Non-adiabatic electron charge pumping in coupled semiconductor quantum dots

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The possibility of non-adiabatic electron pumping in the system of three coupled quantum dots attached to the leads is discussed. We have found out that periodical changing of energy level position in the middle quantum dot results in non zero mean tunneling current appeared due to non-adiabatic non-equilibrium processes. The same principle can be used for fabrication of a new class of semiconductor electronic devices based on non-stationary non-equilibrium currents. As an example we propose a nanometer quantum emitter with non-stationary inverse level occupation achieved by electron pumping.

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I. INTRODUCTION

Electron pumping in nanoscale structures attracts much attention nowadays [1],[2],[3],[4],[5],[6]. A great deal of the previous research works have been devoted to adiabatic electron pumping, the idea discussed by Thouless [7] rather long time ago. The first (to our knowledge) experiment on electron pumping in single electron device was described in [2]. Then experiments in this direction were continued in a three-junction geometry by Pothier et al. [8]. Two phase shifted rf signals were used to realize a single electron pump: a device with current $I = ef$ at zero bias voltage ($f$-frequency of the rf signal). Adiabatic charge pumping based on periodical variation of the potential barriers formed by the finger gates was also recently investigated in [1]. In these systems quantized current is connected with periodic adiabatic changing of the population of the quantum dot.

Proposed in a number of papers photo-assisted tunneling through coupled quantum dots [9],[10],[11],[12] is also an example of an electron pump. Pumping effect is achieved by applying an oscillating signal to the gate electrode or by irradiating the structure by monochromatic [12] and pulsed [13] microwaves.

It was understood that for practical realization of quantized electron pump the phenomenon of Coulomb blockade is very important [14]. General approach to the pumping through the interacting quantum dots in this regime is based on supposition that the charge relates to instantaneous chemical potential of a dot [15]. Using Coulomb blockade ideas another class of non-adiabatic quantized pumping of electrons in hybrid normal metal-superconductor structures was proposed in [10],[17].

These systems are more like a "turnstile" rather than a "pump" because quantized current directly proportional to gate frequency appears at finite (nonzero) value of applied bias. But at low temperatures these systems are very promising as current standards [18].

Adiabatic charge pumping through three tunnel-coupled quantum dots attached to electron leads in the regime of strong Coulomb blockade was investigated in [19]. Slow variations of coupling strength between the dots lead to adiabatic changes of energy levels in the system. Time dependent charge redistribution caused by energy levels changes results in non-stationary adiabatic tunneling current.

In the present paper we suggest a new type of electron pumping also in a system with three quantum dots, but based on non-equilibrium non-stationary tunneling currents. The proposed device requires only a single ac gate signal contrary to other semiconductor devices which require at least two rf signals with a definite phase shift. Mean current appears in our model due to non-adiabatic changing of electron level in a single quantum dot.

II. THREE DOT MODEL OF ELECTRON PUMP

We investigate non-stationary currents which flow in a three dot system shown in Fig.1. The left and right dots have energy levels $\varepsilon_2$ and $\varepsilon_3$ constant in time. And the level position of the middle dot $\varepsilon_1$ is modulated by external gate voltage.

Quantum dots with energy levels $\varepsilon_2$ and $\varepsilon_3$ are also coupled to continuous spectrum states - massive leads. Hamiltonian of the system under investigation has the
Action in turn by inverse operators $G^{-1}_{11}, G^{-1}_{22}, G^{-1}_{33}$ this integral equation can be also presented in the equivalent differential form (except for the point $t = t'$):

$$
[i \frac{\partial}{\partial t} - \epsilon_3 + i \gamma_3](i \frac{\partial}{\partial t} - \epsilon_2 + i \gamma_2)(i \frac{\partial}{\partial t} - \epsilon_1) - T_{12}^2 \cdot (i \frac{\partial}{\partial t} - \epsilon_3 + i \gamma_3) - T_{13}^2(i \frac{\partial}{\partial t} - \epsilon_2 + i \gamma_2)G_{11}^R(t, t') = 0
$$

(4)

Consequently, retarded Green’s function which determine spectrum re-normalization due to the tunneling between the quantum dots can be written in the following form:

$$
G_{11}^R(t, t') = i\Theta(t - t')(A_1 e^{-iE_1(t-t')} + A_2 e^{-iE_2(t-t')} + A_3 e^{-iE_3(t-t')})
$$

