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The effect of Coulomb correlations on the two-level quantum dot susceptibility and polarization

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We revealed that susceptibility and polarization of two-level quantum dot (QD) with Coulomb correlations between localized electrons weakly connected to the reservoirs are not determined only by the stationary electron filling numbers difference. We demonstrated that susceptibility and polarization also depend on high-order correlation functions of electrons localized in the QD. We found that susceptibility and polarization can be controlled by applied bias voltage value, Coulomb correlations strength and Rabi frequency. We demonstrated that susceptibility and polarization amplitudes can significantly increase and even change the sign due to the tuning of the QD parameters. Careful analysis of correlated QD susceptibility, polarization and electron filling numbers (occupancies) difference in a wide range of applied bias voltage, Rabi frequency and Coulomb correlations value was performed in terms of pseudo-operators with constraint.

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1. Introduction. Recently great attention is paid to the investigations of nanostructures with strong Coulomb correlations, in connection with possibility of ultra small size electronic devices fabrication with high precision [1, 2]. Optical properties of three-dimensionally confined electrons and holes in semiconductor QDs have been extensively studied in recent years [3]. Spatial confinement and Coulomb interaction play an important role in determining the optical properties of QDs. Now a days semiconductor QDs appear as a promising candidates to achieve large susceptibilities. Moreover the dipole matrix elements in QDs can also be large, with typical dipole matrix element ranging from a fraction of a nanometer to a few nanometers. Consequently, the analysis of coupled QDs polarization is also of great interest. This work is caused by the interest in fundamental physics of finite systems and their potential as efficient nonlinear optical and laser materials [4, 5].

Modern experimental technique gives possibility to create single QDs with a given set of parameters and coupled QDs with different spatial geometry [6–8]. The main effort in the physics of QDs is devoted to the investigation of non-equilibrium charge states and different charge configurations due to the electrons tunneling through the QDs in the presence of strong Coulomb interaction [9–13]. Double QDs systems behavior is recently under careful investigation because of the variable inter-dot tunneling coupling [14, 15], which is the physical reason for non-linearity formation and consequently for existence of such phenomena as bifurcations [16, 17] and bi-stability [18, 19]. This is the reason for QDs possible application for logic gates fabrication based on the effect of ultra-fast switching between intrinsic stable states. Electron transport in QDs is strongly governed by Coulomb interaction between localized electrons, the ratios between tunneling transfer amplitudes, QDs coupling and of course by the initial conditions [17, 20].

Due to the development of light sources the control on electric charge in low-dimensional systems is produced both by gate voltages [21, 22] and laser pulses [23, 24]. Consequently, QDs with several enegy levels are of great interest, because coupling strength between the QD energy levels and reservoirs can be easily tuned "by hands" during the analysis of non-equilibrium charge states and different charge configurations, which determine the optical properties of semiconductor nanostructures [25]. Experiments on QDs in a weak confinement regime revealed distinctly non-bulk like features including enhanced nonlinear optical susceptibility, which depends on the dots size [22]. Theoretically, considerable progress was achieved in the description of the single-particle electronic structure providing a satisfactory framework for describing the optical response of QDs of a radius comparable to or smaller than exciton Bohr radius [26, 27]. In [28] authors studied non-

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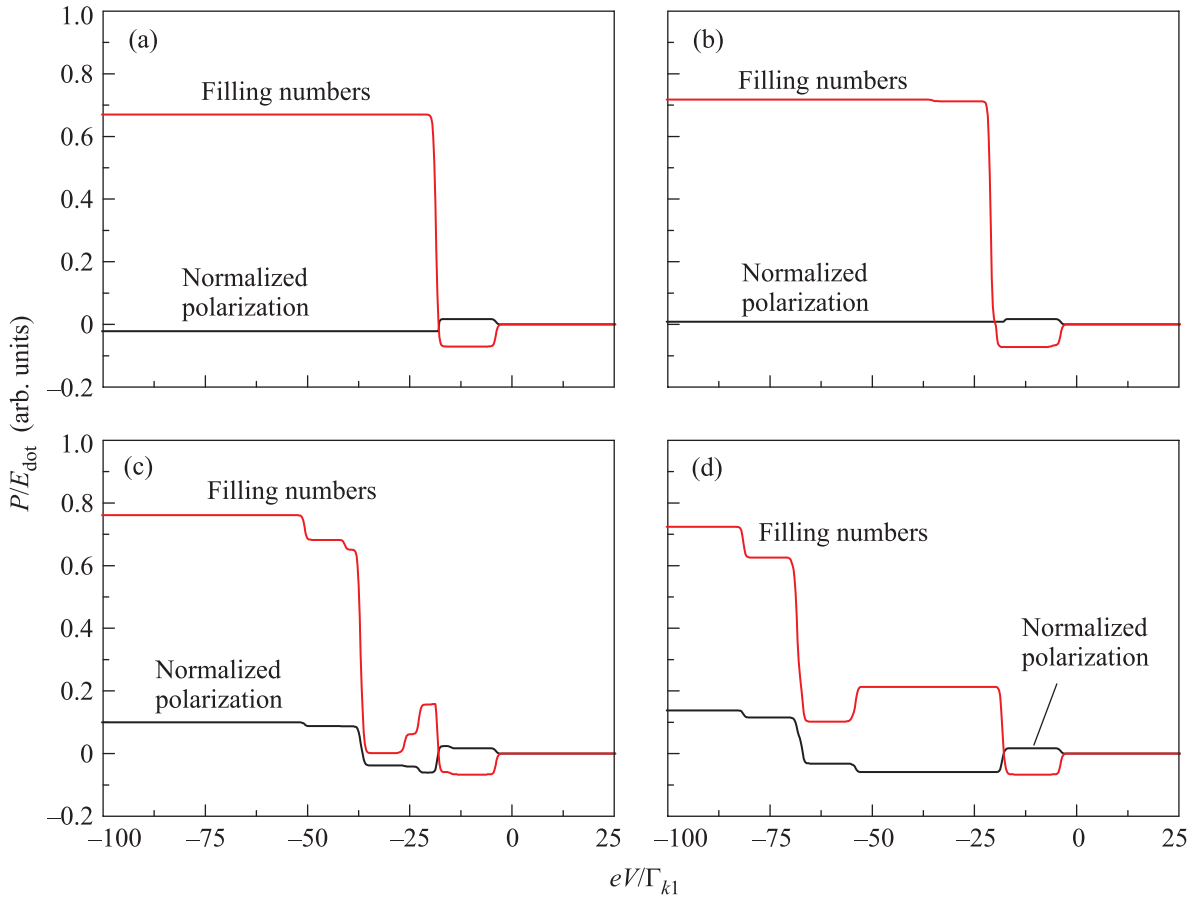

