bl}}$. One finds

$$V^{(\mathrm{cl})}_{\mathrm{bl}}=\min\left[\frac{e}{2C_1},\frac{e}{2C_2}\right] \text{ for } \mathrm{Re}Z(\omega)\ll R_Q \ , \tag{26}$$

$$V^{(\mathrm{cl})}_{\mathrm{bl}}=\frac{e}{2C}=\frac{e}{2}\left[\frac{1}{C_1}+\frac{1}{C_2}\right] \text{ for } \mathrm{Re}Z(\omega)\gg R_Q \ . \tag{27}$$

These alternatives characterize the low- and high-impedance cases, respectively.

V. MASTER EQUATION OF COTUNNELING

Going beyond this approximation,[28,29] where cotunneling via both junctions has to be taken into account, the next relevant term in Eq. (6) is $F^{(4)}_T$ because $F^{(3)}_T$ vanishes. The calculation of cotunneling effects is only interesting in the region of the classical Coulomb blockade where contributions from $F^{(2)}_T$ do not play any role. On the other hand, beyond the classical blockade threshold cotunneling effects are irrelevant because they are strongly suppressed compared with ordinary tunneling. The following considerations are limited to zero temperature without exception.

For the evaluation of matrix elements of $F^{(4)}_T[t;\sigma(t)]$, we will give some hints as to the technical procedure.

(i) It is necessary to calculate the correlation functions of the flux operators.[1,4,5]

They arise from the evaluation of

$$\langle H_{i\pm}(t_1)H_{i\mp}(t_2)\rangle_R \ .$$

Note that $\langle\ \rangle=\mathrm{Tr}_R\{f_R\cdots\}$. In this manner one gets

$$\left\langle \exp\left[\mp\frac{ie}{\hbar}\Phi_j(\tau)\right]\exp\left[\pm\frac{ie}{\hbar}\Phi_j\right]\right\rangle_E=\exp[\kappa_j^2 J(\tau)] \ , \tag{28}$$

where J is given in the zero-temperature case by

$$J(\tau)=\int_{-\infty}^{\infty}\frac{\mathrm{Re}[Z_t(\omega)]}{\omega R_Q}[\cos\omega\tau-1-i\sin\omega\tau]d\omega \ . \tag{29}$$

Z_t denotes the total impedance of the circuit

$$Z_t^{-1}(\omega)=i\omega C+Z^{-1}(\omega)$$

with $1/C=1/C_1+1/C_2$. The averaging procedure $\langle\ \rangle_E$ in Eq. (28) means tracing with respect to the states of the environment. The time dependence of the phase factors correspond to the unitary transformation

$$\exp\left[\mp\frac{ie}{\hbar}\Phi(\tau)\right]=\exp\left[\frac{i}{\hbar}H_E\tau\right]\exp\left[\mp\frac{ie}{\hbar}\Phi\right]\times\exp\left[-\frac{i}{\hbar}H_E\tau\right] \ ,$$

It is assumed that tunneling events at different junctions are not correlated ($i\neq j$),

$$\langle(H_{i\pm}(t_1)H_{j\mp}(t_2)\rangle_R=0 \ . \tag{30}$$

Effects of correlated cotunneling have been investigated by Averin and Nazarov[13,14] using another approach. Depending on a factor Δ/E_C where Δ is the energy splitting of the electron states of the central electrode and $E_C=e^2/(2C_\Sigma)$, they are suppressed against incoherent cotunneling. This is justified for metallic systems with high densities of states. In this way all correlations of higher order with respect to one junction are, in principle, neglected.

(ii) The traces can be carried out following the scheme

$$\mathrm{Tr}_R\{H_{i^\pm}(t)H_{j^\pm}(t')(1-\mathrm{Tr}_R)\{H_{k^\pm}(t'')H_{l^\pm}(t''')f_R\}\}$$
$$=\mathrm{Tr}_R\{H_{i^\pm}(t)H_{j^\pm}(t')H_{k^\pm}(t'')H_{l^\pm}(t''')f_R\times[\delta_{ik}\delta_{jl}(1-\delta_{ij})+\delta_{il}\delta_{jk}(1-\delta_{ij})]$$
$$-H_{i^\pm}(t)H_{j^\pm}(t')f_R\,\mathrm{Tr}_R\{H_{k^\pm}(t'')H_{l^\pm}(t''')f_R\}\delta_{kl}\delta_{ij}\delta_{ki}\}\ .$$