(5)

Where eigenfrequencies $E_{1,2,3}$ can be found from equation (see (4)):

$$
(E - \tilde{\epsilon}_1) \cdot (E - \tilde{\epsilon}_2 + i\gamma_2) \cdot (E - \tilde{\epsilon}_3 + i\gamma_3) - T_{12}^2 \cdot (E - \tilde{\epsilon}_3 + i\gamma_3) - T_{13}^2 \cdot (E - \tilde{\epsilon}_2 + i\gamma_2) = 0
$$

(6)

And coefficients $A_1$ can be evaluated using integral equation for $G_{11}^R$:

$$
A_1 = \frac{E_1(E_1 + E_3 - \tilde{\epsilon}_2 - \tilde{\epsilon}_3) - E_1E_3 + \tilde{\epsilon}_2\tilde{\epsilon}_3}{(E_1 - E_2)(E_1 - E_3)}\tag{7}
$$

$$
A_2 = \frac{E_2(E_2 + E_3 - \tilde{\epsilon}_2 - \tilde{\epsilon}_3) - E_2E_3 + \tilde{\epsilon}_2\tilde{\epsilon}_3}{(E_2 - E_3)(E_2 - E_1)}\tag{8}
$$

$$
A_3 = \frac{E_3(E_3 + E_2 - \tilde{\epsilon}_2 - \tilde{\epsilon}_3) - E_3E_2 + \tilde{\epsilon}_2\tilde{\epsilon}_3}{(E_3 - E_1)(E_3 - E_2)}
$$

$\tilde{\epsilon}_1 = \epsilon_1 - i\gamma_1$

Further on we assume for simplicity that $T_{12} = T_{13} = T$. If $\epsilon_1 = \tilde{\epsilon}_2$ and $\epsilon_1 - \tilde{\epsilon}_3 \gg T$, $\gamma$ coefficients $A_i$ has the form:

$$
A_1 = A_1^0(1 + \frac{E_3 - \tilde{\epsilon}_3}{E_1 - E_3}) = A_1^0(1 + \frac{T^2}{(\epsilon_1 - \epsilon_3)^2})\tag{9}
$$

$$
A_2 = A_2^0(1 + \frac{E_2 - \tilde{\epsilon}_2}{E_1 - E_3}) = A_2^0(1 + \frac{T^2}{(\epsilon_1 - \epsilon_3)^2})\tag{10}
$$

$$
A_3 = -\frac{T^2}{(\epsilon_1 - \epsilon_3)^2}\tag{11}
$$

where $A_1^0 = \frac{E_1 - \tilde{\epsilon}_1}{E_1 - E_3}$, $A_2^0 = -\frac{E_2 - \tilde{\epsilon}_2}{E_1 - E_3}$.

If we disconnect the third quantum dot then coefficients $A_1^0$ and $A_2^0$ give an exact solution for a system of two coupled quantum dots for all energy values $\epsilon_1$ and $\tilde{\epsilon}_2$. 

![Schematic diagram of the three coupled quantum dots with energy level position in the middle quantum dot depending on the time.](image-url)
Time evolution of electron density in the middle dot is determined by the Keldysh Green function $G^<$ \[20]\:

$$G^<_{11}(t, t') = \text{in}_F(t)$$

Equations for the Green functions $G^<_{ii}$ have the form:

$$(G^<_{11})^{-1} - T^{2}_{12}G^R_{12} - T^{3}_{13}G^R_{31} =$$

$$= T_{12}G^A_{22}G^A_{11} + T_{13}G^A_{33}G^A_{11}$$

$$(G^<_{22})^{-1} - T^{2}_{12}G^R_{12} - \sum_{k} T^{2}_{kk}G^R_{kk}G^<_{22} =$$

$$= T_{12}G^A_{22} + \sum_{k} T^{2}_{kk}G^A_{kk}G^<_{22}$$

$$(G^<_{33})^{-1} - T^{3}_{13}G^R_{13} - \sum_{p} T^{2}_{pp}G^R_{pp}G^<_{33} =$$

$$= T_{13}G^A_{33} + \sum_{p} T^{2}_{pp}G^A_{pp}G^<_{33}$$

If $G^<_{22}(0, 0) = \text{in}_F(\varepsilon_2) \simeq 0$, $G^<_{33}(0, 0) = \text{in}_F(\varepsilon_3) \simeq 1$ and $G^<_{11}(0, 0) = n_1(0)$ then Green function $G^<_{11}(t, t)$ is determined by the sum of homogeneous and inhomogeneous solutions. Inhomogeneous solution of the equation can be written in the following way:

