Transport across a carbon nanotube quantum dot contacted with ferromagnetic leads: experiment and non-perturbative modeling

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We present measurements of tunneling magneto-resistance (TMR) in single-wall carbon nanotubes attached to ferromagnetic contacts in the Coulomb blockade regime. Strong variations of the TMR with gate voltage over a range of four conductance resonances, including a peculiar double-dip signature, are observed. The data is compared to calculations in the "dressed second order" (DSO) framework. In this non-perturbative theory, conductance peak positions and linewidths are affected by charge fluctuations incorporating the properties of the carbon nanotube quantum dot and the ferromagnetic leads. The theory is able to qualitatively reproduce the experimental data.

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I. INTRODUCTION

Controlling electronic spin in nano-scale circuits is a long-lasting challenge on the way to fast-switching, energy-efficient building blocks for electronic devices. To this end, spin-dependent transport properties have been investigated in a wealth of low dimensional systems, e.g., mesoscopic magnetic islands [1], 2DEGs [2], InAs nanowires [3], graphene [4] and fullerenes [5]. Carbon nanotubes (CNTs), being thin, durable and high-throughput wiring, allow coherent transport of electronic charge and spin and are promising candidates for future spintronics applications [6]. While control and scalability of CNT-based nanocircuits still pose significant challenges, devices where single carbon nanotubes (CNTs) are contacted to ferromagnetic leads can be produced with standard lithography methods: spin valve experiments were performed on single-wall [7][10] (SWCNT) and multi-wall [11][15] carbon nanotubes in various electronic transport regimes. In most cases, a spatially confined quantum dot is coupled to ferromagnetic electrodes. Electronic transport across CNT quantum dots can take place in different regimes: Depending on the relative magnitude of coupling strength, temperature and charging energy, this ranges from an opaque Coulomb-blockade regime [16][19] to an intermediate coupling regime with lead induced energy level shifts [20][22], to a strongly correlated Kondo regime [23][26]. For highly transparent contacts, in contrast, the dot behaves essentially like an electronic wave guide [27][28].

In our work, we focus on the conductance of a carbon nanotube quantum dot weakly coupled to ferromagnetic contact electrodes, recorded for parallel ($G_p$) and anti-parallel ($G_{ap}$) contact magnetization, respectively. $G_p$ and $G_{ap}$ define the so-called tunneling magneto-resistance (TMR) [27][29]: TMR = ($G_p/G_{ap}$) − 1. Experimentally, the TMR has been shown to be strongly gate dependent [7][30]. We report on shifting and broadening of conductance peaks resulting in specific dip-peak and dip-dip sequences in the TMR gate dependence. Our data covers a range of four Coulomb resonances with extreme TMR values of −20% to +180%.

The pronounced resonant structure of the conductances $G_p$ and $G_{ap}$ leads to large TMR values if the positions and widths of the resonances depend on the magnetization configurations p and ap. Thus, various mechanisms have been proposed which induce a shift of the energy levels of the quantum dot, and thus of the resonance peaks, depending on the magnetization of the contacts. Those are spin-dependent interfacial phase shifts [7] or virtual charge fluctuation processes [21][25]. The effect of spin polarized leads on the resonance width have been described in [31] for a resonant single level junction. Interestingly, a negative TMR is predicted for asymmetric couplings to the leads. An attempt to account for broadening in the presence of Coulomb interactions was discussed within a self-consistent approach based on the equation of motion (EOM) technique [20]. The EOM was applied to model TMR data reported for a SWCNT [7] for a model with spin-dependent interfacial phase shifts.

Here we discuss a transport theory which naturally incorporates the effects of spin polarized leads on the position and width of conductance resonances in the presence of strong Coulomb interactions. It is an extension of the so-called dressed second order (DSO) transport theory, recently developed for normal leads [22], to the case of spin-polarized contacts. This theory accounts for energy renormalization and broadening of the peaks in linear conductance due to charge fluctuation processes. We show that the charge fluctuations also affect transport through excited states in the non-linear conductance regime. This observation is in agreement with previous reports on tilted co-tunneling lines in CNT quantum...
FeMn is sputtered to bias the magnetization of on one of the two contacts, 40 nm of anti-ferromagnetic
µa distance of 1
NiFe (80:20) leads, 20 nm in thickness, are deposited at
nanotube is located by atomic force microscopy and two
nanotube is grown by chemical vapor deposition. The
positively doped silicon substrate capped with 500 nm SiO
lar to that of the measured device. A carbon nanotube on a
devices we realized for this purpose is shown in Fig. 1.
On an oxidized silicon substrate (500 nm SiO
comparable to that of Pd. The structure of one of the
behavior as a function of the applied magnetic field and
spin-valves [33]: the alloy shows a distinct switching be-
is well suited as a material for the electrodes of CNT
contacts.

A qualitative agreement with the experimental
findings is obtained.

This paper is structured as follows. We first present
the measurement details and experimental data in Sec. I
In Sec. III we introduce the so-called dressed second or-
er theory (DSO) [22] in the reduced density matrix
transport framework and address its implications on non-
linear conductance and TMR. Finally, in Sec. IV we pro-
vide a comparison between experimental data and results
from the DSO and draw our conclusions in Sec. V.

II. EXPERIMENT

A. Sample preparation

For the purpose of measuring TMR in CNTs, one needs
to interface the nanotube to two ferromagnetic contacts
with a different switching field. The conductance, being
sensitive to the magnetization in the leads, changes when
the polarization of one of the contacts is reversed by an
external magnetic field. It has been shown that NiFe
is well suited as a material for the electrodes of CNT
spin-valves [33]: the alloy shows a distinct switching be-
havior as a function of the applied magnetic field and
the interface transparency between NiFe and the CNT is
comparable to that of Pd. The structure of one of the
devices we realized for this purpose is shown in Fig. 1.
On an oxidized silicon substrate (500 nm SiO2) a carbon
nanotube is grown by chemical vapor deposition. The
nanotube is located by atomic force microscopy and two
NiFe (80:20) leads, 20 nm in thickness, are deposited at
a distance of 1 µm on top of the nanotube by sputtering.
On one of the two contacts, 40 nm of anti-ferromagnetic
FeMn (50:50) is sputtered to bias the magnetization of
the underlying NiFe contact. The hysteresis loop of this
contact is expected to be shifted with respect to the pure
NiFe contact by virtue of the exchange bias effect [34].
A 20 nm protective layer (Pd) covers the leads from the
top. The switching of the exchange biased contacts was
confirmed independently prior to the measurement using
SQUID and vibrating sample magnetometer techniques.

