Nonlinear cotunneling through an artificial molecule

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We study electron transport through a system of two lateral quantum dots coupled in series. We consider the case of weak coupling to the leads and a bias point in the Coulomb blockade. After a generalized Schrieffer-Wolff transformation, cotunneling through this system is described using methods from lowest-order perturbation theory. We study the system for arbitrary bias voltages below the Coulomb energy. We observe a rich, non-monotonic behaviour of the stationary current depending on the internal degrees of freedom. In particular, it turns out that at fixed transport voltage, the current through the system is largest at weak-to-intermediate inter-dot coupling.

Quantum dots are prototype systems for studying the properties of discrete levels embedded in a solid-state environment. Single dots (“artificial atoms”) can be coupled through quantum point contacts, leading to “artificial molecules”. Indeed it has been shown experimentally, that the eigenstates of double dot systems are coherent molecular superpositions of single dot (atomic) states. Unlike real molecules, these dots are readily contacted and tunable in situ, making them a natural test-bed for molecular transport. Double dots have also been proposed as charge quantum bits. This raises the question, which information on the energy spectrum and the wavefunctions of the dot can be probed by transport measurements. This is only possible if artifacts induced by the coupling to the leads can be sorted out and the double-dot is disturbed as little as possible. This is the case, when the coupling to the outside leads is weak (see Fig. 1) and the gates are tuned to the Coulomb blockade regime. In that regime, only states with a fixed number of electrons are energetically permissible and hence sequential tunneling is suppressed. The leading transport mechanism in this case is cotunneling, the coherent transfer of two electrons via virtual levels in the dots. Our work stands between studies focusing on sequential tunneling and work on linear response in the Kondo regime. The properties of cotunneling currents as a spectroscopic tool for the spectrum of quantum dot system have recently been studied in exquisitely controlled experiments on systems similar to ours.

In this paper, we analyze a serial configuration of lateral quantum dots in the cotunneling regime. We study finite voltages up to the order of the charging energy i.e. do not restrict ourselves to linear response. We find a rich nonmonotonic structure in the current as a function of the dot parameters. In particular, we find a pronounced crossover indicating the opening of an inelastic transport channel, which leads to the surprising result, that a too strong interdot coupling actually inhibits charge transport. We analyze the influence of the asymmetry of the dots on the current.

In the Coulomb blockade regime, the relevant Hilbert space is spanned by four basis states $|i,j\rangle$, $i, j \in \{0,1\}$, which denotes $i$ and $j$ additional electrons (as compared to an appropriate neutral state) on the left and right dot respectively. We study the situation where the gate voltages of the single dots are very close to each other and the inter-dot coupling is, although appreciable, much smaller than the single dot addition energy. Thus, the subspace spanned by the two states $|1,0\rangle$ and $|0,1\rangle$ is energetically most favorable. The next closest states $|v_0\rangle = |0,0\rangle$ and $|v_2\rangle = |1,1\rangle$ are outside the transport window and serve as virtual states. States with higher dipolar moment are energetically even less favorable due to the high charging energy of the individual dots.

The Hamiltonian of this system can be written as

\[ H = H_0 + H_1 \]
\[ H_0 = H_{\text{sys}} + H_{\text{res}} \]
\[ H_{\text{sys}} = \epsilon_{as}(n_l - n_r) - \epsilon_{\alpha} n_v0 + \epsilon_{\beta} n_v2 + \gamma \sum n \left( a_n^{L\dagger} a_n^{R} + a_n^{R\dagger} a_n^{L} \right) \]
\[ H_{\text{res}} = \sum_{\vec{k}} \epsilon_{\vec{k}} b_{\vec{k}}^{L\dagger} b_{\vec{k}}^{L} + \sum_{\vec{k}'} \epsilon_{\vec{k}'} b_{\vec{k}'}^{R\dagger} b_{\vec{k}'}^{R} \]
\[ H_1 = t_c \sum_{\vec{k},n} \left( a_n^{L\dagger} b_{\vec{k}}^{L} + a_n^{L} b_{\vec{k}}^{L\dagger} \right) + t_c \sum_{\vec{k}',n} \left( a_n^{R\dagger} b_{\vec{k}'}^{R} + a_n^{R} b_{\vec{k}'}^{R\dagger} \right). \]