$$G^<_{11}(t, t') = iT_{13} \int_{0}^{t} dt_1 \int_{0}^{t'} dt_2 G^R_{11}(t - t_1) \times$$

$$\times n_F(\varepsilon_3)e^{-i\varepsilon_3(t_1 - t_2)}G^A_{11}(t_2 - t')$$

Homogeneous solution of the differential equation has the form:

$$-iG^<_{11}(t, t') = f_1(t')e^{-iE_1t} + f_2(t')e^{-iE_2t} + f_3(t')e^{-iE_3t}$$

$$[G^<_{11}(t, t')]^* = -G^<_{11}(t', t)$$

Then coefficients $f_i(t')$ can be written as:

$$f_1(t') = Ae^{iE_1t'} + Be^{iE_2t'} + Xe^{iE_3t'}$$

$$f_2(t') = Ce^{iE_2t'} + B^*e^{iE_1t'} + De^{iE_3t'}$$

$$f_3(t') = X^*e^{iE_3t'} + D^*e^{iE_2t'} + Ze^{iE_1t'}$$

Since the solution has to satisfy homogeneous integro-differential equation, we are able to determine all coefficients. After some calculations we obtain that the following proportionality takes place:

$$f_2(t') = F_{21}f_1(t')$$

$$f_3(t') = F_{31}f_1(t')$$

with coefficients $F_{21}, F_{31}$:

$$F_{21} = - [(E_2 - \tilde{\varepsilon}_2)(E_2 - \tilde{\varepsilon}_3)((E_1 - \tilde{\varepsilon}_2)(E_3 - \tilde{\varepsilon}_2) +$$

$$+ (E_1 - \tilde{\varepsilon}_3)(E_2 - \tilde{\varepsilon}_3))] \cdot$$

$$\cdot [(E_1 - \tilde{\varepsilon}_2)(E_1 - \tilde{\varepsilon}_3)(E_3 - \tilde{\varepsilon}_3)(E_2 - \tilde{\varepsilon}_2) +$$

$$+ (\tilde{\varepsilon}_2 - E_3)(E_2 - \tilde{\varepsilon}_2)]^{-1}$$

$$F_{31} = (E_3 - \tilde{\varepsilon}_3)(E_3 - \varepsilon_2) + (E_1 - \varepsilon_2)F_{21}$$

Now we can find all coefficients in \[14\] :

$$A = \frac{n_1(0)}{1 + |F_{21} + F_{31}|^2 + 2Re F_{21} + 2Re F_{31}}$$

$$B = F_{21} \cdot A; \quad C = |F_{21}|^2 \cdot A$$

$$D = F_{31}F_{21} \cdot A; \quad Z = |F_{31}|^2 \cdot A; \quad X = F_{31}^* \cdot A$$

Finally, time dependence of the filling number in the middle quantum dot $n_1(t)$ can be written as:

$$n_1(t) = n_1^0 \cdot (Ae^{-i(E_1 - E_1^*)t} + Ce^{-i(E_2 - E_2^*)t} +$$

$$+ Ze^{-i(E_3 - E_3^*)t} + 2Re(Be^{-i(E_1 - E_3^*)t}) +$$

$$+ 2Re(Xe^{-i(E_2 - E_3^*)t} + 2Re(De^{-i(E_2 - E_3^*)t})$$

We see that there are six typical time scales in the considered system, which are described by the expression \[17\]. Three of them we can identify as three relaxation modes with rates $2ImE_1$, $2ImE_2$ and $2ImE_3$. Three other time scales are determined by the expressions $Re(E_1 - E_2)$, $Re(E_1 - E_3)$ and $Re(E_2 - E_3)$. These time scales are related with charge density oscillations between quantum dots, if the following ratio between $T_{13}$ and $\gamma$ takes place: $T_{13}/\gamma > 1/\sqrt{2}$.

