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Sources of negative tunneling magnetoresistance in multilevel quantum dots with ferromagnetic contacts

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We analyze distinct sources of spin-dependent energy level shifts and their impact on the tunneling magnetoresistance (TMR) of interacting quantum dots coupled to collinearly polarized ferromagnetic leads. Level shifts due to virtual charge fluctuations can be quantitatively evaluated within a diagrammatic representation of our transport theory. The theory is valid for multilevel quantum dot systems and we exemplarily apply it to carbon nanotube quantum dots, where we show that the presence of many levels, among them of excited states, can qualitatively influence the TMR effect.

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I. INTRODUCTION

Recent transport experiments on quantum dots coupled to ferromagnetic leads have demonstrated the existence of spin-dependent energy level shifts through the observation of negative tunnel magnetoresistance (TMR) effects in the single electron tunneling regime and spin splitting in the Kondo regime. So far qualitatively different approaches for explaining the origin of the underlying shifts coexist. For example, negative TMR data from carbon nanotube (CNT) measurements have been fitted with a model relying on spin-dependent interfacial phase shifts picked up by the wave function during multiple reflections at a spin-active interface. The concept is related to that of spin-mixing conductance and, because it only depends on the properties of the spin-active barrier region, is only weakly gate dependent and present in both interacting and noninteracting systems.

In contrast, experiments on CNTs and InAs nanowires in the Kondo regime have demonstrated a combination of gate-dependent and gate-independent contributions to the energy level shifts. Both can be explained in terms of charge fluctuations, whereby electron-electron interactions are responsible for the logarithmic gate dependence, while a Stoner splitting of the energy bands of the magnetically polarized leads accounts for the almost gate-independent part.

While the effects of the energy level shifts in the Kondo regime are by now well understood, a thorough understanding of their influence on the TMR phenomenon in interacting quantum dots is still missing. The negative TMR data have been satisfactorily fitted in terms of a generalized Anderson model already including gate-dependent level shifts. In Ref. a reflection Hamiltonian was included to account for reflection processes at the interface in a second-order sequential tunneling theory.

In this work, we specifically address the TMR phenomenon within a diagrammatic approach to the reduced density matrix of the nanosystem and discuss different level-shift-induced mechanisms for negative TMR. Spin-dependent level shifts can originate to the lowest order in the tunneling Hamiltonian (sequential tunneling limit) if off-diagonal elements of the reduced density matrix participate in the dynamics. This is the case for noncollinearly polarized leads but also for collinearly polarized leads for some double-dot setups. Here we consider situations where coherences of the reduced density matrix do not play a crucial role in the dynamics. We rather identify charge fluctuation terms in all orders in the tunneling Hamiltonian which sum up to a Taylor series yielding intrinsic level shifts.

In particular we discuss two types of spin-dependent level shifts: strongly gate-dependent ones deriving from a net difference of the majority and minority density of states at the Fermi level, and largely gate-independent ones caused by a Stoner shift of the majority and minority bands in the leads. Our results are valid for generic multilevel quantum dots and reproduce the results of Ref. in the case of a simple Anderson model.

Finally, we analyze the TMR of a CNT quantum dot and show that, due to the multilevel spectrum, the intrinsic contributions give rise to a marked gate voltage dependence and a TMR which can indeed become negative. Moreover, we show that charge fluctuations to states including bosonic excitations, which are present in CNTs due to the linearity of the noninteracting spectrum, can largely influence the gate voltage dependence of the level shifts.

The paper is structured as follows: Sec. contains a discussion of possible mechanisms for negative TMR. In Sec. the model Hamiltonian is elucidated, while in Sec. the level shifts are calculated analytically. Results for the conductance and TMR of armchair CNTs are presented in Sec. V, while in Sec. VI conclusions are drawn.

II. MECHANISMS OF NEGATIVE TMR

The basic mechanism underlying a negative TMR is the presence of an effective generalized Zeeman field accounting for both extrinsic (stray fields, applied magnetic field) and intrinsic sources of spin-dependent level shifts. Here \( P/A \) refers to contacts with parallel/antiparallel
effective magnetic field responsible for the effective Zeeman
shift also removes the spin-degeneracy of the ground states by favoring the states with maximum total spin. This situation is illustrated in Fig. 1(d) for the case of a CNT quantum dot, for which we consider the CNT Hamiltonian, Eq. (2).