Fig. 1. (Color online) Normalized polarization $P/E_{\rm dot}$ (black line) and electron filling numbers difference $n_1 - n_2$ (red line) as functions of applied bias voltage calculated for different values of Coulomb correlations in the case when both single-electron energy levels are situated above the sample Fermi level: (a) – $U/\Gamma_{k1} = 0$; (b) – $U/\Gamma_{k1} = 2.5$; (c) – $U/\Gamma_{k1} = 18.75$; (d) – $U/\Gamma_{k1} = 50$. For all the figures parameters values $\varepsilon_1/\Gamma_{k1} = 17.5$, $\varepsilon_2/\Gamma_{k1} = 5$, $\Omega/\Gamma_{k1} = 7.5$, $\Gamma_{k1} = \Gamma_{p2} = 1$, $\Gamma_{k2}/\Gamma_{k1} = \Gamma_{p1}/\Gamma_{k1} = 0.125$ are the same

linear optical response of semiconductor QDs of radii up to 10 Bohr radius with the use of approximation based on exciton-exciton product state basis. Authors calculated the third-order nonlinear optical susceptibility at the exciton resonance and clarified the physics of the observed saturation of the mesoscopic enhancement. Method for calculating the optical spectrum of semiconductor micro-structures which is based on a real-space representation of the Hamiltonian and the time-dependent solution of the Schrödinger equation was presented in [30].

In the present paper we consider electron tunneling through two-level QD with Coulomb correlations weakly coupled to the reservoirs. We analyzed susceptibility, polarization and electron filling numbers difference behavior in terms of pseudo operators with constraint [30–33]. We revealed that susceptibility and polarization of two-level QD weakly connected to the reservoirs are not proportional to the electron filling numbers difference when strong Coulomb correlations are present. Susceptibility and polarization behavior are strongly determined by the single-electron energy levels position, applied bias voltage and Coulomb correlations values.

2. Suggested model. We consider tunneling through the two-level QD with Coulomb interaction of localized electrons connected to the two leads. We assume that the single-particle level spacing in the dot is larger than energy levels width, so that two one-electron spin-degenerate levels of QD spectrum are well resolved. The model system can be described by the Hamiltonian:

$$\widehat{H} = \sum_{\sigma,l=1}^{2} c_{l\sigma}^{+} c_{l\sigma} \widetilde{\varepsilon}_l + \sum_{\sigma,l=1}^{2} U_l \widehat{n}_{l\sigma} \widehat{n}_{l-\sigma} + \\ + \sum_{\sigma} \frac{\Omega}{2} (c_{1\sigma}^{+} c_{2\sigma} + c_{2\sigma}^{+} c_{1\sigma}), \qquad (1)$$

