Spin transport across carbon nanotube quantum dots

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Abstract. We investigate linear and nonlinear transport in interacting single-wall carbon nanotubes (SWCNTs) that are weakly attached to ferromagnetic leads. For the reduced density matrix of a SWCNT quantum dot, equations of motion which account for an arbitrarily vectored magnetization of the contacts are derived. We focus on the case of large diameter nanotubes where exchange effects emerging from short-ranged processes can be excluded and the four-electron periodicity at low bias can be observed. This yields in principle four distinct resonant tunnelling regimes, but due to symmetries in the involved groundstates, each two possess a mirror-symmetry. With a non-collinear configuration, we recover at the $4N \leftrightarrow 4N \pm 1$ resonances the analytical results known for the angular dependence of the conductance of a single level quantum dot or a metallic island. The two other cases are treated numerically and show on the first glance similar, yet not analytically describable dependences. In the nonlinear regime, negative differential conductance features occur for non-collinear lead magnetizations.
1. Introduction

Carbon nanotubes are promising candidates for constituents of tomorrow’s electronic nano-devices [1] and thus are the subject of today’s theoretical [2]–[8] and experimental investigations [9]–[18]. A single-wall carbon nanotube (SWCNT) is just formed by the planar honeycomb lattice of graphene, wrapped along one axis such that a closed, seamless cylindrical surface arises [19]. To guarantee this, the axis and the tube diameter cannot be chosen completely arbitrarily, but in principle there are countless possible configurations. Only special ones, however, inherit the metallic properties of graphene, which stem from the fact that in graphene conduction and valence bands touch at the corner points of the first Brillouin zone. Two of those points are independent and labelled Fermi points $\pm \vec{K}_0$. To be metallic, a SWCNT must contain the Fermi points in its reciprocal lattice of allowed momenta and all armchair type tubes, as well as certain types of zigzag configurations [19] fulfil this condition (but curvature effects yield a small bandgap for all SWCNT types except for the armchair ones). For low-energy processes, only momenta close to the Fermi points can contribute, and as the SWCNT diameter is normally a small fraction of the tube length, the size quantization permits many longitudinal modes, but merely one radial mode. This fact makes the carbon nanotube a one-dimensional (1D) conductor. Moreover, the shape of the graphene bandstructure is very beneficial in the considered region: it develops linearly around the Fermi points, which allows to describe the electronic properties of a nanotube by means of the Tomonaga–Luttinger model for interacting fermions [2, 5], involving a bosonization of the electron operators.
Spin-dependent transport is a rapidly expanding research topic. In particular, recent theoretical works have focused on transport across interacting quantum dots [20]–[22], metallic islands [23] and wires [5] with non-collinearly magnetized leads, offering the prospect of spin-sensitive single electron transistors. It has been already realized that interactions [20, 21, 23] as well as reflection processes at the lead-system-interface [22, 24] can strongly influence the spin-accumulation in the dot (island), and hence the properties of the spin-valve transistor. During recent years, there have been various experimental investigations on spin-dependent transport in SWCNTs [11, 13, 14, 17, 18], and a possible measurement set-up is sketched in figure 1. Notice that we include an arbitrary relative magnetization of the leads and do not restrict ourselves to a common limitation of nowadays experiments on SWCNTs, where merely the two discrete states of parallel (P) and antiparallel (AP) contact magnetization can be realized. There have already been experimental studies on non-collinear spin transport in magnetic multilayers [25], so we can expect that in the near future it will be possible to attach arbitrarily polarized contacts to SWCNTs.

A variable of special interest in spin-dependent transport experiments is the so-called tunnelling magnetoresistance (TMR), which we define for collinear contact magnetizations as \( \text{TMR} = \frac{(G_P - G_{AP})}{G_P} \), where \( G_i \) is meant to denote the conductance in the P \((i = P)\) respectively AP \((i = AP)\) configuration. In [17], the observation of quite regular TMR oscillations with the gate voltage is reported, where large changes of almost 20% in the conductance are reached. Moreover, the TMR acquires negative values, which means that the AP conductance can exceed the P one.

So far, spin-polarized transport in interacting SWCNT quantum dots has been considered only for very long tubes, which do not exhibit level quantization and charging effects [5]. For this latter case, a nontrivial dependence of the current on the interaction strength was predicted. Recently, a single impurity Anderson model with four degenerate orbitals was

\[ \frac{R_{AP} - R_P}{R_P} \] }
proposed [22] as a minimal model to understand the aforesaid negative TMR effect reported in [17]. Particularly, the authors could show that it can originate from multiple reflection processes at the SWCNT–contact interfaces.

In this work, a microscopic treatment of spin-dependent transport in SWCNT quantum dots is presented. Specifically, we focus on the low transparency regime, where a weak coupling between tube and contacts is assumed. Therefore, it is justified to treat all Hamiltonians associated with the tunnelling barrier in lowest nonvanishing order. The opposite case of high transparency, i.e. low-ohmic contacts, has already been studied both experimentally [18] and theoretically [26, 29]; it was found that phase shifts picked up during backscattering events at the tube ends yield a Fresnel/Fabry–Perot interference pattern in $V_{\text{gate}} - V_{\text{bias}}$—TMR plots.

Our model takes into account interface reflections, as well as virtual transition processes. Both are relevant exchange effects for spin-dependent transport [23], inducing a precession of the spin accumulating on the quantum dot.

For strict lowest order perturbation theory, nevertheless, the interface reflections cannot be the source of any negative TMR, as will become clear in the course of this paper, which is structured as follows.

In section 2, we describe the model Hamiltonian we use for our system, section 3 explains how to derive the master equation for the SWCNT density matrix in the presence of arbitrary lead magnetizations. Section 4 introduces the necessary coordinate transformations to proper spin quantization axes and subsequently, section 5 contains the results we acquire for the current in the linear transport regime. Section 6 presents numerical data for the nonlinear case and finally section 8 gives a summary of our achievements. In the appendix, some explicit calculations skipped in section 3 can be found.

2. Model Hamiltonian

To build up a model for spin-dependent transport in the quantum dot regime of a SWCNT, we expand the theoretical work [8] on correlated transport in carbon nanotube quantum dots to include spin polarization of both contacts. The magnetizations $\vec{m}_s$ and $\vec{m}_d$ may enclose an arbitrary angle $\theta$, which is a possibility future experiments are likely to offer (see figure 1).

The Hamilton operator we use for the set-up in figure 1 is

$$\hat{H} = \hat{H}_\odot + \sum_{l=s,d} \hat{H}_l + \hat{H}_{Tl} + \hat{H}_{Rl},$$

where effects of external voltages have been absorbed into $\hat{H}_\odot$ and $\hat{H}_l$. The index $l$ labels the source ($l = s$) and the drain ($l = d$) contacts, which are metallic and thus characterized by a Hamiltonian

$$\hat{H}_l = \sum_{\vec{q}} \left( \epsilon_{\vec{q}} - \text{sgn}(\sigma_l)E_{\text{Stoner}} - eV_l \right) \hat{c}^\dagger_{l\vec{q}} \hat{c}_{l\vec{q}}.$$  

In (2), $\hat{c}_{l\vec{q}}$ is the fermionic annihilation operator for an electron of momentum $\vec{q}$ in lead $l$ with spin $\sigma_l \in \{+, -\}$, where $\pm$ denotes the majority/minority spin species for a quantization along $\vec{q}$. For reasons of simplicity, we just apply the Stoner model for our ferromagnetic contacts, but any other description could be used as well. Both the bias voltage $V_l$ and the Stoner exchange splitting energy $E_{\text{Stoner}}$ for the two spin species in the ferromagnetic leads add a shift to the kinetic energy $\epsilon_{\vec{q}}$ of the particle. $e$ is the electron charge.
The tunnelling processes at contact $l$ are modelled by

$$\hat{H}_{Tl} = \sum_{\sigma_l} \int d^3r \left( T_l(\vec{r}) \hat{\Psi}_l^{\dagger}(\vec{r}) \hat{\Psi}_{\sigma_l}(\vec{r}) + h.c. \right).$$ (3)

The so-called transparency $T_l(\vec{r})$ specifies the tunnelling properties of lead $l$, $\hat{\Psi}_{\sigma_l}(\vec{r})$ and $\hat{\Psi}_{\sigma_l}(\vec{r})$ are the electron annihilation operators in real space representation, for a particle of spin $\sigma_l$ in the tube respectively in lead $l$. For later purposes, it is necessary to know the decomposition

$$\hat{\Psi}_{\sigma_l}(\vec{r}) = \int d\epsilon \left( D_{\sigma_l}(\epsilon) \sum_{\vec{q} \downarrow} \phi_{\epsilon \vec{q}}(\vec{r}) \hat{c}_{\epsilon \vec{q} \sigma_l} \right),$$ (4)

where $D_{\sigma_l}$ is the density of states for carriers of spin $\sigma_l$ in lead $l$, taking as an argument the total energy

$$E_{\text{tot}}(\epsilon) = \epsilon - e V_l - \text{sgn}(\sigma_l) E_{\text{Stoner}}$$

of a particle. $\phi_{\epsilon \vec{q}}(\vec{r})$ is just a wavefunction inside the lead.