B. Measurement

An electronic characterization of the quantum dot
at 300 mK and at zero magnetic field shows a regular
Coulomb blockade behavior (Fig. 2). The data yield a
gate conversion factor α = 0.29 and a charging energy
of $E_c = 6.1 \text{meV}$ (see Eq. (1)). The sample does not
exhibit a clear four-fold symmetry in the peak height or
peak spacing as expected for a carbon nanotube quantum
dot. Consequently, we are not able to label the Coulomb
blockade regions with a value of the electronic shell fill-
ing $n$ in a definite way. The assignment of the number
of electrons to the experimental data in Fig. 2 is done in
agreement with the theoretical predictions in Sec. III E.

Having a closer look at Fig. 2, we can identify an ex-
cited state transition at 1.4 meV parallel to the source
line (left arrow) and at ~ 1.8 meV parallel to the drain
line (right arrow). The energy scale of this excitation
stays approximately constant over a range of at least
six resonances, as can be seen from measurements over
a broader gate range. The quantization energy $\epsilon(n)$
of a CNT shell $n$ is a direct consequence of the electron
confinement along the nanotube. It yields a mean level
spacing $\epsilon_0 = \epsilon(n+1) - \epsilon(n) \propto \hbar v_F/\pi L$, where $L$ is the
CNT length. It is thus reasonable to identify the first
excitation with the confinement energy $\epsilon_0$ equivalent to

\[ 8.12 \quad \frac{\text{di/\text{dV}} (e^2/h)}{\text{bias \ voltage \ (mV)}} \]

\[ 0.5e^{-01} \quad 1.5e^{-01} \quad 8.12 \quad 8.14 \quad 8.16 \quad 8.18 \]

\[ -5.0 \quad -2.5 \quad 0.0 \quad 2.5 \quad 5.0 \]

\[ \text{gate \ voltage \ (V)} \quad \frac{\text{dI/\text{dV}} (e^2/h)}{\text{bias \ voltage \ (mV)}} \]

\[ 4n+1 \quad 4n+2 \quad 4n+3 \]
a lateral confinement of 1.1 \( \mu \)m for a Fermi velocity of 800 km/s [35], a value close to the contact spacing of 1 \( \mu \)m. The asymmetry of the line spacing with respect to source and drain suggests a gate-dependent renormalization [32] of the CNT many-body addition energies in the presence of ferromagnetic contacts. We show in Sec. III D that this can be a direct consequence of charge fluctuations in the presence of contact magnetization.

Electron transport measurements at 300 mK show a significant switching behavior. In Fig. 3 the conductance across the CNT quantum dot is plotted against the magnetic field directed parallel to the stripes, i.e., along their easy axis, as indicated in the inset to the figure. The steps in the signal can be interpreted as the magnetization reversal of the contacts, as sketched in the figure. Sweeping the magnetic field from negative (−100 mT) to positive values, one of the contacts switches at \( H = H_{s,u} \), resulting in a configuration with anti-parallel polarization of the majority spins of the two contacts. This results in a drop of the conductance signal. Upon increasing the field further, the second contact is supposed to switch and the conductance signal and take the average over 100 points (compare Fig. 3). We then identify \( H_{s,d} \) with a height of 0.7 meV. Comparing these values to a solid red curve was recorded with increasing field, the dashed blue curve with decreasing field. Small pictograms indicate possible orientations of the majority spins in the contacts. The switching of one of the two contacts at \( H_{s,u,d} \) is highlighted with arrows at the bottom for both sweep directions. The coercive field is indicated by \( H_e \) and the exchange bias by \( H_{ex} \). Inset: Orientation of the external field \( B \) with respect to the CNT and the leads.

Measurements of the conductance performed at zero magnetic field require \( \Delta t^{fast} \sim 100 \) ms per data point and will be called the fast measurements in the following. Contrarily, in slow measurements, each conductance data point is obtained from magnetic field sweeps with a duration of \( \Delta t^{slow} \sim 20 \) minutes at constant gate voltage (compare Fig. 3). We then identify \( H_{s} \) from a step in the conductance signal and take the average over 100 points on either side of the step to extract the conductance in the parallel and anti-parallel configuration, respectively. This is repeated for 250 values of the backgate potential \( V_{ex} \) in the range between 8.126 V and 8.201 V. In Fig. 4 the TMR as a function of gate voltage is shown together with the conductance at parallel contact polarization. In this slow measurement, we obtain conductance peaks with a height of 0.15e\(^2\)/h and a full width at half maximum (FWHM) of \( \Gamma \sim 0.7 \) meV. Comparing these values to a height of 0.3e\(^2\)/h and a width of 0.4 meV obtained from the fast measurement at \( B = 0 \) we conclude that the peak conductance in the data from the slow measurement is substantially suppressed. We will discuss this deviation in Sec. IV. It is remarkable that besides huge positive (180\%) TMR values, negative regions occur prior to the peak in the TMR curve in the first two resonances while for the last two the value drops again, forming two dips in sequence. Again this will be discussed in more detail in Sec. IV.

![Figure 3](image_url)

**FIG. 3.** (Color online) Differential conductance plotted versus magnetic field at \( V_g = 8.1737 \) V, \( V_d = 0 \) and 300 mK. The solid red curve was recorded with increasing field, the dashed blue curve with decreasing field. Small pictograms indicate possible orientations of the majority spins in the contacts. The switching of one of the two contacts at \( H_{s,u/d} \) is highlighted with arrows at the bottom for both sweep directions. The coercive field is indicated by \( H_e \) and the exchange bias by \( H_{ex} \). Inset: Orientation of the external field \( B \) with respect to the CNT and the leads.

![Figure 4](image_url)

**FIG. 4.** (Color online) Differential conductance and TMR as a function of gate voltage measured over four resonances (slow measurement, see text). The conductance is measured at parallel polarization of the contacts. The TMR graph shows a dip-peak sequence over the first two resonances and a qualitatively different double-dip feature at the last two.

III. THEORETICAL MODELING

We proceed by presenting a theoretical framework capable to reproduce the transport data from the previous section. In particular, the connection between the theory and the resulting shape of the TMR curve will be discussed in detail. In order to be able to account for a gate dependence of the TMR, the transport theory should be able to incorporate the influence of the ferromagnetically polarized leads on the positions of the linear conductance maxima as well as on the width of the conductance peaks.

