Non-equilibrium Coulomb correlations can drastically influence on the local charge distribution in the vicinity of impurity complexes in nanometer tunneling junctions. Coulomb interaction results in significant changes of each localized state electron filling numbers and current-voltage characteristics of impurity complexes. Adjusting parameters of a tunneling contact one can obtain negative tunneling conductivity caused by Coulomb correlations in a certain range of applied bias. There are several experimental situations in which Coulomb interaction values are of the order of electron levels spacing or even strongly exceed this value. It usually takes place if the distance between several impurity atoms or surface defects is comparable with the lattice scale, so coupling between their electronic states can strongly exceed the interaction of these localized states with continuous spectrum. Another possible realization is a quantum dot or two small interacting quantum dots on the sample surface weakly connected with the bulk states. Such systems can be described by the model including several electron levels with Coulomb interaction between localized electrons. 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 Coulomb correlations in non-equilibrium situation result in spatial redistribution of localized charges and possibility of local charge density manipulation governed by Coulomb correlations. In some sense these effects are similar to the "co-tunneling" observed in [1], [2]. Moreover Coulomb interaction of localized electrons can be responsible for inverse occupation of localized electron states and negative local tunneling conductivity in a certain range of applied bias. These effects can be clearly seen if single electron levels have different spatial symmetry.

Great attention was paid to electron transport through a single impurity or a dot in the Coulomb blockade and the Kondo [3] regimes. These effects have been studied experimentally and are up till now under theoretical investigation [4]-[10]. But if tunneling coupling is not negligible the impurity charge is not a discrete value and one has to deal with impurity electron filling numbers (which now are continuous variables) determined from kinetic equations.

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 were described by self-consistent approach based on Keldysh diagram technique in [11], [12]. In the present work we consider the opposite case when Coulomb coupling between localized electron states strongly exceeds tunneling transfer rates.

We suggest theoretical approach based on the Heisenberg equations for localized states electron filling numbers taking into account all order correlators of local electron density [13]. Tunneling current in a two-level system of spinless fermions with infinite value of Coulomb
interaction has been investigated in [14]. But obtained results do not take into account any nontrivial pair correlations in the system for finite Coulomb correlations. If one is interested in kinetic properties for the applied bias range larger than the characteristic energy of correlations between localized and band electrons in the leads then Kondo effect is unimportant. In this case for the finite number of localized electron levels one can obtain closed system of equations for electron filling numbers and all their correlators. It allows to analyze the role of Coulomb correlations in charge redistribution and in formation main features of I-V characteristics.

II. THE SUGGESTED MODEL

We shall analyze tunneling through the two-level system with Coulomb interaction of localized electrons Fig.1. The model system can be described by the Hamiltonian $H$

$$
H = \sum_{i\sigma} \varepsilon_i n_{i\sigma} + \sum_{k\sigma} \varepsilon_k c_{k\sigma}^+ c_{k\sigma} + \\
+ \sum_{\rho\sigma} \varepsilon_{\rho} c_{\rho\sigma}^+ c_{\rho\sigma} + \sum_{i_j\sigma\sigma'} U_{ij} n_{i\sigma} n_{j\sigma'} + \\
+ \sum_{ki\sigma} t_{ki} (c_{k\sigma}^+ c_{i\sigma} + h.c.) + \sum_{n\sigma} t_{ni} (c_{n\sigma} c_{i\sigma} + h.c.)
$$

(1)

FIG. 1. a). Energy diagram of two-level system and b). Schematic spatial diagram of experimental realization. Coulomb energy $U_{ij}$ correspond to the interaction between electrons on different energy levels.

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 localized states with energies $\varepsilon_i$. Operators $c_{k\sigma}^+ / c_{k\sigma}$ correspond to electrons creation/annihilation in the continuous spectrum states and localized states with filling numbers operators $c_{i\sigma}^+ / c_{i\sigma}$ correspond to electrons filling numbers operators as:

$$
\hat{f}_k = c_{k\sigma}^+ c_{k\sigma}
$$

(4)

In order to get equation for the tunneling current one has to multiply equation (3) by combinations of electron filling numbers operators $n_{i(j)\pm\sigma}$ in the following way:

$$
(1 - n_{1\sigma})(1 - n_{2\sigma}) c_{k\sigma}^+ c_{k\sigma} =
$$

$$
= \{(t_{k1} \cdot (n_{1\sigma} - \hat{f}_k) + \sum_{k' \neq k} t_{k1} c_{k\sigma}^+ c_{k'\sigma} + t_{k2} c_{2\sigma}^+ c_{1\sigma}) \cdot (1 - n_{1\sigma})(1 - n_{2\sigma}) \} \cdot \{\varepsilon_1 - \varepsilon_k\}^{-1}
$$

