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Theory of current-voltage asymmetries in double quantum dots

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An explanation for the asymmetries in the current-voltage characteristics in mesoscopic quantum systems weakly coupled to reservoirs is proposed. Thus, it is suggested that inelastic scattering between the states in the interacting region dynamically redistributes the tunneling rate through the system. This redistribution functions non-uniformly for negative and positive bias voltages. Based on a diagrammatic technique for non-equilibrium many-body operator Green functions, an expression is presented that accounts for these scattering effects between the states. The theory is consistent with recent experimental data on mesoscopic systems.

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Current-voltage (J-V) characteristics of mesoscopic quantum systems often display a degree of asymmetry, with respect to the bias voltage.[1–3] This feature becomes particularly apparent in systems where the interacting region is asymmetrically coupled to the left and the right leads. Often the asymmetry is being referred to be an effect of impurities introduced during the growth process or differences in interface roughness between the oxide layers and the interacting region.[4,5] Another source that may introduce asymmetries in the J-V characteristics is unintentional background charges which additionally contribute charging energy to the interacting region.[5] A third argument is that a higher collector barrier enhances the charge storage in the well substantially, thus being responsible for different current amplitudes for the back and forward biased device.[6,7] Theoretically, it has been suggested that inelastic scattering, especially for asymmetric structures, gives different contributions in the back and forward bias direction.[8,9] However, a full understanding of the mechanisms responsible for the observed asymmetries in the J-V characteristics has not yet been put forward.

In this paper, a theoretical many-body model that explains the asymmetry of the J-V characteristics for mesoscopic systems with two or more conducting levels weakly coupled to reservoirs is proposed. Before stepping into the details of the theoretical modeling, the main result of this paper is presented, given in Fig. 1(a) (solid line). The plot displays the J-V characteristics of a double quantum dot (DQD) coupled to external contacts, illustrating the asymmetry of the current with respect the bias voltage. I suggest that this asymmetry arises due to a dynamical process, caused by a reduced probability of the tunneling between the states in the DQD. This prevents the electrons from flowing in the most favorable paths, which leads to a redistribution of the current probabilities, see Fig. 1(b) (solid line).

Although the suggested effect generally appears in mesoscopic quantum systems with two or more conducting levels, its presence is presented in one of the simplest systems, that is, two QDs electrostatically coupled in series and attached to external contacts. The QDs are interacting via an inter-dot Coulomb repulsion U_{AB} and hopping t. The DQD is coupled to external contacts through tunnel barriers, with coupling strength $\Gamma^{L/R}$ between the left/right (L/R) contact and $\mathrm{QD}_{A/B}$. The system is illustrated in Fig. 2. In $\mathrm{QD}_{A/B}$ there is a Coulomb repulsion $U_{A/B} > U_{AB}$. First consider the DQD being detached from the contacts. The energy of the DQD is modeled in Ref. 10, from which one easily finds the empty and the one-, two-, three-, and four-particles state. For the present purpose it is sufficient to work with transitions between the empty state $|0\rangle = |0\rangle_A |0\rangle_B$, at the energy $E_0 = 0$, and the two spin-degenerate one-particle states $|\gamma_{n\sigma}\rangle$, $n = 1,2$, given in Table I. The other states are omitted since they lie out of range of conduction for the parameters used here. This is further discussed in the following.

Now that the exact one-particle states are known, it is advantageous to write the system in diagonal form by introducing the many-body operators $X^{pq} = |p\rangle\langle q|$,[11] describing transitions between the states $|q\rangle$ and $|p\rangle$. Hence, the effective Hamiltonian for the isolated DQD is given by $\mathcal{H}_{\mathrm{DQD}} = \Sigma_{n\sigma} E_{\gamma_{n\sigma}} X^{\gamma_{n\sigma}\gamma_{n\sigma}}$. This description of the DQD is convenient when the contacts are attached to the system. However, it is important to note that the transition matrix elements of the DQD in general are different for the transitions $|\gamma_{1\sigma}\rangle \to |0\rangle$ and $|\gamma_{2\sigma}\rangle \to |0\rangle$. These matrix elements are given by $(a_{\sigma'})^{0\gamma_{n\sigma}} \equiv \langle 0|a_{\sigma'}|\gamma_{n\sigma}\rangle = \delta_{\sigma\sigma'} u^{\sigma}_{n1}$ and $(b_{\sigma'})^{0\gamma_{n\sigma}}$

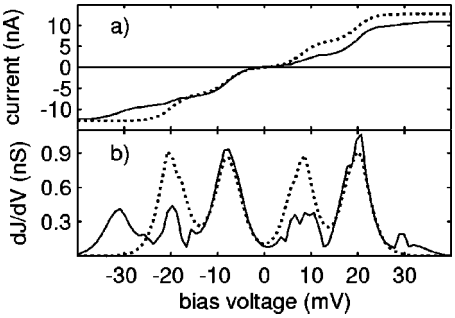

FIG. 1. (a) The current through the DQD calculated with (solid) and without (dotted) the dressed end-factors. (b) The corresponding differential conductances. For the computations the left/right coupling strength $\Gamma^{L/R} = 0.375$ meV and conduction electron band width 2 eV symmetrically around the equilibrium chemical potential $\mu = 0$ at $T = 10$ K were used.