III. THE MODEL HAMILTONIAN

For a quantitative description we consider the transport setup of Fig. 1. In the limit of weak coupling to the leads it can be described by the total Hamiltonian

\[ \hat{H}_{\text{tot}}^{P/A} = \hat{H} + \hat{H}_{\text{ext}}^{P/A} + \sum_{l=x,d} \hat{H}_l + \hat{H}_T, \]

where \(\hat{H} = \hat{H}_0 - e\alpha V_g \hat{N}\) comprises the Hamiltonian \(\hat{H}_0\) of the isolated quantum dot and the effects of a gate voltage (\(\alpha\) is a conversion factor of the order of one). In the case of an armchair nanotube quantum dot of medium-to-large radius far from half-filling it reads\(^{19}\)

\[ \hat{H}_0 = \frac{1}{2} E_c \hat{N}^2 + \frac{1}{2} \sum_{l=a,d} \sum_{r,s} (\epsilon_{0r} \hat{N}_{r\sigma} + \bar{\sigma} \Delta \epsilon) \hat{N}_{s\sigma} + \hat{H}_B, \]

where \(\epsilon_{0r} = \hbar v_F / \pi L\), with \(v_F\) being the Fermi velocity and \(L\) the CNT length, is the level spacing; \(\Delta \epsilon\) is the orbital mismatch; and \(E_c\) is the charging energy. The number of electrons in the orbital band \(\bar{\sigma} = \pm\) with spin \(\sigma\) is determined by the number operator \(\hat{N}_{r\sigma}\) and the total number is determined by \(\hat{N} = \sum_{r\sigma} \hat{N}_{r\sigma}\). Finally,

\[ \hat{H}_B = \sum_{n \neq 0} \sum_{j \in \pi, s} \epsilon_{j,n} \hat{a}^\dagger_{j,n} \hat{a}_{j,n} \]

This requires \(|h^P - h^A|\) of the order of the width \(k_B T\) of the conductance peaks. Second, however, negative TMR can also arise for \(|h^P| = |h^A| \approx k_B T\) [see Fig. 1(c)]. This is because the effective magnetic field responsible for the effective Zeeman magnetization (see Fig. 1). For spin-dependent transport experiments, typically two ferromagnetic contacts of different widths [compare Fig. 1(a)] are utilized to obtain a hysteretic polarization behavior when exposing the setup to a sweep of an external magnetic field. Due to remanent magnetization, the polarization of the contacts is inverted only at a reverted field of some hundreds of millitesla in strength, until the magnetically softer of the two contacts switches first to align with the external field. Thus, for some range of the field, the contacts stay in antiparallel configuration. Intuitively, one would expect that the conductance for contacts polarized in parallel (\(G_P\)) is larger than the one for the antiparallel case (\(G_A\)), yielding positive values of TMR := \((G_P/G_A) - 1\).

Nevertheless, there are at least two different mechanisms which can lead to a negative TMR, as sketched in Figs. 1(b) and 1(c), respectively. Here we consider quantum dots in the single electron tunneling regime and look at the linear conductance peaks, while the peak position signals at which the Fermi level \(\epsilon_F\) of the contacts stay in antiparallel configuration. Intuitively, one would expect that the conductance for contacts polarized in parallel (\(G_P\)) is larger than the one for the antiparallel case (\(G_A\)), yielding positive values of TMR := \((G_P/G_A) - 1\).

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Because the effective Zeeman field leads to corrections to the energy difference \(E_b - E_a := E_{ba}\) associated with the transition from a many-body state \(a\) to a many-body state \(b\), this in turn modifies the position of the conductance peaks.

First, and most obviously, there can be negative TMR if there is a noticeable offset in the conductance peak positions in parallel and antiparallel configuration [see Fig. 1(b)]. This requires \(|h^P - h^A|\) of the order of the width \(k_B T\) of the conductance peaks. Second, however, negative TMR can also arise for \(|h^P| = |h^A| \approx k_B T\) [see Fig. 1(c)]. This is because the effective magnetic field responsible for the effective Zeeman magnetization (see Fig. 1). For spin-dependent transport experiments, typically two ferromagnetic contacts of different widths [compare Fig. 1(a)] are utilized to obtain a hysteretic polarization behavior when exposing the setup to a sweep of an external magnetic field. Due to remanent magnetization, the polarization of the contacts is inverted only at a reverted field of some hundreds of millitesla in strength, until the magnetically softer of the two contacts switches first to align with the external field. Thus, for some range of the field, the contacts stay in antiparallel configuration. Intuitively, one would expect that the conductance for contacts polarized in parallel (\(G_P\)) is larger than the one for the antiparallel case (\(G_A\)), yielding positive values of TMR := \((G_P/G_A) - 1\).

