Non-equilibrium singlet-triplet kondo effect in carbon nanotubes
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Supplementary Discussion

1. Nanotube transport experiments

Our nanotubes were grown by chemical vapor deposition (CVD) on a doped silicon substrate with a 400 nm oxide cap layer. Ferric iron nitrate nanoparticles deposited from a solution in isopropyl alcohol acted as catalyst for the CVD process, which was carried out in a tube furnace by flowing methane and hydrogen over the sample at 900 °C. This process yielded mostly individual single-wall nanotubes as determined by atomic force microscopy. The nanotubes were contacted by thermally evaporated metal electrodes (35 nm Au on 4 nm Cr), spaced by 250 nm and patterned by electron beam lithography. The two-terminal conductance was measured using standard lock-in techniques with $\sim 5 \mu V$ ac excitation and voltage bias $V$ applied to the source with the drain grounded through a low-impedance current amplifier. The effective electron temperature can differ from the measured mixing chamber temperature in the dilution refrigerator. A base electron temperature of $T_{el} \approx 80$ mK was estimated from the temperature dependence of device characteristics.

In the range of gate voltage considered in this paper, the room temperature conductance of the device is around $1.8 e^2/h$ and independent of back-gate voltage $V_g$. At lower gate voltages, a weak $V_g$ dependence at room temperature is seen and can be attributed to a small band gap in the metallic nanotube induced by perturbations. The coupling decreases with $V_g$ and at low $T$ the Coulomb blockade peaks disappear at $V_g \sim -2.5$ V, i.e. around $\Delta N = 40$ electrons away from the present $V_g$ range.
2. Low energy two-particle states and effective Kondo-Hamiltonian

The relevant single-particle states of the nanotube carry spin $\sigma = \uparrow, \downarrow$ and orbital index $i = 1, 2$, and the lowest lying two-particle states are denoted as follows:

$$| s \rangle = (| \downarrow, \uparrow \rangle - | \uparrow, \downarrow \rangle) \otimes | 1, 1 \rangle / \sqrt{2}$$ (1)

$$| -1 \rangle = | \downarrow, \downarrow \rangle \otimes (| 1, 2 \rangle - | 2, 1 \rangle) / \sqrt{2}$$ (2)

$$| 0 \rangle = (| \downarrow, \uparrow \rangle + | \uparrow, \downarrow \rangle) \otimes (| 1, 2 \rangle - | 2, 1 \rangle) / 2$$ (3)

$$| 1 \rangle = | \uparrow, \uparrow \rangle \otimes (| 1, 2 \rangle - | 2, 1 \rangle) / \sqrt{2}$$ (4)

$$| s' \rangle = (| \downarrow, \uparrow \rangle - | \uparrow, \downarrow \rangle) \otimes (| 1, 2 \rangle + | 2, 1 \rangle) / 2$$ (5)

$$| s'' \rangle = (| \downarrow, \uparrow \rangle - | \uparrow, \downarrow \rangle) \otimes | 2, 2 \rangle / \sqrt{2}$$ (6)

These states have energies $E_s, E_{-1,0,1} = E_s + \delta - J, E_s' = E_s + \delta$ and $E_s'' = E_s + 2\delta$, and since $J < \delta$, the singlet $| s \rangle$ is the ground-state. We include only the five lowest lying states and neglect the highest lying singlet, $| s'' \rangle$, altogether. Within the Hilbert-space of these five states, the effective Kondo-Hamiltonian takes the form:

$$H = \sum_{\substack{k,\sigma \\ i=1,2 \atop \alpha=L,R}} (\varepsilon_k - \mu_\alpha) c^\dagger_{\alpha i \sigma} c_{\alpha i \sigma} + H_{\text{int}}$$

where

$$H_{\text{int}} = \frac{1}{2\nu_F} \sum_{\substack{k,k',\sigma,\sigma' \\ i,j=1,2 \atop \alpha,\alpha'=L,R}} \{ g^{ij}_{\alpha' \alpha} \left[ \delta_{ij} \tilde{S} + \tau^3_{ij} \tilde{T} + \tau^1_{ij} \tilde{P} \right] \cdot \tau_{\sigma' \sigma} \\ + p^{ij}_{\alpha' \alpha} \left[ \delta_{ij} |s\rangle\langle s| + \frac{1}{2} (\tau^+_{ij} |s\rangle\langle s'| + \tau^-_{ij} |s'\rangle\langle s|) \right] \delta_{\sigma' \sigma} \\ + q^{ij}_{\alpha' \alpha} \delta_{ij} \sum_{m=-1,0,1} |m\rangle\langle m| \delta_{\sigma' \sigma} \} c^\dagger_{\alpha' k' \sigma'} c_{\alpha j k}$$ (7)