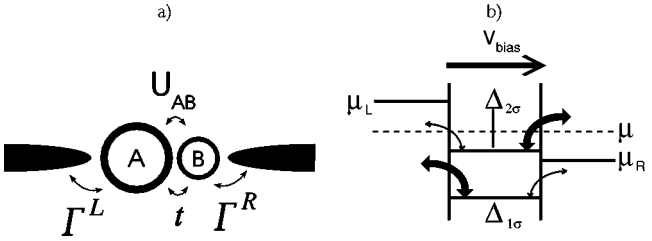

FIG. 2. (a) Schematic drawing of the DQD coupled to external contacts. The QDs interact via the inter-dot Coulomb repulsion $U_{AB}\sim 50$ meV and the hopping $t\sim 2.25$ meV. Each QD is attached to a contact with the coupling strength $\Gamma^{L/R}$. (b) Sketch of the system in the diagonal representation. The arrows signify the strengths of the transition probabilities between the one-particle states in the DQD and the contacts.

$\equiv\langle 0|b_{\sigma'}|\gamma_{n\sigma}\rangle=\delta_{\sigma\sigma'}u^{\sigma}_{n2}$, where a_σ/b_σ annihilates an electron in $\mathrm{QD}_{A/B}$. Hence, $|(a_\sigma)^{0\gamma_{1\sigma}}|^2\neq|(a_\sigma)^{0\gamma_{2\sigma}}|^2$, $|(b_\sigma)^{0\gamma_{1\sigma}}|^2\neq|(b_\sigma)^{0\gamma_{2\sigma}}|^2$ and $|(a_\sigma)^{0\gamma_{n\sigma}}|^2\neq|(b_\sigma)^{0\gamma_{n\sigma}}|^2$, $n=1,2$, whenever $\varepsilon_{A\sigma}\neq\varepsilon_{B\sigma}$, where $\varepsilon_{A\sigma/B\sigma}$ is the single-particle level in $QD_{A/B}$. This situation holds true for most realistic systems since the sizes of the two QDs in general are different. The difference of the transition matrix elements influences the resulting current. Furthermore, through the transition matrix elements one can control whether an electron escapes/enters the DQD to/from the left or the right contact.

The total system, through which the current flows, is now modeled by

$$\mathcal{H}=\sum_{k\sigma\in L,R}\varepsilon_{k\sigma}c^{\dagger}_{k\sigma}c_{k\sigma}+\sum_{n\sigma}E_{\gamma_{n\sigma}}X^{\gamma_{n\sigma}\gamma_{n\sigma}}+\sum_{nk\sigma}(v_{k\sigma}(d_\sigma)^{0\gamma_{n\sigma}}c^{\dagger}_{k\sigma}X^{0\gamma_{n\sigma}}+H.c.),\tag{1}$$

where $c^{\dagger}_{k\sigma}$ ($c_{k\sigma}$) creates (annihilates) an electron in the left/right contact at the energy $\varepsilon_{k\sigma}$, whereas $(d_\sigma)^{0\gamma_{n\sigma}}=(a_\sigma)^{0\gamma_{n\sigma}}$ or $(d_\sigma)^{0\gamma_{n\sigma}}=(b_\sigma)^{0\gamma_{n\sigma}}$ depending on whether the tunneling between the DQD and the contacts occurs to the left or the right, respectively. The current through the system is calculated by the formula[10,12]

$$J=-\frac{e}{2h}\mathrm{Im}\sum_{n\sigma}\int\{[\Gamma^L_{n\sigma}-\Gamma^R_{n\sigma}]G^<_{n\sigma}(\omega)+[f_L(\omega)\Gamma^L_{n\sigma}-f_R(\omega)\Gamma^R_{n\sigma}][G^r_{n\sigma}(\omega)-G^a_{n\sigma}(\omega)]\}d\omega,\tag{2}$$

where

TABLE I. The wave functions and energies of the two spin-degenerate one-particle states. Here $u^{\sigma}_{n1}=(E_{\gamma_{n\sigma}}-\varepsilon_{B\sigma})/\kappa_{1n}$, $|u^{\sigma}_{n2}|^2=1-|u^{\sigma}_{n1}|^2$, $\kappa_{1n}=\sqrt{(E_{\gamma_{n\sigma}}-\varepsilon_{B\sigma})^2+|t|^2}$, $n=1,2$, where $\varepsilon_{A\sigma/B\sigma}$ is a single-particle level in $\mathrm{QD}_{A/B}$.