(5)

$$
= \sum_{\sigma'} n_{1\sigma}(1 - n_{2\sigma}) c_{k\sigma}^+ c_{k\sigma} =
$$

$$
= \{(t_{k1} \cdot (n_{1\sigma} - \hat{f}_k) + \sum_{k' \neq k} t_{k1} c_{k\sigma}^+ c_{k'\sigma} + t_{k2} c_{2\sigma}^+ c_{1\sigma}) \cdot n_{1\sigma}(1 - n_{2\sigma})(1 - n_{2\sigma}) \} \cdot \{\varepsilon_1 - \varepsilon_k + U_{11}\}^{-1}
$$

(6)

$$
= \sum_{\sigma'} n_{2\sigma'}(1 - n_{1\sigma})(1 - n_{2\sigma}) c_{k\sigma}^+ c_{k\sigma} =
$$

$$
= \sum_{\sigma'} \{(t_{k1} \cdot (n_{1\sigma} - \hat{f}_k) + \sum_{k' \neq k} t_{k1} c_{k\sigma}^+ c_{k'\sigma} + t_{k2} c_{2\sigma}^+ c_{1\sigma}) \cdot n_{2\sigma'}(1 - n_{1\sigma})(1 - n_{2\sigma}) \} \cdot \{\varepsilon_1 - \varepsilon_k + U_{12}\}^{-1}
$$

(7)
\[
\sum_{\sigma'} \left\{ (t_{k} \cdot (n_{1\sigma} - \hat{f}_{k}) + \sum_{k' \neq k} t_{k'} c_{k'\sigma} c_{k'\sigma}' + t_{k2} c_{2\sigma} c_{1\sigma}) \cdot n_{1\sigma} n_{2\sigma}' (1 - n_{2\sigma'}) \right\} \cdot \{ \varepsilon_{1} - \varepsilon_{k} + U_{11} + U_{12} \}^{-1}
\]
(8)

\[
\sum_{\sigma'} \left\{ (t_{k} \cdot (n_{1\sigma} - \hat{f}_{k}) + \sum_{k' \neq k} t_{k'} c_{k'\sigma} c_{k'\sigma}' + t_{k2} c_{2\sigma} c_{1\sigma}) \cdot n_{2\sigma} n_{2\sigma}' (1 - n_{1\sigma}) \right\} \cdot \{ \varepsilon_{1} - \varepsilon_{k} + 2U_{12} \}^{-1}
\]
(9)

\[
\sum_{\sigma'} \left\{ (t_{k} \cdot (n_{1\sigma} - \hat{f}_{k}) + \sum_{k' \neq k} t_{k'} c_{k'\sigma} c_{k'\sigma}' + t_{k2} c_{2\sigma} c_{1\sigma}) \cdot n_{1\sigma} n_{2\sigma} n_{2\sigma}' \right\} \cdot \{ \varepsilon_{1} - \varepsilon_{k} + U_{11} + 2U_{12} \}^{-1}
\]
(10)

The relation \( n_{1\sigma}^{2} = n_{1\sigma} \) was used in these equations.

Neglecting changes of electron spectrum and local density of states in the tunneling contact leads caused by the tunneling current we shall uncouple conduction and localized electron filling numbers. This means also that we neglect any correlation effects between localized and band electrons - like the Kondo effect.

It is easy to check:

\[
(1 - n_{1\sigma})(1 - n_{2\sigma})(1 - n_{2\sigma}') + n_{1\sigma}(1 - n_{2\sigma})(1 - n_{2\sigma}') + \sum_{\sigma'} n_{2\sigma'}(1 - n_{1\sigma})(1 - n_{2\sigma'}) + \sum_{\sigma'} n_{1\sigma} n_{2\sigma} n_{2\sigma}' (1 - n_{2\sigma'}) + n_{2\sigma} n_{2\sigma}' (1 - n_{1\sigma}) + n_{1\sigma} n_{2\sigma} n_{2\sigma}' = 1
\]
(11)

Thus summing up the right- and left-hand parts of equations (8-10) we get an equation for \( < c_{1\sigma}^\dagger c_{2\sigma} c_{1\sigma}' > \), which gives us after summation over \( k \) an equation for the tunneling current through the two-level system. Total current is a sum of two contributions:

\[
I_{k\sigma} = I_{k1\sigma} + I_{k2\sigma}
\]
(12)

Where expression for the tunneling current \( I_{k2\sigma} \) can be obtained by changing indexes 1 \( \leftrightarrow \) 2 in the equation for the tunneling current \( I_{k1\sigma} \), which has the following form:

\[
I_{k1\sigma} = \Gamma_{k1\sigma} \left( \langle n_{1\sigma} \rangle - \langle (1 - n_{1\sigma})(1 - n_{2\sigma})(1 - n_{2\sigma'}) \rangle f_{k}(\varepsilon_{1}) - \langle n_{1\sigma} \rangle f_{k}(\varepsilon_{1} + U_{11}) - \langle n_{2\sigma} \rangle f_{k}(\varepsilon_{1} + U_{12}) - \langle n_{2\sigma} \rangle f_{k}(\varepsilon_{1} + U_{12}) - \langle n_{1\sigma} \rangle f_{k}(\varepsilon_{1} + U_{11}) - \langle n_{2\sigma} \rangle f_{k}(\varepsilon_{1} + U_{12}) - \langle n_{1\sigma} \rangle f_{k}(\varepsilon_{1} + U_{11}) - \langle n_{2\sigma} \rangle f_{k}(\varepsilon_{1} + U_{12}) + \right) + \sum_{k' \neq k} t_{k} t_{k'} t_{k2} c_{2\sigma} c_{1\sigma}' + \right)
\]

