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Correlation induced switching of local spatial charge distribution in two-level system

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It was found that tunneling current through a nanometer scale structure with strongly coupled localized states causes spatial redistribution of localized charges induced by Coulomb correlations. We present here theoretical investigation of this effect by means of Heisenberg equations for localized states electron filling numbers. This method allows to take into account pair correlations of local electron density exactly. It is shown that inverse occupation of the two-level system caused by Coulomb correlations appears in particular range of applied bias. Described effects can give a possibility of charge manipulation in the proposed system. We also expect that described results can be observed in tunneling structures with impurities or with small quantum dots.

1. Introduction. Investigation of tunneling properties of interacting impurity complexes in the presence of Coulomb correlations is one of the most important problems in the physics of nanostructures. Tunneling current changes localized states electron filling numbers as a result-the spectrum and electron density of states are also modified due to Coulomb interaction of localized electrons. Moreover the charge distribution in the vicinity of such complexes can be tuned by changing the parameters of the tunneling contact. Self-consistent approach based on Keldysh diagram technique have been successfully used to analyze non-equilibrium effects and tunneling current spectra in the system of two weakly coupled impurities (when coupling between impurities is smaller than tunneling rates between energy levels and tunneling contact leads) in the presence of Coulomb interaction [1]. In the mean-field approximation for mixed valence regime the dependence of electron filling numbers on applied bias voltage and the behaviour of tunneling current spectra have been analyzed in [2].

Electron transport even through a single impurity in the Coulomb blockade and the Kondo regime [3] have been studied experimentally and is up till now under theoretical investigation [4–10]. As tunneling coupling is not negligible the impurity charge is not the discrete value and one has to deal with impurity electron filling numbers (which now are continuous variables) determined from kinetic equations.

Analyzing non-equilibrium tunneling processes through coupled impurities one can reveal switching on and off of magnetic regime (electron filling numbers in the localized states for opposite spins are not equal) on each impurity atom at particular range of applied bias voltage [2].

In the present work we consider the opposite case when coupling between localized electron states strongly exceeds tunneling transfer rates. This situation can be experimentally realized when several impurity atoms or surface defects are situated at the neighboring lattice sites, so coupling between their electronic states can strongly exceeds the interaction of these localized states with continuous spectrum Fig. 1 [11]. Results obtained

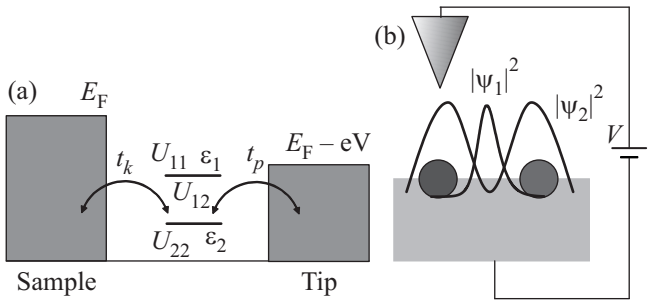

Fig. 1. (a) – Energy diagram of two-level system. (b) – Schematic spatial diagram of experimental realization. Coulomb energy U_{12} correspond to the interaction between electrons on different energy levels

in [11] demonstrate switching on and off of two closely situated impurity atoms in tunneling conductivity due to the Coulomb interaction. Another possible realization is two interacting quantum dots on the sample surface weakly connected with the bulk states. Specific behav-

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iour of such systems results in irregular peaks splitting in tunneling conductivity with the increasing of intradot interaction. Irregular peaks formation takes place due to the interplay between strong Coulomb interaction and resonant tunneling through localized states [12]. At last it could be a single quantum dot with a few lowest electron levels lying in relevant energy range. Such systems can be described by the model including several electron levels with Coulomb interaction between localized electrons [13–15]. The authors of [15] showed that in a two level system Coulomb interaction can cause negative differential conductivity. But they regarded electrons as a spinless particles and assume that Coulomb potential is infinitely large. So they do not take into account any nontrivial pair correlations in the system. If the distance between impurities is smaller than localization radius, strong enough correlation effects arise which modify the spectrum of the whole complex. Electronic structure of such complexes can be tuned both by external electric field which changes the values of single particle levels and by electron correlations of localized electronic states. One can expect that tunneling current induces non-equilibrium spatial redistribution of localized charges and gives possibility of local charge density manipulation strongly influenced by Coulomb correlations. In some sense these effects are similar to the "cotunneling" observed in [16], [17]. Moreover Coulomb interaction of localized electrons can be responsible for inverse occupation of localized electron states. These effects can be clearly seen when single electron levels have different spatial symmetry.