with

$$g^{ii}_{\alpha' \alpha} = \frac{2\nu_F t_{\alpha i} t_{\alpha'}^*}{E_C}, \quad g^{12}_{\alpha' \alpha} = (g^{21}_{\alpha \alpha'})^* = 2\sqrt{2\nu_F t_{1\alpha} t_{2\alpha}^*} / E_C,$$

$$p^{ii}_{\alpha' \alpha} = \tau^3_{ii} g^{ii}_{\alpha' \alpha}, \quad p^{12}_{\alpha' \alpha} = (p^{21}_{\alpha \alpha'})^* = g^{12}_{\alpha' \alpha},$$ (8)

The vector of Pauli-matrices is denoted by $\tilde{\tau}_{ij}$ and all terms in the interaction part have the form of spin, and orbital exchange, except for the two terms in the last line proportional...
to $\delta_{ij}\delta_{\sigma'\sigma}$ which are pure potential scattering terms. Throughout, we use the convention $A_\pm = A_x \pm iA_y$, for any vector-operator $\vec{A}$. The Hamiltonian is expressed in terms of the two-particle vector-operators

$$S^+ = (S^-)^\dagger = \sqrt{2}(|1\rangle\langle 0| + |0\rangle\langle -1|),$$
$$S^z = |1\rangle\langle 1| - |1\rangle\langle -1|,$$
$$P^+_{12} = (P^-_{21})^\dagger = \sqrt{2}|1\rangle\langle s|,$$
$$P^-_{12} = (P^+_{21})^\dagger = -\sqrt{2}|-1\rangle\langle s|,$$
$$P^z_{12} = (P^z_{21})^\dagger = -|0\rangle\langle s|,$$
$$T^+ = (T^-)^\dagger = \sqrt{2}(-|1\rangle\langle s'| + |s'\rangle\langle -1|),$$
$$T^z = |0\rangle\langle s'| + |s'\rangle\langle 0|,$$

which identifies them as generators of the Lie-algebra $SO(5)$. Notice that $T^2 + P^2 + S^2 + M^2 = 4$ is a Casimir-operator, i.e. a constant of motion. The Hamiltonian (7) is manifestly invariant under spatial rotations, but it will posses this abstract $SO(5)$-symmetry only when $\delta = J = 0$ and $t_{1\alpha} = t_{2\alpha}$, in which case one would expect to observe a conventional zero-bias Kondo-peak, characterized by a Kondo-temperature which is enhanced compared to the usual $SU(2)$ Kondo-effect. Notice that an $SO(5)$-Kondo-effect has been discussed earlier in the context of a triple-quantum-dot system$^4$.

As pointed out for a double-dot system studied in Ref. 5, $\vec{S}$ and $\vec{P}$ generate $SO(4)$, and it is the addition of the excited singlet $|s'\rangle$ which adds four new generators in the
present problem. In a double-dot system at even filling, the inter-dot tunneling breaks
the degeneracy between singlet and triplet states and an inelastic cotunneling channel
is generally available. A finite-bias Kondo-resonance in such a double-dot system was
suggested in Ref. 6 and the current due to nonequilibrium cotunneling was calculated in
Ref. 7, but the combined nonequilibrium Kondo-effect has not yet been examined. By
simply leaving out the excited singlet \(|s'\rangle\), our present calculation could readily be applied
to this problem as well.

3. Perturbative renormalization group equations

The (one-loop) perturbative renormalization group (RG) equations satisfied by the fre-
cquency dependent couplings are established in much the same way as explained earlier in
Ref. 8. Here, they take the following form:

$$\frac{\partial g_{\alpha'\alpha}(\omega)}{\partial \ln D} = - \sum_{\alpha'} \left\{ 2 g_{\alpha'\alpha}(\alpha'' V/2) g^0_{\alpha'\alpha}(\alpha'' V/2) \theta(D - |\omega - \alpha'' V/2|) \right. \]
$$

$$\left. + \frac{1}{2} g^0_{\alpha'\alpha}(\alpha'' V/2) g^0_{\alpha'\alpha}(\alpha'' V/2 - \tau^3_{ii} \delta) \theta(D - |\omega + \tau^3_{ii} \delta - \alpha'' V/2|) \right\} , \]

$$\frac{\partial p_{\alpha'\alpha}(\omega)}{\partial \ln D} = - \frac{1}{2} \sum_{\alpha'} \left\{ \left[ 3 g_{\alpha'\alpha}(\alpha'' V/2) + p_{\alpha'\alpha}(\alpha'' V/2) - q^0_{\alpha'\alpha}(\alpha'' V/2) \right] \right. \]