$\lvert\gamma_{n\sigma}\rangle=u^{\sigma}_{n1}\lvert\sigma\rangle_A\lvert 0\rangle_B+u^{\sigma}_{n2}\lvert 0\rangle_A\lvert\sigma\rangle_B$
$E_{\gamma_{n\sigma}}=\frac{1}{2}(\varepsilon_{A\sigma}+\varepsilon_{B\sigma}+(-1)^n\sqrt{(\varepsilon_{A\sigma}-\varepsilon_{B\sigma})^2+4\lvert t\rvert^2})$

$$\Gamma^{L/R}_{n\sigma}=\Gamma^{L/R}|(d_\sigma)^{0\gamma_{n\sigma}}|^2,$$

$$\Gamma^{L/R}=\Gamma^{L/R}(\omega)=2\pi\Sigma_k|v_{k\sigma}|^2\delta(\omega-\varepsilon_{k\sigma}),$$

whereas

$$f_{L/R}(\omega)=f(\omega-\mu_{L/R})$$

is the Fermi function, and $\mu_{L/R}$ is the chemical potential for the left/right contact. In Eq. (2) the lesser, retarded and advanced forms of the Fourier transformed Green function (GF) $G_{n\sigma}(t,t')\equiv(-i)\langle \mathrm{T}X^{0\gamma_{n\sigma}}(t)X^{\gamma_{n\sigma}0}(t')\rangle_U$ appear.[13–15] The subscript U indicates that $G_{n\sigma}(t,t')$ is a non-equilibrium GF subjected to a set of time-dependent source fields $U_\xi(t)$ (not to be confused with the inter- and intra-dot Coulomb repulsions U_{AB} and $U_{A/B}$, respectively). These source fields are introduced in order to generate a diagrammatic expansion in terms of functional derivatives of the DQD GF with respect to $U_\xi(t)$. See Ref. 15 for further reference.

The DQD GF is analyzed by means of equations of motion and a diagrammatic technique for Hubbard operator GF.[15] The exact equations of motion for a single level spin-dependent system are given in Ref. 13. In this context, with two spin-degenerate states, the resulting equations of motion are analogous. The first diagram correction, the *loop correction*,[13] re-normalizes the energy for the transition $|0\rangle\rightarrow|\gamma_{n\sigma}\rangle$ according to $\Delta_{n\sigma}=\Delta^0_{n\sigma}+\delta\Delta_{n\bar{\sigma}}+\Sigma_{\sigma'=\uparrow,\downarrow;m\neq n}\delta\Delta_{m\sigma'}$, where

$$\delta\Delta_{m\sigma'}=\sum_{k\in L,R}|v_{k\sigma'}(d_{\sigma'})^{0\gamma_{m\sigma'}}|^2\frac{f(\varepsilon_{k\sigma'})-f(\Delta_{m\sigma'})}{\varepsilon_{k\sigma'}-\Delta_{m\sigma'}}.\tag{3}$$

Here $\Delta^0_{n\bar{\sigma}}$ is the bare transition energy and $n,m=1,2$, whereas $\bar{\sigma}$ is the opposite spin of σ. The transition energy $\Delta_{n\sigma}$ is renormalized by the other three possible transitions in the DQD. This renormalization arises due to kinematic interactions in the DQD induced by the presence of the contacts.

The renormalization of the transition energy is an important fact, although, it does not explain the degree of asymmetry of the J-V characteristics. This information, however, is contained in the corresponding non-equilibrium spectral weights. The DQD GF is constituted by the product $G_{n\sigma}(t,t')=D_{n\sigma}(t,t')P_{n\sigma}(t')$, where $D_{n\sigma}$ is the locator and $P_{n\sigma}$ is the end-factor, which carry the details of the poles and the spectral weights, respectively. The end-factor $P_{n\sigma}(t')\equiv\langle \mathrm{T}\{X^{0\gamma_{n\sigma}},X^{\gamma_{n\sigma}0}\}(t')\rangle_U$ is interpreted as the sum of the population numbers N_0 and $N_{n\sigma}$ for the states involved in the transition, e.g., $|0\rangle$ and $|\gamma_{n\sigma}\rangle$, respectively. When using the diagrammatic methods in Ref. 15, one has to expand both the locator, which leads to the transition energy shift given by Eq. (3), and the corresponding end-factor. The first diagram correction of $P_{n\sigma}$ generates the (Fourier transformed) dressed end-factor $\mathbb{P}_{n\sigma}(i\omega)=P_{n\sigma}+\delta P_{nn\bar{\sigma}\sigma}(i\omega)+\Sigma_{\sigma',m\neq n}\delta P_{mn\sigma'\sigma}(i\omega)$, where