(13)

In what follows we shall neglect terms \( t_{k1} t_{k2} v_{0k} c_{2\sigma} c_{1\sigma}' \) and terms proportional to \( t_{k1} t_{k2} v_{0k} c_{2\sigma} c_{1\sigma}' \) in the expression (13) as they correspond to the next order perturbation theory in the parameter \( \Gamma_{k} \). Relaxation rates \( \Gamma_{k(p)} = \pi \cdot t_{k(p)}^{2} \cdot \nu_{0} \) are determined by electron tunneling transitions from the two-level system to the leads \( k \) (sample) and \( p \) (tip) continuum states. \( v_{0(k,p)} \)-continuous spectrum density of states. The main equation for the current (13) includes mean electron filling numbers \( n_{1\sigma}, n_{2\sigma} \), pair and triple correlators for the localized states, which have to be determined now. Equations for the total electron filling numbers \( n_{1\sigma}, n_{2\sigma} \), on the levels 1 and 2 can be found from the conditions:

\[
\frac{\partial n_{1\sigma}}{\partial t} = I_{k1\sigma} + I_{p1\sigma} = 0
\]
\[
\frac{\partial n_{2\sigma}}{\partial t} = I_{k2\sigma} + I_{p2\sigma} = 0
\]
(14)

where tunneling current \( I_{p\sigma} \) can be easily obtained from \( I_{k\sigma} \) by changing indexes \( k \leftrightarrow p \).

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

\[
\left( \frac{\partial n_{i\sigma} n_{j\sigma'}}{\partial t} \right) = \left( \frac{\partial n_{i\sigma}}{\partial t} n_{j\sigma'} \right) + \left( \frac{\partial n_{j\sigma'}}{\partial t} n_{i\sigma} \right)
\]
(15)

Full expressions which determine the system of equations for pair filling numbers correlators through the
higher order correlators in the stationary case have the form:

\[
\langle \frac{\partial n_\sigma n_{j\sigma'}}{\partial t} \rangle = (\Gamma_{ki} + \Gamma_{pi} + \Gamma_{kj} + \Gamma_{pj}) \cdot \\
\cdot \langle n_\sigma n_{j\sigma'} \rangle - \\
- (\Gamma_{ki} f_k (\varepsilon_i + U_{ij}) + \Gamma_{pi} f_p (\varepsilon_i + U_{ij})) \cdot \\
\cdot \langle n_{j\sigma'}(1 - n_{j\sigma'}) (1 - n_{i\sigma}) \rangle - \\
- (\Gamma_{kj} f_k (\varepsilon_j + U_{ij}) + \Gamma_{pj} f_p (\varepsilon_j + U_{ij})) \cdot \\
\cdot \langle n_\sigma (1 - n_{i\sigma})(1 - n_{j\sigma'}) \rangle - \\
- (\Gamma_{ki} f_k (\varepsilon_i + U_{ii} + U_{ij}) + \Gamma_{pi} f_p (\varepsilon_i + U_{ii} + U_{ij})) \cdot \\
\cdot \langle n_{i\sigma} (1 - n_{i\sigma})(1 - n_{j\sigma'}) \rangle - \\
- (\Gamma_{kj} f_k (\varepsilon_j + U_{ii} + U_{ij}) + \Gamma_{pj} f_p (\varepsilon_j + U_{ii} + U_{ij})) \cdot \\
\cdot \langle n_{i\sigma} (1 - n_{i\sigma})(1 - n_{j\sigma}) \rangle - \\
- (\Gamma_{kj} f_k (\varepsilon_j + 2U_{ij}) + \Gamma_{pj} f_p (\varepsilon_j + 2U_{ij})) \cdot \\
\cdot \langle n_{i\sigma} n_{j\sigma'} (1 - n_{j\sigma}) \rangle - \\
- (\Gamma_{kj} f_k (\varepsilon_j + U_{jj} + U_{ij}) + \Gamma_{pj} f_p (\varepsilon_j + U_{jj} + U_{ij})) \cdot \\
\cdot \langle n_{i\sigma} n_{j\sigma} (1 - n_{i\sigma}) \rangle - \\
- (\Gamma_{kj} f_k (\varepsilon_j + 2U_{ij}) + \Gamma_{pj} f_p (\varepsilon_j + 2U_{ij})) \cdot \\
\cdot \langle n_{i\sigma} n_{j\sigma} (1 - n_{j\sigma}) \rangle - \\
- (\Gamma_{kj} f_k (\varepsilon_j + U_{jj} + 2U_{ij}) + \Gamma_{pj} f_p (\varepsilon_j + U_{jj} + 2U_{ij})) \cdot \\
\cdot \langle n_{i\sigma} n_{j\sigma} (1 - n_{i\sigma}) \rangle - \\
\cdot \langle n_{i\sigma} n_{j\sigma} n_{i\sigma} n_{j\sigma} \rangle = 0
\]