(iii) Furthermore, the correlations $\langle H_{i^\pm}(\tau)H_{i^\mp}\rangle_R$ can be expressed using the Fourier representation including the imaginary part of the so called "quasiparticle current amplitudes" $\mathrm{Im}I_{q_i}(\omega)$

$$\langle H_{i^\pm}(\tau)H_{i^\mp}\rangle_R=-\frac{\hbar^2}{2\pi e}\int_{-\infty}^{0}e^{i\omega\tau}\mathrm{Im}I_{q_i}(\omega)d\omega e^{\kappa_i^2 J(\tau)}\ . \tag{31}$$

(iv) The density operator $\sigma(t)$ is represented by the diagonal elements

$$\langle n_1,n_2|\sigma(t)|n_2,n_1\rangle\equiv\sigma(n_f,t)\ .$$

n_f is an abbreviation for $\{n_1,n_2\}$. Now, the functional $F_T^{(4)}[t;\sigma(t)]$ can be expressed as the sum of a loss (L) and a gain (G) term and from Eq. (4) one gets the equation of motion

$$\dot\sigma(n_f,t)=F_T^{(4)}[t;\sigma(n_f,t)]_{(L)}+F_T^{(4)}[t;\sigma(n_f-1,t)]_{(G)}\ .$$

It can be shown that, with respect to the nondiagonal elements, there are further autonomous equations of the same type. But it is sufficient to concentrate on the diagonal elements because, according to the hypotheses of the adiabatic switch on of the interaction, the nondiagonal elements vanish due to proper chosen initial conditions. For instance, the loss term can be expressed by

$$\begin{aligned}
F_T^{(4)}&[t;\sigma(n_f,t)]_{(L)}\\
&=-\left[\frac{\hbar^2}{2\pi e}\right]^2\sum_{l,j=1,2}\int_{-\infty}^{\infty}d\omega\int_{-\infty}^{\infty}d\xi\,\mathrm{Im}I_{q_l}(\omega)\,\mathrm{Im}I_{q_j}(\xi)\Theta(-\omega)\Theta(-\xi)\\
&\quad\times\int_{-\infty}^{t}d\tau\int_{-\infty}^{\tau}d\eta\int_{-\infty}^{\eta}d\zeta\,2\,\mathrm{Re}\left\{\exp\left[-\frac{i}{\hbar}[\Delta E(n_f+\delta_{fl};n_f)(t-\tau)+\Delta E(n_f+\delta_{fl}+\delta_{fj};n_f)(\tau-\eta)]\right]\right.\\
&\qquad\times\left[\exp\left[-\frac{i}{\hbar}[\Delta E(n_f+\delta_{fj};n_f)(\eta-\zeta)-\hbar\omega(t-\eta)-\hbar\xi(\tau-\zeta)]\right]\right.\\
&\qquad\left.+\exp\left[-\frac{i}{\hbar}[\Delta E(n_f+\delta_{fl};n_f)(\eta-\zeta)-\hbar\omega(t-\zeta)-\hbar\xi(t-\eta)]\right]\right]\\
&\qquad+\exp\left[-\frac{i}{\hbar}[\Delta E(n_f+\delta_{fl};n_f)(t-\tau)+\Delta E(n_f+\delta_{fl};n_f+\delta_{fj})(\tau-\eta)]\right]\\
&\qquad\times\left[\exp\left[-\frac{i}{\hbar}[\Delta E(n_f;n_f+\delta_{fj})(\eta-\zeta)-\hbar\omega(t-\eta)-\hbar\xi(\zeta-\tau)]\right]\right.\\
&\qquad\left.\left.+\exp\left[-\frac{i}{\hbar}[\Delta E(n_f+\delta_{fl};n_f)(\eta-\zeta)-\hbar\omega(t-\zeta)-\hbar\xi(\eta-\tau)]\right]\right]\right\}\\
&\quad\times\sigma(n_f,\zeta)(1-\delta_{lj})\ .
\end{aligned} \tag{32}$$

The last bracket guarantees cotunneling explicitly because individual tunnel events at single junctions ($l=j$) are forbidden. In the following, only the special cases of low-impedance environment [$\mathrm{Re}Z(\omega)\ll R_Q$] and high-impedance environment [$ReZ(\omega)\gg R_Q$] are considered which can be treated analytically.