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accounts for bosonic charge, $c$, and spin, $s$, excitations. The
energies $\epsilon_{c,n} = n\epsilon_0/g$ are for $n > 0$ depending on the
Coulomb interaction parameter $g \simeq 0.2$, while the others scale only
with the level spacing: $\epsilon_{s,\pm \Delta} = \epsilon_{c,n} \pm |\Delta|/g$. None of
the bosonic excitations influences the particle number in the single
bands and hence one can classify the states of the CNT by a vector
$|N, \vec{m}\rangle$, where $N = (N_+ + N_-, N_+ - N_-)$ determines
the fermionic configuration and $\vec{m}$ determines the bosonic
excitations (see, e.g., Ref. 20).

The ground states $|N, \vec{m}\rangle = 0 : = |N\rangle$ of shell $n$ have $4n$,
$4n + 1$, $4n + 2$, and $4n + 3$ electrons and can be characterized
in terms of the excess spins in band $|\vec{r}, s\rangle$ with respect to the
case of equally filled bands: $|n,n,n,n\rangle := |\uparrow,\downarrow\rangle$, $|n +
1, n,n,n\rangle := |\uparrow,\downarrow\rangle$, or $|n + 1, n + 1, n,n\rangle := |\uparrow,\downarrow\rangle$, etc. (see
Table I). For medium-to-large tube radius and far from the
charge neutrality point, spin-orbit coupling and exchange
effects are not relevant. They can become of interest for large
curvatures and will be investigated elsewhere. $\hat{H}_\text{ext}^{(1)}$ accounts
for external, gate-independent sources of level splitting and
holds, at the least, the contribution arising from the external
field necessary to control the direction of the contact polarizations.

The leads are described by $\hat{H}_l = \sum_{\sigma q} \epsilon_{lq} \hat{c}_{l\sigma q}^\dagger \hat{c}_{l\sigma q}$, with
$\hat{c}_{l\sigma q}$ annihilating an electron of energy $\epsilon_{lq}$ and of spin $\sigma$ in
lead $l$. The density of states in lead $l$, $D_{l\sigma}(\omega)$, is assumed to
be constant over an energy range set by the reduced density matrix
$\hat{\rho}_{\text{tot}}(\omega)$. It determines the leads polarizations $P_l$ at the Fermi level
according to $P_l = (D_{l\uparrow} - D_{l\downarrow})/(D_{l\uparrow} + D_{l\downarrow})$.

On the other hand, in order to account for a Stoner splitting
$\Delta_{\text{St}}$, the range of available energies in the leads is given by
$-(W + \sigma \Delta_{\text{St}}/2) \leq \omega \leq W + \sigma \Delta_{\text{St}}/2$, where $W$
is the bandwidth at zero Stoner splitting.

Finally the perturbative contribution is $\hat{H}_P = \sum_{\sigma l \sigma'} \int d^3r \langle T_l(\bar{r})\hat{\psi}_{\sigma l}^\dagger(\bar{r})\hat{\psi}_{\sigma' l}(\bar{r}) + \text{H.c.}\rangle$,
allowing for tunneling between CNT and leads. Here $T_l(\bar{r})$
is the tunnel kernel, $\hat{\psi}_{\sigma l}(\bar{r})$ is the CNT bulk electron operator as
given in Ref. 19, and $\hat{\phi}_{\sigma l}(\bar{r}) = \int d\bar{r}' D_{l\sigma}(\bar{r}') \sum_{\sigma l'} \hat{\phi}_{\sigma l'}(\bar{r}')\hat{\psi}_{\sigma l'}(\bar{r})$ is the lead
electron operator with $\hat{\phi}_{\sigma l}(\bar{r})$ denoting the corresponding
single-particle wave function.

