Transient current in spin blockade condition

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Abstract. In series double quantum dot with finite bias, strong current suppression by spin-selection rule is expected. Before the current becomes zero starting from a certain initial condition, the transient current flows through the system. We theoretically argue this excess current, under which condition, becomes a simple fraction of charge quantum after being integrated in time. We also propose a scheme to detect this excess current by periodically modulating bias and present the evidence of the fractional excess charge \(e/3\).

1. Introduction

Spin blockade in the linear transport through a quantum dot (QD) had been proposed by Weinmann et al.[1] but had yet not been confirmed experimentally.[2] If one prepares series quantum dots (QDs) with different g-factors or different but parallel local magnetic fields, we may realize a spin filter in low-bias transport by aligning the energy levels of one of the Zeeman sub-levels in both QDs by gate or bias voltage. However, when one applies large source-drain bias, the spin polarized current flows only transiently. This is because there is a finite probability of occupation of an electron with wrong spin (dark state), which will block the flow of the electrons of correct spin (bright state) by Coulomb blockade effect as shown in Fig.1.[3, 4] Before the flow stops, we expect small but finite transient charge transfer, \(\hat{Q}\), whose expectation value is

\[
\langle \hat{Q} \rangle = \sum_{N=1}^{\infty} e P_N = e
\]

where \(N\) is the number of the transfer processes with the probability

\[
P_N = \left(\frac{1}{2}\right)^N
\]

as shown in Fig.1 and \(p = \frac{1}{2}\) is the probability that a correct spin is fed to the QDs. Similar spin blocking function had been known for two electron regime in a series QDs where succeeding current flow is blocked when the spin triplet states (dark states) are realized (Pauli spin blockade).[5] As before, we expect finite charge transfer before the blockade, which is

\[
\langle \hat{Q} \rangle = \sum_{n=1}^{\infty} e \left(\frac{1}{4}\right)^n = \frac{e}{3}
\]

where \(p = \frac{1}{2}\) is the probability that the spin singlet state (bright state) is established.[6] It is interesting to notice that the total transient charge \(\langle \hat{Q} \rangle\) before the spin blockade completes is a simple fraction of the charge quantum.

The motivation of this work is to clarify the condition when above naive arguments are justified and we discuss in detail this transient charge \(\langle \hat{Q} \rangle\) using the master equation. Then we argue the experimental setup to observe it using pump-and-probe method and provide an evidence of the fractional transient charge.

2. Single electron spin blockade

The Hamiltonian of the series double QDs is

\[
H_{QD} = \sum_{\sigma,L,R,\nu} \epsilon_{\nu\sigma} |\nu\sigma\rangle \langle \nu\sigma | - \Omega_c \sum_{\sigma} (|L\sigma\rangle \langle R\sigma| + \text{h.c.})
\]

where \(|\nu\sigma\rangle\) stands for the state of \(\nu = L, R\) QD with spin \(\sigma = \uparrow, \downarrow\) and \(\Omega_c\) is the tunnel
coupling. The level energies are \( \epsilon_{\nu \sigma} = \epsilon_{\nu} - g_{\nu} \mu_{B} B \) with local g-factor \( g_{\nu} \), Bohr’s magneton \( \mu_{B} \) and external magnetic field \( B \). The coupling to the left and right reservoirs are characterized by the line-widths \( \gamma_{L} \) and \( \gamma_{R} \), respectively. We set up the master equation (ME) of the system density matrix \( \hat{\rho} \) assuming large intra- and inter-QD Coulomb interaction where the occupation of more than one electron is excluded. For simplicity, we also concentrate to the large bias limit, \( \mu_{L} \gg \epsilon_{\nu \sigma} \gg \mu_{R} \), where \( \mu_{\nu} \) is the chemical potential of the reservoir \( \nu \). The ME for the density matrix elements \( \rho_{\sigma 0}(\text{left QD occupied}), \rho_{\sigma \sigma}(\text{right QD occupied}), \rho_{LR\sigma}(\text{superposition}) \) is[7, 8]

\[
\frac{d\rho_{\sigma 0}}{dt} = \gamma_{L} \rho_{0} - i\Omega_{c}(\rho_{LR\sigma} - \rho_{RL\sigma}),
\]

\[
\frac{d\rho_{\sigma \sigma}}{dt} = i\Omega_{c}(\rho_{RL\sigma} - \rho_{LR\sigma}) - \gamma_{R} \rho_{0},
\]

\[
\frac{d\rho_{LR\sigma}}{dt} = (-i\delta_{\sigma} - \frac{\gamma_{R}}{2})\rho_{LR\sigma} - i\Omega_{c}(\rho_{0} - \rho_{0}),
\]

where the energy offset of spin \( \sigma \) is \( \delta_{\sigma} \equiv \epsilon_{L\sigma} - \epsilon_{R\sigma} \). The equation for \( \rho_{RL\sigma} \) is obtained by taking complex conjugate of the last equation and the probability of empty QDs \( \rho_{0} \) is obtained from the sum rule \( \rho_{00} + \sum_{\sigma}(\rho_{\sigma 0} + \rho_{0}) = 1 \). The current is obtained by \( I(t) = e\gamma_{R}\sum_{\sigma} \rho_{0} \).