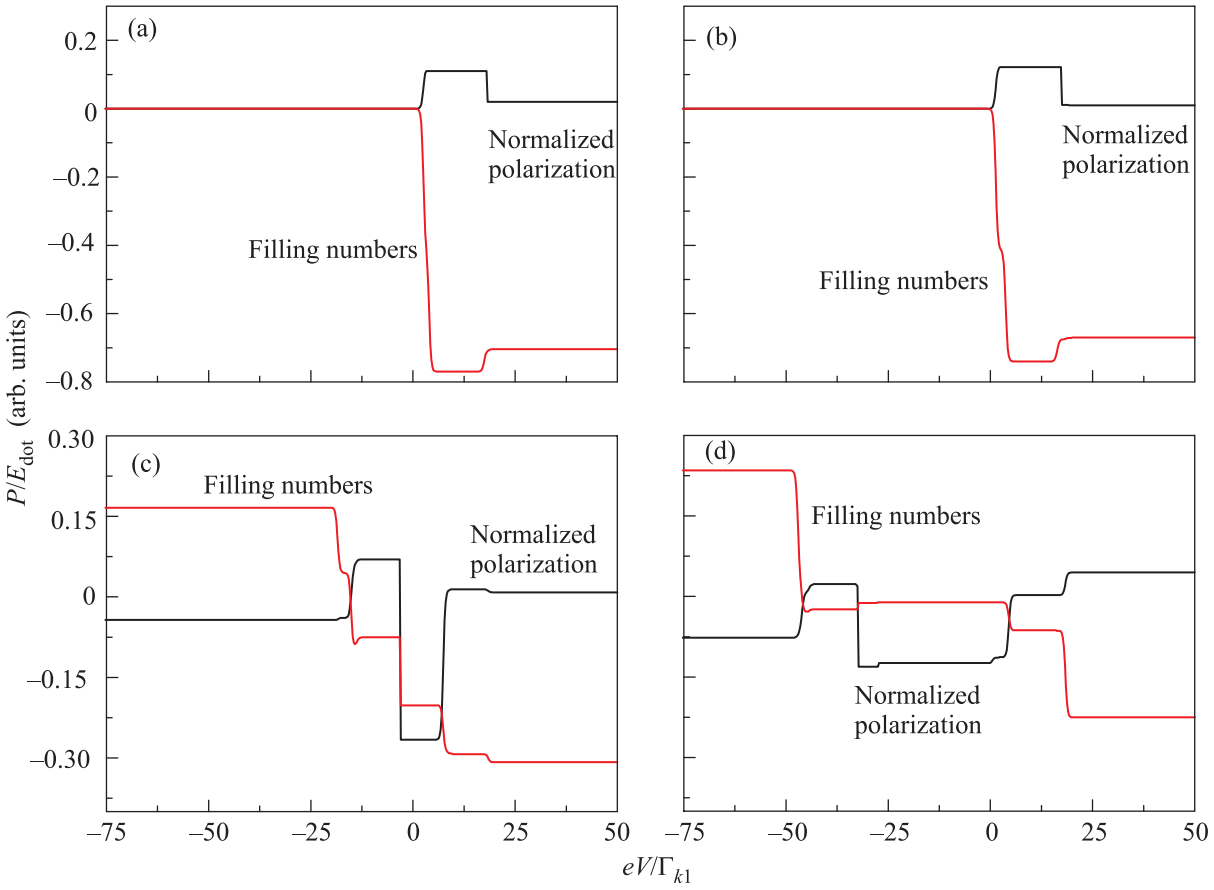

Fig. 2. (Color online) Normalized polarization $P/E_{\rm dot}$ (black line) and electron filling numbers difference $n_1 - n_2$ (red line) as a functions of applied bias voltage calculated for different values of Coulomb correlations in the case when both single-electron energy levels are situated below the sample Fermi level: (a) – $U/\Gamma_{k1} = 0$; (b) – $U/\Gamma_{k1} = 2.5$; (c) – $U/\Gamma_{k1} = 18.75$; (d) – $U/\Gamma_{k1} = 50$. For all the figures parameters values $\varepsilon_1/\Gamma_{k1} = -5$, $\varepsilon_2/\Gamma_{k1} = -17.5$, $\Omega/\Gamma_{k1} = 7.5$, $\Gamma_{k1} = \Gamma_{p2} = 1$, $\Gamma_{k2}/\Gamma_{k1} = \Gamma_{p1}/\Gamma_{k1} = 0.125$ are the same

where operator $c_{l\sigma}$ creates an electron with spin σ on the single energy level $\widetilde{\varepsilon}_l$ and coupling between the energy levels is realized by means of the external field with Rabi frequency Ω, $n_{l\sigma} = c^+_{l\sigma}c_{l\sigma}$, and U_l is the on-site Coulomb repulsion of localized electrons. In our model we neglect Coulomb correlations U_{12} between electrons localized on the different energy levels, as it results only in the on-site Coulomb repulsion renormalization for each [12]. Moreover, the system with inter-dot correlations can be easily reduced to the considered system with the help of the following substitutions: $U_1 = U_{11} - U_{12}$ and $U_2 = U_{22} - U_{12}$. In the case of strongly correlated QD it is reasonable to use the basis of exact eigenfunctions and eigenvalues without taking into account interaction with the leads. In this case all energies of single- and multi-electron states are well known [33, 34].