Using the notations

$$H_0|n_f\rangle=E(n_f)|n_f\rangle\ ,$$
$$\Delta E(n_f;n_f')=E(n_f)-E(n_f')\ ,$$

the energy differences read for an uncharged central electrode ($n_1-n_1=0$) as

$$\mathrm{Re}Z(\omega)\ll R_Q:\ \Delta E(n_1+1,n_2;n_1,n_2)=\frac{e^2}{C_\Sigma}-\kappa_1 eV\ ,$$
$$\Delta E(n_1,n_2+1;n_1,n_2)=\frac{e^2}{C_\Sigma}-\kappa_2 eV\ ;$$
$$\mathrm{Re}Z(\omega)\gg R_Q:\ \Delta E(n_1+1,n_2;n_{1,}n_2)=\frac{e^2}{C_1}-\kappa_1 eV\ ,$$
$$\Delta E(n_1,n_2+1;n_1,n_2)=\frac{e^2}{C_2}-\kappa_2 eV\ .$$

In what follows, both cases are treated together. One should mention that the process is Markovian if $\sigma(n_f,t)$ in the integrand of the integral (32) is only a slowly varying function of time. Then it is possible to write

$$F_T^{(4)}[t;\sigma(n_f,t)]_{(L)}=-r_{(L)}^{(Q)}\sigma(n_f,t)\ .$$

This property is ensured if the quantum rate which is developing from the integration (32) is

$$r_{(L)}^{(Q)}=\left[\frac{\hbar^2}{2\pi e}\right]^2\sum_{\substack{l,j=1,2\\ \alpha=\pm1}}\int_{-\infty}^{\infty}d\omega\int_{-\infty}^{\infty}d\xi\,\mathrm{Im}I_{q_l}(\omega)\,\mathrm{Im}I_{q_j}(\xi)\Theta(-\omega)\Theta(-\xi)(1-\delta_{lj})$$
$$\times\left\{\frac{1}{\omega-\Delta E(n_f+\delta_{fl};n_f)/\hbar+i\alpha\epsilon}\,\frac{1}{\omega+\xi-\Delta E(n_f+\delta_{fl}+\delta_{fj};n_f)/\hbar+i\alpha\gamma}\right.$$
$$\times\left[\frac{1}{\xi-\Delta E(n_f+\delta_{fj};n_f)/\hbar+i\alpha\nu}+\frac{1}{\omega-\Delta E(n_f+\delta_{fl};n_f)/\hbar+i\alpha\nu}\right]$$
$$+\frac{1}{\omega-\Delta E(n_f+\delta_{fl};n_f)/\hbar+i\alpha\epsilon}\,\frac{1}{\omega-\xi-\Delta E(n_f+\delta_{fl};n_f+\delta_{fj})/\hbar+i\alpha\gamma}$$
$$\left.\times\left[\frac{1}{-\xi-\Delta E(n_f;n_f+\delta_{fj})/\hbar+i\alpha\nu}+\frac{1}{\omega-\Delta E(n_f+\delta_{fl};n_f)/\hbar+i\alpha\nu}\right]\right\}$$

satisfies the condition

$$\hbar r_{(L)}^{(Q)}\ll\frac{e^2}{2C_\Sigma}\ . \tag{33}$$

The variables ϵ, γ, and ν take the adiabatic switch on of the interaction into account where the sequence

$$\epsilon>\gamma>\nu\ ,\quad \epsilon\to0^+$$

is valid. During the calculation one repeatedly meets two types of integrals

$$I_1=\int_{-\infty}^{\infty}d\omega\int_{-\infty}^{\infty}d\xi\,\mathrm{Im}I_{q_l}(\omega)\,\mathrm{Im}I_{q_j}(\xi)\Theta(-\omega)\Theta(-\xi)\frac{1}{z_1\omega-a+i\epsilon}\,\frac{1}{z_1\omega+z_2\xi-b+i\gamma}\,\frac{1}{z_2\xi-c+i\nu}\ ,$$
$$I_2=\int_{-\infty}^{\infty}d\omega\int_{-\infty}^{\infty}d\xi\,\mathrm{Im}I_{q_l}(\omega)\,\mathrm{Im}I_{q_j}(\xi)\Theta(-\omega)\Theta(-\xi)\frac{1}{y_1\omega-a+i\epsilon}\,\frac{1}{y_1\omega+y_2\xi-b+i\gamma}\,\frac{1}{y_2\omega-c+i\nu}\ , \tag{34}$$