$$\delta P_{mn\sigma'\sigma}(i\omega) = -\frac{P_{m\sigma'} - P_{n\sigma}}{2\pi} \int \sum_{k\in L,R} \frac{|v_{k\sigma'}(d_{\sigma'})^{0\,\gamma_{m\sigma'}}|^2}{\varepsilon_{k\sigma'} - \omega'} \times [-2\,\mathrm{Im}\, D^r_{m\sigma'}(\omega')] \times \left(\frac{f(\varepsilon_{k\sigma'}) - [n_B(\Delta_{nm\sigma\sigma'}) + 1]}{i\omega - \Delta_{nm\sigma\sigma'} - \varepsilon_{k\sigma'}} - \frac{f(\omega') - [n_B(\Delta_{nm\sigma\sigma'} + 1]}{i\omega - \Delta_{nm\sigma\sigma'} - \omega'} \right) d\omega', \quad (4)$$

and $P_{n\sigma}$ is the bare end-factor. Here, $D^r_{m\sigma'}(\omega')$ is the retarded locator for the transition $|0\rangle \rightarrow |\gamma_{m\sigma'}\rangle$, $n_B(x)$ is the Bose function, whereas $\Delta_{nm\sigma\sigma'} = \Delta_{n\sigma} - \Delta_{m\sigma'}$ is the energy for the tunneling between the states $|\gamma_{m\sigma'}\rangle$ and $|\gamma_{n\sigma}\rangle$. The dressed end-factor clearly is frequency and bias voltage dependent and with a finite imaginary part. More important, though, this function exhibits attributes from inelastic scattering processes between the two one-particle states.

In the present approximation, the equation for the DQD GF, $G_{n\sigma}(i\omega)$, can be written as $(U_{\xi}(t) \rightarrow 0)$

$$(i\omega - \Delta_{n\sigma} - \mathbb{P}_{n\sigma}(i\omega) V_{n\sigma}(i\omega)) G_{n\sigma}(i\omega) = \mathbb{P}_{n\sigma}(i\omega), \quad (5)$$

where $V_{n\sigma}(i\omega) = \Sigma_{k\in L,R} |v_{k\sigma}(d_{\sigma})^{0\,\gamma_{n\sigma}}|^2/(i\omega - \varepsilon_{k\sigma})$. Although there is no simple definition of the locator $D_{n\sigma}(i\omega)$ in terms of the Hubbard operators,[15] one may note that its inverse, $D^{-1}_{n\sigma}(i\omega)$, is identified with the expression in brackets on the left-hand side of Eq. (5) in the given approximation. Thus, in order to accurately account for the voltage-dependent effects introduced in the dressed transition energies and end-factors, one has to perform self-consistent calculations of Eq. (3) for each bias voltage. The result is then inserted into a self-consistent loop constituted of Eqs. (4), (5) and the population numbers[16] $N_{n\sigma} = \mathrm{Im} \int G^{<}_{n\sigma}(\omega) d\omega/(2\pi)$ subject to the boundary condition $1 = N_0 + \Sigma_{n\sigma} N_{n\sigma}$. Hence, the obtained result goes far beyond any mean field theory or master equation approach.

In order to understand the asymmetry of the output current one first needs to study the behavior of the system without the dressing of the end-factors. Then, the current can only flow directly through each state, that is *left contact* $\rightarrow |\gamma_{n\sigma}\rangle \rightarrow$ *right contact*. Therefore, for symmetrically coupled systems ($\Gamma^L = \Gamma^R$) the current increases in equal steps as more and more states become resonant, see Fig. 1(a) (dotted), since the *sum* of the transition probabilities to the left and the right contacts is equal for both states. Hence, the *J-V* characteristics become totally symmetric, which is reflected in the corresponding differential conductance dJ/dV, Fig. 1(b) (dotted). An asymmetric coupling ($\Gamma^L \neq \Gamma^R$) will slightly modify this picture, resulting in a non-uniform increase of the current for each state that becomes resonant. However, the effects from this asymmetry are only marginal and cannot explain the large degree of asymmetry observed in experimental *J-V* data.