Furthermore, we allow for boundary backscattering by introducing the ‘reflection’ Hamiltonian

$$\hat{H}_{RL} = -\int d^3r \sum_{\sigma_l} \text{sgn}(\sigma_l) \hat{\Psi}_{\sigma_l}^{\dagger}(\vec{r}) \hat{\Psi}_{\sigma_l}(\vec{r}).$$ (5)

which is equivalent to the momentum space expression used, e.g. in [23] (where also the relation to the mixing conductance introduced in [27] is explained). An electron picks up some phase when it is scattered at the tube ends, which overall results in a certain energy shift, coming with a positive or a negative sign depending on the spin polarization. In other words, being close to a contact, the electron feels a bit of the magnetic field inside it, causing a position-dependent energy splitting $\Delta_l(\vec{r})$ for the two spin species.

We assume the tunnelling barrier to be spin-independent and as there is only weak spin–orbit coupling in clean nanotubes, it is justified not to consider any spin–flip processes. At this stage, we still want to stay general and do not introduce any assumption on the position dependence of $T_l(\vec{r})$ or $\Delta_l(\vec{r})$. Later on, however, we will have to impose the restriction that both parameters are of relevant value only near the contacts.

The most complex contribution to the system Hamiltonian is given by the terms which belong to the SWCNT itself, due to the presence of Coulomb interaction.

The SWCNT Hamiltonian $\hat{H}_C = \hat{T}_C + \hat{V}_C$ reads

$$\hat{H}_C = \hbar v_F \sum_{\vec{r} \sigma_\sigma} \text{sgn}(\vec{r}) \sum_{n \in \mathbb{Z}} n \hat{c}^{\dagger}_{\vec{r} \sigma_\sigma n} \hat{c}_{\vec{r} \sigma_\sigma n}$$

$$+ \frac{1}{2} \sum_{\sigma_\sigma \sigma'} \int d^3r \int d^3r' \hat{\Psi}_{\sigma_\sigma n}^{\dagger}(\vec{r}) \hat{\Psi}_{\sigma_\sigma n}^{\dagger}(\vec{r}') V(\vec{r} - \vec{r}') \hat{\Psi}_{\sigma_\sigma n}(\vec{r}) \hat{\Psi}_{\sigma_\sigma n}(\vec{r}).$$ (6)

Notice that we index the electron operators by a spin $\sigma_\sigma$, $\sigma_\sigma \in \{\uparrow, \downarrow\}$ which refers to an arbitrary, but fixed unique quantization axis inside the tube as e.g. sketched in figure 4.

The first term in (6), $\hat{T}_C$, collects the kinetic energy of particles in the nanotube. To recognize this, one needs some knowledge about the SWCNT bandstructure. We already pointed out in the introduction section that from the graphene lattice, the reciprocal space of an armchair
Figure 2. Bandstructure of a SWCNT with PBC (left) and OBC (right). For (non-interacting) wavefunctions which fulfil OBCs, the spectrum is characterized by two possible bands $\tilde{L}$ and $\tilde{R}$ and momenta $\kappa_n = \hbar v_F n$, $n \in \mathbb{Z}$. The energy levels of left- and right-mover branches can be shifted with respect to each other, which results in a nonzero band offset $\delta$.

SWCNT inherits two Fermi points $\pm K_0$, where valence and conduction bands touch. Transport can take place in the vicinity of these points, around which the bands develop linearly and, due to our restriction to low energies, one-dimensionally. Figure 2 (left) shows the two branches we obtain at each Fermi point when periodic boundary conditions (PBCs) are employed \cite{19}: a left mover band $L$ of negative slope and a right mover band $R$ of positive slope; both slopes have an absolute value $\hbar v_F$, where $v_F$ is the Fermi velocity. By the linear transformations

$$\phi_{\text{OBC}}^\text{OBC} (\vec{r}) := \frac{1}{\sqrt{2}} \left( \phi_{\kappa_n, K_0} (\vec{r}) - \phi_{L[-\kappa_n][-K_0]} (\vec{r}) \right),$$

$$\phi_{\text{OBC}}^\text{L} (\vec{r}) := \frac{1}{\sqrt{2}} \left( \phi_{\kappa_n, K_0} (\vec{r}) - \phi_{R[-\kappa_n][-K_0]} (\vec{r}) \right),$$

wavefunctions $\phi_{rF}$ which fulfil open boundary conditions (OBCs, figure 2 (right))—these are the appropriate ones for the finite-size system the tube represents—are constructed from the usual PBC wavefunctions $\phi_{rF}$ ($r \in \{L, R\}$). The latter can, as worked out in \cite{8}, be freed from the dependence on the momentum $\kappa_n$ by the approximation

$$\phi_{rF} (\vec{r}) \approx e^{i\kappa_n \cdot \vec{x}} \phi_{rF} (\vec{r}).$$

The transformation (7) maps the four $rF$ branches onto two bands $\tilde{L}$ and $\tilde{R}$, whereas the number of admitted momenta $\kappa_n = \hbar v_F n$, $n \in \mathbb{Z}$ doubles \cite{8}.

The second part of (6), $\tilde{V}_\odot$, arises from the strong electron–electron interactions inside the nanotube, where $V (\vec{r} - \vec{r}')$ is the Coulomb potential, including screening. Under the assumption that forward scattering processes are the only relevant ones, this potential part can be simplified enormously. We are allowed to do so if interactions within one of the two graphene sublattices, so-called intra-lattice interactions, cannot be distinguished from inter-lattice interactions. Such an approximation holds for tubes with sufficiently large diameters ($\gtrsim 5$ lattice constants) and length ($\gtrsim 100$ nm) \cite{28}.

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By combining the OBCs with the expression (8) and the linear character of the bands around the Fermi points, the electron operator

$$\hat{\psi}_{\uparrow \sigma n}(\vec{r}) = \sum_{\vec{r} \in [L,R]} \sum_n \psi_{\text{OBC}}^{\vec{r} \sigma n} \hat{c}_{\vec{r} \sigma n}$$

can be expressed in terms of the 1D operator $$\hat{\psi}_{\uparrow \sigma n}(x) = \frac{1}{\sqrt{L_t}} \sum_n \epsilon^{\text{sgn}(F)k_x} \hat{c}_{\vec{r} \sigma n}$$ as:

$$\hat{\psi}_{\uparrow \sigma n}(\vec{r}) = \sqrt{L_t} \sum_{r \in F} \text{sgn}(F) \delta_{r,\vec{r}} \varphi_{\text{sgn}(F)r}(\vec{r}) \hat{\psi}_{\uparrow \sigma n}(x),$$

(9)

with $$r \in \{L, R\}$$, $$+r = r$$, $$-r = \vec{r}$$ ($$\vec{L} = R$$, $$\vec{R} = L$$), $$\delta_{r,\vec{r}} \neq 0$$ if $$(r, \vec{r}) \in \{(L, \vec{L}), (R, \vec{R})\}$$ and $$L_t$$ being the SWCNT length.

Introducing $$\hat{\rho}_{\uparrow \sigma n}(x) := \hat{\psi}^\dagger_{\uparrow \sigma n}(x) \hat{\psi}_{\uparrow \sigma n}(x)$$, $$\hat{V}_\uparrow$$ can be rewritten as:

$$\hat{V}_\uparrow = \frac{1}{2} \sum_{r \neq \vec{r}} \sum_{F \sigma \sigma_0} \int_0^{L_t} \int_0^{L_t} dx \, dx' \, \varphi_{\text{sgn}(F)r}(x) \varphi_{\text{sgn}(F)^{\dagger} \sigma_0}(x') \hat{V}(x, x') \hat{\rho}_{\uparrow \sigma n}(x') \hat{\rho}_{\uparrow \sigma n}(x).$$

(10)

The 1D density operator $$\hat{\rho}_{\uparrow \sigma n}(x)$$ has the convenient property that from its Fourier components, bosonic operators $$\hat{b}_{\sigma n}$$, $$n \in \mathbb{Z}^\uparrow$$ can be constructed, where the ones indexed with $$n > 0$$ stem from $$\hat{\rho}_{\uparrow \sigma n}(\vec{r})$$, the others with $$n < 0$$ from $$\hat{\rho}_{\downarrow \sigma n}(\vec{r})$$. Within the spin-charge-separation model, a unitary transformation to charge- and spin-like excitations is usually performed:

$$\hat{b}_{\sigma n} := \frac{1}{\sqrt{2}} (\hat{b}_{\uparrow n} + \hat{b}_{\downarrow n}), \quad \hat{b}_{\downarrow n} := \frac{1}{\sqrt{2}} (\hat{b}_{\uparrow n} - \hat{b}_{\downarrow n}).$$

It is possible to reexpress $$\hat{H}_\uparrow$$ in terms of these new quantities and, by means of some more linear transformations ($$\hat{a}_{\uparrow n} \rightarrow \cdots \rightarrow \hat{a}_{\downarrow n}, \text{ } j \in \{c, s\}$$) eventually diagonalizing the SWCNT Hamiltonian.

