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Negative differential resistance in nanoscale transport in the Coulomb blockade regime

Prakash Parida, S Lakshmi¹ and Swapan K Pati

Theoretical Sciences Unit, Jawaharlal Nehru Centre For Advanced Scientific Research,
Jakkur Campus, Bangalore 560064, India

and

DST Unit on Nanoscience, Jakkur Campus, Bangalore 560064, India

E-mail: pati@jncasr.ac.in

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Abstract

Motivated by recent experiments, we have studied the transport behavior of coupled quantum dot systems in the Coulomb blockade regime using the master (rate) equation approach. We explore how electron–electron interactions in a donor–acceptor system, resembling weakly coupled quantum dots with varying charging energy, can modify the system’s response to an external bias, taking it from normal Coulomb blockade behavior to negative differential resistance (NDR) in the current–voltage characteristics.

The switching and negative differential resistance (NDR) behavior of nanoscale systems has gained a lot of interest in the last decade, owing to the potential applications in single-molecule electronics and has been observed in a variety of experimental systems, especially in the widely studied Tour molecules [1, 2]. There have been many theoretical studies to understand this phenomenon mainly through the one-electron picture [3–7]. There have also been a number of theoretical studies on donor–acceptor double quantum dot systems, where strong rectification has been observed [12], and others which showed NDR with variation in the dot–electrode coupling [8, 13, 14] or due to a detuning of the dot levels [13]. Another recent study has attempted to establish the conditions obeyed by the parameters involved, to find such a collapse in the current magnitude [15]. Some recent experiments on double quantum dots also showed an NDR feature [16, 17] and has rekindled interest in the phenomenon occurring in the low temperature weak-coupling limit. Theoretical studies of NDR in this single-electron charging limit is now gaining prominence and attracting a lot of research [9–11]. This regime, where mean-field descriptions usually fail, is one where electron charging energies are very high compared to the broadenings due to average coupling, and are particularly

important for small molecules which behave more like a quantum dot than a wire [18, 19]. Since mean-field methods combined with standard non-equilibrium Green’s function (NEGF) [20–23] treatment of transport is perturbative in the interaction parameter, it cannot capture the transitions between the spectrum of neutral and excited states, which can lead to a variety of interesting features in the current–voltage characteristics. The formalism that has now come to be used widely to capture molecular transport in the Coulomb blockade regime is the master or rate equation method [24].

In this paper, we use the above formalism to study a two-dot system consisting of a donor and an acceptor (see the schematic given in figure 1) in the Coulomb blockade regime. Taking our cue from our previous mean-field transport studies on two-level systems which showed interesting nonlinear behavior in their current–voltage characteristics [6], here we explore the role of strong correlations in affecting their transport behavior. This study becomes interesting, especially in the context of the difference in their low-lying excitations, which would play a very important role in their low-bias current–voltage characteristics. The rate equation formalism describes transport through a correlated system with many-body eigenstates. The presence of Coulomb interactions results in occupation probabilities of each many-body state that cannot be factorized as the product of the occupation probabilities of each single-electron level. Hence, in this case, the full

¹ Present address: Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, D-01062 Dresden, Germany.

rate equation problem, where the occupation probability of each many-body state is treated as an independent variable, is solved, neglecting off-diagonal coherences. In this method, the transition rate, $\Sigma_{s' \rightarrow s}$, from the many-body state s' to s , differing by one electron, is calculated up to linear order in Γ (which is the bare electron tunneling rate between the system and the electrodes), using Fermi's golden rule as [25]

$$\begin{aligned} \Sigma_{s' \rightarrow s}^{L+} &= \Gamma f_L(E_s - E_{s'}) \sum_{\sigma} |\langle s | C_{1\sigma}^{\dagger} | s' \rangle|^2 \\ \Sigma_{s' \rightarrow s}^{R+} &= \Gamma f_R(E_s - E_{s'}) \sum_{\sigma} |\langle s | C_{N\sigma}^{\dagger} | s' \rangle|^2 \end{aligned} \quad (1)$$