$$\left. \times g_{\alpha'\alpha}(\alpha'' V/2 + \tau^3_{ii} \delta) \theta(D - |\omega - \tau^3_{ii} \delta - \alpha'' V/2|) \right\} , \]

$$\frac{\partial q_{\alpha'\alpha}(\omega)}{\partial \ln D} = - 2 \tau^3_{ii} \sum_{\alpha'} g_{\alpha'\alpha}(\alpha'' V/2) g^0_{\alpha'\alpha}(\alpha'' V/2 - \tau^3_{ii} \delta) \theta(D - |\omega + \tau^3_{ii} \delta - \alpha'' V/2|) , \]

$$\frac{\partial p_{\alpha'\alpha}(\omega)}{\partial \ln D} = \frac{\partial q_{\alpha'\alpha}(\omega)}{\partial \ln D} , \]

$$\frac{\partial q_{\alpha'\alpha}(\omega)}{\partial \ln D} = - \frac{1}{2} \frac{\partial p_{\alpha'\alpha}(\omega)}{\partial \ln D} , \]

with the shorthand notation \(\theta_x = \theta(D - |x|)\), \(\bar{1} = 2\) and \(\bar{2} = 1\). For a given set of initial
values (at scale \(D = D_0\), parametrized by the bare tunneling amplitudes \(t_{i\alpha}\) according to
(8), these equations are readily solved numerically for arbitrary \(\omega\) and \(D\). Taking the limit
of \(D \to 0\), we obtain the renormalized coupling functions used in the Golden Rule expression
for the current.
FIG. 1: Inverse couplings vs. bandwidth $D$. The inverse couplings all vanish at the same energy-scale defining $T_K$. The couplings shown here are based on the same bare parameters as were used for the low temperature fit in the main paper: $\{t_{L1}, t_{L2}, t_{R1}, t_{R2}\} = \{0.032, 0.028, 0.108, 0.063\} \sqrt{E_C/\nu_F}$, $J = 0$ and $D_0 = 1$ eV, which implies that $T_K \approx 2$ mK.

As usual, when deriving the Kondo-model from an Anderson model, it is convenient to introduce two angles, $(\cos \phi_i, \sin \phi_i) = (t_{iL}, t_{iR})/\sqrt{t_{iL}^2 + t_{iR}^2}$, and parameterize the exchange couplings as

$$\{g_{ij}^{\alpha'\alpha}, p_{ij}^{\alpha'\alpha}, q_{ij}^{\alpha'\alpha}\} = \{g_{ij}, p_{ij}, q_{ij}\} \begin{pmatrix} \cos \phi_i \\ \sin \phi_i \end{pmatrix} \begin{pmatrix} \cos \phi_j & \sin \phi_j \end{pmatrix}_{\alpha'\alpha}$$

(16)

with initial conditions $g_{ij} = 2(\nu_F/E_C)\sqrt{(1 + \tau_0^i)(t_{iL}^2 + t_{iR}^2)(t_{jL}^2 + t_{jR}^2)}$, $p_{ii} = \tau_0^3 g_{ii}$, $p_{12} = p_{21} = g_{12}$ and $q_{ij} = 0$. The L/R matrix-structure of the couplings is now exterior, in the sense that the RG-equations are identical for every $(\alpha', \alpha)$ component.

For $D \gg \delta, V, T, \omega$, the RG-equations simplify to describe the flow of coupling constants $(g, p, q)_{ij}$ with no dependence on frequency, $\omega$, nor lead-index, $\alpha = L, R$. The difference in tunneling-strengths to the two orbitals still plays an important role, and enters via the initial conditions. From the RG-equations it is clear that all the different couplings diverge at the same energy-scale and it is this scale which we henceforth refer to as the Kondo-temperature, $T_K$ (see Fig. 1). The numerical solution of these high-D RG-equations allows us to study the dependence of $T_K$ on the three independent initial values $g_{11}$, $g_{22}$ and $D_0$.  

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This reveals the following nearly perfect interpolation-formula for $T_K$:

$$T_K \approx D_0 e^{-1/(2(g_{11}+g_{22})-0.84(g_{11}^{-1}+g_{22}^{-1})^{-1})},$$  \hspace{1cm} (17)

where 0.84 is a good approximation to a universal number describing this particular Kondo-effect. For $g_{11} = g_{22}$ this reduces to $T_K \approx D_0 \exp(-0.28/g_{11})$. If we leave out the singlet state $|s\rangle$, the RG-equations are modified accordingly and we find that the factor of 0.28 is replaced by 0.36, as found earlier in a study of vertical quantum dots\(^9\)(cf. also Refs. 10,11).