The final form of $$\hat{H}_\uparrow = \hat{T}_\uparrow + \hat{V}_\uparrow$$ is:

$$\hat{H}_\uparrow = \frac{1}{2} E_c \hat{N}_\uparrow^2 + \epsilon_0 \sum_{\sigma \sigma_0} \left( \hat{N}_{\sigma \sigma_0}^2 \right)_{\text{Pauli}} + \delta(\text{sgn}(\vec{r})) \hat{N}_{\uparrow \sigma n} + \sum_{n \neq 0} \epsilon_{j n} \hat{a}_{j n}^\dagger \hat{a}_{j n} .$$

(11)

The first three contributions (11) contain are purely fermionic, as the operator $$\hat{N}_{\uparrow \sigma n}$$ is just defined to count the particles in the band $$\vec{r} \sigma n \in \{ \vec{L}, \vec{L} \downarrow, \vec{R} \uparrow, \vec{R} \downarrow \}$$ and $$\hat{N}_\uparrow \equiv \sum_{\sigma \sigma_0} \hat{N}_{\sigma \sigma_0}$$.

The three summands, as implied in (11), are the charging energy, a term accounting for the Pauli principle if more and more electrons are filled in the same band, and a correction for a potential band offset $$\delta$$, as the energy levels of the $$\vec{L}$$- and $$\vec{R}$$-band might be shifted with respect to each other (illustrated in figure 2). Here, $$\epsilon_0 = \pi \hbar v_f / L_t$$ is the level spacing and $$E_c = W_{00}$$, where $$W_{nn} := \frac{1}{2 \sqrt{2} \pi} \int_0^{L_t} dx \int_0^{L_t} dx' \varphi \cos(n(x + x')) + \cos(n(x - x'))$$. For a typical SWCNT, $$\epsilon_0$$ and $$E_c$$ are both of the order of some meV.

The last term counts the energies of collective, bosonic excitations. The energies $$\epsilon_{cn}$$ are for $$n > 0$$ dependent on the interaction, $$\epsilon_{c+|n|} = |n| \epsilon_0 \sqrt{1 + 8W_{nn} \epsilon_0^{-1}}$$, and are thus called charged modes, while the other neutral modes only scale with the level spacing: $$\epsilon_{c \pm |n|} = \epsilon_c - |n| \epsilon_0$$.

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3. Equations of motion for the reduced density matrix (RDM)

We want to investigate the time evolution of our system consisting of SWCNT quantum dot and leads by using the Liouville equation for its density matrix $\dot{\rho}^l(t)$ in the interaction picture. This representation is well-suited, because we intend to treat both the tunnelling and the reflection Hamiltonian as perturbations $\hat{H}_I = \hat{H}_T + \hat{H}_R$ to $\hat{H}_0 := \hat{H}_\odot + \sum_l \hat{H}_l$. Indeed it is a critical question whether or not $\hat{H}_R$ should be included in $\hat{H}_I$. Considerations in [23] show that the values of the phase shifts picked up during boundary reflections are of the same order as the transmission coefficients, such that a weak conductive coupling will bring about a weak ferromagnetic coupling.

In [22], an Anderson model is used and both the reflection and the tunnelling are treated non-perturbatively.

The equation of motion reads:

$$i\hbar \frac{\partial \hat{\rho}^l(t)}{\partial t} = \left[ \hat{H}^l_I(t), \hat{\rho}^l(t) \right],$$  \hspace{1cm} (12)

where $\hat{H}_I$ had to be transformed into the interaction picture according to $H^l_I(t) = e^{i\hat{H}_0(t-t_0)} \hat{H}_I(t_0) e^{-i\hat{H}_0(t-t_0)}$.

Our final interest is dedicated to transport through the SWCNT quantum dot, thus we would not mind losing all the information about the contacts contained in $\hat{\rho}^l(t)$: it is sufficient to calculate a RDM $\hat{\rho}_\odot^l(t)$, where the lead degrees of freedom have been traced out:

$$\hat{\rho}_\odot^l(t) := \text{Tr}_{\text{leads}} \left( \hat{\rho}^l(t) \right), \hspace{1cm} \text{yielding} \hspace{1cm} i\hbar \frac{\partial \hat{\rho}_\odot^l(t)}{\partial t} = \text{Tr}_{\text{leads}} \left[ \hat{H}^l_I(t), \hat{\rho}^l(t) \right].$$  \hspace{1cm} (13)

3.1. Perturbation theory in the tunnelling and the reflection Hamiltonian

In general, the contacts can be considered as large systems compared to the SWCNT. Besides, in our case of low transparency, the influence of the nanotube on the leads is marginal. That is why they can approximately be treated as reservoirs which stay in thermal equilibrium all the time, and the following ansatz is valid:

$$\hat{\rho}^l(t) = \hat{\rho}_\odot^l(t) \hat{\rho}_s \hat{\rho}_d + \mathcal{O}(T_s, T_d) + \mathcal{O}(\Delta_s, \Delta_d),$$  \hspace{1cm} (14)

where $\hat{\rho}_l$ ($l = s, d$) are the thermal equilibrium density matrices of the source and the drain contact. Putting (14) into (13), we are allowed to neglect the corrections to the factorization, because in the product with $\hat{H}_I$ they all produce a higher order remainder.

Now $\hat{H}_I^l(t)$ contains operators belonging to the contacts: either $\hat{\Psi}_{l\sigma} (\vec{r})$ or $\hat{\Psi}_{l\sigma}^\dagger (\vec{r})$ takes part in each term of $\hat{H}_I^l$ (see equation (3)), while $\hat{H}_R^l$ (equation (5)) involves no lead electron operator at all. Since $\hat{\rho}_s$ and $\hat{\rho}_d$ appear under the trace, they give us the thermally-averaged
Removing the commutators, we obtain small, we may safely average know that the contacts impose large reservoirs to the SWCNT and assume that they induce fast

\[ \hat{I}  \]

with \( \hat{I} \) we have

The time \( t_0 \) is chosen to be the time where the interaction is switched on, such that for \( t' < t_0 \) we have \( \hat{I}(t') = \hat{I}(t') = \hat{I}(t') \equiv 0 \) and hence \( \hat{I}(t') = \rho_{I}(t') \) holds true accurately.

Equation (15) is exact and we are allowed to reinsert it into (12). As we are comfortable with \( [\hat{I}(t), \hat{I}(t)] \), we only replace \( \hat{I}(t) \) in the commutator term involving \( \hat{I}(t) \) and find:

\[
\hat{I}(t) = \hat{I}(t) - \frac{i}{\hbar} \int_{t_0}^{t} dt_{1} \left[ \hat{I}(t_1), \hat{I}(t_1) \right].
\]

The approximation is also valid when respecting the fact that we examine a static dc circuit, where the detailed dynamics on short time intervals need not to be taken into account.

We arrive at the equation of motion we want to work with when we send \( t_0 \rightarrow -\infty \) (because we are merely interested in the long-term behaviour of the system) and use the abbreviation \( t_2 = t - t_1 \):

\[
\hat{I}(t) = \hat{I}(t) - \frac{i}{\hbar} \int_{0}^{t} dt_{2} \left[ \hat{I}(t), [\hat{I}(t), \hat{I}(t), \hat{I}(t)] \right].
\]

Removing the commutators, we obtain

\[
\hat{I}(t) = - \frac{i}{\hbar} \int_{0}^{t} dt_{2} \left[ \hat{I}(t), [\hat{I}(t), \hat{I}(t), \hat{I}(t)] + h.c. \right].
\]

We can now substitute the explicit form of \( \hat{I}(t) \) (given in equation (3)), and of \( \hat{I}(t) \) (from equation (5)) into equation (17) and get rid of the traces. The terms containing \( \hat{I}(t) \) involve no lead operators, such that the trace just makes the product \( \hat{I}(t) \) vanish. For the contributions with \( \hat{I}(t) \), the lead operators must be brought next to the density matrix. This is done by using the cyclic property of the trace and commuting them past two SWCNT operators in each term; this, remarkably, means that it does not play any role whether the operators are commuting or anticommuting.