with a corresponding equation for $\Sigma_{s \rightarrow s'}^{L-}$ and $\Sigma_{s \rightarrow s'}^{R-}$ obtained by replacing $f_{L,R}(E_s - E_{s'})$ by $(1 - f_{L,R}(E_s - E_{s'}))$. Here, $+/-$ correspond to the creation/annihilation of an electron inside the dot due to electron movement from/to the left (L) or right (R) electrode. $C_{1\sigma}^{\dagger}$ and $C_{N\sigma}^{\dagger}$ are the creation operators for electrons of spin σ at the first and N th sites, respectively. We have also assumed that the creation and annihilation happen only at the terminal sites. The total transition rate is then obtained as $\Sigma_{s \rightarrow s'} = \Sigma_{s \rightarrow s'}^{L+} + \Sigma_{s \rightarrow s'}^{R+} + \Sigma_{s \rightarrow s'}^{L-} + \Sigma_{s \rightarrow s'}^{R-}$. The non-equilibrium probability P_s of occurrence of each many-body state s is obtained by solving the set of independent rate equations defined by $\dot{P}_s = \sum_{s'} (\Sigma_{s' \rightarrow s} P_{s'} - \Sigma_{s \rightarrow s'} P_s)$ through the stationarity condition $\dot{P}_s = 0$ at steady state. This results in a homogeneous set of equations of the size of the many-body space. Taking advantage of the normalization condition $\sum_s P_s = 1$, we obtain linear equations, which can be solved using well-known linear algebraic methods. The steady state probabilities are then used to obtain the terminal current as

$$I_{\alpha} = \frac{e}{\hbar} \sum_{s,s'} \Sigma_{s' \rightarrow s}^{\alpha+} P_{s'} - \Sigma_{s \rightarrow s'}^{\alpha-} P_s \quad (2)$$

where $\alpha = L/R$. Using the above prescribed method, we study a two-site system described by the Hamiltonian:

$$\begin{aligned} H &= \sum_{i=1}^2 (\epsilon_i - eW_g) a_i^{\dagger} a_i + \sum_{\sigma=\uparrow,\downarrow} -t (a_{1\sigma}^{\dagger} a_{2\sigma} + \text{h.c.}) \\ &+ U \sum_{i=1}^2 n_{i\uparrow} n_{i\downarrow} + V_{12} (n_1 - \bar{n})(n_2 - \bar{n}) \end{aligned} \quad (3)$$

where t is the hopping strength between the sites with the same spin (σ), $\epsilon_{1,2}$ are the on-site energies, U is the Hubbard interaction between electrons at the same site, V_{12} is the nearest-neighbor Coulomb interaction and W_g is the external gate bias. The average charge (\bar{n}) is assumed to be unity here [26]. \bar{n} actually gives a constant shift to the energy levels with a fixed number of electrons. For two sites with two electrons, the energy levels are negatively shifted by an amount V_{12} . Note that there exists two quantum phases in this model with the variation of interaction parameters. For the half-filled ground state, with zero on-site energies, while $U > V_{12}/2$ represents a spin density wave (SDW) phase, $U < V_{12}/2$ corresponds to the charge density (CDW) phase in the thermodynamic limit [27, 28]. However, in our case with two sites, while for $U < V_{12}$, the half-filled ground state gives higher preference to the state with two electrons of opposite

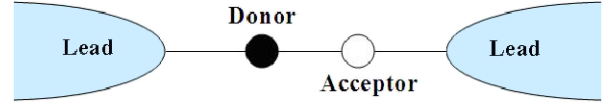


Figure 1. A schematic representation of a two-dot system consisting of a donor and an acceptor coupled to two electrodes.

spins at one site, for $U > V_{12}$ the state with one electron at each site is more preferred.

To study the transport properties through a double quantum dot system comprising of a donor and an acceptor in the weak-coupling regime, we parameterize the different coupling strengths in the total system (system + leads). For perturbation theory to be valid at temperature T , we ensure that $\Gamma \ll k_B T$. More specifically in our calculations, we use the value of $\Gamma = 0.25$ meV for $T = 300$ K and $\Gamma = 0.01$ meV for $T = 0.66$ K, which are also much smaller than the corresponding charging energies, e.g. Hubbard U . As our primary interest focuses on the NDR effect in the system, we choose the asymmetry in the on-site energy ($\Delta\epsilon = \epsilon_2 - \epsilon_1$) to be larger than the interdot hopping parameter (t), and vary the Hubbard U around $\Delta\epsilon$.