It is convenient to convert this ME into matrix form defining matrix element vector \( \vec{\rho} = (\rho_{00}, \rho_{01}, \rho_{0\bar{1}}, \rho_{10}, \rho_{1\bar{1}}, \rho_{\bar{1}0}, \rho_{\bar{1}\bar{1}}, \rho_{\bar{1}\bar{1}})^{\dagger} \) and vectors \( \vec{\rho} = (\gamma_{L}, \gamma_{L}, 0, 0, 0, 0, 0, 0, 0)^{\dagger} \), \( \vec{\rho} = (0, 0, \gamma_{R}, \gamma_{R}, 0, 0, 0, 0, 0)^{\dagger} \) where "\( \dagger \)" stands for transpose. The ME reads \( d\vec{\rho}/dt = \hat{M}\vec{\rho} + \vec{u} \), where \( 8 \times 8 \) matrix \( \hat{M} \) is obtained from Eqs.(1,2,3). Steady state condition is \( d\vec{\rho}_{st}/dt = 0 \) and \( \vec{\rho}_{st} = - \hat{M}^{-1}\vec{u} \). Then the steady state current and its polarization \( P \) using \( I_{st\sigma} = e\gamma_{R}\rho_{0}\) are

\[
I_{st} = e\vec{v}^{\dagger}\vec{\rho}_{st} = \frac{2e\gamma_{L}\gamma_{R}\Omega_{c}^{2}}{\Omega_{c}^{2}(\gamma_{R} + 4\gamma_{L}) + \gamma_{L}(\frac{\gamma_{R}^{2}}{4} + \delta_{1}^{2} + \delta_{2}^{2})},
\]

\[
P = \frac{I_{st\uparrow} - I_{st\downarrow}}{I_{st\uparrow} + I_{st\downarrow}} = \frac{\delta_{1}^{2} - \delta_{2}^{2}}{\delta_{1}^{2} + \delta_{2}^{2} + 2\Omega_{c}^{2} + \frac{\gamma_{L}^{2}}{4}}.
\]

It can be seen that if one of the spin channels is far from the resonant condition: for example \( |\delta_{1}| \gg \Omega_{c}, \gamma_{\nu} \) as shown in Fig.1, the current is suppressed like \( 2e\gamma_{R}\Omega_{c}^{2}/\delta_{1}^{2}[3, 4] \), with almost full polarization \( |P| \sim 1 \).

Calculation of the transient current is slightly involved. We first solve the eigenvalue problem of the matrix \( \tilde{M} \) s.t., \( \hat{M}\tilde{\xi}_{n} = \lambda_{n}\tilde{\xi}_{n} \) for \( n = 1 \ldots 8 \). All the eigenvalues satisfy \( \lambda_{n} < 0 \). We define a unitary matrix \( \hat{U} \equiv (\tilde{\xi}_{1}, \ldots, \tilde{\xi}_{8}) \) and a diagonal matrix \( \hat{\Lambda} \equiv \text{diag}(\lambda_{1}, \ldots, \lambda_{8}) \). Further defining \( \tilde{\sigma} \equiv \hat{U}^{\dagger}\hat{\rho} \hat{U} \), \( \tilde{\omega} \equiv \hat{U}^{\dagger}\hat{\omega} \hat{U} \), we found the ME becomes eight independent differential equations: \( d\tilde{\sigma}/dt = \hat{\Lambda}\tilde{\sigma} + \tilde{\omega} \), which has a general solution with an initial condition \( \sigma(0), \sigma(t) = (e^{\hat{M}t} - \hat{1})\hat{\Lambda}^{-1}\tilde{\omega} + e^{\hat{M}t}\tilde{\sigma}(0) \). Getting back to the original density matrix and starting from the empty QD \( (\rho_{00} = 1 \) and others are zero; \( \rho(0) = 0 \) ), we have \( \tilde{\rho}(t) = \hat{U}e^{\hat{M}t}\hat{\Lambda}^{-1}\hat{U}^{\dagger}\rho_{0} + \rho_{st} \). We plot the current \( I(t) = e\vec{v}^{\dagger}\tilde{\rho}(t) \) for two values of \( \Omega_{c} \) in Fig.2. When the tunnel coupling \( \Omega_{c} \) is relatively strong, we can observe sub-peaks corresponding to the number of charge transfer \( N \). Transient excess charge is defined as \( Q(t) = e\int_{0}^{\tau}d\tau'\vec{v}^{\dagger}(\tilde{\rho}(\tau) - \tilde{\rho}_{st}) \) and the total transient excess charge is

\[
\frac{Q(\infty)}{e} = \vec{v}^{\dagger}\int_{0}^{\infty}d\tau\hat{U}e^{\hat{M}\tau}\hat{\Lambda}^{-1}\hat{U}^{\dagger}\hat{\omega} = -\vec{v}^{\dagger}\hat{M}^{-2}\hat{\omega} \rightarrow \frac{2S}{T^{2}},
\]