Noticeably, the commonly used perturbative description of the Coulomb resonances predicts temperature broadened peaks and maxima whose positions are solely
FIG. 5. (Color online) Left panels: Schematic drawing of the lead induced, polarization dependent, modification of position (a) and width (b) of a peak in the conductance across a quantum dot as a function of the gate voltage. Right panels: As a consequence of the level shift (a) and level broadening (b), the corresponding TMR signal exhibits a characteristic dip-peak (c), or dip-peak-dip (d) feature.

is the Hamiltonian of an ensemble of non-interacting electrons in the leads $l = s/d$ with wave vector $k$ and spin $\sigma$. The operator $\hat{c}^{\dagger}_{l\sigma k} (\hat{c}_{l\sigma k})$ annihilates (creates) an electron with energy $\epsilon_{l\sigma k}$. The second part,

$$\hat{H}_D = \frac{1}{2} E_c \hat{N}^2 + \sum_{n\sigma} \left[ \epsilon(n) + \tau \sigma \frac{\Delta_{SO}(n)}{2} \right] \hat{N}_{n\sigma}$$

$$- e\alpha V_g \hat{N} + \hat{H}_{ext}$$

(1)

describes the electrons on the CNT quantum dot in terms of the quantum numbers $n$ (shell), spin $\sigma$ and valley $\tau$. Here we used $\hat{N}_{n\tau\sigma} = \hat{d}_{n\tau\sigma}^{\dagger} \hat{d}_{n\tau\sigma}$, with the fermionic dot operator $\hat{d}_{n\tau\sigma}$ and $\hat{N} = \sum_{n\tau\sigma} \hat{N}_{n\tau\sigma}$, the total dot occupation. For our purposes, it is sufficient to account for Coulomb interaction effects in terms of a capacitive charging energy $E_c$. Short range exchange contributions are neglected here. The symbols $\tau$ and $\sigma$ represent the eigenvalues $\pm 1$ of the states with quantum numbers $K, K'$ and $\uparrow, \downarrow$, respectively. In the CNT, a non-zero spin-orbit coupling $\Delta_{SO}$ can lead to the formation of degenerate Kramer pairs [20]. Notice that, for simplicity, a valley mixing contribution is not included in Eq. (1), as it would not affect the main conclusions drawn in this work. Hence, the valley degree of freedom is a good quantum number to classify the CNT’s states [36]. The next to last part of the Hamiltonian $\hat{H}_D$ models the effect of an electrostatic gate voltage $V_g$ scaled by a conversion factor $\alpha$. Finally, $\hat{H}_{ext}$ accounts for external influences on the dot potential, e.g., stray fields from the contacts and the external magnetic field used to switch the contact polarization.

The ground states of shell $n$ have $4n + a$ ($0 \leq a \leq 3$) electrons and will in the following be characterized by the quantum numbers of the excess electrons with respect to the highest filled shell $n - 1$. For instance, the quantum dot state labeled by $|K\uparrow; n\rangle$ contains $4n$ electrons plus one additional electron in the $(K, \uparrow)$ state. Including states with $4n - 1$ and $4n + 5$ electrons we end up with 6 ground states with different degeneracies (see Tab. 1 left column). In total we consider a Fock space of dimension 24 if the four-fold degeneracy is not lifted by a sufficiently large spin-orbit coupling $\Delta_{SO}$. The extra states with occupation $4n - 1$ and $4n + 5$ are included to allow for charge fluctuations in and out of the shell $n$ under consideration. Conversely, for large enough spin-orbit coupling the dimension of the Fock space is reduced to 10, see Tab. 1 right column. Judging from the stability diagram in Fig. 2 and from data over a greater gate range where we see no two-fold pattern in the spacing of the excited state lines, we consider the configuration on the left side in Tab. 1 to be more likely. For a compact notation, the shell number will in the following be neglected from the state ket if not necessary.

Quantum dot and metallic leads are coupled perturba-
tively by a tunneling Hamiltonian
\[
\hat{H}_T = \sum_{lkn\sigma\tau} T_{lkn\sigma\tau} \hat{d}_l^{\dagger \sigma} c_{l\tau\sigma} + \text{h.c.},
\tag{2}
\]
with a tunnel coupling \(T_{lkn\sigma\tau}\) generally dependent on the quantum numbers of both leads and quantum dot. In the following, for simplicity, we assume that \(T_{lkn\sigma\tau} = T_l\).

**B. The reduced density matrix within the dressed second order (DSO) approximation**

We describe the state of our system by the reduced density matrix \(\tilde{\rho} = \text{Tr}_R\{\hat{\rho}_{\text{tot}}\}\), obtained by tracing over the possible configurations of states in the reservoirs, assuming that they are in thermal equilibrium. For the quantum dot itself we suppose that it reaches a steady state characterized by \(\hat{\rho} = 0\). The corresponding stationary Liouville equation reads \([22]\)
\[
0 = -i \sum_{a} \delta_{ab} \delta_{a'b'}(E_a - E_{a'}) \rho_{aa'} + \sum_{a} K_{bb'}^{aa'} \rho_{aa'},
\tag{3}
\]
in terms of matrix elements \(\rho_{ab} = \langle a | \hat{\rho} | b \rangle\) of \(\hat{\rho}\) in the eigenbasis of the quantum dot. The superoperator \(K\) connects initial states \(|a\rangle, |a'\rangle\) to final states \(|b\rangle\) and \(|b'\rangle\) at a certain order in the perturbation \(\hat{H}_T\).

The calculation of the kernel elements is performed along the lines of Ref. [22]. As an example, the element connecting the states \(|b\rangle, |b'\rangle = |b\rangle\) and \(|a\rangle, |a'\rangle = |a\rangle\) is given in second order by
\[
K_{bb}^{aa} = \sum_l \Gamma_{l,bb}^p = \sum_l \frac{i}{\hbar} \lim_{\lambda \to 0} \int d\epsilon \frac{\gamma_{ba}(\epsilon) f_l^p(\epsilon)}{E_b^0 - \epsilon + i\lambda} + \text{h.c.},
\]
where \(\Gamma_{l,bb}^p\) is the corresponding tunneling rate. The function \(f_l^p(\epsilon)\) with \(p = \pm\) is defined as \(f_l^\pm(\epsilon) = \left[1 + \exp(-\beta(\epsilon - \mu_l))\right]^{-1}\), where \(\beta\) is the inverse temperature and \(\mu_l\) the lead’s chemical potential. Hence, \(f_l^\pm(\epsilon)\) is the Fermi function and describes the occupation probability in lead \(l\). In general, \(p = \pm 1\) if the final state \(|b\rangle\) has one electron more/less than the initial state \(|a\rangle\). The energy difference between final and initial dot configuration is given by \(E^a_b = E_b - E_a = E_b - E_a - e\alpha V_b(N_b - N_a)\). Finally,
\[
\gamma_{ba}^{\pm}(\epsilon) = \gamma_{a(b,a)}(\epsilon) = |T_l|^2 D_{l\sigma}(\epsilon)
\]
is a spin-dependent linewidth defined in terms of the tunneling amplitude \(T_l\) and of the spin-dependent density of states \(D_{l\sigma}(\epsilon)\). A Lorentzian provides a cut-off for the density of states at a bandwidth \(W\). The notation \(\sigma(a,b)\) indicates that the spin \(\sigma\) of the electron tunneling out of/onto lead \(l\) depends on the spin configuration of the initial state \(|a\rangle\) and the final state \(|b\rangle\) of the quantum dot. It is convenient to introduce the spin-resolved density of states of lead \(l\) at the Fermi energy
\[
D_{l\sigma}(\epsilon) = D_{l\sigma}(\epsilon_F) = D_0(1 + \sigma P_l)/2
\tag{4}
\]
where \(P_l = (D_{l\uparrow} - D_{l\downarrow})/(D_{l\uparrow} + D_{l\downarrow})\) is the polarization of lead \(l\). The couplings \(|T_l|^2\) we define in the same spirit as
\[
|T_{s/dl}|^2 = |T_0|^2(1 \pm a)/2,
\tag{5}
\]
using the parameter \(a\) to tune the asymmetry in the coupling to the leads. We will in the following use the factorization
\[
\gamma_{a(b,a)}(\epsilon_F) = \gamma_0 K_{l\sigma},
\tag{6}
\]
where we collect the lead and spin independent prefactors in an overall coupling strength \(\gamma_0 = D_0|T_0|^2\) and include the dependence on spin and lead index in the dimensionless parameter \(K_{l\sigma}\), where \(\sum_{l\sigma} K_{l\sigma} = 1\). Note that \(\gamma_0\) is related to the level broadening \(\Gamma_0\) by \(\Gamma_0 = 2\pi\gamma_0\).