We adopt the well-known exact diagonalization (ED) method to solve the Hamiltonian in equation (3) for the system containing two sites. As the total number of electrons, N , and the z component of the total spin, S_z , commute with the Hamiltonian (H) and can be considered as conserved quantities, the H matrix can be diagonalized for a particular charge and spin sector. The Fock space can then be factored into many blocks, with the largest block consisting of four states with quantum numbers, number of electrons (N) = 2 and $S_z = 0$. The ground state energies for $N = 1$ (E_{1e}), $N = 2$ (E_{2e}) and $N = 3$ (E_{3e}) with on-site energies $\epsilon_1 = \epsilon_2 = 0$ can be easily found ($\bar{n} = 1$):

$$\begin{aligned} E_{1e} &= -t - W_g \\ E_{2e} &= \frac{U - V_{12}}{2} - \sqrt{\frac{(U - V_{12})^2}{4} + 4t^2} - 2W_g \\ E_{3e} &= U - t - 3W_g. \end{aligned} \quad (4)$$

Thus, the gate bias window ($\Delta W_g = E_{3e}^0 + E_{1e}^0 - 2E_{2e}^0$) over which the $N = 2$ (half-filled) state becomes the lowest energy state can be estimated to be

$$\Delta W_g = -2t + V_{12} + \sqrt{(U - V_{12})^2 + 16t^2} \quad (5)$$

which strongly depends on the parameters involved. E_{1e}^0 , E_{2e}^0 and E_{3e}^0 are the ground state energies for 1e, 2e and 3e states, respectively, in the absence of gate bias. However, with inclusion of asymmetric on-site energies, the general analytical expression for the energies assumes a complicated form. For chosen on-site energy values considering donor and acceptor sites, $\epsilon_2 = -\epsilon_1 = 2.0$ eV, we plot in figure 2 the number of electrons in the lowest energy state as a function of gate bias. This is obtained by calculating the many-body states with minimum energy at every value of gate bias W_g as $\text{Min}(E_s)$.

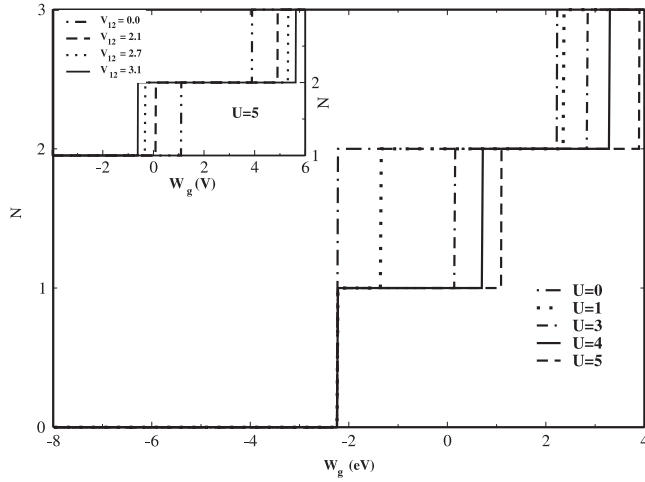


Figure 2. The number of electrons (N) in a two-site donor–acceptor system with variation of gate bias of the system (W_g) for various values of the Hubbard parameter U . The inset shows the same for $U = 5$ eV with different values of nearest-neighbor Coulomb interaction parameter, V_{12} . Here, $\epsilon_2 = -\epsilon_1 = 2.0$ eV, $t = 1.0$ eV and all the values of U and V_{12} are in eV ($\bar{n} = 1$).

For $U < 4$ eV, with the increase in on-site electron–electron interaction, there is a reduction in gate bias over which the two-electron state is stable, while, in contrast, for $U > 4$ eV, the gate bias window increases with the increase in on-site electron–electron interaction. Furthermore, for V_{12} values closer to or greater than $U/2$, the bias range over which the two-electron state is the lowest energy state increases with the increase in V_{12} . This happens because an increase in U by 1 eV causes no change to E_{1e}^0 , an increase of 1 eV to E_{3e}^0 , but an increase of more than 0.5 eV to E_{2e}^0 for $U < 4$ eV and an increase of less than 0.5 eV to E_{2e}^0 for $U > 4$ eV. This is due to the fact that, for $U < 4$ eV, the two-electron ground state gives higher preference to the state with two electrons of opposite spins at the site with lower on-site energy. However, for $U > 4$ eV, it prefers the state with one electron each at the donor and at the acceptor. Hence for $U < 4$ eV, an increase in U value by 1 eV causes an increase in the value of $2E_{2e}^0$ by more than 1 eV and an increase of 1 eV to E_{3e}^0 , so effectively reducing the value of ΔW_g . However, for $U > 4$ eV, the increase in the value of $2E_{2e}^0$ is always less than 1 eV and hence ΔW_g increases with the increase in U value.