Then one can introduce the correlation functions

\[
\left\{ \psi_{j}^{\dagger} \psi_{m} \right\}_{\text{th}} := \text{Tr}_{\text{leads}} \left( \psi_{j}^{\dagger} \psi_{m} \hat{I} \right),
\]

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and exploit

\[ \text{Tr}_{\text{leads}} \left( \hat{\Psi}_{l \sigma} \hat{\Psi}_{l' \sigma'} \hat{\rho} \hat{\rho}_d \right) = \text{Tr}_{\text{leads}} \left( \hat{\Psi}_{l \sigma}^\dagger \hat{\Psi}_{l' \sigma'}^\dagger \hat{\rho} \hat{\rho}_d \right) = 0, \]

With the abbreviation

\[ \mathcal{E}_{l \sigma}(\vec{r}, \vec{r}', t_2) := T_1(\vec{r}) T_1^*(\vec{r}') \left\{ \hat{\Psi}_{l \sigma}^\dagger(\vec{r}, t) \hat{\Psi}_{l \sigma}(\vec{r}, t) \hat{\rho}_1^\circ(t) - \text{h.c.} \right\}, \]

\[ \mathcal{F}_{l \sigma}(\vec{r}, \vec{r}', t_2) := T_1^*(\vec{r}) T_1(\vec{r}') \left\{ \hat{\Psi}_{l \sigma}^\dagger(\vec{r}, t) \hat{\Psi}_{l \sigma}(\vec{r}, t - t_2) \hat{\rho}_1^\circ(t) \right\} + \text{h.c.} \]

\[ \mathcal{F}_{l \sigma}^*(\vec{r}, \vec{r}', t_2) \hat{\Psi}_{l \sigma}(\vec{r}, t) \hat{\rho}_1^\circ(t) \hat{\Psi}_{l \sigma}(\vec{r}', t - t_2) - \text{h.c.} \]

equation (17) is changed to:

\[ \dot{\hat{\rho}}_1^\circ(t) = \frac{i}{\hbar} \sum_{l \sigma} \int d^3r \Delta_l(\vec{r}) \text{sgn}(\sigma_l) \left( \hat{\Psi}_{l \sigma}^\dagger(\vec{r}, t) \hat{\rho}_1^\circ(t) \hat{\Psi}_{l \sigma}(\vec{r}, t) - \text{h.c.} \right) \]

\[ - \frac{1}{\hbar^2} \sum_{l \sigma} \int d^3r \int d^3r' \int_0^\infty dt_2 \left\{ \mathcal{E}_{l \sigma}(\vec{r}, \vec{r}', t_2) \hat{\Psi}_{l \sigma}^\dagger(\vec{r}, t) \hat{\Psi}_{l \sigma}(\vec{r}, t) \hat{\rho}_1^\circ(t) \right\} + \text{h.c.} \]

Moreover, we are able to write out the time dependences of the operators according to

\[ \hat{\Psi}_f(t) = e^{i(\hbar/\gamma)\hat{H}_0(t-t')} \hat{\Psi}_f(t') e^{-i(\hbar/\gamma)\hat{H}_0(t-t')} = e^{i(\hbar/\gamma)\hat{H}_f(t-t')} \hat{\Psi}_f(t') e^{-i(\hbar/\gamma)\hat{H}_f(t-t')}, \]

independently of all indices but \( f \in \{ s, d, \circ \} \), because all parts of \( \hat{H}_0 \) besides \( \hat{H}_f \) commute with \( \hat{\Psi}_f \).

3.2. Equations of motion in the SWCNT energy basis

The starting point for this subsection is equation (20), which is used to derive the equations of motion for a single element \( (\hat{\rho}_1^\circ(t))_{nm} \) of the RDM in the SWCNT energy basis.

At first, we apply two more approximations that will ease our tasks considerably.

1. We presume that a SWCNT generally is in a pure charge state, i.e. it is filled with a certain number of electrons \( N \) and thus density matrix elements between states of distinct electron numbers are set to zero (we deal with bias voltages not exceeding the height of the Coulomb diamonds and permanently measure \( N \) in our circuit via the gate voltage).

2. Being interested in the static behaviour of our system, we can neglect fast oscillating terms arising from the exponentials in (21) for \( f = \circ \). This secular approximation completely decouples the time evolution of matrix elements between states degenerate in energy from all matrix elements between states non-degenerate in energy (figure 3).
To calculate the current, merely the diagonal elements of the density matrix, the populations, are required, and as a consequence it is actually sufficient to deal with block matrices $\hat{\rho}_{\Theta N}$, which are restricted to the Hilbert space spanned by states

$$j \in |E_N N| := \left\{ |\vec{N}'\rangle : |\vec{N}'\rangle = N, \langle \vec{N}' | \hat{H} \otimes |\vec{N}'\rangle = E_N \right\}$$

with fixed energy $E_N$ and particle number $N$ (dark squares in figure 3).

Equation (20) then reads, introducing Bloch–Redfield tensors in order to simplify the notation,

$$\left( \frac{1}{\hbar} \frac{\partial}{\partial t} \hat{\rho}_{\Theta N}(t) \right)_{nm} = -\sum_{jj'} R_{nm,jj'}^{NN} \left( \hat{\rho}_{\Theta N}(t) \right)_{jj'} + \sum_{kk'} R_{nm,kk'}^{NN+1} \left( \hat{\rho}_{\Theta E_{N+1}}(t) \right)_{kk'} + \sum_{ii'} R_{nm,ii'}^{NN-1} \left( \hat{\rho}_{\Theta E_{N-1}}(t) \right)_{ii'}.$$

The sums in (22) run over states with fixed particle numbers: $j, j' \in |E_N N\rangle$, $k, k' \in |N+1\rangle$, $i, i' \in |N-1\rangle$, where it is the secular approximation which additionally fixes the energy of $j, j'$.

The Bloch–Redfield tensors are defined as follows:

$$R_{nm,jj'}^{NN} := \sum_{l} \left( \delta_{m,j} \Gamma^{(+)}_{l(nn)} + \delta_{n,j} \Gamma^{(-)}_{l(ji)} \right) + \sum_{l} \sum_{k \in |N+1\rangle} \left( \delta_{m,j} \Gamma^{(+)}_{l(ak)} + \delta_{n,j} \Gamma^{(-)}_{l(jk)} \right),$$

$$R_{nm,ss'}^{NN\pm 1} := \sum_{l} \left( \Gamma^{(+)}_{l(nn)} + \Gamma^{(-)}_{l(nn)} \right),$$

Figure 3. The RDM acquires a block diagonal form. Light squares: particle numbers fixed. Dark squares: also degenerate in energy.
with the rates
\[
\Gamma^{(\sigma)NN+1}_{l(nkk'j)} := \frac{1}{\hbar^2} \sum_{\sigma_l} \int d^3 r \int d^3 r' \left( \hat{\psi}^\dagger_{\sigma_l} (\vec{r}) \right)_{nk} \left( \hat{\psi}^\dagger_{\sigma_l} (\vec{r}') \right)_{k'j} \int_0^\infty dt_2 \mathcal{F}_{l\sigma_l} (\vec{r}, \vec{r}', t_2) e^{i(h/\hbar)(E_j - E_k)t_2},
\]
\[
\Gamma^{(\sigma)NN-1}_{l(nii'j)} := \frac{1}{\hbar^2} \sum_{\sigma_l} \int d^3 r \int d^3 r' \left( \hat{\psi}^\dagger_{\sigma_l} (\vec{r}) \right)_{ni} \left( \hat{\psi}^\dagger_{\sigma_l} (\vec{r}') \right)_{i'j} \int_0^\infty dt_2 \mathcal{E}_{l\sigma_l} (\vec{r}, \vec{r}', t_2) e^{i(h/\hbar)(E_j - E_k)t_2} - \alpha \frac{i}{\hbar} \sum_{\sigma_l} \int d^3 r \Delta_l(\vec{r}) \sgn(\sigma_l) \left( \hat{\psi}^\dagger_{\sigma_l} (\vec{r}) \right)_{ni} \left( \hat{\psi}^\dagger_{\sigma_l} (\vec{r}) \right)_{i'j}. \tag{24}
\]

We would like to point out that up to now, the relations we deduced are very general ones, as we did not exploit any property specific for SWCNTs. Equation (22) already shows the features that are important for us: the time evolution of the diagonal matrix elements of \( \hat{\rho}_{\sigma E N N} \) will couple to some elements of \( \hat{\rho}_{\sigma E N -1} \) and \( \hat{\rho}_{\sigma E N +1} \). Which elements are actually involved can be mapped out by transforming the expression (24) further. First, the explicit form of the correlation functions (18) needs to be determined. This and some consecutive steps are carried out in the appendix, and as a result the rates are changed to (A.2):

\[
\Gamma^{(\sigma)NN+1}_{l(nkk'j)} := \frac{\pi L}{\hbar} \sum_{\tilde{r}} \sum_{\sigma_l} \Phi_l \left( \hat{\psi}^\dagger_{\tilde{r}\sigma_l} \right)_{nk} \left( \hat{\psi}^\dagger_{r\sigma_l} \right)_{k'j} \times \left[ D_{l\sigma_l} (E_{kj}) f_l (E_{kj}) + \alpha \frac{i}{\pi} \left( \int_{-\infty}^\infty \frac{d\epsilon}{\epsilon - E_{kj}} D_{l\sigma_l} (\epsilon) f_l (\epsilon) \right) \right],
\]
\[
\Gamma^{(\sigma)NN-1}_{l(nii'j)} := \frac{\pi L}{\hbar} \sum_{\tilde{r}} \sum_{\sigma_l} \Phi_l \left( \hat{\psi}^\dagger_{\tilde{r}\sigma_l} \right)_{ni} \left( \hat{\psi}^\dagger_{r\sigma_l} \right)_{i'j} \times \left[ D_{l\sigma_l} (E_{ji}) (1 - f_l (E_{ji})) - \alpha \frac{i}{\pi} \left( \int_{-\infty}^\infty \frac{d\epsilon}{\epsilon - E_{ji}} D_{l\sigma_l} (\epsilon) (1 - f_l (\epsilon)) + \frac{1}{\Phi_l} R_l \right) \right]. \tag{25}
\]

Here
\[
f_l (E_{tot}^{(\sigma)l}) = \left( 1 + \exp \frac{E_{tot}^{(\sigma)l} + eV_l - \tilde{E}_{E,l}^{(\sigma)f}}{k_B T} \right)^{-1}
\]

is the Fermi function in lead \( l \) and has arisen from the correlation functions. \( \tilde{E}_{E,l}^{(\sigma)f} \) is the common Fermi level for the two spin species \( \sigma_l = +1 \) and \( \sigma_l = -1 \) in contact \( l \) without any bias voltage applied (for calculations we will assume that source and drain are made of the same metal, which allows to shift the energy scale such that \( \tilde{E}_{E,l}^{(\sigma)f} = \tilde{E}_{E,d}^{(\sigma)f} = 0 \)).