If we neglect for a moment the tunneling from the middle dot to the right one, then for the system of two coupled quantum dots ($T_{13} = 0$) only three time scales appear and the equations \[15], \[16], \[17\] are transformed in the following way:

$$\frac{f_1(t')}{f_2(t')} = -\frac{\varepsilon_2 - E_1 - i\gamma}{\varepsilon_2 - E_2 - i\gamma}$$

Time dependence of the filling numbers in the first quantum dot $n_1(t)$ can be written as:

$$n_1(t) = n_1^0 \cdot \left[Ae^{-i(E_1 - E_1^*)t} + 2Re(Be^{-i(E_1 - E_3^*)t}) +$$

$$+ Ce^{-i(E_2 - E_2^*)t}\right]$$

where coefficients $A$, $B$ and $C$ are determined as:
and coefficients $X$, $Z$ and $D$ are equal to zero.

In this situation we can distinguish two relaxation rates $\gamma_{\text{res}}$ and $\gamma_{\text{nonres}}$ which characterises charge relaxation through an intermediate quantum dot in resonant and nonresonant cases:

$$\gamma_{\text{res}} = \frac{2T^2}{\gamma}, \quad \gamma_{\text{nonres}} = \gamma_{\text{res}} \frac{\gamma^2}{(\varepsilon_1 - \varepsilon_2)^2}$$

As we consider $\varepsilon_1 - \varepsilon_3 \gg T$, $\gamma$ then $\gamma_{\text{res}} \gg \gamma_{\text{nonres}}$ and small parameter $\gamma_{\text{nonres}}/\gamma_{\text{res}}$ exists in the theory. This allows us to write the following approximate relations for the system of three coupled quantum dots in the case $\varepsilon_1 \simeq \varepsilon_2$ valid in the first order of the small parameter $\varepsilon_1 - \varepsilon_3$:

$$E_1 - E_1^* = -i\gamma_{\text{res}} \left[1 + \frac{\gamma^2}{(\varepsilon_1 - \varepsilon_3)^2}\right]$$

$$E_2 - E_2^* = -2i\gamma \left[1 - \frac{T^2}{\gamma^2} + \frac{T^2}{\gamma^2 (\varepsilon_1 - \varepsilon_3)^2}\right]$$

$$E_3 - E_3^* = 2i\gamma \left[1 + \frac{T^2}{(\varepsilon_1 - \varepsilon_3)^2}\right]$$

$$E_2 - E_3^* = \varepsilon_2 - \varepsilon_3 - 2i\gamma \left[1 + \frac{T^2}{2\gamma^2} - \frac{T^2}{(\varepsilon_1 - \varepsilon_3)^2}\right]$$

$$E_1 - E_3^* = \varepsilon_1 - \varepsilon_3 - i\gamma_{\text{res}} \left(\frac{\gamma^2}{(\varepsilon_1 - \varepsilon_3)^2} - i\gamma \left[1 - \frac{T^2}{(\varepsilon_1 - \varepsilon_3)^2}\right]\right)$$

$$E_1 - E_2^* = i\gamma + \frac{2T^2}{\varepsilon_1 - \varepsilon_3} - i\gamma \left[\frac{\gamma^2}{(\varepsilon_1 - \varepsilon_3)^2} + \frac{T^2}{(\varepsilon_1 - \varepsilon_3)^2}\right]$$

When $\varepsilon_1 = \varepsilon_2$ and $\varepsilon_1 - \varepsilon_3 \gg T$, $\gamma$ the exact equations can be transformed in the following way:

$$F_{21} \simeq - \frac{T^2}{\gamma^2} \left[1 + \frac{\gamma}{\varepsilon_1 - \varepsilon_3} + \frac{T^2}{\gamma^2 (\varepsilon_1 - \varepsilon_3)}\right]$$

$$F_{31} \simeq \frac{T^2}{\gamma^2} \left[\frac{T^2}{(\varepsilon_1 - \varepsilon_3)^2} + i\frac{T^2}{(\varepsilon_1 - \varepsilon_3)^2}\right]$$

So, coefficients $D$, $Z$ and $X$ which are resposible for the "reverse" current to the right lead are much smaller than $A$, $B$ and $C$, which correspond to "direct" current to the left, due to the appearance of the parameter $\frac{T^2}{(\varepsilon_1 - \varepsilon_3)^2}$. For simplicity we omit the terms with coefficients $D$, $Z$ and $X$ in equation (17) which determine time evolution of localized charge in the middle quantum dot. For any concrete system the accuracy of this approximation can be easily estimated from the exact equations.