(16)

High order correlators can be found in the similar way:

\[
\langle \frac{\partial n_{j\sigma} n_{i\sigma} n_{j\sigma'}}{\partial t} \rangle = \langle \frac{\partial n_{j\sigma} n_{i\sigma} n_{j\sigma'}}{\partial t} \rangle + \\
+ \langle \frac{\partial n_{i\sigma} n_{j\sigma} - n_{j\sigma'} n_{i\sigma}}{\partial t} \rangle n_{i\sigma} n_{j\sigma}
\]

(17)

So expressions for high order correlations have the form:

\[
\langle n_{j\sigma} n_{i\sigma} n_{j\sigma'} \rangle = \{\Gamma_{kj} \cdot f_k (\varepsilon_j + U_{jj} + 2U_{ij}) \cdot \\
\cdot (\langle n_{i\sigma} n_{j\sigma} \rangle + \langle n_{i\sigma} n_{j\sigma} \rangle) + \\
+ \Gamma_{ki} \cdot f_k (\varepsilon_i + 2U_{ij}) \cdot \langle n_{j\sigma} n_{j\sigma} \rangle + \\
+ \Gamma_{pj} \cdot f_p (\varepsilon_j + U_{jj} + 2U_{ij}) \cdot \\
\cdot (\langle n_{i\sigma} n_{j\sigma} \rangle + \langle n_{i\sigma} n_{j\sigma} \rangle) + \\
+ \Gamma_{pi} \cdot f_p (\varepsilon_i + 2U_{ij}) \cdot \langle n_{j\sigma} n_{j\sigma} \rangle \} \cdot \\
\cdot \{\Gamma_{ki} \cdot \{3 + f_k (\varepsilon_i + 2U_{ij}) \} - \\
- f_k (\varepsilon_i + U_{ii} + 2U_{ij}) \} + \\
+ \Gamma_{pi} \cdot \{3 + f_p (\varepsilon_i + 2U_{ij}) \} - \\
- f_p (\varepsilon_i + U_{ii} + 2U_{ij}) \} \}^{-1}
\]

(18)

We consider here paramagnetic situation \(n_{i\sigma} = n_{i\sigma} n_{j\sigma} = \langle n_{i\sigma} n_{j\sigma} \rangle \) and \( \langle n_{i\sigma} n_{i\sigma} n_{j\sigma} \rangle \) = \( \langle n_{i\sigma} n_{i\sigma} n_{j\sigma} \rangle \).

(19)

(Note that the system of equations (14)-(19) allows to analyze magnetic regime with \(n_{i\sigma} \neq n_{i\sigma} \) as well. So the system of equations for the pair correlators \(K_{11} = \langle n_{1\sigma} n_{1\sigma} \rangle, K_{22} = \langle n_{2\sigma} n_{2\sigma} \rangle \) and \(K_{12} = \langle n_{1\sigma} n_{2\sigma} \rangle \) after substitution of equation (18) to (19) has the form:

\[
\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
\]

(20)

with coefficients \(a_{ij}\):

\[
a_{11} = 1
\]
\[
a_{12} = 2 \cdot n_{1T} (\varepsilon_1 + U_{11}) - n_{1T} (\varepsilon_1 + U_{11} + U_{12}) - \\
- 2 \cdot \frac{\Gamma_2}{\Gamma_1} \cdot n_{2T} (\varepsilon_2 + U_{22} + U_{12}) \cdot \Phi_1
\]
\[
a_{13} = -n_{1T} (\varepsilon_1 + 2 \cdot U_{12}) \cdot \Phi_1
\]
\[
a_{21} = -n_{2T} (\varepsilon_2 + 2 \cdot U_{12}) \cdot \Phi_2
\]
\[
a_{22} = 2 \cdot n_{2T} (\varepsilon_2 + U_{22}) - n_{1T} (\varepsilon_2 + U_{22} + U_{12}) - \\
- 2 \cdot \frac{\Gamma_1}{\Gamma_2} \cdot n_{1T} (\varepsilon_1 + U_{11} + U_{12}) \cdot \Phi_2
\]
\[
a_{23} = 1
\]
\[
a_{31} = \frac{\Gamma_2}{\Gamma_1 + \Gamma_2} \cdot (n_{2T} (\varepsilon_2 + U_{12}) - \\
- n_{2T} (\varepsilon_2 + 2 \cdot U_{12}) \cdot (1 + 2 \cdot A_2))
\]
\[
a_{32} = 1 + \frac{\Gamma_1}{\Gamma_1 + \Gamma_2} \cdot (n_{1T} (\varepsilon_1 + U_{12}) - \\
- n_{1T} (\varepsilon_1 + U_{11} + U_{12}) \cdot (1 + 4 \cdot A_2))
\]
\[
+ \frac{\Gamma_2}{\Gamma_1 + \Gamma_2} \cdot n_{1T} (\varepsilon_2 + U_{12}) - \\
- n_{1T} (\varepsilon_2 + U_{22} + U_{12}) \cdot (1 + 4 \cdot A_1))
\]
\[
a_{33} = \frac{\Gamma_1}{\Gamma_1 + \Gamma_2} \cdot (n_{1T} (\varepsilon_1 + U_{12}) - \\
- n_{1T} (\varepsilon_1 + 2 \cdot U_{12}) \cdot (1 + 2 \cdot A_1))
\]