The dynamical (voltage-dependent) dressing of the end-factors ruins this scenario quite substantially, since it contains information of inelastic scattering between the two one-particle states in the DQD. For instance, consider negative biases and the correction $\delta P_{21\sigma\sigma}$ to $\mathbb{P}_{1\sigma}$, cf. Eq. (4). In equilibrium, the most favorable current path is *right contact* $\rightarrow |\gamma_{2\sigma}\rangle \rightarrow |\gamma_{1\sigma}\rangle \rightarrow$ *left contact*. The first term of $\delta P_{21\sigma\sigma}$ is peaked around $\Delta_{1\sigma}$ and logarithmically diverging around $\Delta_{12\sigma\sigma} + \mu_L$. However, the importance of this correction becomes significant for bias voltages such that $\mu_L \approx \Delta_{1\sigma}$, since then both the transitions corresponding to the energies $\Delta_{1\sigma}$ and $\Delta_{12\sigma\sigma}$ are resonant.[17] This results in that the population number $N_1 = \Sigma_{\sigma} N_{1\sigma}$ decreases as an effect of the scattering processes between the two one-particle states included into $\delta P_{21\sigma\sigma}$, hence, the transition $|0\rangle\langle\gamma_{1\sigma}|$ becomes less available for conduction of electrons through the DQD. Thus, for negative bias voltages the current through the path *right contact* $\rightarrow |\gamma_{2\sigma}\rangle \rightarrow |\gamma_{1\sigma}\rangle \rightarrow$ *left contact* is decreased, leading to a flattening of the current step as the state $|\gamma_{1\sigma}\rangle$ becomes resonant, see Fig. 1(a) (solid). The same argument holds for the correction $\delta P_{12\sigma\sigma}$ to $\mathbb{P}_{2\sigma}$. However, since only the state $|\gamma_{2\sigma}\rangle$ is resonant for voltages such that $\mu_L \approx \Delta_{2\sigma}$, the effects from the redistribution of the transition probabilities become negligible.

For positive voltages, the current path *left contact* $\rightarrow |\gamma_{1\sigma}\rangle \rightarrow |\gamma_{2\sigma}\rangle \rightarrow$ *right contact* is the most favorable. This fact leads to the current being small until $|\gamma_{1\sigma}\rangle$ becomes resonant. At voltages such that $|\gamma_{2\sigma}\rangle$ becomes resonant, the current increases in a small step, see Fig. 1 (solid), which is related to a decreased tunneling probability through the DQD via this state due to the scattering effects between the one-particle states.

The above arguments hold both for symmetrically ($\Gamma^L = \Gamma^R$) and asymmetrically ($\Gamma^L \neq \Gamma^R$) coupled systems. However, depending on whether the coupling to the left is stronger or weaker than that to the right, the asymmetry in the resulting *J-V* characteristics becomes either amplified or reduced, respectively.

It remains to discuss the influence of transitions to the states with two or more electrons in the DQD. The two-particle states in the system consist of a triplet at the energy $E_{\mathrm{triplet}} = \varepsilon_{A\sigma} + \varepsilon_{B\sigma} + U_{AB}$ and three non-degenerate states of which the lowest lies around $(E_{\mathrm{trip}} - |t|^2/10)/2$, since $t \ll |U_{AB} - U_A|, |U_{AB} - U_B|$. The energy required for an additional electron to enter the DQD is, thus, in the order of E_{triplet}. Therefore, by restricting the bias voltage applied to the system to be less than this energy in the present example, states with two or more electrons can safely be omitted.

It should be noted that no discussion about Kondo physics has been included into the present case. However, as was discussed by Lacroix[18] such effects are of main importance for temperatures below the Kondo temperature, which is not the case here. Second, possible (small) contributions from the Kondo effect cannot by themselves introduce the large degree of asymmetry often observed in experiments, since any Kondo resonance is suppressed by the application of a bias voltage.[19] Nevertheless, for low fields such contributions may slightly amplify the asymmetry introduced by the mechanism proposed in this paper.

In conclusion, a theoretical explanation is proposed for the often observed asymmetries in mesoscopic quantum systems with two or more conducting levels weakly coupled to

reservoirs. The theory suggests that the asymmetries arise due to a dynamical process, caused by a reduction of the most favorable tunneling between different states in the interacting region. In order to theoretically understand this process one has to perform highly accurate calculations, involving loop corrections to the end-factors of the QD GF.

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[17] Here, a *resonant transition* refers to that the corresponding transition energy lies in the interval between μ_L and μ_R.

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