IV. TRANSPORT THEORY

The current, as any other observable of the transport setup,
can be calculated by a trace over the associated operator multiplied
by the reduced density matrix $\hat{\rho}(t) = \text{Tr}_T \{\hat{\rho}_{\text{tot}}(t)\}$. Being
obtained from the density matrix $\hat{\rho}_{\text{tot}}$ of the total setup
upon tracing out the lead degrees of freedom, $\hat{\rho}(t)$ stores the
full information about the state of the dot in the presence of the
tunnel coupling to the leads. The time evolution of $\hat{\rho}(t)$ follows
the Liouville equation, and it can for the stationary state ($\hat{\rho}(t) = 0$) be cast into the form (see, e.g., Ref. 21)

$$0 = -i \sum_{a,a'} \delta_{a,b} \delta_{a',b'} (E_a - E_{a'}) \rho_{a'a} + \sum_{a,a'} K_{a'bb'}^{bb'} \rho_{a'a},$$

taking matrix elements with respect to the many-body eigenstates of the CNT: $\rho_{a'a} := \langle a|\hat{\rho}|a'\rangle$ and $E_a := \langle a|\hat{H}|a\rangle$. Furthermore $K_{bb'}^{aa} := \langle b|\hat{K}|a\rangle\langle a'|\hat{K}|b'\rangle$, with the kernel superoperator $K$ arising from the perturbation, i.e., the tunnel coupling between quantum dot and leads. Depending on up to which order $2n$ in the tunnel coupling $K$ is calculated, one takes into account effects from $2n$ correlated tunnel events. The most involved part of a perturbative transport calculation is the determination of the kernel $K$.

A fully diagrammatic representation of $K$ has first been proposed in Ref. 15. For example, one contribution to the second-order kernel ($K^{(2)}_{ab}^{bb'}$) is shown in Fig. 2(a). It relates to the tunnel process $|a\rangle \rightarrow |b\rangle$, represented by the solid line connecting upper and lower parts of the diagram contour. In the following we identify specific terms in all orders of the perturbation series as charge fluctuation processes, which sum up to a Taylor series yielding the intrinsic level renormalization $E_{ba} \rightarrow E_{ba} + h_{\text{int}}^{ba}$ to the energy difference $E_{ba} = E_b - E_a$, where $h_{\text{int}}^{ba}$ is given in Eq. (7) below.

A. Diagrammatic evaluation of the intrinsic level renormalization

Recently, it has been recognized that certain fourth-order
diagrams can be related to charge fluctuations in the initial or
final state during a tunnel event.

Here we identify a specific class of these charge fluctuation
diagrams which, summed up in all orders, yield a whole
Taylor series and therewith an actual level shift. Figure 2(b)
shows a diagram of order $2n + 2$, contributing to $(K^{(2n+2)}_{ab}^{bb'})$, which dresses Fig. 2(a) by $k$ charge fluctuations in the final state $b$ ($“bubbles“$ on the upper part of the contour) and $n$–$k$ charge fluctuations in the initial state $a$ ($“bubbles“$ on the lower part of the contour). We thereby look at fluctuations isolated, in the sense of separated in time, from each other: each bubble must start and end at consecutive
times $\tau_i$ and $\tau_{i+1}$ ($1 \leq i \leq 2n - 1$). The electron transfer event is initialized at the earliest time $\tau = \tau_0$ and ends at the latest time $\tau_{2n+1} = \tau_1$. This gives ($\binom{n}{i}$) possibilities for the time ordering of the bubbles among the upper and lower parts of the contour. Summing all those plus their hermitian conjugates, the total contribution is, as obtained by applying diagrammatic rules.
Here \( c_j \) and \( c_j' \) serve as placeholders for the possible intermediate states; \( f_j^\beta(\omega) := f_c(\beta \omega + \beta e V_i), p = \pm, \) where we defined \( f^+(\omega) = f(\omega) = 1/(1 + e^{\omega}) \), denoting the Fermi function, while \( f^-(\omega) = f(\omega) \). The lead indices \( l, l', l_j \in \{s, d\} \) are summed over, and the lead energies \( \omega, \omega_0, \) and \( \omega_j \) are integrated over. The values of \( p \) and \( \sigma \) are set by fixing the initial and final states \( a \) and \( b \). Likewise the values of the spin indices \( \sigma_j \) and \( \sigma \) and of the Fermi function labels \( p_j \) and \( p_i \) depend on the intermediate states \( c_j \) and \( c_j' \) that can be reached from \( b \) and \( a \), respectively, and are summed over. Finally, there are tunnel matrix elements characterizing transitions on the dot, \( T_{l\sigma, \sigma}(b, a) \sim T_{l}(\tilde{r}_i)(b)|\psi(\tilde{r}_i)(a) \rangle \) (\( \tilde{r}_i \) characterizes the position of lead \( l \)). A multitude of terms emerges from the expression Eq. (6) when applying the decomposition