where $z_{1,2}=\pm1$, $y_{1,2}=\pm1$ and a, b, and c are real numbers. One is only interested in the imaginary parts of these integrals since the real parts do not contribute to the quantum rates. The integrations which can be done analytically yield

$$\mathrm{Im}I_1=-\frac{\pi}{a+c-b}\left\{\mathrm{Im}I_{q_j}(b-a)\,\mathrm{Im}I_{q_l}(a)z_1\left[\Theta(z_1z_2)\Theta(-z_2b)\ln\left|\frac{b-a}{a}\right|\right.\right.$$
$$\left.+\Theta(-z_1z_2)\ln|\max[z_2(b-a),-z_2a]|-\Theta(-z_1a)\ln|b-a|\right]$$
$$+\mathrm{Im}I_{q_j}(c)\,\mathrm{Im}I_{q_l}(b-c)z_1\left[\Theta(z_1z_2)\Theta(-z_2b)\ln\left|\frac{b-c}{c}\right|\right.$$
$$-\Theta(-z_1z_2)\ln|\max[z_2c,-z_2(b-c)]|$$
$$\left.-\Theta(-z_2c)z_1z_2\ln|b-c|\right]$$
$$\left.+\mathrm{Im}I_{q_j}(c)\,\mathrm{Im}I_{q_l}(a)[\Theta(-z_2c)z_2\ln|a|+\Theta(-z_1a)z_1\ln|c|]\right\},$$

and

$$\mathrm{Im}I_2=-\frac{\pi}{a-c}\left\{\mathrm{Im}I_{q_j}(b-a)\,\mathrm{Im}I_{q_l}(a)y_1y_2\left[\Theta(y_1y_2)\Theta(-y_1b)\ln\left|\frac{a}{b-a}\right|\right.\right.$$
$$\left.+\Theta(-y_1y_2)\ln|\max[y_1a,y_2(b-a)]|+\Theta(-y_1a)y_1y_2\ln|b-a|\right]$$
$$-\mathrm{Im}I_{q_l}(c)\,\mathrm{Im}I_{q_j}(b-c)y_1y_2\left[\Theta(y_1y_2)\Theta(-y_1b)\ln\left|\frac{c}{b-c}\right|\right.$$
$$+\Theta(-y_1y_2)\ln|\max[y_1c,y_2(b-c)]|$$
$$\left.\left.+\Theta(-y_1c)y_1y_2\ln|b-c|\right]\right\}.$$

Note that the logarithms of type $\ln|d|$ are singular objects in the sense of $\lim_{x\to\infty}\ln|d/x|$. But further analysis shows that due to the presence of the unit-step function $\Theta(x)$ these terms vanish for $V<V_{\mathrm{bl}}^{(\mathrm{cl})}$. Finally, the quantum rate is given by the expression

$$r_{(L)}^{(Q)}=\frac{\Theta(-b)\hbar}{2\pi\frac{e^4}{C_\Sigma}}\left\{\ln\frac{|b-a|}{|a|}\left\{2\,\mathrm{Im}I_{q_l}(a)\,\mathrm{Im}I_{q_2}(b-a)-\frac{e^2}{\hbar C_\Sigma}[\mathrm{Im}I'_{q_1}(a)\,\mathrm{Im}I_{q_2}(b-a)-\mathrm{Im}I_{q_1}(a)\,\mathrm{Im}I'_{q_2}(b-a)]\right\}\right.$$
$$+\ln\frac{|b-c|}{|c|}\left\{2\,\mathrm{Im}I_{q_1}(b-c)\,\mathrm{Im}I_{q_2}(c)-\frac{e^2}{\hbar C_\Sigma}[\mathrm{Im}I_{q_1}(b-c)\,\mathrm{Im}I'_{q_2}(c)-\mathrm{Im}I'_{q_1}(b-c)\,\mathrm{Im}I_{q_2}(c)]\right\}$$
$$\left.+\frac{e^2b}{\hbar C_\Sigma}\left\{\mathrm{Im}I_{q_1}(a)\,\mathrm{Im}I_{q_2}(b-a)\frac{1}{a(b-a)}+\mathrm{Im}I_{q_1}(b-c)\,\mathrm{Im}I_{q_2}(c)\frac{1}{c(b-c)}\right\}\right\}. \tag{35}$$