For obtaining the current, for every value of U and V_{12} , the Fermi energy (E_F) is chosen as the value of the gate bias which ensures that the two-electron state is the ground state. The Fermi energy is also placed in such a way that we observe the transition from the ground state to the state with one less electron. After fixing the Fermi energy, we have studied the current as a response of source–drain bias (V) in all our calculations. In figure 3, we have plotted the I – V characteristics of the system for a range of U and V_{12} values at room temperature. As can be seen clearly, low values of U result in step-like features in I – V characteristics, while with an increase in U , a rise and fall in current (an NDR feature) is observed for positive values of source–drain bias. Interestingly, with inclusion of the nearest-neighbor Coulomb interaction, V_{12} , the I – V characteristics show a wide plateau region before

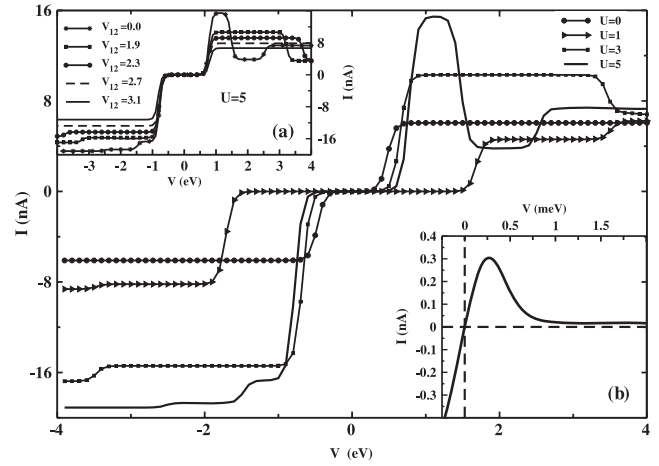


Figure 3. Current (I)–source–drain voltage (V) characteristics of the two-site donor–acceptor system for various values of U and the inset (a) represents the same for $U = 5$ eV with various V_{12} values for $\epsilon_2 = -\epsilon_1 = 2.0$ eV, $t = 1$ eV, $\Gamma = 0.25$ meV and temperature (T) = 300 K. The inset (b) shows the same for $\epsilon_2 = -\epsilon_1 = 2$ meV, $t = 0.2$ meV, $U = 4$ meV, $\Gamma = 0.01$ meV and temperature (T) = 0.66 K.

showing an NDR feature. However, the height of the NDR peak decreases in the positive source–drain bias region with an increase in V_{12} . To compare our results with the experimental findings at low temperature, in the inset (b) of figure 3, we have plotted the low temperature behavior of I – V characteristics. Note that the NDR peak together with the overall I – V feature compare fairly well with the experimental results obtained by Tarucha *et al* on GaAs-based double quantum dots [17]. We also note that there is, in fact, no qualitative change in the I – V characteristics except for a constant shift in bias, if we change the average dot charge ($\bar{n} = 1$) from one to zero in the Hamiltonian in equation (3).

The step-like feature in I – V is well understood in the literature as due to Coulomb repulsions [15, 29]. However, to understand the NDR feature, we analyze the probabilities of the occurrence of various many-body states. We find that the NDR occurs when the source–drain bias drives the system from the 1e doublet to a higher excitation of the 2e state, namely the triplet states, instead of to the state with zero electrons. It is because, when U is small, the ground state gives higher preference to the state with two electrons of opposite spins at the site with lower on-site energy. This allows annihilation of an electron by the electrode followed by one more annihilation leading to a transition from the 2e singlet to the 1e doublet and then to the state with zero electrons. However, when U increases, the ground state gives more weight to the state with one electron at the donor and one at the acceptor. This allows for one-electron annihilation from the ground state to the 1e doublet state, followed by a creation of an electron from the same electrode to the 2e triplet state, which has the same energy as the zero-electron state. Since the current at any electrode is calculated at steady state as the difference between the outgoing and incoming current, this transition results in a reduction in current, leading to the negative differential resistance (NDR) peak in the large U