To understand such correlation induced "charge" switching it's sufficient to analyze Heisenberg equations for localized states total electron filling numbers taking into account pair correlations of local electron density [13]. If one is interested in kinetic properties and changes of local charge density for the applied bias range higher than the value of energy levels tunneling broadening modification of initial density of states due to the Kondo effect can be neglected. In this case for the finite number of localized electron levels one can obtain closed system of equations for electron filling numbers and their higher order correlations.

2. Suggested model. We shall analyze tunneling through the two-level system with Coulomb interaction Fig. 1. The model system can be described by the Hamiltonian $\hat{H}$:

$$\hat{H} = \sum_{i\sigma} \varepsilon_i n_{i\sigma} + \sum_{k\sigma} \varepsilon_k c^+_{k\sigma} c_{k\sigma} + \sum_{p\sigma} \varepsilon_p c^+_{p\sigma} c_{p\sigma} + \\ + \sum_{ij\sigma\sigma'} U_{ij}^{\sigma\sigma'} n_{i\sigma} n_{j\sigma'} + \sum_{ki\sigma} t_k c^+_{k\sigma} c_{i\sigma} + \\ + \sum_{pi\sigma} t_p c^+_{p\sigma} c_{i\sigma} + \text{h.c.} \quad (1)$$

Indices k and p label continuous spectrum states in the left (sample) and right (tip) leads of tunneling contact respectively; $t_{k(p)}$ – tunneling transfer amplitudes between continuous spectrum states and two-level system with elctron levels ε_i and we assume here that $t_{1k(p)} = t_{2k(p)}$. Operators $c^+_{k(p)}/c_{k(p)}$ correspond to electrons creation/annihilation in the continuous spectrum states $k(p)$; $n_{i\sigma} = c^+_{i\sigma} c_{i\sigma}$ – two-level system electron filling numbers, where operator $c_{i\sigma}$ destroys electron with spin σ on the energy level ε_i; $U_{ij}^{\sigma\sigma'}$ is the on-site Coulomb repulsion of localized electrons.

Tunneling current through the two-level system can be written as

$$I = I_{k\sigma} = \sum_{k\sigma} \dot{n}_{k\sigma} = \sum_{ki\sigma} t_k (\langle c^+_{k\sigma} c_{i\sigma} \rangle - \langle c^+_{i\sigma} c_{k\sigma} \rangle). \quad (2)$$

Let us consider $\hbar = 1$ further on. We use Heisenberg motion equation to calculate $\langle c^+_{k\sigma} c_{i\sigma} \rangle$:

$$i\frac{\partial c^+_{k\sigma} c_{i\sigma}}{\partial t} = (\varepsilon_i - \varepsilon_k) c^+_{k\sigma} c_{i\sigma} + U_{ii} n_{i-\sigma} c^+_{k\sigma} c_{i\sigma} + \\ + U_{ij}(n_{j\sigma} + n_{j-\sigma}) c^+_{k\sigma} c_{i\sigma} - t_k (n_{i\sigma} - \widehat{f}_k) + \\ + \sum_{k' \neq k} t_{k'} c^+_{k\sigma} c_{k'\sigma} + \sum_{i \neq j} t_k c^+_{j\sigma} c_{i\sigma} = 0, \quad (3)$$

where

$$\widehat{f}_k = c^+_{k\sigma} c_{k\sigma}. \quad (4)$$

Neglecting changes of electron spectrum and local density of states in the tunneling contact leads due to the tunneling current we uncouple conduction and two-level system electron filling numbers. This approximation means that we neglect the level width, compared to bias scale, Coulomb repulsion and other energy parameters. In this approximation we also lost any correlations between localized and conduction band electrons (like Kondo effect), but these effects are out of the scope of the paper. On the other hand this approximation allows us to take into account all correlations between localized electrons exactly. Then expression for the tunneling current can be derived with the help of Eq. (3) by the following way. We multiply Eq. (3) by various combinations of the operators $n_{i\sigma}, n_{j\sigma}$ and use relations $n^2_{i\sigma} = n_{i\sigma}$, $n_{i\sigma}(1 - n_{i\sigma}) = 0$. That gives us closed system of equations for correlators $\langle c^+_{k\sigma} c_{i\sigma} \rangle$, $\langle n_{i-\sigma} c^+_{k\sigma} c_{i\sigma} \rangle$ and so on. After summation over k one can get an equation which describe tunneling current $I_{k1\sigma}$ from the left lead to the first level in the two-level system:

$$
\begin{aligned}
I_{k1\sigma} = \Gamma_k \Big\{ & \langle n_{1\sigma}\rangle + \sum_{j\neq i}\langle c^+_{j\sigma}c_{i\sigma}\rangle - \\
& - \langle (1-n_{1-\sigma})(1-n_{2-\sigma})(1-n_{2\sigma})\rangle f_k(\varepsilon_1) - \\
& - \langle n_{1-\sigma}(1-n_{2-\sigma})(1-n_{2\sigma})\rangle f_k(\varepsilon_1+U_{11}) - \\
& - \sum_{\sigma'}\langle n_{2\sigma'}(1-n_{2-\sigma'})(1-n_{1-\sigma})\rangle f_k(\varepsilon_1+U_{12}) - \\
& - \sum_{\sigma'}\langle n_{1-\sigma}n_{2\sigma'}(1-n_{2-\sigma'})\rangle f_k(\varepsilon_1+U_{11}+U_{12}) - \\
& - \langle n_{2\sigma}n_{2-\sigma}(1-n_{1-\sigma})\rangle f_k(\varepsilon_1+2U_{12}) - \\
& - \langle n_{1-\sigma}n_{2-\sigma}n_{2\sigma}\rangle f_k(\varepsilon_1+U_{11}+2U_{12}) \Big\} + \\
& + \sum_{k'\neq k}\langle t_k t_{k'} c^+_{k\sigma}c_{k'\sigma}\rangle \times \\
& \times \left\{ \left\langle \frac{(1-n_{1-\sigma})(1-n_{2-\sigma})(1-n_{2\sigma})}{\varepsilon_1-\varepsilon_k}\right\rangle + \right. \\
& + \left\langle \frac{n_{1-\sigma}(1-n_{2-\sigma})(1-n_{2\sigma})}{\varepsilon_1+U_{11}-\varepsilon_k}\right\rangle + \\
& + \left\langle \frac{\sum_{\sigma'} n_{2\sigma'}(1-n_{1-\sigma})(1-n_{2-\sigma'})}{\varepsilon_1+U_{12}-\varepsilon_k}\right\rangle + \\
& + \left\langle \frac{\sum_{\sigma'} n_{1-\sigma}n_{2\sigma'}(1-n_{2-\sigma'})}{\varepsilon_1+U_{11}+U_{12}-\varepsilon_k}\right\rangle + \\
& + \left\langle \frac{n_{2-\sigma}n_{2\sigma}(1-n_{1-\sigma})}{\varepsilon_1+2U_{12}-\varepsilon_k}\right\rangle + \\
& \left. + \left\langle \frac{n_{1-\sigma}n_{2-\sigma}n_{2\sigma}}{\varepsilon_1+U_{11}+2U_{12}-\varepsilon_k}\right\rangle \right\}. \qquad (5)
\end{aligned}
$$

The total current is a sum of currents through each level:

$$I_{k\sigma} = I_{k1\sigma} + I_{k2\sigma}. \qquad (6)$$

Where expression for the tunneling current $I_{k2\sigma}$ can be obtained by changing indexes $1 \leftrightarrow 2$ in equation for the tunneling current $I_{k1\sigma}$.

In what follows we shall neglect terms $t_k c^+_{i\sigma}c_{j\sigma}$ and $t_k c^+_{k\sigma}c_{k'\sigma}$ in expression (5) as they correspond to the next order perturbation theory in the parameter Γ_i/ε_i. Relaxation rates $\Gamma_{k(p)} = \pi\, t^2_{k(p)}\,\nu_0$ are determined by electron tunneling transitions from two-level system to the leads k (sample) and p (tip) continuum states; ν_0 – continuous spectrum density of states.

Now the problem reduced to calculation of $\langle n_{i\sigma}\rangle$ and various $n_{i\sigma}$ correlators which are not trivial due to Coulomb interaction. Equations for filling numbers $n_{1\sigma}$ and $n_{2\sigma}$ can be found from the conditions:

$$
\begin{aligned}
\frac{\partial n_{1\sigma}}{\partial t} &= I_{k1\sigma} + I_{p1\sigma} = 0, \\
\frac{\partial n_{2\sigma}}{\partial t} &= I_{k2\sigma} + I_{p2\sigma} = 0, \qquad (7)
\end{aligned}
$$

where tunneling current $I_{p\sigma}$ can be easily determined from $I_{k\sigma}$ by changing indexes $k \leftrightarrow p$.