The prime means differentiation with respect to the argument. In the case $n_1-n_2=0$ (cotunneling), the coefficients are given by

$$\hbar a=\Delta E(n_1+1,n_2;n_1,n_2)$$
$$=\frac{e^2}{2C_\Sigma}-eV\kappa_1+\phi\frac{C_2}{C_1}\frac{e^2}{2C_\Sigma}, \tag{36}$$

$$\hbar_c=\Delta E(n_1,n_2+1;n_1n_2)$$
$$=\frac{e^2}{2C_\Sigma}-eV\kappa_2+\phi\frac{C_1}{C_2}\frac{e^2}{2C_\Sigma}, \tag{37}$$

$$\hbar b=\Delta E(n_1+1,n_2+1;n_1,n_2)$$
$$=-eV+\phi\left[\frac{C_1}{C_2}+\frac{C_2}{C_1}\right]\frac{e^2}{2C_\Sigma}. \tag{38}$$

The variable ϕ means

$$\phi=\begin{cases}0 \text{ for } \mathrm{Re}Z(\omega)\ll R_Q\\ 1 \text{ for } \mathrm{Re}Z(\omega)\gg R_Q\end{cases}.$$

The gain term $F_T^{(4)}[t;\sigma(n_f,t)]_{(G)}$ can be calculated in the same manner. Finally, the master equation reads as

$$\dot{\sigma}(n_f,t) = -r^{(Q)}_{(L)}\sigma(n_f,t) + r^{(Q)}_{(G)}\sigma(n_f-1,t) \ . \tag{39}$$

VI. PROPERTIES OF COTUNNELING

The formulas (35)–(37) show that the cotunneling transition $n_1, n_2 \to n_1+1, n_2+1$ can be split into two subprocesses,

(1) $n_1,n_2 \to n_1+1,n_2 \to n_1+1,n_2+1$

(2) $n_1,n_2 \to n_1,n_2+1 \to n_1+1,n_2+1$,

where the first subprocess is described by those terms in $r^{(Q)}_{(L)}$ containing the frequency a. This corresponds to the virtual generation of an electron on the central electrode. In the opposite case the other subprocess which is connected with the virtual generation of a hole corresponds to the frequency term c. Furthermore, the equivalence of the transitions

$$(n_1-1,n_2-1) \to (n_1,n_2) \ ,$$

$$(n_1,n_2) \to (n_1+1,n_2+1) \ ,$$

is expressed by the equality

$$r^{(Q)}_{(L)} = r^{(Q)}_{(G)} \equiv r^{(Q)} \ .$$

The current amplitudes $\mathrm{Im} I_{q_j}$ describe how tunnel channels are available for the tunneling process. The quantum rate $r^{(Q)}$ depends of the product of the current amplitudes of both junctions. This corresponds to an incoherent tunneling process which is alone under consideration. One notes that

$$r^{(Q)} = 0 \text{ for } V < \phi \left[\frac{C_1}{C_2} + \frac{C_2}{C_1} \right] \frac{e}{2C_\Sigma}$$

independently of the specific structure of the current amplitudes. Consequently, for $\phi=1$ there is the "quantum Coulomb blockade"

$$V^{(Q)}_{\mathrm{bl}} = \frac{e}{2C_\Sigma} \left[\frac{C_1}{C_2} + \frac{C_2}{C_1} \right] \ . \tag{40}$$

Using the Ohmic approximation

$$\mathrm{Im} I_{q_{1,2}} = \frac{\hbar\omega}{eR_{1,2}} \ , \tag{41}$$

and the abbreviations

$$\tilde{V} = V - \phi \frac{e}{2C_\Sigma} \left[\frac{C_1}{C_2} + \frac{C_2}{C_1} \right] \ ,$$

$$E_1 = \hbar a \ , \quad E_2 = \hbar c \ ,$$

one gets

$$r^{(Q)} = \frac{1}{(2\pi)^3} \frac{R_Q^2}{R_1 R_2} \Theta(\tilde{V}) \frac{e\tilde{V}}{\hbar} \times \left\{ \left[1 + \frac{2}{e\tilde{V}} \frac{E_1 E_2}{E_1 + E_2 + e\tilde{V}} \right] \times \sum_{i=1}^{2} \ln \left| 1 + \frac{e\tilde{V}}{E_i} \right| - 2 \right\} \ , \tag{42}$$