\[
\lim_{\eta \to 0} \int d\bar{\omega} \frac{D_{l\sigma}(\bar{\omega}) f^\beta(\bar{\omega})}{\bar{\omega} - \bar{\mu} + i\eta} = \int d\omega \frac{D_{l\sigma}(\omega) f^\beta(\omega)}{\omega - \tilde{\mu}} - i\pi \frac{D_{l\sigma}(\tilde{\mu}) f^\beta(\tilde{\mu})}{\omega - \tilde{\mu}},
\]

for all fractions and expanding the product \( \langle \cdots \rangle \) denotes a principal part integration; \( \bar{\omega} \in \{\omega, \omega_0, \omega_j\} \), etc.). We want to focus on a certain contribution, namely the combination where the fraction containing merely \( \omega \),

\[
\left( \frac{1}{p\omega - E_{ba} + i\eta} \right)^{n+1} = \frac{1}{n!} \frac{d}{d\omega} \left( \frac{1}{\omega - E_{ba} + i\eta} \right),
\]

has been replaced by the \( \delta \) function and all others by their principal parts.

In terms of \( f^{(\sigma)}(\omega) = \frac{d}{d\omega} f(\omega) \) it reads

\[
\frac{2\pi}{h} n! \int f^{(\sigma)}(\beta E_{ba} + \beta e V_i) D_{l\sigma} \left| T_{l\sigma}(b, a) \right|^2 \beta^\sigma \binom{n}{k} \times \text{Re} \left[ \prod_{j=1}^{k} \int \frac{d\omega_j}{\omega_j - \beta E_{c_j - p_j e V_i}} \left| T_{l\sigma}(c_j, b) \right|^2 \right] \times \text{Re} \left[ \prod_{j=1}^{n-k} \int \frac{d\omega_j}{\omega_j - \beta E_{a_j' + p_j e V_i}} \left| T_{l\sigma}(c_j', a) \right|^2 \right].
\]

Exhausting all possibilities of choosing the states \( c_j \) and \( c_j' \) as well as setting \( k = 1, \ldots, n \), it is clear that one generates all kinds of terms appearing in an expansion of the power \( n \) of a quantity \( h_{ba}^c \) defined as

\[
h_{ba}^c = \sum_{l} \left( \sum_{c} \left| T_{l\sigma}(c, b) \right|^2 \int d\omega - \beta E_{ac} + p\beta e V_i \right),
\]

where \( c \) and \( c' \) run over all states connected to \( b \), respectively, \( |a\rangle \) via a charge fluctuation. In total, we obtain the \( n \)th term \( \frac{2\pi}{h} n! \int f^{(\sigma)}[\beta(E_{ba} + e V_i)]\left| T_{l\sigma}(b, a) \right|^2 D_{l\sigma}(\beta h_{ba}^c) \) in the Taylor expansion of a Fermi function \( \frac{2\pi}{h} \int f^{(\sigma)}[\beta(E_{ba} + e V_i + h_{ba}^c)]\left| T_{l\sigma}(b, a) \right|^2 D_{l\sigma} \). So, effectively, the considered contribution of the initial and final state charge fluctuations renormalizes any energy difference \( E_{ba} \) to \( E_{ba} + h_{ba}^c \). From comparison with Eq. (6), we can extract a renormalization to a many-body energy \( E_b \) by looking at all the possible fluctuations on the upper contour:

\[
\delta E_b^{(c)} = \sum_{l} \sum_{c} \left| T_{l\sigma}(c, b) \right|^2 \int d\omega - \frac{f^{(\sigma)}(\omega) D_{l\sigma}(\omega)}{p\omega - E_{ch}},
\]

where the values of \( p \) and \( \sigma \) are settled\(^{22} \) once the state \( c \) is assigned. For the case of the single impurity Anderson model this expression reduces to the result for the level shift obtained in perturbation theory.\(^{10,11} \) Hence Eq. (8) provides a diagrammatic interpretation of these results. As temperature goes to zero, these diagrams diverge logarithmically at the charge-degeneracy points, which gives a parametric selection of these specific diagrams.