(21)

where \(\Gamma_i = \Gamma_{ki} + \Gamma_{pi}\). If we introduce tunneling filling numbers in the absence of Coulomb interaction \(n_{iT} (\varepsilon_i)\), \(n_{iT} (\varepsilon_i + U_{ij})\):

\[
n_{iT} (\varepsilon) = \frac{\Gamma_{ki} f_k (\varepsilon) + \Gamma_{pi} f_p (\varepsilon)}{\Gamma_{ki} + \Gamma_{pi}}
\]

(22)

then coefficients \(\Phi_i\) and \(A_i\) can be found as:
In this case the final equations for higher order correlators, obtained from equation (16) and the leads and back. To improve the results it is necessary to mention that without Coulomb interaction one can find filling numbers for both electron levels to be step functions which correspond to the reentrant character. When applied bias increases two possibilities for charge accumulation for large values of Coulomb energies \( U_{ij} \) are realized in turn. Charge can be localized on both electron levels equally \( n_1 = n_2 \) or mostly accumulated on the lower energy level \( n_1 < n_2 \). In the particular range of applied bias \( \varepsilon_2 < \varepsilon_1 + U_{12} < eV \) for large values of the Coulomb energies. Decreasing of the Coulomb energies leads to the situation where charge is mostly accumulated on the lower energy level \( n_1 \neq 0 \). In the particular range of applied bias \( \varepsilon_2 < \varepsilon_1 + U_{12} < eV \) for large values of the Coulomb energies. Decreasing of the Coulomb energies leads to the situation where charge is mostly accumulated on the lower energy level \( n_1 \neq 0 \). 

Pair correlators \( K_{ij} \) can be expressed through \( n_{i(j)} \) from equations (19, 25). Substituting the solution for higher order correlators, obtained from equation (16) and II to equation (14) one can find \( < n_{i\sigma} \) and finally the tunneling current. 

The determinant of the system (19) can turn to zero or even becomes negative for some choice of the parameters and consequently electron filling numbers of the two-level system can get negative values at some ranges of applied bias voltage. Such invalid system behaviour is the result of our approximation because we neglected the interaction between the two localized electronic states due to the electron transitions to the continuous spectrum states in the leads and back. To improve the results it is necessary to take into account corrections which can be found using next order perturbation theory in the parameter \( \Gamma_i \), retaining the terms \( t_{k1}t_{k2}\mu_{\alpha}c_{k\sigma}c_{i\sigma} \) in equation (23). In this case the final equations for \( n_{i\sigma} \) have additional nonlinear terms and can be schematically written as:

\[
\begin{align*}
\Phi_i &= \frac{n_i^T(\varepsilon_i + U_{ii}) - n_i^T(\varepsilon_i + U_{ii} + 2U_{ij})}{3 + n_i^T(\varepsilon_i + 2U_{ij}) - n_i^T(\varepsilon_i + U_{ii} + 2U_{ij})} + \frac{n_i^T(\varepsilon_i + U_{ii} + 2U_{ij})}{3 + n_i^T(\varepsilon_i + 2U_{ij}) - n_i^T(\varepsilon_i + U_{ii} + 2U_{ij})}
A_i &= \frac{n_i^T(\varepsilon_i + 2U_{ij}) - n_i^T(\varepsilon_i + U_{ii})}{3 + n_i^T(\varepsilon_i + 2U_{ij}) - n_i^T(\varepsilon_i + U_{ii} + 2U_{ij})} + \frac{n_i^T(\varepsilon_i + U_{ii}) - n_i^T(\varepsilon_i + U_{ii} + 2U_{ij})}{3 + n_i^T(\varepsilon_i + 2U_{ij}) - n_i^T(\varepsilon_i + U_{ii} + 2U_{ij})}
F &= \left( \frac{n_1^T(\varepsilon_1 + U_{11}) \cdot n_{1\sigma}}{1 + n_1^T(\varepsilon_1 + U_{12})}, \frac{n_2^T(\varepsilon_2 + U_{22}) \cdot n_{2\sigma}}{1 + n_2^T(\varepsilon_2 + U_{12})} \right)
\end{align*}
\]

Pair correlators \( K_{ij} \) can be expressed through \( n_{i(j)} \) from equations (19, 25). Substituting the solution for higher order correlators, obtained from equation (16) and II to equation (14) one can find \( < n_{i\sigma} \) and finally the tunneling current. 