Equation (42) can be approximated for small voltages. In the low-impedance case ($\phi=0$, $\tilde{V}=V \ll V^{(\mathrm{cl})}_{\mathrm{bl}}$), one gets[11,12]

$$r^{(Q)} = \frac{R_Q^2}{R_1 R_2} \frac{C_1 C_2}{\pi^3 e\hbar} V^3 \ . \tag{43}$$

In the high-impedance case ($\phi=1$, $\tilde{V}=V-V^{(Q)}_{\mathrm{bl}}$) $\ll V^{(\mathrm{cl})}_{\mathrm{bl}} - V^{(Q)}_{\mathrm{bl}}$), the analogous approximtion reads as

$$r^{(Q)} = \frac{R_Q^2}{R_1 R_2} \Theta(\tilde{V}) \frac{C_1 C_2}{48\pi^3 e\hbar} \frac{C_1^3 C_2^3}{C^6} \tilde{V}^3 \ . \tag{44}$$

In the special case of symmetric junction capacities ($\phi=1, C_1=C_2$) the quantum blockade is just the half of the classical blockade

$$V^{(\mathrm{cl})}_{\mathrm{bl}} = \frac{e}{C_1} \ , \quad V^{(Q)}_{\mathrm{bl}} = \frac{e}{2C_1} \ .$$

In the other special case of extreme asymmetric junction capacities ($\phi=1, C_1 \ll C_2$), both blockade voltages are equal

$$V^{(\mathrm{cl})}_{bl} = \frac{e}{2C_1} \ , \quad V^{(Q)}_{\mathrm{bl}} = \frac{e}{2C_1} \ .$$

In general, one can state that $V^{(Q)}_{\mathrm{bl}} \le V^{(\mathrm{cl})}_{\mathrm{bl}}$ but both are of the same order of magnitude.

VII. REGULARIZATION OF LOGARITHMIC SINGULARITY

Equation (42) shows the remarkable property of a logarithmic singularity at $\min(E_1,E_2)=0$ which corresponds to the value of the classical Coulomb blockade (26) and (27). Of course, the cotunneling current will be finite at the threshold where the current of first order starts. This singularity is an artifact of perturbation theory and means that Eq. (35) is wrong in the vicinity of $V^{(\mathrm{cl})}_{\mathrm{bl}}$. But beyond this blockade the tunneling current will be dominated in any case by the first-order current. Note that the second-order contribution is smaller than the first-order one by a factor of $R_Q/R_{1/2}$. Nevertheless, it is interesting to estimate a finite value of the cotunneling current for a consistency check. A complicated regularization procedure has been suggested,[30–32] but another way is to use a simple linewidth method of quantum theory.

This approach can be demonstrated by starting from the golden rule formulation[13,14] which is equivalent to our master-equation procedure. For the sake of simplicity, the quantum rate in the low-impedance case ($\phi=0$) in the zero-temperature limit is considered. It reads in the

Ohmic case (41) as

$$r^{(Q)}=\frac{\hbar}{2\pi e^4 R_1 R_2}\int_{-\infty}^{\infty} d\epsilon_1 d\epsilon_2 d\epsilon_3 d\epsilon_4 \Theta(-\epsilon_1)\Theta(\epsilon_2)\Theta(-\epsilon_3)\Theta(\epsilon_4)$$
$$\times\left|\frac{1}{E_1+\epsilon_2-\epsilon_1}+\frac{1}{E_2-\epsilon_3+\epsilon_4}\right|^2 \delta(eV+\epsilon_1-\epsilon_2+\epsilon_3-\epsilon_4)\ . \tag{45}$$