In this paper we shall analyze the situation when Coulomb energy values are large and condition $U_{ij} \gg \varepsilon_{i/j}$ can be taken into account. It means that if one has to calculate tunneling current through such system it is necessary to find all pair filling numbers correlators in the energy range $\varepsilon_i + U_{ij}$. So we retain the terms containing $f_{k(p)}(\varepsilon_i + U_{ij})$ and neglect all higher orders correlators and pair correlators which contain $f_{k(p)}(\varepsilon_i + U_{ij} + U_{kl})$.

Pair filling numbers correlators can be found in the following way:

$$\left\langle \frac{\partial n_{i\sigma}n_{j\sigma'}}{\partial t}\right\rangle = \left\langle \frac{\partial n_{i\sigma}}{\partial t}n_{j\sigma'}\right\rangle + \left\langle \frac{\partial n_{j\sigma'}}{\partial t}n_{i\sigma}\right\rangle. \qquad (8)$$

For short we consider here the paramagnetic situation: $\langle n_{i\sigma}\rangle = \langle n_{i-\sigma}\rangle$ and also $\langle n_{i\sigma}n_{j\sigma}\rangle = \langle n_{i\sigma}n_{j-\sigma}\rangle$. So a system of equations for pair correlators $K_{11} \equiv \langle n_{1\sigma}n_{1-\sigma}\rangle$, $K_{22} \equiv \langle n_{2\sigma}n_{2-\sigma}\rangle$ and $K_{12} \equiv \langle n_{1\sigma}n_{2\sigma}\rangle$ for large Coulomb energies $U_{ij} \gg \varepsilon_{i/j}$ is closed if we neglect triple correlators:

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \times \begin{pmatrix} K_{11} \\ K_{12} \\ K_{22} \end{pmatrix} = F, \qquad (9)$$

where

$$
\begin{aligned}
a_{11} &= a_{23} = 1, \\
a_{13} &= a_{21} = 0, \\
a_{12} &= 2n^{\mathrm{T}}(\varepsilon_1 + U_{11}), \\
a_{22} &= 2n^{\mathrm{T}}(\varepsilon_2 + U_{22}), \qquad (10)
\end{aligned}
$$

$$
\begin{aligned}
a_{31} &= \frac{1}{2}n^{\mathrm{T}}(\varepsilon_2 + U_{12}), \\
a_{32} &= 1 + \frac{1}{2}n^{\mathrm{T}}(\varepsilon_1 + U_{12}) + \frac{1}{2}n^{\mathrm{T}}(\varepsilon_2 + U_{12}), \\
a_{33} &= \frac{1}{2}n^{\mathrm{T}}(\varepsilon_1 + U_{12}) \qquad (11)
\end{aligned}
$$

and

$$F = \begin{pmatrix} n^{\mathrm{T}}(\varepsilon_1 + U_{11})n_{1\sigma} \\ n^{\mathrm{T}}(\varepsilon_2 + U_{22})n_{2\sigma} \\ \frac{1}{2}n^{\mathrm{T}}(\varepsilon_1 + U_{12})n_{2\sigma} + \frac{1}{2}n^{\mathrm{T}}(\varepsilon_2 + U_{12})n_{1\sigma} \end{pmatrix}. \qquad (12)$$

Where we introduced tunneling filling numbers $n^{\mathrm{T}}(\varepsilon_i)$, $n^{\mathrm{T}}(\varepsilon_i + U_{ij})$ and $\tilde{n}^{\mathrm{T}}_{ij}$ which have the form:

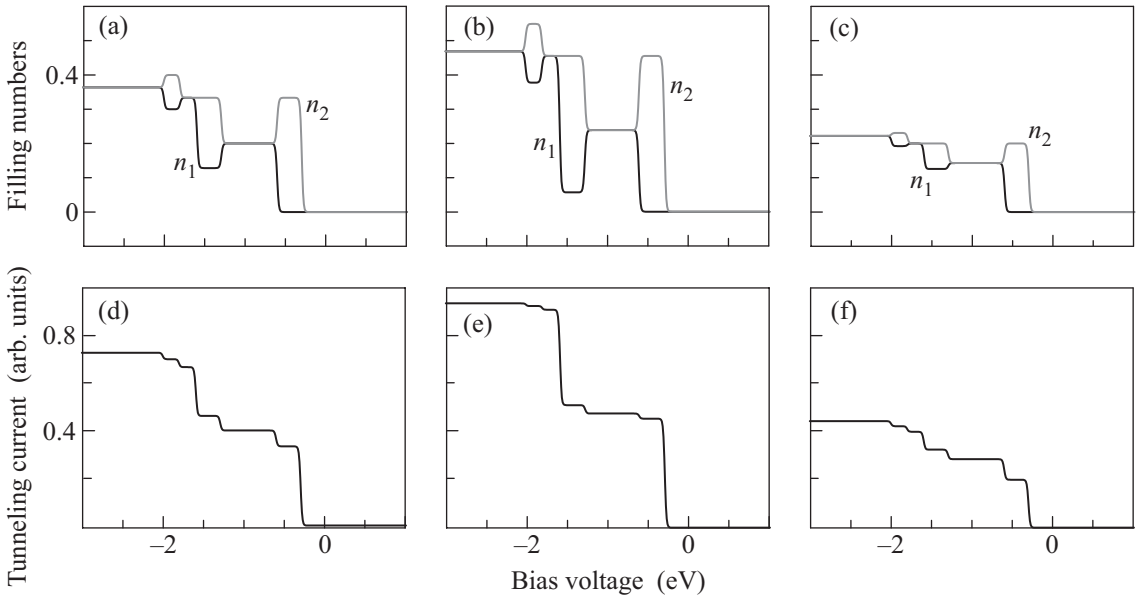

Fig. 2. Two-level system filling numbers (a)–(c) and tunneling current (d)–(f) as a function of applied bias voltage in the case when both energy levels are situated above the sample Fermi level. Parameters $\epsilon_1 = 0.6$, $\epsilon_2 = 0.3$, $U_{12} = 1.0$, $U_{11} = 1.4$, $U_{22} = 1.5$ are the same for all the figures. (a), (d) – $\Gamma_k = 0.01$, $\Gamma_p = 0.01$. (b), (e) – $\Gamma_k = 0.05$, $\Gamma_p = 0.01$. (c), (f) – $\Gamma_k = 0.01$, $\Gamma_p = 0.03$

$$n^{\mathrm{T}}(\varepsilon_i) = \frac{\Gamma_k f_k(\varepsilon_i) + \Gamma_p f_p(\varepsilon_i)}{\Gamma_k + \Gamma_p},$$
$$n^{\mathrm{T}}(\varepsilon_i + U_{ij}) = \frac{\Gamma_k f_k(\varepsilon_i + U_{ij}) + \Gamma_p f_p(\varepsilon_i + U_{ij})}{\Gamma_k + \Gamma_p},$$
$$\tilde{n}^{\mathrm{T}}_{ij} = \frac{\Gamma_k \tilde{f}_{kij} + \Gamma_p \tilde{f}_{pij}}{\Gamma_k + \Gamma_p}, \qquad (13)$$

where

$$\tilde{f}_{kij} = f_k(\varepsilon_i) - f_k(\varepsilon_i + U_{ij}). \qquad (14)$$

The final expression for the tunneling current in terms of the correlators K, determined from Eq. (9), has the form:

$$I_{k1\sigma} = \Gamma_k\{\langle n_{1\sigma}\rangle - (1 - \langle n_{1\sigma}\rangle - 2\langle n_{2\sigma}\rangle + K_{22} + 2K_{12})\times$$
$$\times f_k(\varepsilon_1) - (\langle n_{1\sigma}\rangle - 2K_{12}) f_k(\varepsilon_1 + U_{11}) -$$
$$- 2(\langle n_{2\sigma}\rangle - K_{12} - K_{22}) f_k(\varepsilon_1 + U_{12}). \qquad (15)$$

Eqs. (7) and (9) allows to determine the filling numbers fron the following system, since the correlators K have been calculated:

$$n_{1\sigma}(1 + \tilde{n}^{\mathrm{T}}_{11}) + n_{2\sigma}\cdot 2\tilde{n}^{\mathrm{T}}_{12} - K_{22}[n^T(\varepsilon_1) -$$
$$- 2n^{\mathrm{T}}(\varepsilon_1 + U_{12})] + 2K_{12}[-n^{\mathrm{T}}(\varepsilon_1) +$$
$$+ n^{\mathrm{T}}(\varepsilon_1 + U_{11}) + n^{\mathrm{T}}(\varepsilon_1 + U_{12})] = n^{\mathrm{T}}(\varepsilon_1),$$
$$n_{2\sigma}(1 + \tilde{n}^{\mathrm{T}}_{22}) + n_{1\sigma}\cdot 2\tilde{n}^{\mathrm{T}}_{21} - K_{11}[n^{\mathrm{T}}(\varepsilon_2) -$$
$$- 2n^{\mathrm{T}}(\varepsilon_2 + U_{12})] + 2K_{12}[-n^{\mathrm{T}}(\varepsilon_2) +$$
$$+ n^{\mathrm{T}}(\varepsilon_2 + U_{22}) + n_{\mathrm{T}}(\varepsilon_2 + U_{12})] = n^{\mathrm{T}}(\varepsilon_2). \qquad (16)$$

Let us give an answer for two particular cases. The first one is when all Coulomb energies are extremely large $U_{ij} \to \infty$. In this situation expressions for filling numbers will have the following form:

$$n_{1\sigma} = \frac{n^{\mathrm{T}}(\varepsilon_1)[1 - n^{\mathrm{T}}(\varepsilon_2)]}{[1 + n^{\mathrm{T}}(\varepsilon_1)][1 + n^{\mathrm{T}}(\varepsilon_2)] - 4n^{\mathrm{T}}(\varepsilon_1)n^{\mathrm{T}}(\varepsilon_2)},$$
$$n_{2\sigma} = \frac{n^{\mathrm{T}}(\varepsilon_2)[1 - n^{\mathrm{T}}(\varepsilon_1)]}{[1 + n^{\mathrm{T}}(\varepsilon_1)][1 + n^{\mathrm{T}}(\varepsilon_2)] - 4n^{\mathrm{T}}(\varepsilon_1)n^{\mathrm{T}}(\varepsilon_2)}. \qquad (17)$$

And the second one is when energy levels are very close to each other (for example two degenerate in orbital quantum number states) $\varepsilon_1 = \varepsilon_2 = \varepsilon$ and almost the same Coulomb repulsion for any states $U_{ij} = U$. In this case the filling numbers have the form:

$$n_\sigma = \frac{n^{\mathrm{T}}(\varepsilon)}{1 + 3n^{\mathrm{T}}(\varepsilon)}. \qquad (18)$$

3. Main results and discussion. The behaviour of non-equilibrium electron filling numbers with changing of applied bias and tunneling conductivity characteristics obtained from equations (9)–(15) are depicted in Figs. 2–4.

We consider different experimental realizations: both energy levels are situated above the sample Fermi level (Fig. 2); both levels below sample Fermi level (Fig. 3) and one of the energy levels is located above