The determinant of the system (19) can turn to zero or even becomes negative for some choice of the parameters and consequently electron filling numbers of the two-level system can get negative values at some ranges of applied bias voltage. Such invalid system behaviour is the result of our approximation because we neglected the interaction between the two localized electronic states due to the electron transitions to the continuous spectrum states in the leads and back. To improve the results it is necessary to take into account corrections which can be found using next order perturbation theory in the parameter \( \Gamma_i \), retaining the terms \( t_{k1}t_{k2}\mu_{\alpha}c_{k\sigma}c_{i\sigma} \) in equation (23). In this case the final equations for \( n_{i\sigma} \) have additional nonlinear terms and can be schematically written as:

\[
\begin{align*}
n_{1\sigma} \cdot (A_{11} + \mu_1 n_{1\sigma}^2) + n_{2\sigma} \cdot (A_{12} + \mu_2 n_{2\sigma}^2) &= n_T(\varepsilon_1) \\
n_{2\sigma} \cdot (A_{22} + \nu_2 n_{2\sigma}^2) + n_{1\sigma} \cdot (A_{21} + \nu_1 n_{1\sigma}^2) &= n_T(\varepsilon_2)
\end{align*}
\]

Coefficients \( A_{ij} \), \( \mu_i \) and \( \nu_i \) have rather simple but cumbersome form and depend only on the tunneling filling numbers and parameters of the tunneling contact.

In this paper we shall not regarded this case.

III. MAIN RESULTS AND DISCUSSION

The behaviour of \( n_{i\sigma} \) and I-V characteristics strongly depends on the parameters of the tunneling system. At first let us analyze the situation in which tunneling rates from both localized states to the leads are approximately equal \( t_{k(p)1} = t_{k(p)2} \). Figures 2,7 demonstrate behaviour of filling numbers and tunneling current obtained from kinetic equations for the different values of Coulomb energies \( U_{ij} \) and various electron levels location relative to the sample Fermi level in symmetric \( \Gamma_{ki} \sim \Gamma_{pi} \) and asymmetric \( \Gamma_{ki} < \Gamma_{pi} \) tunneling contact taking into account all order correlators. The bias voltage in our calculations is applied to the sample. So if both levels are above(below) the Fermi level all the specific features of charge distribution and tunneling current characteristics can be observed at the negative(positive) values of \( eV \).

In the case of both energy levels situated above (Fig.2) or below (Fig.3, Fig.4) the sample Fermi level we observe charge redistribution between electron levels of reentrant character. When applied bias increases two possibilities for charge accumulation for large values of Coulomb energies \( U_{ij} \) are realized in turn. Charge can be localized on both electron levels equally \( n_1 = n_2 \) or mostly accumulated on the lower energy level \( n_1 < n_2 \). In the particular range of applied bias \( \varepsilon_2 < \varepsilon_1 + U_{12} < eV \) for large values of the Coulomb energies. Decreasing of the Coulomb energies leads to the situation where charge is mostly accumulated on the lower energy level \( n_1 \neq 0 \). In the particular range of applied bias \( \varepsilon_2 < \varepsilon_1 + U_{12} < eV \) for large values of the Coulomb energies. Decreasing of the Coulomb energies leads to the situation where charge is mostly accumulated on the lower energy level \( n_1 \neq 0 \). Taking into account all order correlators gives us an opportunity to investigate tunneling through the two-level system in the case of small Coulomb energies \( U_{ij} \sim \varepsilon_{i(j)} \). Figure 5 demonstrates how filling numbers and tunneling current dependencies change due to decrease of Coulomb energies for the symmetric tunneling contact \( \Gamma_{ki} = \Gamma_{pi} \) (asymmetric contacts show the same tendencies). We demonstrate the case of both electron levels localized above the sample Fermi level.

If Coulomb interaction is of the order of single electron energies, three ranges of applied bias appear, where inverse occupation takes place: \( n_1 > n_2 \) (Fig.5b) \( \varepsilon_2 + U_{12} < eV < \varepsilon_1 + U_{11} < \varepsilon_2 + U_{12} < eV < \varepsilon_1 + U_{12} \) and \( \varepsilon_1 + U_{11} + U_{12} < eV < \varepsilon_2 + U_{12} + 2U_{12} \). Such situation exists due to the condition that system configuration with two electrons on the upper level and one electron on the lower level has lower energy than configuration with one electron on the upper level and two electrons on the lower level for the parameters shown in Fig.5. Further decreasing of the Coulomb energies (Fig.5c) reduces the effect of inverse occupation and finally local charge mostly accumulates on the lower energy level as it should be.

We obtain that the effects of reentrant charge redistribution is more pronounced for asymmetric contact if tunneling rates to the sample are larger than tunneling rates to the tip.

It is necessary to mention that without Coulomb interaction one can find filling numbers for both electron levels to be simple step functions which correspond to the tunneling filling numbers \( n_i^T(\varepsilon_i) \) shifted from each other on the value \( \varepsilon_1 - \varepsilon_2 \).
The effect of inverse occupation due to the Coulomb correlations is more pronounced in a system with electron levels positioned on the opposite sides of the sample Fermi level. (Fig.4a,b) Without Coulomb interaction, when $\Gamma_{k(p)1} = \Gamma_{k(p)2}$, the difference of the two levels occupation numbers ($n_1 - n_2 \sim \Gamma_{k1} \Gamma_{k2} - \Gamma_{p1} \Gamma_{k2}$) turns to zero. Taking into account Coulomb correlations of localized electrons in the two-level system results in inverse occupation of the two levels at the wide range of applied bias voltage (Fig.4c,d).