When correlated two-level QD is connected with the tunneling contact leads the number of electrons in the dot changes due to the tunneling processes. Transitions between the states with different number of electrons in the QD can be analyzed in terms of pseudo-particle operators with constraint on the physical states (the number of pseudo-particles) [30–33]. Consequently, the electron operator $c^+_{\sigma l}$ $(l = 1, 2)$ can be written in terms of pseudo-particle operators as:

$$c^+_{\sigma l} = \sum_i X^{\sigma l}_i f^+_{\sigma i} b + \sum_{j,i} Y^{\sigma-\sigma l}_{ji} d^{+\sigma-\sigma}_j f_{i-\sigma} + \\ + \sum_i Y^{\sigma\sigma l}_i d^{+\sigma\sigma} f_{i\sigma} + \sum_{m,j} Z^{\sigma\sigma-\sigma l}_{mj} \psi^+_{m-\sigma} d^{\sigma-\sigma}_j + \\ + \sum_m Z^{\sigma-\sigma-\sigma l}_m \psi^+_{m\sigma} d^{-\sigma-\sigma} + \sum_m W^{\sigma-\sigma-\sigma l}_m \varphi^+ \psi_{m\sigma}, \quad (2)$$

where $f^+_\sigma(f_\sigma)$ and $\psi^+_\sigma(\psi_\sigma)$- are pseudo-fermion creation (annihilation) operators for the electronic states with one and three electrons correspondingly; $b^+(b)$, $d^+_\sigma(d_\sigma)$, and $\varphi^+(\varphi)$ are slave boson operators, which correspond to the states without any electrons, with two

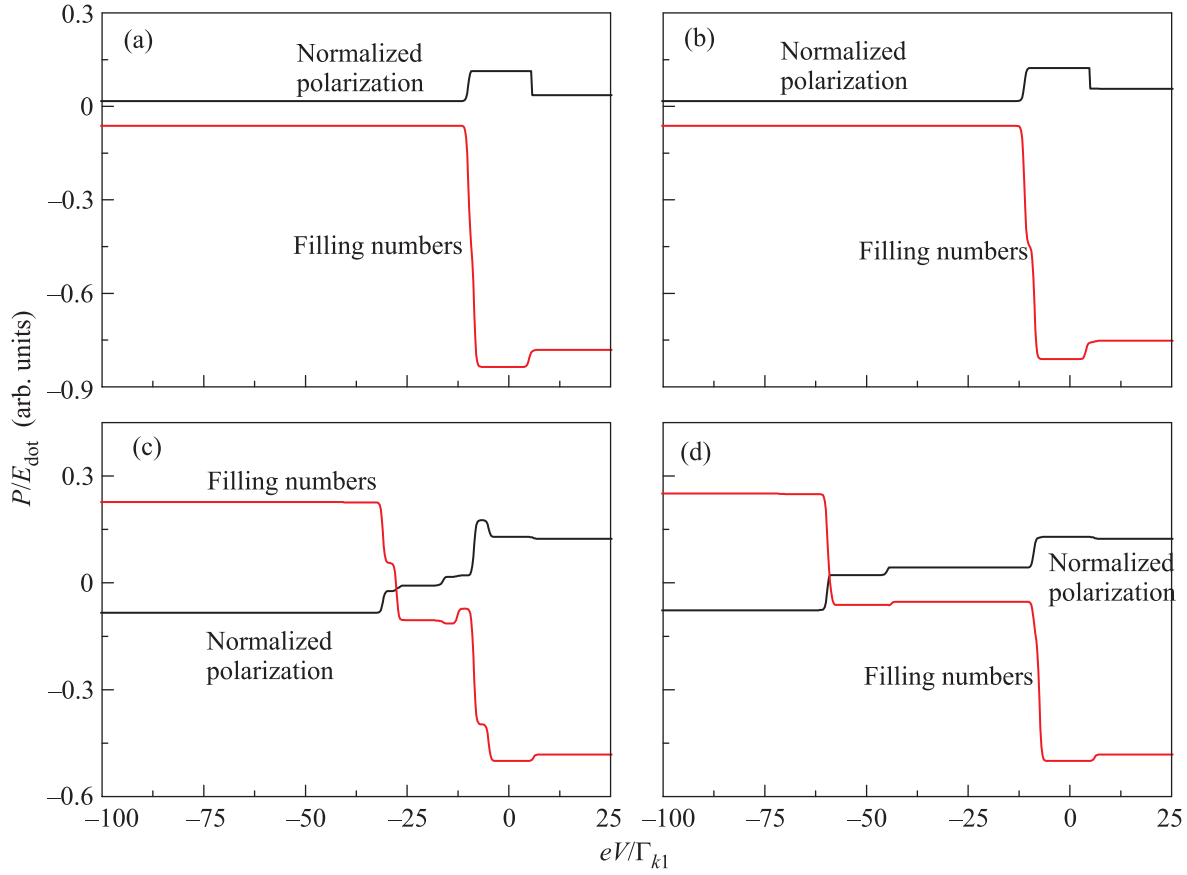

Fig. 3. (Color online) Normalized polarization $P/E_{\rm dot}$ (black line) and electron filling numbers difference $n_1 - n_2$ (red line) as a functions of applied bias voltage calculated for different values of Coulomb correlations in the case when one of the single-electron energy levels is situated above and another one below the sample Fermi level: (a) – $U/\Gamma_{k1} = 0$; (b) – $U/\Gamma_{k1} = 2.5$; (c) – $U/\Gamma_{k1} = 18.75$; (d) – $U/\Gamma_{k1} = 50$. For all the figures parameters values $\varepsilon_1/\Gamma_{k1} = 7.5$, $\varepsilon_2/\Gamma_{k1} = -5$, $\Omega/\Gamma_{k1} = 7.5$, $\Gamma_{k1} = \Gamma_{p2} = 1$, $\Gamma_{k2}/\Gamma_{k1} = \Gamma_{p1}/\Gamma_{k1} = 0.125$ are the same

or four electrons correspondingly. Operators $\psi^{+}_{m-\sigma}$ describe system configuration with two spin up electrons σ and one spin down electron $-\sigma$ in the symmetric and asymmetric states.