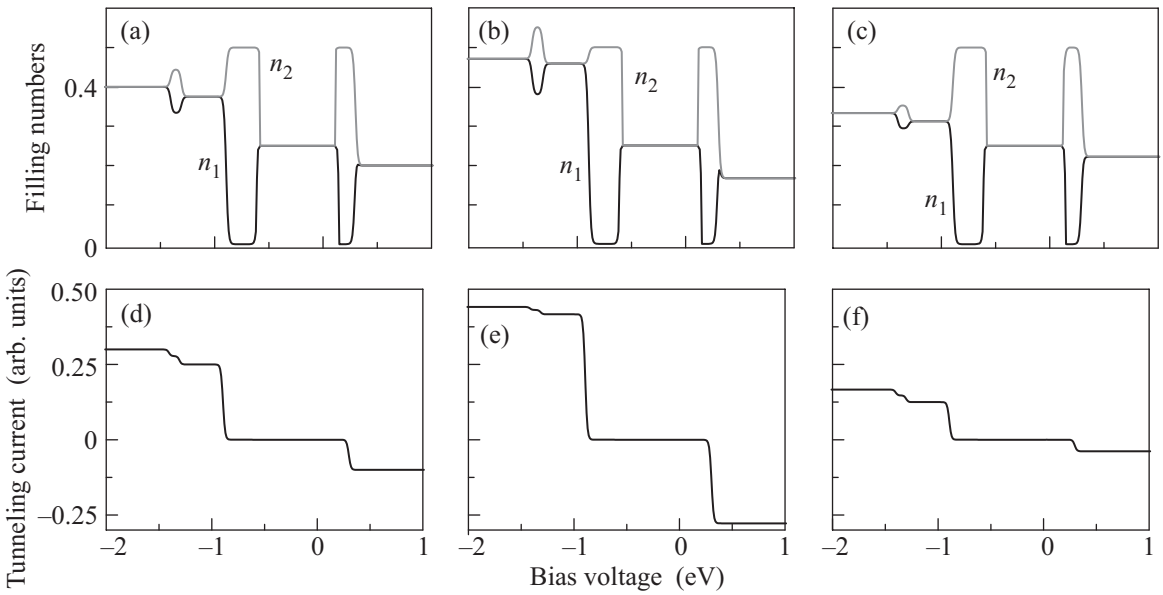

Fig. 3. Two-level system filling numbers (a)–(c) and tunneling current (d)–(f) as a function of applied bias voltage in the case when both energy levels are situated below the sample Fermi level. Parameters $\epsilon_1 = -0.1$, $\epsilon_2 = -0.3$, $U_{12} = 1.0$, $U_{11} = 1.5$, $U_{22} = 1.6$ are the same for all the figures. (a), (d) – $\Gamma_k = 0.01$, $\Gamma_p = 0.01$. (b), (e) – $\Gamma_k = 0.05$, $\Gamma_p = 0.01$. (c), (f) – $\Gamma_k = 0.01$, $\Gamma_p = 0.03$

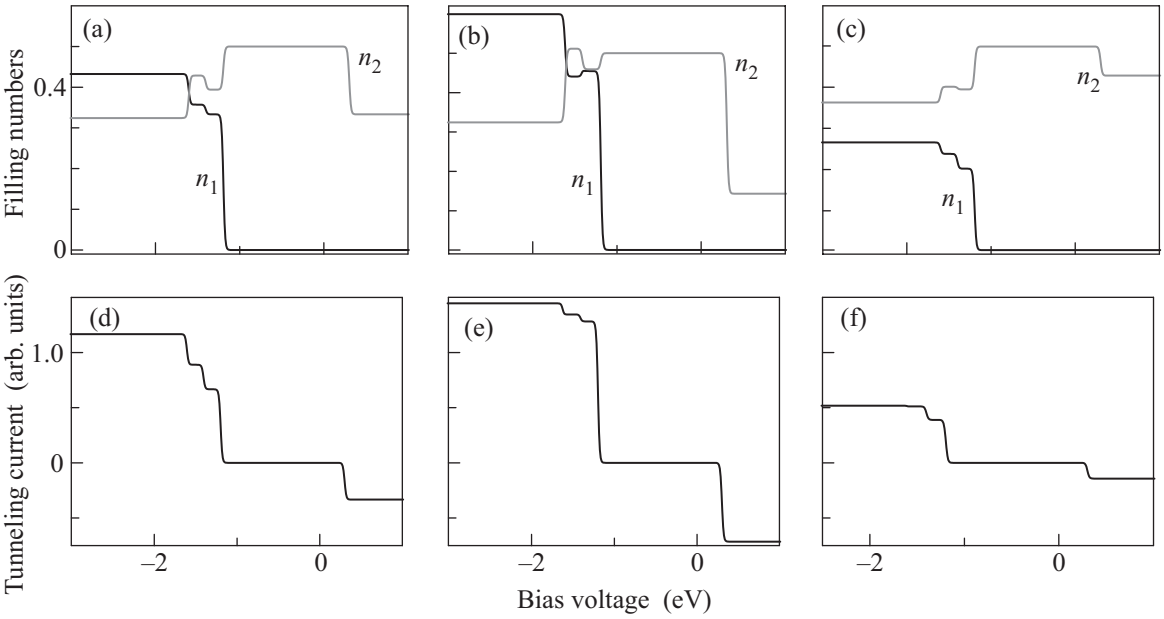

Fig. 4. Two-level system filling numbers (a)–(c) and tunneling current (d)–(f) as a function of applied bias voltage in the case when one energy level is situated above and another one below the sample Fermi level. Parameters $\epsilon_1 = 0.2$, $\epsilon_2 = -0.3$, $U_{12} = 1.0$, $U_{11} = 1.4$, $U_{22} = 1.7$ are the same for all the figures. (a), (d) – $\Gamma_k = 0.01$, $\Gamma_p = 0.01$. (b), (e) – $\Gamma_k = 0.05$, $\Gamma_p = 0.01$. (c), (f) – $\Gamma_k = 0.01$, $\Gamma_p = 0.03$

the Fermi level and another one below the Fermi level (Fig. 4). Applied bias is assumed to act on the right lead (tip), moving it's Fermi level relatively to the Fermi level of the left lead (sample).