Pumping of electrons takes place if energy level $\varepsilon_1(t)$ is a function of time and changes periodically (Fig. 1). We shall describe the most favorable case with $T \ll \gamma$.

For current calculation we consider the situation of periodically switching the position of level $\varepsilon_1$ by external gate:

$\varepsilon_1(t) = \varepsilon_3$ in the interval $0 < t < t_0$ it means resonant tunneling between energy levels $\varepsilon_1$ and $\varepsilon_3$

$\varepsilon_1(t) = \varepsilon_2$ in the interval $t_0 < t < 2t_0$ - resonance between energy levels $\varepsilon_1$ and $\varepsilon_2$

Time evolution of local electron density $n_1(t)$ in the central quantum dot can be determined from equation (17) (Fig. 2).

When $0 < t < t_0$

$$n_1(t) = n_1^0 \left[\left(1 + \frac{\gamma_{\text{res}}}{\gamma}\right) e^{-\gamma_{\text{res}}t} - \frac{\gamma_{\text{res}} e^{-\gamma t}}{\gamma}\right]$$

and when $t_0 < t < 2t_0$

$$n_1(t) = n_1^0 \left[\left(1 + \frac{\gamma_{\text{res}}}{\gamma}\right) e^{-\gamma_{\text{res}}(t - t_0)} - \frac{\gamma_{\text{res}} e^{-\gamma(t - t_0)}}{\gamma}\right] + \left[1 - \left(1 + \frac{\gamma_{\text{res}}}{\gamma}\right) e^{-\gamma_{\text{res}}(t - t_0)} + \frac{\gamma_{\text{res}} e^{-\gamma(t - t_0)}}{\gamma}\right]$$

Taking into account periodicity condition $n_1(2t_0) = n_1^0$, one can find $n_1^0$:

$$n_1^0 = \frac{1}{1 + \left(1 + \frac{\gamma_{\text{res}}}{\gamma}\right) e^{-\gamma_{\text{res}}t_0} - \frac{\gamma_{\text{res}} e^{-\gamma t_0}}{\gamma}}$$

Results for $n_1(t)$ are shown in Fig.2. Situation when frequency $\Omega \equiv 1/2t_0$ of the level $\varepsilon_1(t)$ switching is higher than tunneling rates $\gamma_{\text{res}}, \gamma$ is depicted by grey line. Black line corresponds to the case when frequency $\Omega$ is lower than $\gamma_{\text{res}}, \gamma$. It is clear that with the increasing of frequency the value of $n_1(t)$ always tends to the value 1/2 and is almost independent on time at high gate frequencies.

For low frequencies if $\gamma_{\text{res}}t_0 \gg 1$ the value $n_1^0$ is almost equal to 1. The energy level $\varepsilon_1$ is filled up almost completely during the pumping cycle (for considered situation when energy level $\varepsilon_2$ is well above and energy level $\varepsilon_3$ is well below the Fermi energy). Non-stationary tunneling current through the system appears for zero applied bias:

$$e \frac{\partial}{\partial t} n_1(t) = I(t)$$
FIG. 2: Time evolution of local electron density $n_1(t)$ in the central quantum dot in the system of three coupled quantum dots if energy level position in the middle quantum dot depends on time. Black line corresponds to the case when frequency of energy level $\varepsilon_1(t)$ switching is lower than tunneling rates and grey line corresponds to the case when frequency is higher than tunneling rates.