In Fig.4(a,b) there are three ranges of applied bias where inverse occupation takes place ($\varepsilon_1 + U_{11} < eV < \varepsilon_2 + 2U_{12}$, $\varepsilon_1 + 2U_{12} < eV < \varepsilon_2 + U_{22} + U_{12}$ and $\varepsilon_1 + U_{11} + U_{12} < eV$). It is clearly evident (Fig.4a,b) that when applied bias doesn’t exceed the value $\varepsilon_1 + U_{12}$ the whole charge is localized on the lower energy level ($n_1 = 0$). With the increasing of applied bias inverse occupation takes place and localized charge redistributes. The effect of inverse occupation strongly depends on relation between tunneling rates. It is most pronounced in asymmetric contact with more strong tunneling coupling to the lead $k$ (sample). But we have not found inverse occupation if the two-level system strongly coupled with tunneling contact lead $p$ (tip) (Fig.4c). In this case with the increasing of applied bias upper electron level charge increases but local charge still mostly accumulated on the lower electron level.

Decreasing of the Coulomb energies results in disappearing of the inverse occupation (Fig.4a,c) and local charge mostly accumulates on the lower energy level. This clearly demonstrates the role of Coulomb interaction in described charge distribution effects.

Tunneling current as a function of applied bias voltage for different level’s positions is depicted in (Fig.4a,f) (tunneling current amplitudes are normalized on $2\Gamma_k$). 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). If both levels are situated below the Fermi level (Fig.4c-f) upper electron level doesn’t appear as a step in current-voltage characteristics but charge redistribution takes place due to Coulomb correlations.

For approximately equal tunneling rates for both localized levels current-voltage characteristics are mostly monotonous functions. But some new peculiarities appear if tunneling rates are essentially different. In Fig.4d we show some results for the case $t_{k(p)1} \neq t_{k(p)2}$. In this case an interplay between “single electron” nonequilibrium occupation effects and Coulomb correlation effects exists and at certain bias charge redistribution is accompanied by negative differential conductivity.

The case of both energy levels situated above the sample Fermi level is shown in Fig.4b. If the tunneling transfer rate from the sample to the lower energy level is the largest in the system and the tunneling transfer amplitude from the lower energy level to the tip is the lowest one (Fig.4c,d), we see, that local charge in the system is mostly accumulated on the lower energy level. Vice versa if the tunneling transfer rate from the sample to the upper energy level is the largest one and from the upper energy level to the tip is the lowest in the system (Fig.4b,d), one can find that local charge is mainly accumulated on the upper energy level and consequently inverse occupation takes place. But due to the Coulomb interaction three ranges of applied bias exist where local charge is mostly localized on the lower energy level $\varepsilon_2 < eV < \varepsilon_1$, $\varepsilon_2 + U_{12} < eV < \varepsilon_1 + U_{12}$ and $\varepsilon_2 + U_{22} + U_{12} < eV < \varepsilon_1 + U_{11} + U_{12}$.

Inverse occupation also takes place when energy levels are positioned on the opposite sides of the sample Fermi level (Fig.4b) or when both energy levels are situated below the Fermi level (Fig.4b). In any case Coulomb interaction modifies single electron occupation behavior, changing with applied bias normal occupation to inverse one or vice versa.

If we look at Fig.4b, we find several ranges of applied bias where the charge is distributed differently. These intervals depend on Coulomb interaction values: the whole charge is accumulated on the lower energy level ($n_1 = 0$) for $eV < \varepsilon_1 + U_{12}$; inverse occupation exists (local charge is mostly localized on the upper energy level) for $\varepsilon_1 + U_{12} < eV < \varepsilon_2 + U_{22} + U_{12}$ and $\varepsilon_1 + U_{11} + U_{12} < eV$; charge is equally accumulated on both electron levels $n_1 = n_2$ if $\varepsilon_2 + U_{22} + U_{12} < eV < \varepsilon_1 + U_{11} + U_{12}$.

If both energy levels are situated below the Fermi level (Fig.4b) there are similar ranges of applied bias in which charge is distributed differently (equally for $\varepsilon_1 < eV < \varepsilon_2 + U_{12}$, inverse occupation if $\varepsilon_1 + U_{12} < eV < \varepsilon_2 + U_{22} + U_{12}$ and $\varepsilon_1 + U_{11} + U_{12} < eV$ and so on).

The appearance of negative conductivity regions is the most essential feature of the tunneling characteristics, depicted in Figs.4c,d, 4b,d). We want to stress once more that formation of negative conductivity is an interplay between non-equilibrium effects connected with the tunneling current and Coulomb correlations.