Notice that in the standard Kondo-effect involving a zero-bias conductance peak, the Kondo-temperature can be estimated directly from the width of the conductance peak. In the present problem, however, $T_K$ is of little physical significance insofar as the non-linear conductance is characterized mainly by the spin-relaxation rate $\Gamma$ (see sec. 5) and the subband-mismatch $\delta$. Nevertheless, $T_K$ still encodes a scaling-property of the RG-solutions and shows how our rather arbitrary choice of bare bandwidth $D_0 = 1$ eV is linked to the values of the bare couplings. In spite of the fact that the Kondo-temperature in this problem is enhanced over that of the standard SU(2) symmetric Kondo-model, our fitting parameters imply that $T_K \approx 2$ mK, which is much smaller than the value of 1 K found for the neighboring Coulomb-blockade valley having an odd number of electrons on the tube (cf. Fig. 2b in main text). Estimating the effective coupling in the neighboring valley from Eq. (17) with $g_{22} = 0$, we find that this big difference in Kondo-temperature corresponds to a mere reduction of the total hybridization to orbital 1 ($\sqrt{t_{11}^2 + t_{12}^2}$) by roughly 30% when changing from $N=1$ to $N=2$.

4. Nonequilibrium distribution functions

Using the renormalized coupling-functions, we may calculate the transition rates between the various two-particle states using the Golden Rule expression

$$W_{\gamma'\gamma}(V, \delta, T) = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} d\omega \sum_{\sigma,\sigma'=\uparrow,\downarrow} |g_{\alpha'\alpha,\sigma';\alpha,\sigma}(\omega)|^2 f(\omega - \mu_L)(1 - f(\omega + \varepsilon_{\gamma} - \varepsilon_{\gamma'} - \mu_R)),$$ \hspace{1cm} (18)

where $f$ denotes the Fermi-function. The nonequilibrium distribution functions for the two-particle states, $n_{\gamma}$, are then found by solving the steady-state quantum Boltzmann equation

$$\sum_{\gamma'} W_{\gamma'\gamma} n_{\gamma'} = \sum_{\gamma'} W_{\gamma'\gamma} n_{\gamma},$$ \hspace{1cm} (19)
together with the constraint $\sum_{\gamma=s,-1,0,1,s'} n_\gamma = 1$, ensuring that the two electrons in the half-filled shell on the nanotube occupy exactly one of the five lowest two-particle states. These voltage-dependent distribution functions are subsequently plugged into the Golden Rule expression for the current.

5. Spin-relaxation

As demonstrated in Ref. 12, a finite bias gives rise to Korringa-like spin-relaxation via inter-lead particle-hole excitations and for a single spin-1/2 the logarithmic singularities were found to be cut off by the (V-dependent) spin-relaxation rates $1/T_{1,2}$. To be more precise, it is the broadening of the transverse dynamical susceptibility, $1/T_2$, which cuts off the log-renormalization of spin-flip exchange couplings, whereas the renormalization of non-spin-flip exchange couplings are contained by the broadening of the longitudinal susceptibility, $1/T_1$. Diagrammatically, these rates arise from a combination of both self-energy, and vertex corrections to the spin-susceptibility bubble and a simplification which simply omits the vertex corrections will lead to serious mistakes.

In the present problem, the inter-orbital transition-rates (from excited to ground-state) are strongly enhanced by the large available phase-space. $W_{s,m}$ ($m = -1, 0, 1$) and $W_{s,s'}$ turn out to be larger than all other transition-rates by roughly an order of magnitude and the self-energy broadenings or line-widths of the excited states are therefore much larger than that of the ground-state. At least one of these large self-energy broadenings will contribute to the cut-off in the log-renormalization of all exchange couplings except one. $p_{ii}$ couples the ground-state to itself and the relevant self-energy broadening is very weak. For this particular coupling, vertex corrections will now play the dominant role and they supply additional terms in the total broadening involving the large $W_{s,m}$ and $W_{s,s'}$. All log-singularities are therefore cut off by rates of the order of these dominant line-widths of the excited states.

From the expression for the current (cf. main text Eq.(1)), it is clear that the conductance peak at $V \sim \delta$ is mainly determined by the inter-orbital exchange-couplings $g^{\vec{a}}$ and $p^{\vec{a}}$. Furthermore, these couplings turn out to be larger than the others and altogether the main influence of spin-relaxation on the physical current is therefore via these inter-orbital couplings. In other words, broadening all log-renormalization by a single effective spin-relaxation rate estimated from an inter-orbital susceptibility is expected to produce only
minute errors.