where the last expression is obtained by setting \( \gamma_{L} = \gamma_{R} = \gamma \) and \( \delta_{1} = 0 \) since general formula is too complicated to present, where \( S = 2(\frac{\delta_{1}}{\gamma})^{4} + (2(\frac{\delta_{1}}{\gamma})^{2} - 7)(\frac{\Omega_{c}}{\gamma})^{2} - 8(\frac{\Omega_{c}}{\gamma})^{4} \) and
\[ T \equiv 2\left(\frac{\delta_1}{\gamma}\right)^2 + 1 + 10\left(\frac{\Omega}{\gamma}\right)^2 \]. It can be seen \( Q(\infty) \) is slightly negative for \( |\delta_1| \ll \gamma \) but approaches \( e \) for \( |\delta_1| \gg \gamma \) as is argued in Sec.1. Similarly, we can define the spin polarization of total excess charge, which approaches unity for \( |\delta_1| \gg \gamma \),

\[ P_Q \equiv \frac{Q_1(\infty) - Q_{\uparrow} (\infty)}{Q_1(\infty) + Q_{\downarrow} (\infty)} \rightarrow \left(\frac{\delta_{\downarrow}}{\gamma}\right)^2 \frac{T}{S}. \] (7)

**Figure 1.** Top: The mechanism of the spin blockade in the series double QD under finite bias with different g-factors. Bottom: First three series of processes of transport of spin up electrons before the spin down electron is occupied in the left dot.

**Figure 2.** Transient current as a function of time starting from empty QD for two different inter-dot tunnel couplings. We set \( \gamma_L = \gamma_R \equiv \gamma \) and \( \delta_{\uparrow} = 0, \delta_{\downarrow} = 20\gamma \).

3. Pauli spin blockade

ME for the transport involving one and two electron states can be setup up analogously[6, 9] We obtain qualitatively similar behavior of the transient charge as in Sec.2 with a slight modification in the number of dark states. Please note that the Pauli spin blockade under zero magnetic field (also if we can neglect the effect of Overhauser field by nuclear spins), the transient current and charge are spin unpolarized.

In the following, we propose an experimental setup to observe this transient charge. In a series double QD with an asymmetry in level energies at zero bias[5], we observe Pauli spin blockade (SB) only for one of the polarities of the bias \((V_s, 1)\) as shown in Fig.3(a). In the other polarity, we observe narrow region of current suppression by Coulomb blockade (CB). When the bias is at the edge of CB condition \((V_s, 2)\), the system is in the one-electron ground state, and hence we can use this condition \((V_s, 2)\) as an initialization stage of the process. We change the bias periodically between \(V_s, 1\) for duration \(\tau_s, 1\) and \(V_s, 2\) for duration \(\tau_s, 2\) and observe the current in average. We change the duty ratio \(\tau_s, 2/\tau_s, 1\) as shown in Fig.3(c) where \(\tau_s, 1 \equiv \tau_s, 1 + \tau_s, 2\) is the period. When the period is much longer than tunneling time, the current as a function of duty ratio is expected simple linear interpolation between the current \(I_{V_s, 1}\) and \(I_{V_s, 2}\) as shown in the dashed line in Fig.4(left). The vertical series double QD device structure is similar to that found in Ref.[5] but we replaced with a semi-insulating substrate to obtain much higher cut-off frequency (> 40GHz). Measurement is performed at 1.5K and the DC pulse from the pulse-generator is applied to the source-drain terminal via bias-Tee and semi-rigid cables. The observed currents as a function of duty ratio for various period shown in Fig.4(left) are clearly deviating from
the dashed line. This negative excess current can be attributed to the transient current before the SB completed. We plot this excess current as a function of repetition frequency \( (f \equiv 1/\tau_p) \) in Fig.4(right) with a solid line of ideal excess current obtained by \( I = Q(\infty)f \), where \( Q(\infty) = \frac{e}{3} \). The deviation of the data points from the ideal line can be understood that the period is too short to transfer all the excess charge, which takes time roughly \( \max(\Omega_c^{-1}, \gamma^{-1}) \).

**Figure 3.** (a) Bias dependence of the current through series vertical QD at the Pauli spin blockade condition. (b) Magnified plot of (a) showing two bias points \( V_s1, V_s2 \) we set. (c) Schematics of the periodic modulation of bias with three different duty ratios.

**Figure 4.** (left) Duty ratio dependence of the average current for four different periods and the dashed line showing classical interpolation between 0% and 100%. (right) Excess current deviated from the interpolation (dashed line) as a function of repetition frequency. The solid line is the ideal dependence of the excess current on the frequency.

### 4. Conclusion
We argued the excess current/charge theoretically and experimentally before the spin blockade completed in series double quantum dot with finite bias. We clarify the condition of the blockade and polarization of the excess current/charge by using master equation. We also propose a scheme to detect this excess current by periodically modulation of bias and present the evidence of the fractional excess charge \( e/3 \).

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