Matrix elements $X_i^{\sigma l}$, $Y_{ji}^{\sigma -\sigma l}$, $Y_{ji}^{\sigma\sigma l}$, $Z_{mj}^{\sigma\sigma -\sigma l}$, $Z_{mj}^{\sigma -\sigma -\sigma l}$, and $W_m^{\sigma -\sigma -\sigma l}$ can be evaluated as:

$$\begin{aligned} X_i^{\sigma l} &= \langle \psi_i^{\sigma} | c_{\sigma l}^{+} | 0 \rangle, \\ Y_{ji}^{\sigma -\sigma l} &= \langle \psi_j^{\sigma -\sigma} | c_{\sigma l}^{+} | \psi_i^{-\sigma} \rangle, \\ Y_{ji}^{\sigma\sigma l} &= \langle \psi_j^{\sigma\sigma} | c_{\sigma l}^{+} | \psi_i^{\sigma} \rangle, \\ Z_{mj}^{\sigma\sigma -\sigma l} &= \langle \psi_m^{\sigma\sigma -\sigma} | c_{\sigma l}^{+} | \psi_j^{\sigma -\sigma} \rangle, \\ Z_m^{\sigma -\sigma -\sigma l} &= \langle \psi_m^{\sigma -\sigma -\sigma} | c_{\sigma l}^{+} | \psi_j^{-\sigma -\sigma} \rangle, \\ W_m^{\sigma -\sigma -\sigma l} &= \langle \psi^{\sigma\sigma -\sigma -\sigma} | c_{\sigma l}^{+} | \psi_m^{\sigma -\sigma -\sigma} \rangle, \end{aligned} \qquad (3)$$

where ψ_i^{σ}, $\psi_j^{(\sigma\sigma)\sigma -\sigma}$, $\psi_m^{(\sigma -\sigma -\sigma)\sigma\sigma -\sigma}$, and $\psi^{\sigma\sigma -\sigma -\sigma}$ are wave functions for one, two, three, and four electrons in the system [33, 34].

Our approach leads to constraint on the possible physical system states: states with only one pseudo-particle are allowed have to be taken into account:

$$\widehat{n}_b + \sum_{i\sigma} \widehat{n}_{fi\sigma} + \sum_{j\sigma\sigma'} \widehat{n}_{dj}^{\sigma\sigma'} + \sum_{m\sigma} \widehat{n}_{\psi m\sigma} + \widehat{n}_{\varphi} = 1. \qquad (4)$$

Electron filling numbers in the two-level QD can be expressed in the terms of the pseudo-particles filling numbers:

$$\begin{aligned} \widehat{n}_{\sigma}^{el} = \sum_{l} c_{\sigma l}^{+} c_{\sigma l} = \sum_{i,l} |X_i^{\sigma l}|^2 \widehat{n}_{fi\sigma} + \sum_{i,j,l} |Y_{ji}^{\sigma -\sigma l}|^2 \widehat{n}_{dj}^{\sigma -\sigma} + \\ + \sum_{i,l} |Y_{ji}^{\sigma\sigma l}|^2 \widehat{n}_{dj}^{\sigma\sigma} + \sum_{m,j,l} |Z_{mj}^{\sigma\sigma -\sigma l}|^2 \widehat{n}_{\psi m-\sigma} + \\ + \sum_{m,l} |Z_{mj}^{-\sigma -\sigma\sigma l}|^2 \widehat{n}_{\psi m\sigma} + \sum_{m,l} |W_m^{\sigma -\sigma -\sigma l}|^2 \widehat{n}_{\varphi}. \end{aligned} \qquad (5)$$

In expression (5) the terms containing only one pseudo-particle filling number are present due to constraint (4). Consequently, the Hamiltonian of the system