For a multilevel quantum dot, the summation over the intermediate virtual states \( c_j \) implies that the amplitude of the level shifts is expected to vary from a resonance peak to the other, as also shown in Figs. 4, 5, and 6. Notice also that for the CNT used in our calculations, the summation over the virtual states also includes states with bosonic excitations as described by the bosonic Hamiltonian \( \hat{H}_B \) in Eq. (3).

### B. Flat band with Stoner shift

In order to better analyze the contribution of majority and minority spins, we consider for the evaluation of Eq. (7) the case of a flat band including a Stoner splitting \( \Delta_S \), i.e., \( D_{l\sigma}(\omega) \) as given in Eq. (4). Notice that in order to retain holomorphic functions, one can make use of the Fermi function representation of the step functions, \( \theta(\pm x) = \lim_{\beta \to \infty} f^\pm(\beta x) \). Using
then the residue calculus, one obtains
\[ h_{\text{int}}^{ba} = \sum_l \left[ \sum_c D_{1\sigma} \left| T_{1\sigma}^{\text{ib}}(c,b) \right|^2 \tilde{\Psi}_{\sigma,1}^{(0)}(\beta E_{\text{cb}} + p\beta eV_l) \right. \]
\[ \left. - \sum_{c'} D_{1\sigma} \left| T_{1\sigma}^{\text{ib}}(c',a) \right|^2 \tilde{\Psi}_{\sigma,1}^{(0)}(\beta E_{\text{ac}} - p\beta eV_l) \right] \]
where the upper/lower boundary \( \pm W - \sigma \Delta_{\text{St}}/2 \) of the integration enters via
\[ \tilde{\Psi}_{\sigma,1}^{(0)}(x) = \text{Re}\left[ \Psi^{(0)}(0.5 + i\frac{x \pm \beta(W - \sigma \Delta_{\text{St}}/2)}{2\pi}) \right] \]
where \( \Psi^{(0)} \) is the digamma function. An approximation of this result is obtained elegantly when using Eq. (4) to split the integration range of the integrals in Eq. (7) as
\[ \int_{-W - \Delta_{\text{St}}/2}^{W + \Delta_{\text{St}}/2} d\omega = \int_{-W - \Delta_{\text{St}}/2}^{-W + \Delta_{\text{St}}/2} d\omega + \int_{W + \Delta_{\text{St}}/2}^{W - \Delta_{\text{St}}/2} d\omega \]
and noting that in the region at the top/bottom of the band is \( f(\omega) = 0/1 \). Here we get the result
\[ h_{\text{int}}^{ba} = \sum_l \left[ \sum_c D_{1\sigma} \left| T_{1\sigma}^{\text{ib}}(c,b) \right|^2 \tilde{\Psi}_{\sigma,1}^{(0)}(\beta E_{\text{cb}} + p\beta eV_l) \right. \]
\[ \left. - \sum_{c'} D_{1\sigma} \left| T_{1\sigma}^{\text{ib}}(c',a) \right|^2 \tilde{\Psi}_{\sigma,1}^{(0)}(\beta E_{\text{ac}} - p\beta eV_l) \right] \]
\[ \left. + \sum_{l,c} D_{1\sigma} \left| T_{1\sigma}^{\text{ib}}(c,b) \right|^2 \ln \left( \frac{W_l + E_{\text{cb}} + p\beta eV_l}{W_l + E_{\text{cb}} + p\beta eV_l} \right) \right] \]
\[ + \sum_{l,c} D_{1\sigma} \left| T_{1\sigma}^{\text{ib}}(c',a) \right|^2 \ln \left( \frac{W_l - E_{\text{ac}} + p\beta eV_l}{W_l - E_{\text{ac}} + p\beta eV_l} \right) \]
FIG. 3. (Color online) Parallel (\(G_P\)) and antiparallel (\(G_A\)) conductance along with the resulting tunneling magneto-resistance (TMR) for a CNT of 500 nm in length (\(\varepsilon_0 = 3.35\) meV) and charging energy \(E_c = 6.7\) meV. The thermal energy was set to \(k_B T = 0.3\) meV, the lead polarization to \(P = 0.4\), the tunnel broadening to \(\hbar/\Gamma_1 = 3\) \(\mu\)eV, and the bandwidth to \(W = 3\) eV. (a) For a full fourth-order calculation a mirror-symmetric TMR slightly oscillating around a value of 20% is obtained. (b) An equal splitting \(h_{PE} = h_{AE} = 0.4\) meV causes gate asymmetry and negative values of the TMR by the mechanism shown in Fig. 1(c). (c) For \(h_{PE} = 2 h_{AE} = 0.8\) meV, the regularity of the curve is broken and the negative TMR mechanism shown in Fig. 1(b) comes into play. (d) TMR and conductance for \(\Delta_{St} = 0\). We find electron-hole symmetry perfectly preserved, also upon inclusion of virtual excited states (with cutoff set to \(E_0\) and \(2E_0\)). (e) Same as panel (d) but for \(\Delta_{St} = 0.2W\), which breaks electron-hole symmetry. Including the influence of the virtual excited states negative TMR can be reached. (f) Combining intrinsic shifts with external splitting. An asymmetric coupling to the leads can enlarge \((\gamma < 1)\) or diminish \((\gamma > 1)\) the TMR effect.