One can find for $0 < t < t_0$

$$I(t) = e n_1^0 \gamma_{res} \left[ (1 + \gamma_{res} \gamma) e^{-\gamma_{res} t} e^{-\gamma t} \right]$$ \hspace{1cm} (28)

Mean tunneling current value can be found as:

$$< I > = e \frac{1}{2t_0} \int_0^{t_0} I(t) dt = e \frac{1}{2t_0} n_1^0 \cdot \left[ 1 - (1 + \gamma_{res} \gamma) e^{-\gamma_{res} t_0} + \gamma_{res} \gamma e^{-\gamma t_0} \right]$$ \hspace{1cm} (29)

If $\Omega \equiv 1/2t_0 \ll \gamma_{res}$ tunneling current mean value can be written as $< I > = e \Omega$ since $n_1^0 = 1$ for such frequencies. This is the regime, when the device operates like a current standard: the current is directly proportional to the gate frequency. This regime has exponential accuracy which is governed by the second and third terms in the square brackets in expression (29). Note, that even for very unsuitable case if $|\varepsilon_2 - \varepsilon_3| \gg \gamma \gg T$ the pumping effect still remains, and the current is proportional to the frequency, though its value is suppressed compared to the ideal relation $< I > = e \Omega$.

For high frequencies of the gate voltage in the region $\gamma \gg \Omega \equiv 1/2t_0 \gg \gamma_{res}$ tunneling current average value is almost independent on the frequency and equal to:

$$< I > = e \gamma_{res} / 4 \left( \gamma_{res} = 2 T^2 \right)$$

With further frequency increase ($\Omega \gg \gamma \gg \gamma_{res}$) mean current value decreases to $< I > = e \gamma_{res}^2 / 4 \gamma$.

The non-stationary mean tunneling current value has non-monotonic dependence on the gate frequency with maximum at $\Omega \approx \gamma_{res}$ (Fig 3). This effect can be used for frequency stabilization in nanoelectronics.

The parameters of devices based on quantum dots depend on the size of quantum dots, tunneling transfer rates, energy levels positions and distances between them. Estimation of tunneling parameters for achievable setup gives us characteristic frequencies and currents for such devices:

FIG. 3: Frequency dependence of the mean tunneling current for the system of three coupled quantum dots (gate frequency $\Omega = 1/2t_0$). Different frequency scales are presented.

$$T \approx 1 \text{ meV}, \quad \gamma \approx 1 \div 10 \text{ meV} \Rightarrow \gamma_{res} = 2 T^2 / \gamma \approx 0.1 \div 1 \text{ meV} \approx 10^{10} \div 10^{11} \text{ e/sec}$$

where parameters $T$ and $\gamma$ are determined by the widths and heights of the barriers. For such values of the tunneling rates we need to have quantum dots of tens of nanometers size, for which quantum size quantization energies are not less than the tunneling width of levels. Such devices could operate at gigahertz and subgigahertz frequencies at nano and subnanoampere currents ($1 nA \approx 6 \cdot 10^9 e/\text{sec}$).

The difference between tunneling rates in resonant and non resonant cases can be used also for creation of inverse occupation in quantum emitter based on the system of coupled quantum dots. An example of such device is shown in Fig 4. In the system of two coupled quantum dots we switch the level $\varepsilon$ by applying gate voltage to the second quantum dot between two levels of the first quantum dot directly coupled to the lead. If the third non-resonant level exists in the same quantum dot between the lowest and the highest levels, then charge pumping from the initially filled state $\varepsilon_3$ to the highest level $\varepsilon_1$...
creates inverse occupation of the states $\varepsilon_2$ and $\varepsilon_1$. So some nanometer quantum generators can be fabricated on this principle.

### III. CONCLUSION

We investigated electron pumping ability of a system of three tunnel coupled quantum dots attached to the leads. Periodical changing of energy level position in the middle quantum dot by gate voltage leads to nonzero tunneling current even if applied to the structure bias is equal to zero.

Our calculations of the mean current are based on accurate analysis of relaxation processes in quantum dots in non-adiabatic regime. Exact equations allows to investigate various regimes of the device and estimate the mean current value and accuracy of it’s operation as a current standard. For very small dots with pronounced size effect a possibility of room temperature electron pumping is opened. We should like to stress that the ideas discussed in the paper can be used for fabrication of a new type of electronic devices based on non-equilibrium non-stationary tunneling currents. As an example we proposed in the paper nanometer quantum emitter based on two coupled quantum dots.

### IV. ACKNOWLEDGEMENTS

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