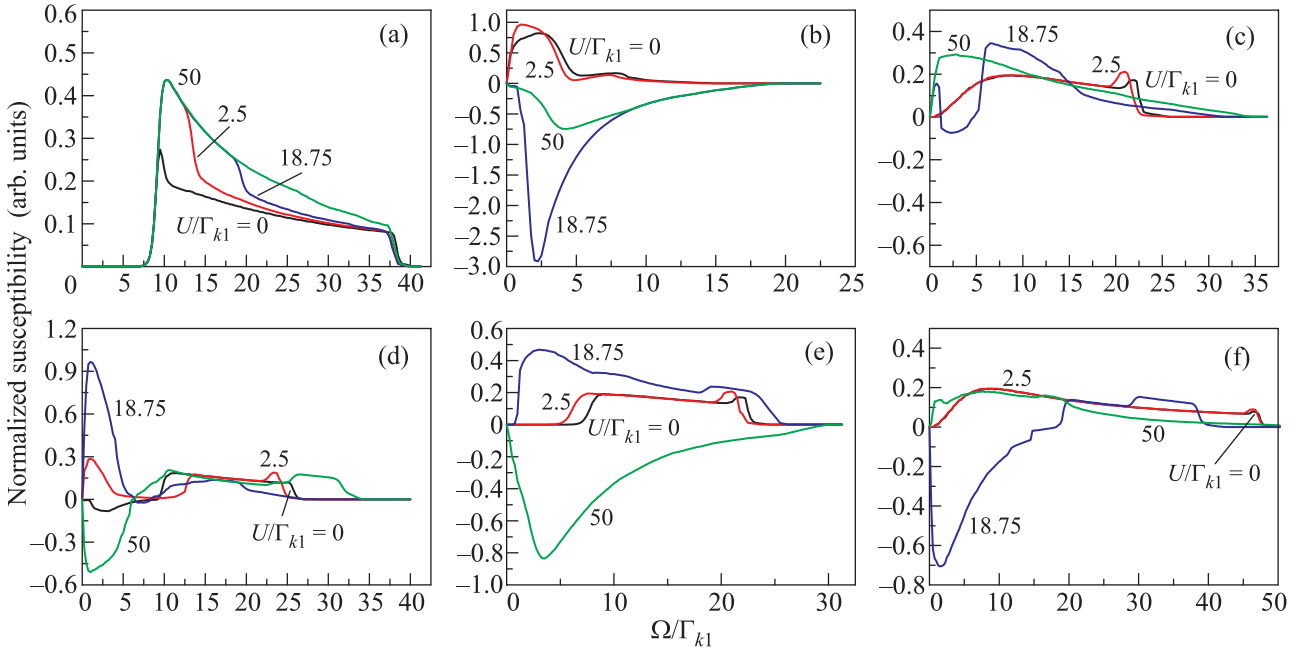

Fig. 4. (Color online) Normalized susceptibility $\mathrm{Im}(\chi)\Gamma_{k1}/U$ as a function of Rabi frequency for different values of Coulomb correlations, applied bias voltage and energy levels position: (a) – $eV/\Gamma_{k1} = 50$; (b) – $eV/\Gamma_{k1} = 3.75$; (c) – $eV/\Gamma_{k1} = -25$; (d) – $eV/\Gamma_{k1} = -37.5$; (e) – $eV/\Gamma_{k1} = -12.5$; (f) – $eV/\Gamma_{k1} = -50$; (a, d) – $\varepsilon_1/\Gamma_{k1} = 17.5$, $\varepsilon_2/\Gamma_{k1} = 5$; (d, e) – $\varepsilon_1/\Gamma_{k1} = -5$, $\varepsilon_2/\Gamma_{k1} = -17.5$; (c, f) – $\varepsilon_1/\Gamma_{k1} = 7.5$, $\varepsilon_2/\Gamma_{k1} = -5$. For all the figures parameters values $\Gamma_{k1} = \Gamma_{p2} = 1$, $\Gamma_{k2}/\Gamma_{k1} = \Gamma_{p1}/\Gamma_{k1} = 0.125$ are the same

can be re-written in the terms of the pseudo-particle operators:

$$\hat{H} = \hat{H}_0 + \hat{H}_{\mathrm{tun}},$$
$$\hat{H}_0 = \sum_{i\sigma} \varepsilon_i f^+_{i\sigma} f_{i\sigma} + \sum_{j\sigma\sigma'} E^{\sigma\sigma'}_{\mathrm{II}j}\, d^{+\sigma\sigma'}_j d^{\sigma\sigma'}_j + {}$$
$$+ \sum_{m\sigma} E^{m\sigma}_{\mathrm{III}} \psi^+_{m\sigma}\psi_{m\sigma} + E_{\mathrm{IV}}\varphi^+_\sigma\varphi_\sigma + {}$$
$$+ \sum_{k\sigma} (\varepsilon_{k\sigma} - eV) c^+_{k\sigma} c_{k\sigma} + \sum_{p\sigma} \varepsilon_{p\sigma} c^+_{p\sigma} c_{p\sigma}, \qquad (6)$$
$$\hat{H}_{\mathrm{tun}} = \sum_{k\sigma} T_{k1}(c^+_{k\sigma} c_{\sigma 1} + c^+_{\sigma 1} c_{k\sigma}) + {}$$
$$+ \sum_{p\sigma} T_{p1}(c^+_{p\sigma} c_{\sigma 1} + c^+_{\sigma 1} c_{p\sigma}) + (1 \leftrightarrow 2),$$