FIG. 4. (Color online) Gate-voltage dependence of the intrinsic level shift \(h_{ba}\), Eq. (7), for exemplary electron-hole symmetric transitions, see state labels in panel (a). The electron-hole symmetry is nicely reflected in the curves for \(\Delta_{St} = 0\). Moreover, it becomes obvious that \(\Delta_{St}\) acts opposite to an external Zeeman splitting, i.e., increases (decreases) the energy difference for \(\uparrow\) (\(\downarrow\)) transitions. Raising the energy cutoff for the inclusion of excited states, more and more transitions to virtual states will be contributing to the charge fluctuations for the exemplary transitions. (a) Only fluctuations to ground states. (b) Fluctuations to all states within the range \(E_0\). (c) All states within the range \(2E_0\).
can be identified. Note that going beyond a cutoff of 2 of energy shifts at the cutoff of 2 excited states for $\Delta_0$.

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conductances and the TMR curve for former contributions by preserving the mirror symmetry in the virtual states including bosonic excitations.

spin $\pm 1$ (c). Notice the pronounced influence of virtual states including bosonic excitations.

excited states for $\Delta_{St} = 0$ and $\Delta_{St} = 0.2W$, respectively. In the latter case we find that inclusion of virtual transitions to excited states can even yield negative TMR. Notice that, due to the presence of both fermionic and bosonic excitations, the number of available states increases rapidly with energy. For excitations with energy up to 2$\varepsilon_0$ one has already 716 possible states. In Fig. 3(d), we find the TMR already nicely converged at the cutoff of 2$\varepsilon_0$, while in Fig. 3(e) the trend to convergence can be identified. Note that going beyond a cutoff of 2$\varepsilon_0$ is numerically very demanding. Interestingly, the intrinsic gate-dependent contribution, Fig. 3(d), distinguishes itself from all former contributions by preserving the mirror symmetry in the conductances and the TMR curve for $\Delta_{St} = 0$. The reason is that a given electron-hole symmetry in the tunneling results in an electron-hole symmetry for any charge fluctuation (see Sec. IV). This is no longer true if $\Delta_{St} \neq 0$, Fig. 3(e), where we observe a behavior similar to that of a constant external shift but of opposite sign. i.e., $\Delta_{St} \neq 0$ induces a gate constant positive (negative) shift, for $\uparrow$ ($\downarrow$)-mediated transitions, which also breaks the electron-hole symmetry. These statements are confirmed by Figs. 4(a)–4(c) which show the shifts $\delta E_{St}^{\text{in}}$ (Eq. (7), the quantity determining the shift of position of the conductance peaks) for selected electron-hole symmetric transitions $|b\rangle \rightarrow |a\rangle$. Due to the orbital degeneracy of the CNT, the shifts are seen to differ from peak to peak. The trend to convergence is obvious, in particular, around the resonances the respective transitions belong to (curves thickened in the respective gate voltage region).