Note, that the sum over dot states $n$ only runs over the restricted Hilbert space described above. $H_0$ describes the isolated double-dot ($H_{\text{sys}}$) and the leads ($H_{\text{res}}$), whereas the tunneling part $H_1$ describes the coupling of each dot to its lead and will be treated as a perturbation. $n_{vL/R}$ are the number operators counting additional electrons on either dot. The asymmetry energy $\epsilon_{as} = (\epsilon_L - \epsilon_R)/2$ describes half of the difference between the energy level for the additional electron in left dot ($\epsilon_L$) and the corresponding energy level in the right dot ($\epsilon_R$), which can be tuned through the gate voltages. $\gamma$ is the tunable inter-dot coupling strength. The $a_{\alpha}^{L/R}b_{\vec{k}}^{L/R}$ denote electron creation/annihilation operators in the dots and leads. In
The stationary density matrix is found using the well-established and controlled Bloch-Redfield theory\(\textsuperscript{15}\). This is a systematic technique for deriving generalized master equations within Born approximation in \(\hat{H}_I\), eq. \(\text{(6)}\), which includes all relevant non-markovian parts. This approach has been shown\(\textsuperscript{15}\) to be numerically equivalent to formally exact path integral methods for the Spin-Boson model in the weak-coupling limit. The Redfield equations\(\textsuperscript{16}\) for the elements of the reduced density matrix \(\rho\) in the molecular basis read

\[
\dot{\rho}_{nm}(t) = -i\omega_{nm}(t)\rho_{nm}(t) - \sum_{k,l} R_{nmkl}\rho_{kl}(t),
\]

where \(\omega_{nm} = (E_n - E_m)/\hbar\) are the appropriate energy splittings and \(R_{nmkl}\) are the elements of the Redfield tensor. They are composed out of golden rule rates involving \(\hat{H}_I\) from eq. \(\text{(6)}\). \(n, m, k\) and \(l\) can be either + (molecular excited state) or − (molecular ground state). The \(E_s\) are the eigenenergies of the two molecular states. Due to the lack of symmetry, this leads to a huge number of processes contributing to each term\(\textsuperscript{18}\). We are only interested in stationary solutions here. A full treatment of the simple case with \(\gamma = 0\) can be found in Ref.\(\textsuperscript{17}\).

The current is derived from the standard formula\(\textsuperscript{10}\)

\[
I(t) = -\frac{e}{\hbar} \int_{-\infty}^{t} dt' \langle [\hat{N}_L(t'), \hat{H}_I(t')] \rangle,
\]

where \(\hat{N}_L\) is the particle number operator on the left dot in the interaction representation and the transformed interaction Hamiltonian \(\hat{H}_I\) from eq. \(\text{(6)}\) is also taken in the interaction picture. Carrying out the integration in equation \(\text{(8)}\) and rotating back to the Schrödinger picture, we get a time-independent expression for the current \(I\). Using the stationary occupation probability of the molecular ground (\(\rho_{-,-}\)) or excited state (\(\rho_{+,-}\)), we obtain for the expectation value of the stationary current

\[
I_{st} = \text{tr}(\rho_{st} I) = \rho_{+++} I_{++} + \rho_{-,-} I_{--},
\]

where we find from balancing relaxation processes in the Bloch-Redfield equation, eq. \(\text{(7)}\)

\[
\rho_{++,-} = \frac{R_{++,-}}{R_{+++} - R_{ +++}} , \quad \rho_{-,-} = \frac{R_{-,-}}{R_{-,-} - R_{--}} .
\]

The current amplitudes \(I_{++}\) and \(I_{--}\) in eq. \(\text{(8)}\) are of the same form as the contributions to the Redfield tensor. We emphasize, that the choice of processes from all possibilities is very distinct. As an example, Fig. \(\text{2}\) displays a variety of possible processes in such a double-dot system. Processes of the type displayed in Fig. \(\text{2}\) (a) contribute to the relaxation but do not carry current, (b) shows a process which carries current but does not relax the state, and (c) relaxes and carries current. The phase information of the quantum state is lost in all three pictures of Fig. \(\text{2}\) Consequently, one must not confuse cotunneling rates with relaxation rates.
We now turn to the discussion of the results. All internal energies $\varepsilon_{\text{as}}$ and $\gamma$ are normalized in units of the bias voltage $V$, the stationary current $I_{\text{st}}$ in terms of $I_0 = e\Gamma$.

In Fig. 2 the current at fixed bias voltage as a function of the inter-dot coupling is shown. The sign of $\varepsilon_{\text{as}}$ plays a role, as one can see above, for an intermediate $\varepsilon_{\text{as}}$-regime. This effect is more pronounced in $I(V)$, see Ref. 19. Close to $\gamma = 0$, the curves all turn to zero, because at that point the dots are disconnected and no current can flow. However, a number of curves, the ones with $2\varepsilon_{\text{as}}/V < 1$, exhibit an intermediate maximum at low $\gamma$ next to a very sharp minimum at $\gamma = 0$, which sometimes is hardly resolved. At high $\gamma \gg V$, the stationary current saturates into a value, which for our parameters turns out to be about $I_{0,\text{st}}/I_0 = 7.5 \cdot 10^{-7}$. Remarkably, this is half the value of the current at the aforementioned low-$\gamma$-Maximum. This is the central result of this paper.