where ε_i, $E^{\sigma\sigma'}_{\mathrm{II}j}$, $E^{m\sigma}_{\mathrm{III}}$, and E_{IV} are the energies of the single-, double-, triple-, and quadri-electron states; $\varepsilon_{k(p)\sigma}$ is the energy of the conduction electrons in the states k and p correspondingly; $c^+_{k(p)1(2)\sigma}/c_{k(p)\sigma}$ are the creation (annihilation) operators in the leads of the tunneling contact; $T_{k(p)1(2)}$ are the tunneling amplitudes, which we assume to be independent on momentum and spin. Indexes $k(p)$ mean only that tunneling takes place from the QD to the continuous spectrum states with momentum k and p correspondingly. The tunneling coupling strength $\Gamma_{k(p)i}$ is defined as $\Gamma_{k(p)i} = \pi\nu_0 T^2_{k(p)i}$, where ν_0 is a constant density of states in the reservoir (which is not a function of energy). Depending on the tunneling barrier width and height typical tunneling coupling strength $\Gamma_{k(p)i}$ can vary from 10 μeV [35] to 1–5 meV [36]. Further we'll analyze the situation when each energy level is strongly coupled with only one of the tunneling contact leads. For energy level ε_1: $T_{k1} \gg T_{p1}$ and for energy level ε_2: $T_{p2} \gg T_{k2}$. In the proposed approach strongly interacting electrons in QD can now be described as non-interacting pseudo-particles. Interaction with the tunneling contact leads continue being weak, but becomes more complicated and non-linear.

Bilinear combinations of pseudo-particle operators are closely connected with the density matrix elements. So, similar expressions can be obtained from equations for the density matrix evolution but method based on the pseudo particle operators is more compact and convenient. The polarization $P/E_{\mathrm{dot}} \sim c^+_{1\sigma} c_{2\sigma}$, where $E_{\mathrm{dot}} \sim e/a^2$ is intradot electric field strength (e – electron charge and a – typical QDs size) and susceptibility $\mathrm{Im}\chi \sim \frac{U c^+_{1\sigma} c_{2\sigma}}{\Omega}$ ($U \sim e^2/a$ has typical value about 150 meV for 10 nm QDs) of the proposed system are directly determined by the following expression, written in the terms of pseudo-particle operators:

$$\sum_{\sigma} c_{1\sigma}^{+} c_{2\sigma} = \sum_{i} X_i^{\sigma 1} X_i^{\sigma 2 *} \widehat{n}_{fi\sigma} + \\ + \sum_{ij} Y_{ji}^{\sigma -\sigma 1} Y_{ji}^{\sigma -\sigma 2 *} \widehat{n}_{dj}^{\sigma -\sigma} + \sum_{ij} Y_{ji}^{\sigma\sigma 1} Y_{ji}^{\sigma\sigma 2 *} \widehat{n}_{dj}^{\sigma\sigma} + \\ + \sum_{mj} Z_{mj}^{\sigma\sigma -\sigma 1} Z_{mj}^{\sigma\sigma -\sigma 2 *} \widehat{n}_{\psi m -\sigma} + \\ + \sum_{m} W_{m}^{\sigma -\sigma -\sigma 1} W_{m}^{\sigma -\sigma -\sigma 2 *} \widehat{n}_{\varphi}. \qquad (7)$$

We set $\hbar = 1$. Stationary system of equations can be obtained for the pseudo particle filling numbers n_{fi}, $n_{dj}^{\sigma -\sigma}$, $n_{d}^{\sigma\sigma}$, $n_{\psi m}$, and n_{φ}, which determine the normalized susceptibility and polarization of the correlated QD [13, 33, 34]. We'll neglect the non-diagonal averages of pseudo-particle operators such as $\langle f_{\sigma}^{+} b f_{-\sigma}^{+} d \rangle$ etc. These terms are of the next order in small parameter $\Gamma_{k(p)}/\Delta\varepsilon$ where $\Delta\varepsilon$ is the energy difference between any energy states in the QD. We consider the paramagnetic situation, when conditions $n_{fi\sigma} = n_{fi-\sigma}$, $n_{\psi m\sigma} = n_{\psi m-\sigma}$, $n_{k\sigma} = n_{k-\sigma}$ and $n_{dj}^{-\sigma-\sigma} = n_{dj}^{\sigma\sigma}$ are fulfilled. Consequently, one must solve the linear system, which allows to determine pseudo particle filling numbers, electron filling numbers $n_{el}(eV)$ QDs normalized susceptibility and polarization as a functions of applied bias voltage, Coulomb correlations value, single-electron energy levels positions [13, 33, 34]. Obtained results are discussed in the next section.

3. Main results and discussion. Behavior of the single-electron energy levels filling numbers difference $n_1 - n_2$, QD normalized susceptibility $\mathrm{Im}(\chi)\Gamma_{k1}/U$ and normalized polarization P/E_{dot} with the changing of applied bias voltage and Rabi frequency both in the presence and in the absence of Coulomb correlations are depicted in the Figs. 1–4. We consider different experimental realizations: both single-electron energy levels are situated above the sample Fermi level (Figs. 1 and 4); both energy levels are localized below the sample Fermi level (Figs. 2 and 4); one of the energy levels is situated above and another one below the sample Fermi level (Figs. 3 and 4).

Let us start from the analysis of QD normalized polarization P/E_{dot} and electron filling numbers difference $n_1 - n_2$ (Figs. 1–3) as a functions of applied bias voltage.