Further, we have abbreviated \( E_{ab} \equiv E_a - E_b \) and \( \Phi_l, R_l \) (A.1) are real values into which the integrations over space, the tunnelling and reflection parameters as well as the wavefunctions from the decompositions (4) and (9) of the electron operators have been absorbed. To do so, the assumption that both \( T_l (\tilde{r}) \) and \( \Delta_l (\tilde{r}) \) are small away from the tunnelling contacts was needed. The new parameters \( \Phi_l \) and \( R_l \) scale with the strength of the tunnelling and the reflection, respectively.

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Finally, \( \oint \) denotes a principal part integration.

Now let us have a closer look at the rates (25). As indicated by the notation, \( \Gamma_{NN+1} \) is related to transitions \( N \to N + 1 \); that is why its real part involves the product of the density of states \( D_{l\sigma} \) and the Fermi function \( f_l \), which refers to the number of electrons that can potentially leave the contact. \( \Gamma_{NN-1} \) correspondingly contains \( 1 - f_l \), which accounts for vacancies in the lead.

The imaginary parts also include a product of Fermi function and density of states, but the energy arguments of the functions are integrated over: there is no restriction to energetically permitted transitions. That is why we call these virtual; the real parts, however, represent processes where the energy is conserved.

Still, the electron operators in equations (25) are given for spins along the quantization axes of the two distinct lead coordinate systems. To go on, we must reexpress them in a common basis.

4. Spin quantization axes

4.1. Non-collinear magnetizations

A clever choice for the SWCNT spin quantization axis \( z_\circ \) is the direction perpendicular to the plane spanned by \( \vec{m}_s \) and \( \vec{m}_d \) [21]: using the coordinate systems drawn in figure 4, we will get particularly nice matrices \( U_{l,\circ} \), which transfer the electron operators \( \hat{\psi}_{\sigma}^\dagger \) to their representation \( \hat{\psi}_{\sigma_0}^\dagger \), where the spin is quantized along \( z_\circ \).

To calculate \( U_{l,\circ} \), we rewrite the 3D coordinate system basis \( \{ \vec{x}_l, \vec{y}_l, \vec{z}_l \} \) of contact \( l \) in the nanotube basis:

\[
(\vec{e}_l)_\circ = U_{l,\circ} (\vec{e}_l)_l = U_{l,\circ} (\vec{e}_\circ)_l,
\]

where \( \vec{e} \in \{ \vec{x}_l, \vec{y}_l, \vec{z}_l \} \). \( U_{l,\circ} \in SO(3) \) and \( (\vec{e})_{l/\circ} \) indicates that \( \vec{e} \) is given in the lead/SWCNT coordinate representation.

The second equality makes clear that the coordinate transformation matrix \( U_{l,\circ} \) is just the 3D rotation matrix that rotates the basis \( \{ \vec{x}_\circ, \vec{y}_\circ, \vec{z}_\circ \} \) of the nanotube coordinate system on to that of lead \( l \), \( \{ \vec{x}_l, \vec{y}_l, \vec{z}_l \} \).

Figure 4. The \( x_\circ \)-axis is chosen such that it bisects the angle \( \theta \) between \( \vec{m}_s \) and \( \vec{m}_d \). In each case, \( z \) is the quantization axis in the corresponding part of the system.
Operator quantum mechanics, we obtain the corresponding transformation matrices for the electron operator $\hat{\psi}_{\vec{r}\sigma}$:

\[
U_{l,0} = U_{\vec{x}_{0}}(\theta_l)U_{\vec{y}_{0}}(\pi/2) \quad \text{with} \quad \theta_l := \begin{cases} \frac{-\theta}{2} & l = s, \\ \frac{+\theta}{2} & l = d. \end{cases}
\]

Here $\vec{x}_{0}$ denotes the $\vec{x}_{0}$ axis, having undergone the first rotation. With some basic quantum mechanics, we obtain the corresponding transformation matrices for the electron operator $\hat{\psi}_{\vec{r}\sigma}$:

\[
U_{l,0} = \begin{pmatrix} 1 & e^{i\theta l/2} & e^{-i\theta l/2} \\ e^{-i\theta l/2} & 1 & e^{i\theta l/2} \\ e^{i\theta l/2} & e^{-i\theta l/2} & 1 \end{pmatrix}, \quad \text{such that}
\]

\[
\begin{pmatrix} \hat{\psi}_{\vec{r}+1} \\ \hat{\psi}_{\vec{r}-1} \end{pmatrix} = U_{l,0}^{-1} \begin{pmatrix} \hat{\psi}_{\vec{r}+1} \\ \hat{\psi}_{\vec{r}-1} \end{pmatrix}.
\]

Using (27) it is straightforward to evaluate

\[
\sum_{\sigma} D_{l\sigma\sigma} \hat{\psi}_{\sigma} \hat{\psi}_{\sigma} = \frac{1}{2} \left( D_{l+1} + D_{l-1} \right) \left[ \hat{\psi}_{\uparrow} \hat{\psi}_{\uparrow} + \hat{\psi}_{\downarrow} \hat{\psi}_{\downarrow} \right] + \frac{1}{2} \left( D_{l+1} - D_{l-1} \right) \left[ e^{i\theta} \hat{\psi}_{\uparrow} \hat{\psi}_{\uparrow} + e^{i\theta} \hat{\psi}_{\downarrow} \hat{\psi}_{\downarrow} \right],
\]

where all uninvolved indices and arguments have been dropped for clarity. Defining

\[
\Phi_{l\sigma\sigma'} := \begin{cases} 1 & \sigma_{0} = \sigma'_{0} \\ e^{i\theta l} & \sigma_{0} = \uparrow, \sigma'_{0} = \downarrow \\ e^{-i\theta l} & \sigma_{0} = \downarrow, \sigma'_{0} = \uparrow \end{cases}
\]

and $D_{l\sigma\sigma'} := \begin{cases} D_{l+1} + D_{l-1} & \sigma_{0} = \sigma'_{0} \\ D_{l+1} - D_{l-1} & \sigma_{0} \neq \sigma'_{0} \end{cases}$

the rates in equation (25) are reformulated for one last time:

\[
\Gamma_{i(\alpha)N+1}^{(\alpha)N+1} \mid_{l(\alpha\alpha')} := \frac{\pi L}{\hbar} \sum_{\vec{r}} \sum_{\sigma\sigma'} \Phi_{l\sigma\sigma'} \left( \hat{\psi}_{\vec{r}\sigma_{0}} \right)_{nk} \left( \hat{\psi}_{\vec{r}\sigma'} \right)_{k'j} \left[ D_{l\sigma\sigma'}(E_{kj}) f_{i}(E_{kj}) 
\right.
\]

\[
+ \alpha \frac{i}{\pi} \left( \int_{-\infty}^{\infty} \frac{D_{l\sigma\sigma'}(\epsilon) f_{i}(\epsilon)}{\epsilon - E_{kj}} \right),
\]

\[
\Gamma_{i(\alpha)N+1}^{(\alpha)N+1} \mid_{l(\alpha\alpha')} := \frac{\pi L}{\hbar} \sum_{\vec{r}} \sum_{\sigma_{1}} \Phi_{*l\sigma\sigma'} \left( \hat{\psi}_{\vec{r}\sigma_{0}} \right)_{n'i} \left( \hat{\psi}_{\vec{r}\sigma'} \right)_{i'j} \left[ D_{l\sigma\sigma'}(E_{ji}) (1 - f_{i}(E_{ji})) 
\right.
\]

\[
- \alpha \frac{i}{\pi} \left( \int_{-\infty}^{\infty} \frac{D_{l\sigma\sigma'}(\epsilon) (1 - f_{i}(\epsilon))}{\epsilon - E_{ji}} + \frac{1}{\Phi_{l}} R_{i} \left( \delta_{\sigma_{0} \uparrow} \delta_{\sigma'_{1} \downarrow} + \delta_{\sigma_{0} \downarrow} \delta_{\sigma'_{1} \uparrow} \right) \right).
\]

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4.2. Collinear magnetizations

Before we explain how to solve the final master equation, we should have a look at the special case of collinear contact configurations, where it is clever to use the common quantization axis of the leads also for the coordinate system inside the SWCNT (figure 5).