In Fig. [3a], a diagrammatic representation of one contribution to the second order kernel is shown for the case of \(|a\rangle = |0\rangle\) and \(|b\rangle = |\tau\sigma\rangle\). The fermionic line connecting the lower to the upper contour carries indices \(l, \epsilon, \sigma\) which fully characterize the nature of the electron tunneling between lead \(l\) and quantum dot. The direction of the arrow further specifies if the electron tunnels out of (towards lower contour) or onto (towards upper contour) the dot.

Beside this lowest (second) order contribution, we consider all diagrams of the structure shown in Fig. [3b]. The selected diagrams contain arbitrary numbers of uncorrelated charge fluctuation processes (bubbles in
According to Tab. I, the CNT spectrum can be spin and valley degenerate. However, the tunneling Hamiltonian conserves the spin during tunneling, and thus spin coherences are not present in the dynamics. Here, for simplicity, orbital coherences are neglected as well [37].

C. Current within the DSO

The current through the terminal $l$ can be written in terms of the difference of in- and out-tunneling contribu-
lations at the junction [38]:
\[ I_I(V_b) = \frac{e}{2\pi\hbar} \sum_{\epsilon \in C_C} \int d\epsilon \left[ P_a(V_b)f_{I}^{+}(\epsilon) - P_c(V_b)f_{I}^{-}(\epsilon) \right] \nu_{I}^{ca}(\epsilon, V_b), \]
(13)
where \( V_b \) is the bias voltage applied between the two contacts, and \( C \) is the set of all possible configurations (see Tab. [1]). In general, the populations can be expressed in terms of rates via the Liouville equation [3] and a closed form for the current and, consequently, for the conductance can be found. This is straightforward if two states are connected by pairwise gain-loss relations [38]. For the case of the single impurity Anderson model, for example, a compact notation of the conductance can be given [22].

The width of a resonance in conductance with respect to the gate potential is determined by the populations, the TDOS which has a form similar to a Lorentzian, and the derivative of the Fermi functions. Note that the populations are themselves a function of the rates and therefore also governed by the resonance conditions of the rates. The DSO theory has been proven to be quantitatively valid down to temperatures 4k_B T \sim \gamma_0, in the single electron transistor [22]. Upon decreasing the temperature below \( \gamma_0/4 \), a quantitative description of the transition rate \( \Gamma_{\text{inc}} \) would require to calculate \( \Sigma \) beyond the lowest order in \( \gamma_0 \). In the regime where temperature and coupling are of comparable magnitude, the width and position of the Coulomb blockade peaks in a gate trace are strongly influenced by the TDOS and, more precisely, by the self energy \( \Sigma \). The role of \( \text{Re}(\Sigma) \) is to influence the positions of the Coulomb blockade peaks: In the rate for the transition \( a \) to \( b \), the real part appears next to the energy difference \( E_a^b \) of the transition in the denominator. Hence, due to this contribution the resonant level is shifted depending on the configuration of the leads.

D. Renormalization of excited states

In the stability diagram in Fig. [2] we observe an asymmetry in the spacing of lines associated with excited states connected to one charging state, as drawn schematically in Fig. [3]. The line \( 0 \rightarrow 1' \) meets the diamond at bias voltage \( V_{b1} \). Measured along the bias voltage axis, this value is larger than the energy difference \( V_{bg} \) associated with the line \( 2 \rightarrow 1' \) on the right. A similar behavior has been discussed previously for the co-tunneling regime [32]. As noted by these authors, the asymmetry cannot be explained within the sequential tunneling picture but can be attributed to the renormalization of the excitation energies \( E_a^b \) in Eq. (1) due to virtual tunneling processes. Although the framework in Ref. [32] is different, the evaluation of \( \text{Re}(\Sigma_{\text{exc}}) \) is similar to that in our model. The condition for a resonance for a transition between states \( a \) and \( b \) is given by
\[ \epsilon \pm eV_b/2 + e\alpha V_g - \tilde{E}_{a}^{b} + \text{Re}(\Sigma_{\text{exc}}) = 0, \]
(14)
where \( \epsilon \) is the energy of the tunneling electron with respect to the chemical potential of the unbiased contact \( \mu_0 \). Note that this condition can be fulfilled for different transitions at the same time, a situation that occurs at any point where two lines in a stability diagram intersect. In order to interpret the observed shift of the excited state line in the differential conductance data in Fig. [7] it is illuminating to study the contribution from \( \text{Re}(\Sigma) \) at points \( (V_{g1}, V_{b1}) \) and \( (V_{g2}, V_{b2}) \) marked by a dot and a circle, respectively, in Fig. [5]. We consider an exemplary set of states \( 0 = |0; n⟩, 1 = |K; n⟩, 1'_1 = |[K; n⟩, 1'_2 = |K; K; n⟩, 2 = |K; K; n⟩ \) and \( 2 = |K; K; n⟩ \). A similar analysis can be carried out for other states with \( 4n + 1 \) and \( 4n + 2 \) electrons. The quantum numbers in round
FIG. 8. Schematic drawing of the conductance lines in the vicinity of the charging state with $4n + 1$ electrons in Fig. 2. The first visible excitation is shifted upwards on the left and downwards on the right side of one charging diamond by $e(V_{b1} - V_{b2})/2 = -\delta_1$. The corresponding energies in Fig. 2 are $eV_{b1}/2 \approx 2\text{meV}$ and $eV_{b2}/2 \approx 1.4\text{meV}$. For our analysis we choose bias and gate voltages close to the filled dot for the first transition $0 \to 1$’ and to the empty circle for the second transition $2 \to 1$’.