The generalization to the high-impedance case is straightforward and leads only to an energy shift. It is obvious that the singular denominators are the reason for the occurrence of the logarithm singularities. The term $\Delta E_j-\epsilon_k+\epsilon_p$, for instance, corresponds to the energy difference connected with tunneling from the left electrode to the island. Following, e.g., the arguments of Messiah,[33] the finite lifetime of the island states has to be taken into account by adding to the island energy an energy-dependent complex term $\Delta_j-i\Gamma_j$ which itself can be determined by perturbation theory. Consistency is already guaranteed if this term is taken in first-order perturbation theory at a representative energy scale. Here, only the imaginary part is considered which is essential for regularization. Because of a higher degree of generality, two different linewidth terms are introduced. Hence, with the substitutions ($j=1,2$)

$$E_j \rightarrow E_j - i\Gamma_j\ ,$$

the integrations yield the result

$$r^{(Q)}=\frac{\hbar}{2\pi e^4 R_1 R_2}\left\{\sum_{j=1}^{2}\frac{1}{2}\ln\left[\frac{(E_j+eV)^2+\Gamma_j^2/4}{E_i^2+\Gamma_j^2/4}\right]\mathrm{Im}F_j+\left[\arctan\frac{2(E_j+eV)}{\Gamma_j}-\arctan 2E_j\Gamma_j\right]\mathrm{Re}F_j\right\}\ , \tag{46}$$

where the complex functions F_j ($j,k=1,2,k\neq j$) are given by

$$F_j=\frac{(\Gamma_j+i2E_j)[\Gamma_j+i2(E_j+eV)][\Gamma_1+\Gamma_2-i2(E_1+E_2+eV)]}{2\Gamma_j[\Gamma_k-\Gamma_j-i2(E_1+E_2+eV)]}\ .$$

Note that $\mathrm{Im}F_1=\mathrm{Im}F_2$. Now the singularity at $V_{\mathrm{bl}}^{(\mathrm{cl})}$ has disappeared. The calculation of Γ_j in the relevant order of perturbation theory shows that they correspond up to a factor $\hbar$ to the golden rule expressions of the tunneling rates at the respective single junctions in first order. Therefore, the terms Γ_j can be approximated by the ordinary tunneling rates

$$\Gamma_j \approx \hbar\frac{1}{e^2 R_j}\frac{e^2}{2C_\Sigma}\ .$$

Taking into account that the logarithm is not very sensitive concerning the exact value of large arguments, a rough approximation gives the following value of the quantum rate at the classical Coulomb blockade:

$$r^{(Q)}\approx\frac{1}{(2\pi)^3}\frac{R_Q^2}{R_1R_2}\frac{eV_{\mathrm{bl}}}{\hbar}\ln\left[2\pi\frac{R_{1,2}}{R_Q}\right]\quad \text{for } V\rightarrow V_{\mathrm{bl}}^{(\mathrm{cl})}\ . \tag{47}$$

The notation $R_{1,2}$ means that one has to take R_1 if $C_1>C_2$ and vice versa. The dependence of $r^{(Q)}$ on the logarithm $\ln[2\pi(R_{1,2}/R_Q)]$ at the classical Coulomb blockade [cf. Eq. (47)] shows that only the logarithms in Eq. (46) have to be taken into account. Only there is the linewidth mechanism effective.

Concerning symmetric conditions $R_1=R_2=R_T, C_1=C_2$, the renormalized and unrenormalized mean currrents $\langle I\rangle=er^{(Q)}$ read in the low-impedance case [$z=eV/(E_C)$, $\alpha=R_Q/(2\pi R_T)$] as

$$\langle I\rangle_{\mathrm{reg}}=\frac{\hbar}{2\pi e^3 R_1 R_2}E_C\left\{(\tfrac{1}{4}+z^2+\alpha^2)\ln\left[\frac{(\frac{1}{2}+z)^2+\alpha^2}{(\frac{1}{2}-z)^2+\alpha^2}\right]-\arctan\left[\frac{2\alpha z}{\frac{1}{4}-z^2+\alpha^2}\right]\frac{\frac{1}{4}-z^2+\alpha^2}{\alpha}\right\}\ , \tag{48}$$

$$\langle I\rangle=\frac{\hbar}{\pi e^3 R_1 R_2}E_C\left\{(\tfrac{1}{4}+z^2)\ln\left[1+\frac{2z}{\frac{1}{2}-z}\right]-z\right\}\ . \tag{49}$$