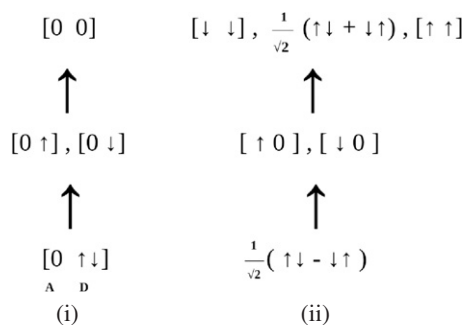


Figure 4. A schematic describing the transitions between the states of the donor–acceptor system: (i) in the small U regimes, (ii) in the large U regimes. The arrow direction indicates the states to which the transition occurs.

limit. A schematic figure describing the states involved with an increase in positive source–drain bias for small and large U limits are shown in figure 4. Note that, with inclusion of V_{12} , particularly for large V_{12} values, the charge density modulated state gets prominence, similar to the ground state electronic configuration as in the small U limit. Thus, with the increase in V_{12} , the NDR feature gets suppressed. Also, since with the inclusion of V_{12} , the gate bias range over which the 2e state remains the ground state differs, we pin the electrode’s Fermi energy in such a way that the transition from 2e singlet to 1e doublet state occurs at the same values of V (see the inset (a) of figure 3) for a range of V_{12} values. However, with an increase in positive bias, the electrochemical potential at the left electrode (μ_L) moves down and that at the right electrode (μ_R) moves up, causing the transport channel $\epsilon = E_T - E_{1e}$ to be in resonance with the levels of the electrodes, where E_T and E_{1e} are the energy levels associated with the 2e triplet state and 1e doublet state, respectively. With large V_{12} values, this channel width causes the plateau in I – V characteristics to be wider before showing NDR.

To understand the NDR feature more clearly, and to estimate the height of the peak value in the I – V plot in figure 3, we calculate the probability of occurrence of the 2e triplet state with the increase in V for a range of Hamiltonian parameters. In figure 5, we have plotted the variation of current and the occupation probability of the 2e triplet state with source–drain bias for $U = 5$ eV. It is clear that, when the occupation probability of the 2e triplet state starts increasing appreciably, the current decreases in magnitude, however, only to a nonzero value. The main point is that the triplet state being the blocking state suppresses the current: however, since its probability of occurrence does not increase more than 30%, there is still some finite current (leakage current) which flows through the system. Note that the I – V characteristics are asymmetric because of the inherent asymmetry in the system comprising of a donor and an acceptor with different site energy (ϵ) values.

In conclusion, we have studied transport behavior of the donor–acceptor system in the Coulomb blockade regime through the rate equation approach. Our study shows how the variation in the on-site Coulomb repulsions can influence the system’s response to an external source–drain bias. A strong Coulombic repulsion even results in NDR for positive values

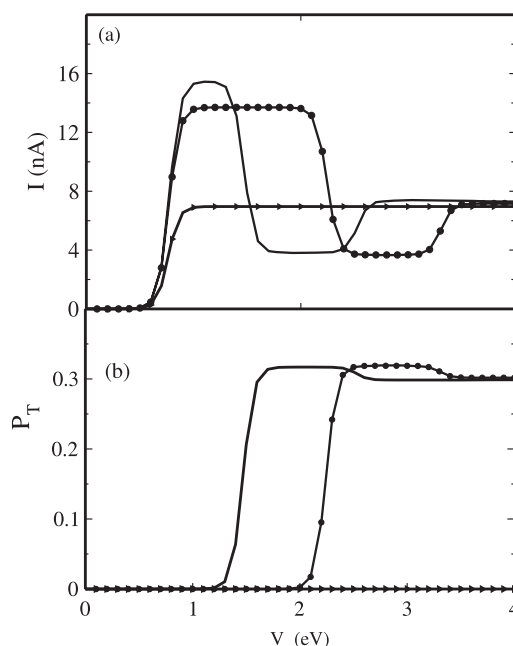


Figure 5. The variation of (a) current (I) and (b) occupation probability (P_T) of the 2e triplet state with source–drain bias, corresponding to $U = 5$ eV and varying V_{12} values: $V_{12} = 0$ eV (solid line), $V_{12} = 1$ eV (circle) and $V_{12} = 3$ eV (triangle).

of source–drain bias in the I – V characteristics. Also, a strong nearest-neighbor Coulomb interaction suppresses the NDR-like feature, taking the system back to the normal Coulomb staircase regime.

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