brackets denote a missing electron of shell $n-1$ whereas the square brackets indicate a state of shell $n + 1$. For each of the highlighted points in Fig. 8 two conditions in the form of Eq. (14) can be given. Subtracting them pairwise we are left with

\[
\begin{align*}
V_{b1} - E_1' &+ [\text{Re}(\Sigma_1^{1,0}) - \text{Re}(\Sigma_1^{1,0})] = 0, \quad (15) \\
V_{b2} - E_1' &+ [\text{Re}(\Sigma_2^{1,1}) - \text{Re}(\Sigma_2^{1,1})] = 0, \quad (16)
\end{align*}
\]

where the self energy contributions depend on bias and gate voltage. To lowest order in $\gamma_0$ we analyze the differences in $\text{Re}(\Sigma)$ using $eV_{b1/2} = E_1'$ and $e\delta_0 E_1^{1/2} = E_0 / 2$ at $\epsilon = 0$. In order to calculate $\text{Re}(\Sigma)$ we have to analyze the contributions from all accessible states in Eq. (10). In principle there are arbitrarily many states that can be reached by a charge fluctuation. However, we assume that the available energy interval for charge fluctuation processes is given by $\max(eV_{b1}, \Gamma_0, 3 - 4k_B T)$ and contributions beyond this scale are suppressed. Numerical results using a larger bandwidth can be found in Sec. A of the appendix.

For our considerations we assume that the spin orbit coupling of our CNT quantum dot is small, i.e., $\Delta_{SO} < \max(k_B T, \Gamma)$. Otherwise we would expect to see a twofold symmetry in the spacing of the excited state lines in the stability diagram in Fig. 2. The other important scales - charging energy, shell spacing and linewidth - are related in the way $E_c > \epsilon_0 \gg \max(k_B T, \gamma_0)$. Within this choice of parameters the difference of the self energy corrections for the resonant transition can be calculated by $15 - 16 = 0$, i.e.,

\[
\delta_1 \equiv \left[ \text{Re}(\Sigma_1^{1,0}) - \text{Re}(\Sigma_1^{1,0}) \right] - \left[ \text{Re}(\Sigma_2^{2,1}) - \text{Re}(\Sigma_2^{2,1}) \right] \approx \gamma_0 \{-1 + 2\bar{\kappa}_d - \bar{\kappa}_d + \bar{\kappa}_d + \bar{\kappa}_d \} \Psi_R^0(\epsilon_0/2).
\]

where we used the abbreviation $\Psi_R^0(\epsilon) = \text{Re}[\Psi^0(1/2 + i\epsilon/2\pi k_B T)]$ and a bar denotes a summation over indices, e.g., $\bar{\kappa}_d = \sum_{\sigma} \kappa_{d\sigma}$. A detailed derivation of these quantities is given in the appendix, Sec. A. Similar calculations are performed for the excited states in the $n+2$ and $n+3$ diamonds, yielding

\[
\delta_2 \approx \gamma_0 \{\bar{\kappa}_d - \bar{\kappa}_d + \bar{\kappa}_d + \bar{\kappa}_d \} \Psi_R^0(\epsilon_0/2),
\]

\[
\delta_3 \approx \gamma_0 \{1 + \bar{\kappa}_d - 2\bar{\kappa}_d + \bar{\kappa}_d + \bar{\kappa}_d \} \Psi_R^0(\epsilon_0/2),
\]

where the states with three electrons are chosen to be electron-hole symmetric with respect to the state with one electron. Note that for the case of symmetric couplings the shifts reflect the electron-hole symmetry of the system while a choice of $a \neq 0$ (Eq. 5) breaks this symmetry. For highly asymmetric couplings $|a| \sim 1$ the shifts are comparable to those in Ref. [32]. Note that the effective change of the resonance with respect to the energy difference has a negative sign (compare Eq. 14). The resonance marked by the left arrow in Fig. 3 is situated above the resonance marked by the right arrow. The experimental data thus corresponds to a negative shift. We therefore assume an asymmetric coupling to the leads with a dominant coupling to the drain contact, i.e., $\kappa_d < \kappa_t$, $-1 < a < 0$. Using the parameters from a fit to the data in Sec. V, i.e., $a = -0.7$ and $\epsilon_0 = 1.4\text{meV}$ we obtain $\delta_1 \approx -0.2\text{meV}$ and $\delta_2 \approx -0.1\text{meV}$.

Compared to the shifts in the experimental data, these values are too small by a factor of 2-3. We expect that additional states may contribute to the charge fluctuations that are not considered within this approximation.

E. Tunneling magneto-resistance

Corrections to the conductance peak width are given by $\text{Im}(\Sigma)$. Because $\text{Re}(\Sigma)$ and $\text{Im}(\Sigma)$ both depend on the different magnetic properties of the source and drain leads as well as on the dot’s configuration, the resulting impact on the TMR is quite intricate. Thus we analyze the contributions to the self energy in the light of different configuration of the lead’s polarizations. We focus on the last resonance, i.e., the transitions $|0, n+1 \rangle \rightleftarrows |(\sigma\tau), n+1 \rangle$ where the TMR graph in Fig. 4 exhibits a double dip structure. The back-gate voltage is tuned such that

\[
\epsilon + e\alpha V_g - \tilde{E}_{(\tau\sigma)} = \text{Re}(\Sigma(\sigma\tau)) = 0,
\]

and the quantum numbers in round brackets $(\tau\sigma)$ denote a missing electron of shell $n+1$. At lowest order in the tunnel coupling $\gamma_0$ we approximate $\alpha V_g = E_{(\tau\sigma)}$ when we calculate $\text{Re}(\Sigma(\sigma\tau))$. From Eq. (12) we list the
imaginary part of the self energy for this transition, i.e.,

$$\text{Im}(\Sigma^0(\tau\sigma)) = \pi \gamma_0 \sum_l \left\{ \sum_{c' \in C_{\tau\sigma}^c} \kappa_{l\sigma(c')} f_l^+(E_{l\tau\sigma} - \epsilon) + \sum_{c' \in C_{\tau\sigma}^c} \kappa_{l\sigma(c')} f_l^-(\epsilon - E_{c'\tau\sigma}) \right\}.$$  

$$+ \sum_{c \in C_{\tau\sigma}^c} \kappa_{l\sigma(c)} f_l^+(\epsilon - E_c^0) + \sum_{c' \in C_{\tau\sigma}^c} \kappa_{l\sigma(c')} f_l^-(E_{c'}^0 - \epsilon) \right\}.$$

The magnitude of the energy difference of the virtual state with respect to the state on the other contour determines whether a possible charge fluctuation contributes to the renormalization of the self energy or not: a contribution $f_l^+(E_c - \epsilon)$, e.g., is exponentially suppressed in the vicinity of the resonance. Therefore, knowing the arguments in the step functions $f^\pm$, we can simplify the result significantly. Close to the resonance where $|\epsilon| < \max(k_B \Gamma, \gamma_0)$, the fluctuations with an energy cost of the charging energy $E_c$ or of the shell spacing $\epsilon_0$, e.g., the states that can be reached by out-tunneling from the state $(\tau\sigma)$ can be neglected.