The matrices we need for replacing equation (26) when we work with the coordinate systems sketched in figure 5 are

\[
U_{l,\circ} = \begin{cases} 
1 \cos(0) + i\sigma_x \sin(0) & l = d, \text{ or } l = s \wedge \theta = 0, \\
1 \cos(-\pi/2) + i\sigma_x \sin(-\pi/2) & l = s \wedge \theta = \pi.
\end{cases}
\]

(31)

The electron operators transform accordingly,

\[
\begin{pmatrix}
\hat{\psi}^+_{r,+} \\
\hat{\psi}^+_{r,-}
\end{pmatrix} = \begin{cases} 
\begin{pmatrix}
\hat{\psi}^+_{r[\sigma_\circ = \uparrow]} \\
\hat{\psi}^+_{r[\sigma_\circ = \downarrow]}
\end{pmatrix} & l = d, \text{ or } l = s \wedge \theta = 0, \\
-i \begin{pmatrix}
\hat{\psi}^+_{r[\sigma_\circ = \downarrow]} \\
\hat{\psi}^+_{r[\sigma_\circ = \uparrow]}
\end{pmatrix} & l = s \wedge \theta = \pi,
\end{cases}
\]

(32)

and instead of (28) we obtain

\[
\sum_{\sigma_l} D_{l \sigma_l} \hat{\psi}^+_{\sigma_l} \hat{\psi}_{\sigma_l} = \begin{cases} 
D_{l,+} \hat{\psi}^+_{+} \hat{\psi} + D_{l,-} \hat{\psi}^+_{-} \hat{\psi} & l = d, \text{ or } l = s \wedge \theta = 0, \\
D_{l,-} \hat{\psi}^+_{-} \hat{\psi} + D_{l,+} \hat{\psi}^+_{+} \hat{\psi} & l = s \wedge \theta = \pi.
\end{cases}
\]

(33)
The corresponding rates are not much different from (25):

\[
\Gamma^{(\alpha)NN+1}_{I(nkk')j} := \frac{\pi L}{\hbar} \sum_{\tilde{r}} \sum_{\sigma_{\alpha}} \Phi_l \left( \hat{\psi}_{\tilde{r}\sigma_{\alpha}} \right)_{n_k} \left( \hat{\psi}_{\tilde{r}\sigma_{\alpha}}^\dagger \right)_{k'j} \left[ D_{l\sigma_{\alpha}}(E_{kj}) f_l(E_{kj}) \right. \\
+ \alpha \frac{i}{\pi} \left( \int_{-\infty}^{\infty} d\epsilon \frac{D_{l\sigma_{\alpha}}(\epsilon) f_l(\epsilon)}{\epsilon - E_{ji}} \right) \left. \right],
\]

\[
\Gamma^{(\alpha)NN-1}_{I(nii')j} := \frac{\pi L}{\hbar} \sum_{\tilde{r}} \sum_{\sigma_{\alpha}} \Phi_l \left( \hat{\psi}_{\tilde{r}\sigma_{\alpha}} \right)_{n_i} \left( \hat{\psi}_{\tilde{r}\sigma_{\alpha}}^\dagger \right)_{i'j} \left[ D_{l\sigma_{\alpha}}(E_{ji}) \right. \\
- \alpha \frac{i}{\pi} \left( \int_{-\infty}^{\infty} d\epsilon \frac{D_{l\sigma_{\alpha}}(\epsilon) \left(1 - f_l(\epsilon)\right)}{\epsilon - E_{ji}} + \frac{(-1)^{\gamma_l(0)}}{\Phi_l} R_l \right) \left. \right],
\]

with \( \gamma_l(\theta) := \delta_{l,s} \delta_{\theta,\pi} \) and

\[
D_{l\sigma_{\alpha}} = \begin{cases} 
D_{l+} & \sigma_{\alpha} = \uparrow \\
D_{l-} & \sigma_{\alpha} = \downarrow \\
D_{l-} & \sigma_{\alpha} = \uparrow \\
D_{l+} & \sigma_{\alpha} = \downarrow 
\end{cases} \quad l = d, \ \text{or} \quad l = s \land \theta = 0,
\]

(35)

The important point is that in equation (33), no terms with mixed spins occur at all, which will make setting up the necessary equations easier than in the general case for an arbitrary \( \theta \). In particular, all coherences drop out and for this reason, also the imaginary parts the rates (34) still contain will vanish: in our regime of weak coupling, neither the virtual transitions nor the boundary reflections have any influence for collinear configurations.

Now we can start an analytical examination of the low-bias regime, i.e. the linear transport.

5. Linear transport

What we are going to do analytically is to consider linear transport, where \( eV_I \ll k_B T \), at thermal energies \( k_B T \ll \epsilon_0 \leq \epsilon_{jn} \ \forall \ jn \). In this case, both fermionic\(^4\) and bosonic excitations can only be created virtually. That is why \( |\tilde{N}, \tilde{m}\rangle = |\tilde{N}, 0\rangle \) is certainly true for all states contributing to transport in our limit, and caring only about different fermionic configurations we symbolize the associated eigenstates by \( |\tilde{N}\rangle \). Due to the absence of any excitations, the four bands of the SWCNT have to be populated as equally as possible, i.e. their electron fillings \( N_\tilde{L}_1, N_\tilde{L}_2, N_\tilde{R}_1, N_\tilde{R}_2 \) may at most differ by one.

Involved states \( |\tilde{N}\rangle \) are thus for fixed \( |\tilde{N}| \) characterized by the energy

\[
E_{\tilde{N}} = E_N^{(0)} := \min\{E_N : |\tilde{N}| = N\}.
\]

They are the groundstates, i.e. the states with lowest possible energy for a fixed tube filling, and therefore we call them lowest energy (ground)states (LEGs). For a SWCNT without a band

\(^4\) In the presence of fermionic excitations, the four \( |\tilde{r}\sigma_{\alpha}| \) bands are no longer filled as equally as possible: there are at least two bands whose electron fillings differ by more than one. Still, however, it is the lowermost states of each band which must be populated. ‘Holes’ can solely be produced by bosonic excitations.
All possible lowest energy (ground)states for a SWCNT without band offset, filled with $4N + \Delta$ electrons.

Figure 6. All possible lowest energy (ground)states for a SWCNT without band offset, filled with $4N + \Delta$ electrons.

offset $\delta$, figure 6 shows all these LEGs which can, under the given conditions, a priori be involved in transport. We will soon explain that also for the virtual processes, only transitions between the LEGs contribute in the end.

At low-bias voltages, the current flow across the SWCNT is Coulomb blocked, unless the applied gate voltage $V_{\text{gate}}$ aligns the states $|E_N^{(0)} N\rangle$ and $|E^{(0)}_{N+1} N+1\rangle$: resonant tunnelling then permits to add or remove the $N + 1$th inside the SWCNT without any energy cost.

In this paper, we will just consider the situation $\delta = 0$ as shown in figure 6, because this is sufficient for describing a SWCNT also in the case of a finite band offset $\delta > k_B T$ completely. Namely, if the thermal energy cannot overcome the energy gap between the $\tilde{R}$- and the $\tilde{L}$-band, resonant tunnelling can solely be allowed for one of those two bands: at each value of the gate voltage, we just deal with a specific of two independent single level quantum dots, while the other one is not important. Any regime of such a system can be mapped on either the resonance $\Delta = 0 \leftrightarrow \Delta = 1$ or $\Delta = 3 \leftrightarrow \Delta = 0$ for a zero band mismatch.

To calculate the current, we need the populations of the LEGs,

$$P_N = \sum_{n \in \{|E^{(0)}_N N\rangle\}} \left( \hat{\rho}_{E^{(0)}_N N} (t) \right)_{nn}, \quad (36)$$

which represent the probabilities to find the SWCNT filled with $N$ electrons. As we want to examine the dc long-term behaviour of the system, we claim that the populations do not change with time. If the value of $V_{\text{gate}}$ is tuned to admit resonant tunnelling, this stationarity demands:

$$0 = P_N = - \sum_{l=s,d} A_{l}^{N \rightarrow N+1} P_N + \sum_{l=s,d} A_{l}^{N+1 \rightarrow N} P_{N+1}, \quad (37)$$
Here, \( A_{l}^{N \rightarrow N+1} / A_{l}^{N+1 \rightarrow N} \) denote the rates by which electrons tunnel into \((N \rightarrow N+1)\) out of \((N+1 \rightarrow N)\) the nanotube at contact \(l\).

The question is now to which elements of \( \hat{\rho}_{\Omega \Omega}^{1}(t) \), \( \hat{\rho}_{\Omega \Omega+1}^{1}(t) \), \( \hat{\rho}_{\Omega \Omega-1}^{1}(t) \) the time evolutions of the populations, \( P_{N} \) and \( P_{N+1} \), couple. For an answer, the rates—either equation (30) or (34)—must be consulted: we find that their real parts always include Fermi functions with an energy difference as an argument. While \( V_{\text{gate}} \) is chosen such that

\[
\mu_{N+1} := E_{0}^{(0)} - E_{N}^{(0)} < k_{B} T,
\]

all other energy differences are at least of the order of the charging energy \( E_{c} \) and due to the condition \( k_{B} T \ll \epsilon_{0} \approx E_{c} \), the real parts of the corresponding rates are suppressed enormously. Not the rates, but the Bloch–Redfield coefficients (23) enter the equation of motion for the density matrix. By its definition, \( R_{\Omega \Omega+1}^{N \Omega \Omega} \) is real and thus the only nonvanishing contribution is the one of \( R_{\Omega \Omega}^{N \Omega \Omega} \) with \( k, k' \) being LEGs; merely elements of the density matrix \( \hat{\rho}_{\Omega \Omega}^{1}(t) \) are involved. For \( \hat{\rho}_{\Omega \Omega}^{1}(t) \), the Kronecker deltas in \( R_{\Omega \Omega \Omega}^{N \Omega \Omega} \) demand either \( n = j \) or \( m = j' \).