Finally, Fig. 3(f) combines the impact of the intrinsic shifts with an equal extrinsic splitting $|\delta h_{ext}^{\text{in}}| = |\delta h_{ext}^{\text{out}}|$. The intrinsic effects suffice to change the TMR curve, Fig. 3(b), to a shape observed in experiments, though due to our limitation to the weak coupling regime, quantitative agreement cannot be expected. An asymmetric coupling is found to affect the curve quantitatively, while the qualitative shape is retained. Thereby, $\gamma < 1$ ($\gamma > 1$) relates to an increased (decreased) coupling to the drain contact—the one in which the parallel and antiparallel configurations differ from each other [see Fig. 1(a)]—and enhances (suppresses) the TMR effect. Even for the very small values of $\Gamma_{St}$, used here to justify lowest-order perturbation theory, the difference between Figs. 3(b) and 3(f) reveals a marked influence of the intrinsic, tunneling induced, level shifts.

The dramatic influence of the excited states, and among these, in particular, the bosonic excitations, is further demonstrated in Figs. 5 and 6.

In Fig. 5 the difference of energy shifts $\delta E_{St}^{\text{in}} - \delta E_{St}^{\text{out}}$ [see Eq. (8)] is plotted versus gate voltage for the same states as those used in Fig. 4. Notice that this quantity vanishes for states with total spin 0. For nonzero spin, the shifts exhibit a characteristic gate dependence, which proves to be most sensitive to the presence of bosonic excitations: omitting them (Fig. 5, magenta dotted + cyan dash-dotted) influences the curves not only quantitatively but also qualitatively. Compared to that effect, it plays only a minor role whether the cutoff is chosen to be $\varepsilon_0$ (blue solid) or 2$\varepsilon_0$ (red dashed), and the curves for $\Delta_{St} = 0$ and $\Delta_{St} \neq 0$ practically fall upon one another. In perfect similarity to an externally induced splitting, there is a
factor of 2 between the differences of the energy shifts for the spin-1 and the spin-1/2 states.

In Fig. 6 the quantity $\delta E_b - \delta E_c$ is shown for the case of parallel configuration, for the two states $b = |↑, \downarrow\rangle$ and $b' = |↓, ↓\rangle$ (upper plot) and $b = |↑, ↓\rangle$ and $b' = |↑, ↓\rangle$ (lower plot). These shifts for states with 1 and 3 excess electrons are those which have been measured, e.g., in the Kondo experiments of Refs. 3 and 5. For $\Delta_{S1} = 0$ the result is already converged, as the curves for cutoff energies $E_0$ and $2E_0$ are mostly identical. For $\Delta_{SL} = 0.2W$ the energy difference acquires a negative offset.

VI. CONCLUSIONS

We analyzed the impact of different kinds of effective Zeeman shifts in magnetically coupled multilevel quantum dots, obtaining a characteristic gate dependence and the possibility of negative TMR. In particular, we have provided a systematic way of including the important effects of tunneling-induced level shifts in a transport calculation by identifying a subclass of diagrams, to all orders in the tunneling Hamiltonian, describing charge fluctuations. This infinite class of diagrams is seen to correspond to a perturbative renormalization of the dot spectrum, an effect which is indeed expected on physical grounds. In conventional Feynman diagrammatics, these terms arise from summing up the second-order dot electron self-energy in the Dyson equation. Importantly, this concept generalizes to higher orders: virtual charge fluctuations during inelastic cotunneling (appearing first in sixth order) give a renormalization of the inelastic cotunneling threshold, as already experimentally observed in Ref. 26. In general, a TMR signal will be influenced by many parameters relevant to the given device. Nevertheless, following the lines of the analysis given here for a CNT, it should be possible to disentangle the importance of the various contributions.

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22$T_{ac}^{\mu}(c,b)$ is zero unless the electronic configurations of $b$ and $c$ differ by one electron of spin $\sigma$. Then, $p = sgn(N_r - N_b)$, with $N_\sigma := |\langle a|\hat{N}|\rangle|$
23Equivalently to $\int d\omega f^*(\omega)D_{\sigma}(\omega)/(\omega - \mu)$ one can consider $\lim_{N} \int_{- \infty}^{\infty} d\omega D_{\sigma}(\omega)\int_{- \infty}^{\infty} f_0^1(\beta\omega)e^{i\omega\Gamma}d\omega$
24Notice that $\Gamma_i \sim D_i T_i^2$ and $\Gamma = \sum_i \Gamma_i$