Focusing on the resonant contributions, we are left with

$$\frac{\text{Im}(\Sigma^0(\tau\sigma))}{\pi \gamma_0} \simeq \sum_l \left\{ \kappa_{l\sigma} f_l^+(\epsilon) + \sum_{\tau'\sigma'} \kappa_{l\sigma} f_l^-(\epsilon - E_{l\tau'\sigma'}^0) \right\}.$$

It is clear from this result that the broadening of the TDOS peak does depend on the lead configuration $\kappa_{l\sigma}$. Let the majority spins be polarized such that $\sigma = +1$ in the layout with parallel lead polarization. The sum over the leads is then given by $\sum_{l} \kappa_{l\sigma} = (1 + \sigma P)/4$ and $\sum_{l} \kappa_{l\sigma} = (1 + \sigma Pa)/4$ for parallel and anti-parallel polarizations, respectively. Let us first consider the case of zero effective Zeeman splitting, i.e., $E_{\sigma}^p = E_{\sigma}^a = 0$. The difference of $\text{Im}(\Sigma)$ for the two configurations then reads

$$\text{Im} \left[ \Sigma_p^{0,(\tau\sigma)} - \Sigma_a^{0,(\tau\sigma)} \right] = \delta \text{Im} = \pi \gamma_0 \left( 1 - a \right) f^+(\epsilon).$$

Note that the validity of this result depends on the ratio of linewidth and level spacing, namely that $\gamma_0 \ll \epsilon_0$ such that only the selected small set of charge fluctuations contribute. The sign of the difference in Eq. (19) is determined by $\sigma$, a result which is intuitively clear since the sum over the couplings will be greater for the spin-up transition ($\sigma = 1$) in the parallel case and for the spin-down transition in the anti-parallel one ($\sigma = -1$), as shown schematically in Fig. 9(a). For zero energy splitting $E_{\sigma}^{(\tau\sigma)}$ we would expect a broadening of the peak associated with the transition $0 \Rightarrow (\uparrow)$ for the parallel configuration and a broadening of the peak in $G^{ap}$ for the transition $0 \Rightarrow (\downarrow)$. Note, however, that the second effect will not be visible since the TMR ratio will be dominated by the spin up transition. Hence, we will observe a TMR signal as depicted in Fig. 9(b).

Now let us assume a non-zero effective Zeeman splitting $E_{\sigma}^{\uparrow} = E_{\sigma}^{\downarrow} = g_B h p_{\uparrow}/p_{\downarrow}$ of states with quantum numbers $\sigma = \uparrow/\downarrow$. This splitting also depends on the magnetization state $p$ (parallel) or $ap$ (anti-parallel) of the contact electrodes. The energy difference is expressed in terms of the effective magnetic fields $g_B h p$ and $g_B h_{ap}$. We assume that this field is non-zero for both polarizations. $\text{Im}(\Sigma)$ as well as the TMR are very sensitive to the choice of the shifts, the couplings and the polarization. The mechanism we want to discuss can be observed for different parameter regimes, but for the sake of the argument it is sufficient to present one possible set that we deduce from the experiment and the line of reasoning that goes with it. In the last part of Sec. III D we argue that couplings $\kappa_{c} < \kappa_{c'}$ or, similarly, $0 < a < 0$ are needed to explain the shift of the excited state lines in Fig. 2. Furthermore we point out that the peaks in conductance in Fig. 4 are descending in height as we fill the shell. In our model the drain lead switches polarization upon interaction with
external magnetic field while the density of states in the weakly coupled source contact remains unaltered. Given that the spin transport is more sensitive to the bottleneck (source) contact, it is plausible to assume that the shifts are such that the majority spins tunnel first on the quantum dot, namely spin up electrons in both configurations. These considerations favor a choice of negative shifts \( g\mu_B h \) such that \( g\mu_B h < -k_B T \). The second pair of resonances is then dominated by spin down electrons and the respective contributions \( f^-(\epsilon + g\mu_B h_{\text{pol}}) \) in Eq. (18) are suppressed. Conversely, for spin up electrons \( f^-(\epsilon - g\mu_B h_{\text{pol}}) = 1 \). In the resonant case, \( |\epsilon| \lesssim k_B T \), the imaginary part of the self-energy for the conductance data from the measurement yields conductance peaks that fit for the parallel configuration (compare Fig. 3). The data Sec. II B. Note that it provides only conductance data for fields \( g\mu_B h_{\text{pol}} = -40 \, \mu eV \) and \( g\mu_B h_{\text{pol}} = -80 \, \mu eV \). In the panels on the left side, the polarization is varied keeping \( a = -0.8 \) fixed. We see that the right shoulder in the TMR curve (c) is lifted upwards with increasing polarization. On the right panels in Fig. 10 we increase the coupling to the source contact which is proportional to \( a \). While the conductance is decreased for asymmetric choices of \( a \) in both configurations (see (d) and (e)), the magnitude of the peak in \( G_{\text{pol}} \) is not symmetric with respect to the coupling to source and drain. The TMR in Fig. 10 (c) can be related to Eq. (20): the shoulders for \( a = 0.8 \) turn into dips approaching \( a = -0.8 \). Please keep in mind that this discussion is simplified since we do not account for the fact that the relative position of the peaks changes, too, as we vary the parameters \( a \) and \( P \) (compare Re(\( \Sigma \)) and Im(\( \Sigma \)) plotted in Fig. 12 in the appendix Sec. A).

### IV. COMPARISON

**a. Conductance in the experiment and in the model**

In Fig. 11 (a) (blue circles) we show the conductance \( G^{\text{fast}}_P \) obtained at \( B = 0 \) performing a fast measurement, i.e., sweeping the gate voltage \( V_g \) at zero bias voltage, see Sec. II B. Note that it provides only conductance data for the parallel configuration (compare Fig. 3). The data from this measurement yields conductance peaks that fit to Lorentzian curves with an average FWHM of 0.3 meV. Adapting our model parameters to the data of \( G^{\text{fast}}_P \), we obtain the continuous lines in Fig. 11 (a,b,d). The conductance data from the slow measurement (compare Sec. II B) for the two configurations, \( G^{\text{slow}}_P \) and \( G^{\text{slow}}_G \), are shown in Fig. 11 (a,b) (green crosses). The shape of the conductance peaks turns out to be non-Lorentzian, with the peak height in the conductance data limited to \( \sim 0.1 \epsilon^2/h \). While the flanks of the peaks match for the first three resonances in the data from the slow and from the fast measurement, the maximum conductance values deviate by a factor of three. So far no full explanation for the suppression of the peak conductance was found.