Hence, \( E_{n} = E_{m} = E_{N}^{(0)} \) and consequently \( E_{j} = E_{j}' = E_{N}^{(0)} \) no elements other than the ones of \( \hat{\rho}_{\Omega \Omega}^{1}(t) \) itself are participating.

With the help of figure 6, finally the concerned Bloch–Redfield coefficients can be figured out. Before, however, the role of the electron operators must be revealed. From [8], it can be extracted that:

\[
\begin{pmatrix}
\psi_{\tilde{r} \sigma_{\alpha}} \\
\psi_{\tilde{r} \sigma_{\alpha}'}
\end{pmatrix}_{ab} \begin{pmatrix}
\psi_{\tilde{r} \sigma_{\beta}} \\
\psi_{\tilde{r} \sigma_{\beta}'}
\end{pmatrix}_{bc} = \begin{cases}
\pm 1 / \sqrt{2L_{t}} & \text{if } \tilde{N}_{c} - \tilde{N}_{a} = \tilde{e}_{\tilde{r} \sigma_{\alpha}} - \tilde{e}_{\tilde{r} \sigma_{\alpha}'}, \\
0 & \text{otherwise.}
\end{cases}
\]  

(38)

If nonzero, the sign of (38) depends on whether an even (+) or an odd (−) number of electrons with the same energy sits in the bands prior to \( \tilde{r} \sigma_{\alpha} \). To define prior, one has to choose an arbitrary but fixed ordering of the bands, e.g. \( \tilde{L} \) be prior to \( \tilde{r} \) and \( \tilde{r} \downarrow \) be prior to \( \tilde{r} \uparrow \).

For our case, \( a, b, c \in \{ |E_{N}^{(0)}| \} \) in equation (38). What about \( b \in \{ |N+1| \} \)? Still there is a sum over all \( \Gamma_{niim} \) and \( \Gamma_{nkkm} \) in \( R_{nm, jj}^{N \Omega \Omega} \), with \( i \in \{ |N-1| \} \) and \( \Gamma_{nkkm} \) with \( k \in |N+1| \). For the real parts of the rates, the energy argument of the Fermi functions restricts these \( i \) and \( k \) to LEGs, but for the imaginary parts, this energy argument is integrated over and so there is no forbiddance for fermionic or bosonic excitations a priori. After some closer examination, however, it turns out that really all non-LEG contributions of the four rates \( R_{nm, jj}^{N \Omega \Omega} \) contains drop out exactly. Especially, one has to comprehend that if, for example, in

\[
(\psi_{\tilde{r} \sigma_{\alpha}})_{jk}(\psi_{\tilde{r} \sigma_{\alpha}'}_{kj}'),
\]

\( k \in \{ N+1 \} \) was a non-LEG while \( j \) and \( j' \) are LEGs, the expression can be nonzero only for \( \tilde{r} \sigma_{\alpha} = \tilde{r} \sigma_{\alpha}' \); if we create an electron to reach an excited state and wish to return to a LEG, there is actually no other possibility than to remove again the electron we added. Of course, \( j = j' \) then, such that the concerned rates \( \Gamma_{(+)N+1}^{l(nkk)} \) and \( \Gamma_{(-)N+1}^{l(nkk)} \) in equation (23) become \( \Gamma_{l(nkk)}^{(+)N+1} \) and \( \Gamma_{l(nkk)}^{(-)N+1} \) the imaginary parts of the latter, however, cancel each other.
After all, we can set-up equations (37) for the four different tunnelling regimes $\Delta \leftrightarrow \Delta + 1$, where $\Delta := N \mod 4$. Once we have done so, it is easy to extract the current by just omitting the sum over one of the leads:

$$|I_{\Delta+1}| = e \left| A_\Delta^{\Delta \rightarrow \Delta+1} P_\Delta - A_\Delta^{\Delta \rightarrow \Delta} P_{\Delta+1} \right| = e \left| A_\Delta^{\Delta \rightarrow \Delta+1} P_{\Delta} - A_\Delta^{\Delta \rightarrow \Delta} P_{\Delta+1} \right|. \quad (39)$$

Notice that from here on, we replace the indices $N$ by $\Delta$, because for all the properties we will study just $\Delta := N \mod 4 = \Delta$ matters, and not the actual value of $N$.

For the calculations we state

$$D_{l_{\alpha+}}(\mu_\Delta) + D_{l_{\alpha-}}(\mu_\Delta) =: D_l(\mu_\Delta) \equiv D_l;$$

this is justified since the densities have to be taken at the Fermi edge $E_{F,l}$ of the lead metal and $E_{F,l} \gg \mu_\Delta$, so that we can assume them to be approximately constant within the range we consider.

Moreover we define the contact polarization:

$$\mathcal{P}_l = \frac{D_{l_{\alpha+}} - D_{l_{\alpha-}}}{D_l}.$$  

5.1. Resonant tunnelling regime $\Delta = 0 \leftrightarrow \Delta = 1$

If $\Delta = 0$, there is only one LEG, with all bands equally occupied (see figure 6). We have

$${\hat{\rho}}_{\Delta=0}^1(t) = (\hat{\rho}_{\Delta=0}^1(t))_{11} =: P_0,$$

which is the probability to find the SWCNT filled with $N = 4\tilde{N}$, $\tilde{N} \in \mathbb{N}$ electrons. Note that for the new variables, we drop the argument $(t)$ to save some space in the following.

For $\Delta = 1$, things are a bit more complicated, because the LEG is fourfold degenerate: the excess electron can be placed in any one of the four bands $\hat{L} \uparrow$, $\hat{L} \downarrow$, $\hat{R} \uparrow$, $\hat{R} \downarrow$; consequently, the density matrix consists of 16 elements. Eight of them, however, can immediately be set to zero, as we know that our system is unpolarized with respect to the $\hat{L}$- and $\hat{R}$-bands. This forbids any transitions and thus coherences between those two bands:

$$\hat{\rho}_{\Delta=1}^1(t) = \begin{pmatrix} 0 & \hat{\rho}^1_{\Delta=1,\hat{L}}(t) \\ \hat{\rho}^1_{\Delta=1,\hat{R}}(t) & 0 \end{pmatrix}.$$  

Here, due to the indistinguishability of the $\hat{L}$- and $\hat{R}$-bands, we are allowed to set

$$\hat{\rho}^1_{\Delta=1,\hat{L}}(t) = \hat{\rho}^1_{\Delta=1,\hat{R}}(t) = \begin{pmatrix} P_\downarrow^{(1)} e^{i\vartheta^{(1)}} \\ P_\downarrow^{(1)} e^{-i\vartheta^{(1)}} \end{pmatrix},$$

where $P_\downarrow^{(1)}$, $P_\uparrow^{(1)}$ are the probabilities to find the SWCNT in a single-electron spin-down ($\downarrow$) respectively spin-up ($\uparrow$) state. Correspondingly,

$$P_1 := 2P_\downarrow^{(1)} + 2P_\uparrow^{(1)}$$

is the total occupation probability for one electron.

Furthermore, the density matrix is Hermitian and that is why we could define

$$(\hat{\rho}^1_{\Delta=1,\hat{R}}(t))_{12} = (\hat{\rho}^1_{\Delta=1,\hat{R}}(t))_{21}^* =: P_1^{(1)} e^{i\vartheta^{(1)}}$$

for the off-diagonal elements.
The meaning of these quantities is revealed when we extract the information about the average spin $\vec{S}^{(1)}$ on the quantum dot from $\hat{\rho}_{\Delta=1}$; a single spin-1/2 particle that can either be in the spin-up or the spin-down state would be described by a $2 \times 2$ density matrix and the average spin would be given by a trace with the Pauli matrices. Now we have additionally the two $\vec{r}$-bands, but those decouple completely, such that we can obtain the components of the average spin by

$$ S_j^{(1)} = \frac{1}{2} \text{Tr} \left( \left( \begin{array}{cc} \sigma_j & 0 \\ 0 & \sigma_j \end{array} \right) \hat{\rho}_{\Delta=1} \right), \quad j \in \{x, y, z\}, $$

where $\sigma_j$ are the Pauli matrices, and therefore

$$ S_x^{(1)} = \frac{1}{2} \left( 2p^{(1)} e^{i \omega t^{(1)}} + 2p^{(1)} e^{-i \omega t^{(1)}} \right) = 2p^{(1)} \cos(\alpha^{(1)}), $$

$$ S_y^{(1)} = \frac{1}{2} \left( 2p^{(1)} e^{i \omega t^{(1)}} - 2p^{(1)} e^{-i \omega t^{(1)}} \right) = -2p^{(1)} \sin(\alpha^{(1)}), $$

$$ S_z^{(1)} = \frac{1}{2} \left( 2P^{(1)} - 2P^{(1)} \right) = P^{(1)} - P^{(1)}. $$

If we chose our spin quantization axis inside the SWCNT such that $\vec{z}_0 \parallel \vec{S}^{(1)}$, then $S_x^{(1)}$ and $S_y^{(1)}$ and with them all off-diagonal elements of $\hat{\rho}_{\Delta=1}$ would vanish: we had a coordinate system where the Hermitian density matrix is diagonalized. For arbitrary $\theta$, we are not able to give the direction of $\vec{S}$ a priori and thus cannot make any use of this insight. For $\theta = 0$ and $\theta = \pi$, however, we have already found the right axis and could obtain diagonal equations when using the rates (34) instead of (30). But actually the $\Delta = 0 \leftrightarrow \Delta = 1$ regime is analytically accessible for any $\theta$ and as it is instructive, we want to set-up the general equations, in terms of the physical quantities $P_0$, $P_1$, $S_x$, $S_y$ and $S_z$, which are five independent variables determining the density matrices $\hat{\rho}_{\Delta=0}$ and $\hat{\rho}_{\Delta=1}$.