In terms of a generalized ss'-susceptibility, we determine the physical spin-relaxation-rate \( \Gamma_{ss'} \) as the sum of self-energy, and vertex corrections:

\[
\Gamma_{ss'} = \Gamma_{v} + \frac{1}{2} \left( \sum_{\gamma' \neq s} W_{\gamma',s} + \sum_{\gamma' \neq ss'} W_{\gamma',ss'} \right), \tag{20}
\]

where \( \Gamma_{v} \) is the contribution from vertex corrections. Just as the vertex-corrections to the transverse spin-susceptibility for a single spin-1/2 include only non-spin-flip processes\(^{12}\), the correction \( \Gamma_{v} \) picks up only intra-orbital transition-rates and is therefore negligible compared to the dominant term \( W_{ss'} \) coming from the self-energy broadening of the excited state. \( \Gamma_{sm} (m = -1, 0, 1) \) is determined in a similar way and since these two different inter-orbital relaxation rates turn out to be very close in magnitude as well as in \( V \)-dependence we define a single effective spin-relaxation rate as their average:

\[
\Gamma = \frac{1}{2} (\Gamma_{ss'} + \Gamma_{s1}). \tag{21}
\]

The spin-relaxation mechanism is incorporated in the RG-equations by replacing \( \theta_{x} \) by \( \theta(x) \) in Eqs. (11-15). Furthermore, all Fermi-functions occurring in the transition-rates and the current are effectively smeared by \( \Gamma \), by replacing the energy-conserving \( \delta \)-functions appearing in the Golden rule expressions with Lorentzians of width \( \Gamma \) (i.e. spin-susceptibilities \( \text{Im}[\chi^R] \)). For example, we evaluate the transition rate \( W_{ST} \) as follows:

\[
W_{ST} (V, \delta, T) = \frac{2\pi}{\hbar} \sum_{\alpha, \sigma' = \pm 1} \int_{-\infty}^{\infty} d\omega \int_{-\infty}^{\infty} d\varepsilon \left| g^{s_{1}}_{\omega', \sigma'; \alpha, \sigma} (\omega) \right|^2 f(\omega - \mu_{\alpha})(1 - f(\omega + \varepsilon - \mu_{\alpha'})) \text{Im}[\chi^R_{ST} (\varepsilon)]
\]

\[
\approx \frac{2\pi}{\hbar} \sum_{\alpha, \sigma' = \pm 1} \int_{-\infty}^{\infty} d\omega \int_{-\Lambda+\delta}^{\Lambda+\delta} d\varepsilon \left( \frac{1}{\mu_{\alpha} - \mu_{\alpha'} + \varepsilon} \int_{\mu_{\alpha'} - \varepsilon}^{\mu_{\alpha}} d\omega' \left| g^{s_{1}}_{\omega', \sigma'; \alpha, \sigma} (\omega') \right|^2 \right)
\times f(\omega - \mu_{\alpha})(1 - f(\omega + \varepsilon - \mu_{\alpha'})) \frac{\Gamma / \pi}{(\varepsilon - \delta)^2 + \Gamma^2}
\]

\[
\approx \frac{2\pi}{\hbar} \sum_{\alpha, \sigma' = \pm 1} \int_{\mu_{\alpha'} - \delta}^{\mu_{\alpha}} d\omega' \left| g^{s_{1}}_{\omega', \sigma'; \alpha, \sigma} (\omega') \right|^2 \int_{-\Lambda}^{\Lambda} d\varepsilon (1 + N(\varepsilon + \mu_{\alpha} - \mu_{\alpha'} + \delta)) \frac{\Gamma / \pi}{\varepsilon^2 + \Gamma^2}, \tag{22}
\]

where \( N \) is the Bose-function and \( \Lambda = \sqrt{\delta^2 + J^2 + V^2 + T^2} \) is an ultra-violet cut-off on the spin-susceptibility ensuring convergence of the integral over \( \varepsilon \). Notice that, in order to speed up the numerical evaluation, we have replaced the square of the coupling-function
by its average over the window set by the Fermi-functions. The error introduced by this approximation is estimated to be subleading in the small parameter $1/\ln(\delta/T_K)$.

We find that $\Gamma(V = \delta) \approx 250$ mK (varying between 220 mK and 360 mK when changing $V$ over the measured range) and the data clearly sample the full crossover from low to high temperatures with $T_{\text{el}}^{\text{lowest}} \approx 81$ mK $< \Gamma < 687$ mK $\approx T_{\text{el}}^{\text{highest}}$.

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