**IV. CONCLUSION**

We investigated tunneling through the two-level system with strong Coulomb interaction between localized electrons taking into account all order correlators of local electron density. It was shown that charge redistribution between electron states is strongly governed by the Coulomb correlations and is of reentrant type. Electron filling numbers dependence on applied bias becomes quite different from that for non interacting electrons. Existence of the charge redistribution effects means that adjusting the applied bias one can control spatial redistribution of localized charges. So wide possibilities for local charge accumulation and charge switching exist for such systems.

Besides this, at certain values of Coulomb interaction of localized electrons one can obtain correlation induced
inverse occupation of the two-level system in different ranges of applied bias. Inverse occupation is mostly pronounced in asymmetric contacts with different tunneling rates to the sample and to the lead, and when one energy level lies below the Fermi level and another one - above.

Changing the parameters of the tunneling contact (tunneling rates of each level to the leads) we can observe interplay between two mechanisms responsible for non-equilibrium occupation of each level: tunneling current induced inverse occupation of two-level system at particular ratio between tunneling rates (which exists in the absence of Coulomb interaction) and inverse occupation connected only with Coulomb interaction of localized electrons.

We revealed that for some parameter range system demonstrates negative tunneling conductivity in certain ranges of applied bias voltage. Negative tunneling conductivity is revealed in asymmetric case $\Gamma_{ki} \neq \Gamma_{pi}$ (Fig.8 and Fig.9) and is more pronounced if both energy levels are situated above the Fermi level. When energy levels are situated on the opposite sites of Fermi level negative tunneling conductivity is much weaker and when both of them are positioned below the Fermi level it is negligible.

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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). $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$; b),e). $\Gamma_{k1} = \Gamma_{k2} = 0.03$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$; c),f). $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.03$.

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). $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$; b),e). $\Gamma_{k1} = \Gamma_{k2} = 0.03$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$; c),f). $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.03$;
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). $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$; b),e). $\Gamma_{k1} = \Gamma_{k2} = 0.03$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$; c),f). $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.03$.

FIG. 5: 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 $\epsilon_1 = 0.6$, $\epsilon_2 = 0.3$, $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$ are the same for all the figures. a),d). $U_{12} = 0.7$, $U_{11} = 1.0$, $U_{22} = 1.1$; b),e). $U_{12} = 0.35$, $U_{11} = 0.5$, $U_{22} = 0.9$; c),f). $U_{12} = 0.35$, $U_{11} = 0.45$, $U_{22} = 0.55$. 
FIG. 6: 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 $\epsilon_1 = -0.1$, $\epsilon_2 = -0.3$, $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$ are the same for all the figures. a),d). $U_{12} = 1.0$, $U_{11} = 1.5$, $U_{22} = 1.6$; b),e). $U_{12} = 0.4$, $U_{11} = 0.5$, $U_{22} = 0.65$; c),f). $U_{12} = 0.15$, $U_{11} = 0.25$, $U_{22} = 0.4$

FIG. 7: 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 $\epsilon_1 = 0.2$, $\epsilon_2 = -0.3$, $\Gamma_{k1} = \Gamma_{k2} = 0.01$, $\Gamma_{p1} = \Gamma_{p2} = 0.01$ are the same for all the figures. a),d). $U_{12} = 1.0$, $U_{11} = 1.4$, $U_{22} = 1.7$; b),e). $U_{12} = 0.1$, $U_{11} = 0.25$, $U_{22} = 0.8$; c),f). $U_{12} = 0.1$, $U_{11} = 0.15$, $U_{22} = 0.25$
FIG. 8: Two-level system filling numbers a).-b). and tunneling current c).-d). as a function of applied bias voltage in the case when both energy levels are situated above the sample Fermi level for different values of tunneling rates. Parameters $e_1 = 0.6, e_2 = 0.3, U_{12} = 1.0, U_{11} = 1.5, U_{22} = 1.6$ are the same for all the figures. a),c).$\Gamma_{k1} = 0.06, \Gamma_{p1} = 0.05, \Gamma_{k2} = 0.15, \Gamma_{p2} = 0.005$; b),d).$\Gamma_{k1} = 0.15, \Gamma_{p1} = 0.005, \Gamma_{k2} = 0.06, \Gamma_{p2} = 0.05$
FIG. 9: Two-level system filling numbers a).-b). and tunneling current c).-d). as a function of applied bias voltage in the case when one energy level is situated above and another one below a).c). and both energy levels are situated below b).d). the sample Fermi level for different values of tunneling rates. Parameters $\Gamma_k = 0.15, \Gamma_p = 0.005, \Gamma_{k2} = 0.06, \Gamma_{p2} = 0.05$ are the same for all the figures. a),c). $\epsilon_1 = 0.2$, $\epsilon_2 = -0.3$, $U_{12} = 1.0$, $U_{11} = 1.4$, $U_{22} = 1.7$; b),d). $\epsilon_1 = -0.1$, $\epsilon_2 = -0.3$, $U_{12} = 1.0$, $U_{11} = 1.5$, $U_{22} = 1.6$