When both single-electron energy levels are localized above the Fermi level and Coulomb interaction is absent (Fig. 1a), electrons filling numbers difference and QD polarization accept values with opposite signs. For the large negative values of applied bias voltage ($eV \ll 0$) electrons filling numbers difference is positive ($n_1 - n_2 > 0$), consequently inverse occupation takes place, and QD polarization is negative ($P/E_{\mathrm{dot}} < 0$). Electrons filling numbers difference and QD polarization rapidly change signs when $\varepsilon_a \le eV \le \varepsilon_s$, where $\varepsilon_a = \frac{\varepsilon_1+\varepsilon_2+\sqrt{(\varepsilon_1-\varepsilon_2)^2+4(\Omega/2)^2}}{2}$ and $\varepsilon_s = \frac{\varepsilon_1+\varepsilon_2-\sqrt{(\varepsilon_1-\varepsilon_2)^2+4(\Omega/2)^2}}{2}$. The presence of Coulomb correlations drastically change the system behavior (Figs. 1b–d). One can easily find that even in the case of weak Coulomb interaction ($U/\Gamma_{k1} = 2.50$ in the Fig. 1b) QD polarization adopts positive value simultaneously with the presence of inverse occupation in particular range of applied bias. This is the direct manifestation of Coulomb correlations role in the QD response. When Coulomb correlations are present polarization is not proportional to electrons filling numbers difference. Both of them can simultaneously adopt positive value. This effect becomes more pronounced in the case of strong Coulomb correlations ($U/\Gamma_{k1} = 18.75$, $U/\Gamma_{k1} = 50.00$ in the Figs. 1c and d).

We revealed another interesting effect in the case when both energy levels are situated below the Fermi level (Fig. 2). When Coulomb correlations are weak or even absent electrons filling numbers difference and QD polarization accept values with opposite signs or turn to zero for the same values of applied bias voltage (Figs. 2a and b). In the case of strong Coulomb correlations ($U/\Gamma_{k1} = 18.75$, $U/\Gamma_{k1} = 50.00$ in the Figs. 2c and d) electrons filling numbers difference and polarization of QD accept values with opposite signs for the large negative values of applied bias voltage ($eV \ll 0$). Electrons filling numbers difference is positive ($n_1 - n_2 > 0$) and consequently inverse occupation occurs and polarization is negative ($P/E_{\mathrm{dot}} < 0$). The increasing of applied bias voltage leads to the multiple switching of electrons filling numbers difference and QD polarization between positive and negative values. The direct manifestation of Coulomb correlations can be clearly seen in the Fig. 2c, when electrons filling numbers difference and QD polarization are both negative for the same value of applied bias voltage. Moreover, due to the Coulomb correlations, maximum negative value of QD polarization strongly exceeds maximum positive polarization value observed in the system.

In the case, when one of the energy levels is localized above and another one below the Fermi level and Coulomb interaction is absent or weak (Figs. 3a and b), QD demonstrates normal occupation and positive polarization for all the values of applied bias voltage. Strong Coulomb correlations ($U/\Gamma_{k1} = 18.75, U/\Gamma_{k1} = 50.00$ in the Figs. 3c and d) result in formation of inverse occupation for the large negative values of applied bias voltage. Simultaneously with the presence of inverse occupation, QD reveals negative polarization. With the increasing of applied bias voltage, sign changing of electrons filling

numbers difference and QD polarization occurs. Electrons filling numbers difference becomes negative and polarization becomes positive and vice verse.

Let us now analyze the behavior of QD normalized susceptibility ($\mathrm{Im}(\chi)\Gamma_{k1}/U$) as a function of Rabi frequency for different values of Coulomb correlations and applied bias voltage (Fig. 4). The general features of obtained results are non-monotonic behavior of susceptibility with the increasing of Coulomb correlations value and susceptibility switching between negative and positive values with the increasing of Rabi frequency in the case when strong Coulomb correlations are present. Non-monotonic behavior of susceptibility with the increasing of Coulomb correlations (Figs. 4b–f) is the result of the following effect: with the increasing of Coulomb correlations value energy level $\varepsilon_1 + U$ starts to exceed eV and energy level $\varepsilon_2 + U$ continue being lower than eV. Further increasing of Coulomb correlations value leads to the situation when both energy levels exceed the value of applied bias voltage.

We would like to stress that one can tune the QD susceptibility – change it's sign and amplitude by means of system parameters changing: Rabi frequency, Coulomb correlations value, applied bias voltage, and energy levels position.

4. Conclusion. Careful analysis of correlated two-level QD susceptibility, polarization and electron filling numbers difference in a wide range of applied bias voltage and Rabi frequency was performed in terms of pseudo-operators with constraint. It was demonstrated that susceptibility and polarization of correlated two-level QD weakly connected to the reservoirs are not determined only by the stationary electron filling numbers difference, but also depend on the high order Coulomb correlation functions for electrons localized in QD. We demonstrated that susceptibility and polarization can significantly increase and even change the sign depending on the applied bias voltage, Rabi frequency values and Coulomb correlations strength.

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