**b. Model parameters** A bare coupling of \( \gamma_0 = 80 \, \mu eV \) is found to optimize the fit to \( G^{\text{fast}}_P \). The thermal energy is chosen as \( k_B T = 40 \, \mu eV \) (400 mK), close to

![FIG. 10. (Color online) Conductance and TMR calculations in the vicinity of the resonance \( \{0, n+1\} \) for different polarizations \( P \) (panels (a)-(c), \( a = -0.8 \)) and coupling asymmetry \( \gamma_0 = 100 \, \mu eV \) (d),(e) applied in the parallel configuration for effective Zeeman splitting \( g\mu_B h = -40 \, \mu eV \) and \( g\mu_B h = -80 \, \mu eV \). (a),(b): increasing the polarization reduces the peak width and height of both \( G_P \) and \( G_{\text{pol}} \). (c): In the TMR curve, the shoulder on the left at \( P = 0.2 \) is shifted to the right for \( P = 0.6 \). (d),(e): The coupling asymmetry \( \gamma_0 = 0 \) diminishes the peak heights of the conductance for both configurations of the leads. Note that in the anti-parallel case shown in (e) the symmetry between the contacts is broken and the peak height is sensitive to the variation of the dominating coupling. (f): The TMR curve exhibits a double dip feature for values \( -1 \leq a < 0 \). It is transformed to a double peak for \( 0 < a \leq 1 \). All plots are calculated at a temperature corresponding to \( 40 \, \mu eV \) and a coupling \( \gamma_0 = 160 \, \mu eV \).
the base temperature (300 mK). For the quantum dot parameters we set $E_c = 6.1$ meV and a shell spacing $\epsilon_0 = 1.4$ meV as inferred from Sec. II. The shell number $n \sim 40$ is estimated from the distance to the bandgap. We assume asymmetric contacts with $a = -0.7$ and polarization $P = 0.4$. For the calculation of the charge fluctuations we include all states within an energy interval of $3\epsilon_0$ (see Sec. A in the appendix). The effective Zeeman shifts for the model output in Fig. 11 are $g\mu_B h_p = -0.12$ meV and $g\mu_B h_{ap} = -0.16$ meV.

c. Discussion If only features of the leads density of states at the Fermi energy are included, compare Eq. [1], the DSO preserves particle-hole symmetry by construction [22]. To break this symmetry, a Stoner-shift of the majority band with respect to the minority band should be included [24], whose effect is analogous to that of an effective Zeeman field [21]. Such effective fields have also been used to model the effects of coherent reflections at the magnetic interfaces in double barrier systems [7]. Since the data in Fig. 11(a-c) do not reflect particle hole symmetry, we use effective Zeeman splittings to break the particle-hole symmetry and reproduce the observed magnitude of the TMR effect. The splittings are of similar magnitude as those used in Ref. [23] ($g\mu_B h_p = 0.25$ meV and $g\mu_B h_{ap} = 0.05$ meV) to explain the experimental TMR data of Ref. [7].

In case of non-zero spin-orbit coupling [40, 41], we would expect a splitting of the excited state lines in the stability diagram in Fig. 2. This is not resolved in our experimental data. For simplicity we therefore here assume $\Delta_{SO} = 0$. Model calculations with non-zero spin orbit coupling can be found in the appendix, Sec. C.

From the conductance traces calculated within our model, Fig. 11(a,b) (continuous lines), the TMR, Fig. 11(d), is obtained. The data and the model calculation agree in the decay of the TMR amplitude within a sequence of four charging states including the “double dip” feature in the last two resonances at $V_g = 8.17$ V and $V_g = 8.19$ V. This indicates that the sequence in Fig. 11 represents one shell, i.e., charging states $4n+1$ to $4(n+1)$. We note that in the model output the last resonance is dominated by a peak while the dips are more prominent in the experimental data.

In the vicinity of all conductance peaks (at $V_g = 8.13$ V, $V_g = 8.15$ V, $V_g = 8.17$ V and $V_g = 8.19$ V) an additional small shoulder around TMR = 0 occurs in the data of Fig. 11(c). These shoulders are likely related to the aforementioned suppression of the peak conductance in the slow measurement (see Fig. 11(a,b)). We recall that the TMR is calculated from the ratio $G_p/G_{ap}$ (com-
pare also Fig. 5 and Fig. 9: in the regions where the peaks are cut off, the ratio $G_{p}^{\text{low}}/G_{ap}^{\text{low}}$ is smaller than it is in the same region in the model output, where steep peak flanks lead to a larger ratio $G_{p}/G_{ap}$.

V. SUMMARY

The tunneling magneto-resistance of a carbon-nanotube based quantum dot with ferromagnetic leads has been explored both experimentally and theoretically. The experimental data shows a distinct variation of the tunneling magneto-resistance (TMR) lineshapes within a single quadruplet of charging states.

To model the data we apply the dressed second-order (DSO) framework based on the reduced density matrix formalism. This theory accounts for charge fluctuations between the quantum dot and the ferromagnetic contacts. Thereby, it goes beyond the sequential tunneling approximation which can only account for a positive and gate-independent TMR. When the charge fluctuation processes are summed to all orders in the coupling to the leads according to the DSO scheme, they yield tunneling rates where the Lamb shift and the broadening of the resonances are given by the real and imaginary parts of the self energy, respectively. This is a nontrivial result which yields the tunneling rates for an interacting quantum dot in the intermediate parameter regime $E_{c} \gg k_{B}T \sim \Gamma$ depending on the polarization of the contacts.

We explicitly compare the DSO self energy for different contact magnetizations and show that the DSO modeling can account both for the renormalization of excited states and the specific structures observed in the TMR gate dependence. A comparison of the TMR obtained from the model and from the experimental data shows a qualitative agreement.

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Appendix A: Contribution of other excited states to the renormalization of the self energy

When we discuss the effect of the charge fluctuations in Sec. III D and Sec. III E of the main text, we always focus on the most resonant transitions (see Fig. 7) that are energetically favorable, i.e., on transitions in Eq. (12) with an energy difference $E_{n/c/b}^{c'/b}$ of the order of the effective line-width or below. At zero bias this is the largest available energy scale in the system. Nevertheless it is interesting to see how the outcome is affected by increasing the bandwidth and allowing excited states of the neighboring shells to contribute to the charge fluctuation channels. In terms of an effective energy shift in a multi-level quantum dot the renormalization due to excited states was also discussed in Ref. 21. To illustrate the effect of such a modification we plot the real and imaginary parts of the self energy $\Sigma$ in the vicinity of the transition $|\langle K \downarrow, n \rangle \Rightarrow |, n + 1 \rangle$ for different sets of charge fluctuations within energy ranges of $\gamma_{0}, \epsilon_{0}$, $2\epsilon_{0}$ and $3\epsilon_{0}$ in Fig. 12.