These regimes can be classified in terms of the level splitting $2\varepsilon_{\text{as}}$. At $V < 2\delta$, the energy $V$ supplied from the leads is only sufficient to use one of the molecular states for transport (elastic cotunneling) whereas at $V > 2\delta$, both states participate and also inelastic processes contribute, i.e. there is a second current channel, which carries the same contribution of $I_0$. The crossover naturally occurs at $\gamma = \sqrt{V^2 - \varepsilon_{\text{as}}^2}$, which can only be reached if $\varepsilon_{\text{as}}/V < 1/2$. As long as $\gamma$ is not too low, the coupling to the leads is the limiting element for the current flow. Only if $\gamma < \varepsilon_{\text{as}}$, the double-dot eigenstates become localized and the inter-dot coupling becomes the current bottleneck. Consequently, associated dips have a half-width of $\varepsilon_{\text{as}}$ for low temperatures and bias voltages and can thus be extremely narrow. We would like to remark that the notion of transport “channels” is appropriate here, because cotunneling is a coherent transport process.

Fig. 4 shows the dependence of the stationary current on $\varepsilon_{\text{as}}/V$. It confirms the interpretation of Fig. 3. The plot is only weakly asymmetric to $\varepsilon_{\text{as}}/V = 0$. At zero asymmetry, $\varepsilon_{\text{as}}/V = 0$, the condition for charge transport is ideal, $\sqrt{V^2 - \varepsilon_{\text{as}}^2}$ has its maximum and therefore the current is only governed by the inter-dot coupling $\gamma/V$, resulting in a zero-asymmetry maximum.

Still, all three transport regimes can be recognized in Fig. 4. The $\gamma/V = 0$ curve shows, that the stationary current $I_{\text{st}}/I_0$ is exactly zero as expected. For growing, but small values of $\gamma/V$, the maximum at $\varepsilon_{\text{as}}/V = 0$ reaches the highest value $I_{\text{st}}/I_0 = 2I_{0,\text{st}}/I_0$ at about $1.5 \cdot 10^{-6}$ (like in Fig. 3), corresponding to two open transport channels (elastic and inelastic). If we raise $\gamma/V$ further, the height of the peak goes down again and saturates at $I_{\text{st}}/I_0 = I_{0,\text{st}}/I_0 \approx 7.5 \cdot 10^{-7}$, corresponding to only the elastic channel being open.

The three transport regimes are summarized in Fig. 5: i) the atomic limit (no transport) $\gamma < \varepsilon_{\text{as}}$; ii) the two-channel case (inelastic cotunneling) $\varepsilon_{\text{as}} < \gamma < \sqrt{V^2 - \varepsilon_{\text{as}}^2}$; and iii) the one-channel case (elastic cotunneling) $\gamma > \sqrt{V^2 - \varepsilon_{\text{as}}^2}$.

These conditions show that indeed cotunneling can be used as a tool for investigating the energy spectrum of an undisturbed artificial molecule. The cross-over between the elastic and the inelastic cotunneling in dependence of the applied bias voltage has recently been observed. A similar conclusion was found in Ref. 20.

Although the notion of (elastic and inelastic) cotunneling was already introduced very early, its consequences for realistic quantum dot systems have only been discussed very recently, along with detailed and accurate experiments on small semiconductor quantum dots.
FIG. 5: Limits for the three transport regimes with $V = 5.170 \mu V$

becoming available. The sharp crossover between elastic and inelastic cotunneling, which we discuss, has been identified in a vertical quantum dot by changing the transport voltage. Ref. 4 studies cotunneling in a parallel double-dot topology, using again cotunneling and the elastic-to-inelastic crossover as a spectroscopic tool and tuning the inter-dot coupling in situ. In both cases, the narrow regime of decoupled dots would not have been accessible through a conductance measurement. Some of the experimental issues have been theoretically addressed in Ref. 20. In that case, however, the behavior of a single multilevel dot system was modelled with phenomenological couplings to the leads, whereas we take a realistic model and only by this manage to predict effects which e.g. depend on the serial dot topology of the sample. Note, that parts of the double-dot literature focus on phonon/photon assisted transport (see e.g. Refs. 21,22 for experiments and Refs. 23,24 for theory). Unlike Ref. 25 we concentrate on the Coulomb blockade regime and do not consider cotunneling at resonance. In Ref. 26 a different approach to the problem was developed, in which the master equation is carried to second order instead of using a Schrieffer-Wolff transformation, and a few setups simpler than ours are studied. Our approach does not require the molecule to be artificial, in principle it can be applied to “real” molecules. In contrast to the approach in Ref. 25 it permits to take into account charging effects, however, the Schrieffer-Wolff transformation is clearly a laborious step for larger systems.

To conclude, we analyzed the stationary coherent cotunneling current $I_{st}$ through a double quantum dot system or artificial molecule. As a function of the inter-dot coupling strength it displays a rich, non-monotonic structure, which enables us to perform “molecular cotunneling spectroscopy”. Strikingly, we have shown that at fixed bias voltage, the current is highest, if the dots are weakly-to-intermediately connected, such that the inter-dot coupling is at least as strong as the coupling to the leads, but the splitting of the molecular wavefunctions is still smaller than the transport voltage.

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