From Figs. 2–4 one can clearly see that sharp charge redistribution between two electron states occurs at some values of applied bias voltage (Figs. 2–4). When both levels are situated above (Fig. 2) or below (Fig. 3)

the sample Fermi level there are two possibilities for charge distribution in the two-level system. The first one corresponds to the case when local charge is mostly accumulated on the lower electron level ε_2, $n_1 < n_2$ ($\varepsilon_2 < eV < \varepsilon_1$, $\varepsilon_2 + U_{12} < eV < \varepsilon_1 + U_{12}$ and $\varepsilon_2 + U_{22} < eV < \varepsilon_1 + U_{11}$ on Fig. 2 and Fig. 3). The second one deals with the case when charge is localized on both levels equally $n_1 = n_2$ ($\varepsilon_1 < eV < \varepsilon_2 + U_{12}$, $\varepsilon_1 + U_{12} < eV < \varepsilon_2 + U_{22}$ and $\varepsilon_1 + U_{11} < eV$ in Fig. 2 and Fig. 3). The most interesting effect is that Coulomb correlations induce sudden jumps of each level filling numbers at certain values of applied bias.

So if electron states have essentially different symmetry one can expect charge accumulation in various spatial areas and thus the possibility of local charge manipulation appears.

When both electron level energies are situated below the sample Fermi level upper electron state becomes empty ($n_1 = 0$) for two ranges of applied bias voltage ($\varepsilon_2 < eV < \varepsilon_1$ and $\varepsilon_2 + U_{12} < eV < \varepsilon_1 + U_{12}$) (Fig. 3).

Described peculiarities take place for all the ratios between tunneling transfer rates Γ_k and Γ_p, though for $\Gamma_k > \Gamma_p$ they are usually more pronounced.

The other interesting effect is the possibility of inverse occupation of the two-level system due to Coulomb interaction in special range of applied bias (Fig. 4). In the absence of Coulomb interaction difference of electron filling numbers are determined by the electron tunneling rates $n_1 - n_2 \sim \gamma_{k1}\gamma_{p2} - \gamma_{p1}\gamma_{k2}$. So without Coulomb interaction, for our choice $\gamma_{k(p)1} = \gamma_{k(p)2}$, difference of the two levels occupation numbers turns to zero. Coulomb interaction of localized electrons in the two-level system results in inverse occupation of the two levels at the wide range of applied bias voltage. This situation is clearly demonstrated in the Fig. 4.

When the applied bias doesn't exceed value $\varepsilon_1 + U_{12}$ all the charge is localized on the lower energy level ($n_1 = 0$). With the increasing of the applied bias inverse occupation takes place and localized charge in the system redistributes. Local charge is mostly accumulated on the upper level when applied bias value exceeds $\varepsilon_1 + U_{11}$. Two-level system demonstrates such behaviour if the system more strongly coupled with tunneling contact lead k (sample) (Fig. 4b). Qualitatively the we see if the tunneling contact is symmetrical. But if tunneling rate to the lead p (tip) exceeds tunneling rate to the lead k (sample), then we have not found inverse occupation. In this case with the increasing of applied bias upper electron state charge also increases but local charge continue being mostly accumulated on the lower electron state. So this effect is essentially of nonequilibrium nature.

Tunneling current as a function of applied bias voltage for different level's positions is shown in (Figs. 2–4d–f). Tunneling current amplitudes are normalized on $2\Gamma_k$ everywhere in the paper. For all the values of the system parameters tunneling current dependence on applied bias has a step structure. Height and length of the steps depend on the parameters of the tunneling contact (tunneling transfer rates and values of Coulomb energies). When both energy levels are above the Fermi level one can find six steps in the tunneling current (Fig. 2d–f). If both levels are situated below the Fermi level there are four steps in tunneling current (Fig. 3d–f) and the upper electron level doesn't appear as a step in current-voltage characteristics but charge redistribution takes place due to Coulomb correlations. One can also find four steps in the case when only lower energy level is situated below the Fermi level (Fig. 4d–f).

4. Conclusion. It was shown that for a two-level system with strong coupling between localized electron states it is necessary to take into account high order correlators of local electron density if one calculates the tunneling current. The importance of these Coulomb correlation effects is different for various electron levels location relative to the sample Fermi level and various parameters of the contact. We revealed that charge redistribution between electron states takes place in the suggested model at some bias voltage when both electron levels are situated above or below the sample Fermi level. Another interesting effect concerned with Coulomb correlations consists in inverse occupation of localized electrons states within some bias range if electron levels are localized on the opposite sites of the sample Fermi level.

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