FIG. 12. (Color online) Re(\Sigma) (a,b) and Im(\Sigma) (c,d) for both lead configurations as a function of energy $\epsilon$ in units of the shell spacing $\epsilon_{0}$. Different lines are plotted for bandwidth $W_{\text{fluc}} = \gamma_{0}$ (green, dotted) to $3\epsilon_{0}$ (red, continuous) in steps of $\epsilon_{0}$. In the vicinity of a few $k_{B}T$ around the resonance ($\epsilon = 0$, gray region) the difference between the graphs for the real part (a,b) is small and for the imaginary part (c,d), it is vanishing.

In this section we perform the calculation of Re(\Sigma') – Re(\Sigma^{10}) as part of the quantity $\delta_{1}$ introduced in Sec. III D of the main text. To this extent we analyze the renormalization of the energy difference $E_{1}^{\prime}$ due to charge fluctuations to and from states $0 = |0; n\rangle$, $1 = |K; n\rangle$ and $1^{'} = |\langle K|; n\rangle$ in more detail. We recall that the real part
of the self energy related to a charge fluctuation to state \( c' \) has the form (see. Eq. [12])

\[- \sum_{l} 2 \gamma_{l}^{(b/a)}(\epsilon) \Psi_{R}^{0}(\mu_{l} \pm p(E_{a/c}^{c'/b} - \epsilon)),\]

where we have to replace \( b = 1' \), \( a = 0 \) or \( b = 1 \) and \( a = 0 \), respectively. Note that the contribution \( \propto \Psi^{00}(W) \) in Eq. [12] does not appear explicitly since it cancels in the difference of the shifts. Next, we have to find all states \( c' \) that contribute within our resonant approximation. We can immediately discard states that can be reached by in-tunneling from \( b \) and by out-tunneling from \( a \), since their energy differences \( E_{a/c}^{c'/b} \) are of the order of the charging energy and thus beyond our charge fluctuation bandwidth of \( W_{c} = \max(\epsilon, V_{b}, k_{B}T, \gamma_{0}) = \epsilon_{0}/2 \). We are left with states that can be reached by in-tunneling into state \( a \) and by out-tunneling from state \( b \). Let us discuss one example for the state \( 1' \). There is one electron in the shell \( n+1 \) (denoted by the brackets \([ \ldots ] \) in the state ket) which can tunnel out and we are left with a state \([ \ldots , n ] \). Actually this state is identical to the state \( 0 \) on the other contour, thus \( E_{0}^{c'/b} = 0 \). We can now evaluate the argument of the digamma function, i.e., \( \mu_{l} - E_{0}^{c'/b} + \epsilon \), for \( \epsilon = 0 \). Since \( \mu_{s}/d = \pm \epsilon_{0}/2 \) and thus \( |\mu_{l}| \leq W_{c}, we have to sum over both leads. The total contribution from fluctuations to \( c' = 0 \) is thus \( -\gamma_{0} \sum_{l} \gamma_{l}^{(b/a)} \Psi_{R}^{0}(\epsilon_{0}/2) \). The other states that can be reached by out-tunneling, e.g., \( [(K^{'}, [K], n) \), yield energy differences of at least \( 3/2 \epsilon_{0} > W_{c} \). Using similar arguments we can collect all relevant contributions to the difference \( \text{Re}(\Sigma^{c'/b}) - \text{Re}(\Sigma^{c/b}) \). In a graphical representation, this can be visualized as

\[
\text{Re}(\Sigma^{c'/b}) - \text{Re}(\Sigma^{c/b}) = \begin{cases} 0 & \text{in to 0} \\ - & \text{out from 1'} \\ + & \text{out from 1} \\ \end{cases}
\]

\[
= 2k_{n} \Psi_{R}^{0}(\epsilon_{0}/2)
\]

where one set of four boxes symbolizes one shell and we use \( E_{a/c}^{c'/b} \) as a label. Fluctuations that cancel are crossed out. Note that for excited states with an energy difference \( E_{a/c}^{c'/b} = \pm \epsilon_{0} \) we add only the contribution from the source(drain) contact where \( |\mu_{l} - E_{a/c}^{c'/b}| < W_{c} \). Similarly we find

\[
\text{Re}(\Sigma^{c'/b}) - \text{Re}(\Sigma^{c/b}) = (1 + k_{d} - k_{t} + k_{i}) \Psi_{R}^{0}(\epsilon_{0}/2),
\]

which leaves us with \( \delta_{l} \) from Eq. [17].

**Appendix C: Spin-orbit coupling and valley polarization**

In Sec. [11A] we discussed the possibility to include spin-orbit interaction effects, as they have been reported to play a prominent role in carbon nanotubes [40, 42]. However, we did not add it in the comparison to the experimental data since they could not be resolved in the transport spectrum (Fig. 2). Nevertheless, values of the order of \( \Delta_{SO} \sim 100 \mu eV \) would still be consistent with the experimental data. Introducing a finite \( \Delta_{SO} \) a priori does not affect the TMR as the Kramers pairs are spin degenerate pairs with anti-parallel and parallel alignment of spin and valley magnetic moments. Yet it has been argued that the two valleys of a CNT can couple differently to the leads [43]. If the valley quantum number is conserved upon tunneling, the mechanism can be understood in terms of a valley polarization. A possible tunneling Hamiltonian that describes this situation can be written as

\[
\hat{H}_{T} = \sum_{l} \delta \tau_{l} \tau_{j} \tau_{l} \tau_{i} \psi_{l} \psi_{i} \text{h.c.} \quad (C1)
\]

with a valley dependent coupling \( \delta \tau_{l} \tau_{j} \tau_{l} \tau_{i} \) and an operator \( \psi_{l} \psi_{i} \) that describes the electrons in the leads (that are also part of the CNT). Including a valley polarization in turn also renders the TMR sensitive to magnetic stray fields \( g_{orb} \mu_{orb} h_{ap}^{orb} \) and \( g_{orb} \mu_{orb} h_{ap}^{orb} \) along the tube axis. The orbital magnetic moments \( g_{orb} \mu_{orb} \) are considered to be larger than \( \mu_{B} \) by one order of magnitude [44]. In Fig. 13 we present a TMR calculation for \( \Delta_{SO} = 100 \mu eV \), orbital polarization \( P_{orb} = 0.6 \) and stray fields \( g_{orb} \mu_{orb} h_{ap}^{orb} = -80 \mu eV \) and \( g_{orb} \mu_{orb} h_{ap}^{orb} = -40 \mu eV \) again combined with the experimental data. The spin-dependent shifts are assumed to be negligible in this setup. We see that the agreement with the experimental data improved slightly in Fig. 13 at the expense...
of additional free parameters. It is, however, outside the scope of this paper to discuss the effect of spin-orbit coupling and the valley polarization in more detail.

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