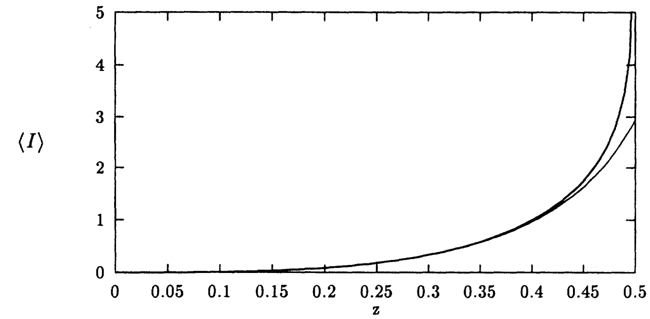

FIG. 2. Plot of the regularized and not regularized currents $\langle I \rangle = er^{(Q)}$ in the low-impedance case in units of $\hbar/(2\pi e^3 R_1 R_2)$ vs the dimensionless voltage $z = V/[e/(2C)]$ up to the classical Coulomb blockade. The parameter α is chosen to be $\alpha = 0.05$.

These currents have been plotted in Fig. 2. In the high-impedance case, the starting point of the curves is, instead of the origin, the "quantum" Coulomb blockade.

VIII. CONCLUSIONS

One should prove the Markov property (33) at the classical blockade voltage. Concerning the quantum rate in the unrenormalized version (42), the question whether the Markov property is guaranteed leads in the low-impedance limit to the condition

$$\ln \frac{V_{\rm bl}^{(\rm cl)}}{V_{\rm bl}^{(\rm cl)} - V} \ll (2\pi)^3 \frac{R_1 R_2}{R_Q^2} . \qquad (50)$$

It is obvious that this condition is violated in the immediate vicinity of the Coulomb blockade. In the renormalized version (46) this condition leads to the inequality

$$\frac{V_{\rm bl}^{(\rm cl)}}{(2\pi)^3} \frac{R_Q^2}{R_1 R_2} \ln \left[2\pi \frac{R_{1,2}}{R_Q} \right] \ll \frac{e}{2C_\Sigma} \qquad (51)$$

which is satisfied due to $R_Q \ll R_{1,2}$. This statement also remains true in the high-impedance case. So far, these results are correct for Ohmic current amplitudes. Therefore, the regularization guarantees not only finite current values but also the validity of the Markov property.

In general, the question whether the Markov property is guaranteed cannot be answered in a simple way. But the observation that only the logarithm terms are dominating at the critical point $V_{\rm bl}^{(\rm cl)}$ allows a rough estimation of the value of the quantum rate $r^{(Q)}$. Using Eq. (35) and taking into account that at the critical point the singular denominators are substituted by the finite linewidth the quantum rate at $V_{\rm bl}^{(\rm cl)}$ can be approximated by

$$r^{(Q)} \approx \frac{1}{(2\pi)^2 e} \frac{R_Q}{R_D|_{V_{\rm bl}^{(\rm cl)}}} {\rm Im} I_q \left[\frac{eV_{\rm bl}^{(\rm cl)}}{\hbar} \right] \times \ln \left| \frac{2\pi}{R_Q} \frac{V_{\rm bl}^{(\rm cl)}}{{\rm Im} I_q \left[\frac{eV_{\rm bl}^{(\rm cl)}}{\hbar} \right]} \right| \qquad (52)$$

where a differential tunnel resistance has been introduced by

$$R_D|_{V_{\rm bl}^{(\rm cl)}} = \frac{\hbar}{e} \frac{1}{{\rm Im} I_q' \left[\frac{eV_{\rm bl}^{(\rm cl)}}{\hbar} \right]} . \qquad (53)$$

For Ohmic amplitudes and $R_Q \ll R_{1,2}$, this expression reduces to Eq. (47). Equation (52) shows that the existence of the Markov property depends essentially on the behavior of ${\rm Im} I_q$. Due to formula (52), one can state that the consistency condition (33) can be fulfilled if the differential resistance (53) is not too small.

It has been shown that SET cotunneling in ultrasmall double junctions, which is a macroscopic quantum effect, can be formulated by means of a quantum master equation (39) including quantum rates proportional to $\hbar$. This approach allows the conclusion that the q-MQT current is connected with a quantum shot noise which could be treated analogously to the standard approach.[34,10]

The investigation of coherent q-MQT tunneling[13,14] using the master-equation approach is, in principle, possible by dropping Eq. (30) but is very inconvenient.

Here, only the special cases of low and high environment impedances have been investigated. The general case characterized by ${\rm Re} Z(\omega) \approx R_Q$ cannot be treated analytically and requests further considerations.

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