The equations for $\dot{P}_0$ and $\dot{P}_1$ are not independent ($\dot{P}_0 = -\dot{P}_1$), but together with the normalization condition

$$ P_0 + P_1 = 1, \quad (40) $$

we have the necessary set of five equations, where the physical meaning of the single terms becomes obvious:

$$ \frac{d}{dt} P_1 = \frac{\pi}{\hbar} \sum_{l=x,d} \Phi_l D_l \left[ \frac{4 f_l(\mu_{\Delta=1}) P_0 - (1 - f_l(\mu_{\Delta=1})) P_1}{\text{electrons tunnelling in}} - \frac{2 P_l (1 - f_l(\mu_{\Delta=1})) (\vec{S}^{(1)} \cdot \vec{m}_l)}{\text{electrons tunnelling out}} \right], $$

$$ \frac{d}{dt} \vec{S}^{(1)} = \frac{\pi}{\hbar} \sum_{l=x,d} \Phi_l D_l \left[ \frac{P_l (2 f_l(\mu_{\Delta=1}) P_0 - \frac{1}{2} (1 - f_l(\mu_{\Delta=1})) P_1 \vec{m}_l)}{\text{spin accumulation}} - \frac{(1 - f_l(\mu_{\Delta=1})) \vec{S}^{(1)}}{\text{spin relaxation}} - \frac{P_l}{\pi} \overline{q}_l(\mu_{\Delta=1}, \mu_{\Delta=2}) (\vec{m}_l \times \vec{S}^{(1)}) \right]. \quad (41) $$

The probability $P_1$ of the SWCNT to be occupied with $4\tilde{N} + 1$ electrons grows with the rate of electrons tunnelling into the tube already containing $4\tilde{N}$ electrons and decays with electrons leaving the quantum dot occupied with $4\tilde{N} + 1$. Additionally, the average spin of the electrons
inside the SWCNT interacts with the magnetic fields in the contacts, which yields a special term accounting for the difference in the chemical potential for spin-up and spin-down electrons.

That is why we have also to solve the second equation for $S^{(1)}$. The time evolution of the average spin is affected by three contributions: the net number of particles accumulating on the tube brings a spin polarized along the lead magnetizations with it; electrons tunnelling out take some spin with them such that the spin inside the tube relaxes, and finally the electrons feel the magnetic field of the contacts, which makes them precess a little bit.

Besides of a factor 2 (due to the fact that we have the distinct $\vec{r}$ bands) and the precise form of $\Psi_l$, these are the very same equations obtained in [21] for a single level quantum dot. The function

$$\Psi_l(\mu_{\Delta=1}, \mu_{\Delta=2}) \equiv \oint d\epsilon \left( \frac{1 - f_l(\epsilon)}{\epsilon - \mu_{\Delta=1}} + \frac{f_l(\epsilon)}{\epsilon - \mu_{\Delta=2}} \right) + \Phi_l^{-1} R_l / \mathcal{P}_l D_l,$$

summarizes all the imaginary contributions the rates contain; this means that it merges our two exchange effects: the interface backscattering processes—which have not been included in [21]—and the virtual transitions. The exchange is responsible for the precession of the total spin on the quantum dot.

The principal part integration appearing in $\Psi_l$ can be evaluated by a trick: for a Lorentzian-like shaped function $L(\epsilon) = L \frac{E_W^2}{\epsilon^2 + E_W^2},$

$$\oint d\epsilon \frac{L(\epsilon) f(\pm \epsilon)}{\epsilon - E} \approx \mp L \ln \frac{E_W}{\max (k_B T, |E|)}.$$

For $E_W \rightarrow \infty$, $L(\epsilon)$ is constant and approximately we may set our function $D_{l+}(\epsilon) = D_{l+}(\epsilon) - D_{l-}(\epsilon) \equiv \mathcal{P}_l D_l$ constant under the principal part integral. This yields for $\Psi_l$:

$$\Psi_l(\mu_{\Delta=1}, \mu_{\Delta=2}) = \ln \frac{\max (k_B T, \mu_{\Delta=2})}{\max (k_B T, \mu_{\Delta=1})} + \frac{R_l}{\Phi_l \mathcal{P}_l D_l}.$$

Why do we employ this estimation for the digamma function [20]? In our analytical results, we will find $\Psi_l$ accompanied by a Fermi function, which at low temperatures dominates the gate voltage evolution of the product. Therefore, we will, though (42) cuts the peaks of the digamma function, without any exception obtain smooth curves (which indicates that we make no noticeable error).

For the simplest case of identical source and drain tunnelling contacts,

$$D_s = D_d =: D, \quad \mathcal{P}_s = \mathcal{P}_d =: \mathcal{P}, \quad \Phi_s = \Phi_d =: \Phi, \quad R_s = R_d =: R,$$

and thus $\Psi_s = \Psi_d =: \Psi$, we can give a nice analytical expression for the low-bias current in dependence on both the magnetization angle $\theta$ and the bias voltage $V_{\text{gate}}$. The zero point of the latter we fix for each resonance $\Delta \leftrightarrow \Delta + 1$ such that $V_{\text{gate}} = 0 \leftrightarrow E_{\Delta+1}^{(0)} - E_{\Delta}^{(0)} = 0$, which means $V_{\text{gate}} = -\mu_{\Delta+1}$:

$$I_{01}(\theta, V_{\text{gate}}, V_{\text{bias}}) = \tilde{I}_{01} \left( 1 - \frac{\mathcal{P}^2 \sin^2 (\theta/2)}{1 + \mathcal{P}^2 [\Phi_s(\mu_{\Delta=1}, \mu_{\Delta=2}) / (\pi^2 f^2(\mu_{\Delta=1}) \cos^2 (\theta/2))]^2} \right) V_{\text{bias}},$$

(43a)
Figure 7. Currents $I_{01}(V_{\text{gate}})$ and $I_{30}(V_{\text{gate}})$ for different values of $\theta$, together with the TMR (44). The symbols represent numerical data, which are perfectly fit by the analytical lines. The corresponding angle-dependent TMR curves are plotted as lines as well.

where $V_d = -V_s = \frac{V_{\text{bias}}}{2}$ and

$$I_{01} = \frac{2\pi^2 e^2}{h k_B T} \Phi D \frac{f(\mu_{\Delta=1}) f(-\mu_{\Delta=1})}{1 + 3 f(\mu_{\Delta=1})},$$

just as was derived in [8] for a SWCNT attached to unpolarized leads. Figure 7(a) shows the $I_{01}$ dependence on $V_{\text{gate}}$ around its resonance for several $\theta$ and the corresponding TMR curves

$$\text{TMR}(\theta) = \frac{I_{\Delta=1}(0, V_{\text{gate}}) - I_{\Delta=1}(\theta, V_{\text{gate}})}{I_{\Delta=1}(0, V_{\text{gate}})}$$

at $\Delta = 0$. Notice that, here, we have defined a TMR for arbitrary angles. Equation (44) expands the definition we gave in the introduction for the special case of $\theta = \pi$.

In figure 8(a), $V_{\text{gate}}$ is fixed at three different values and the normalized current $I_{01}(\theta)/I_{01}(0)$ is plotted against $\theta$. The parameters we employ are given beneath the plots.

Figures 7(b) and 8(b) belong to the $\Delta = 3 \leftrightarrow \Delta = 0$ resonance and are mirror-symmetric to figures 7(a) and 8(a) with respect to $V_{\text{gate}} = 0$. This fact results from the symmetry in the LEGs: for $\Delta = 1$, there is one excess electron that can be put in any of the four bands, and for $\Delta = 3$ it is just the same situation with one hole instead of an electron. The resulting formula for the current is actually:

$$I_{30}(\theta, V_{\text{gate}}, V_{\text{bias}}) = \tilde{I}_{30} \left(1 - \frac{\mathcal{P}^2 \sin^2(\theta/2)}{1 + \mathcal{P}^2 [\mathcal{P}^2(\mu_{\Delta=0}, \mu_{\Delta=1})/\pi^2 f^2(\mu_{\Delta=0})] \cos^2(\theta/2)} \right) V_{\text{bias}},$$

and

$$\tilde{I}_{30} = \frac{2\pi^2 e^2}{h k_B T} \Phi D \frac{f(-\mu_{\Delta=0}) f(\mu_{\Delta=0})}{1 + 3 f(-\mu_{\Delta=0})}.$$
Fig. 8. Normalized currents $I_{01}(\theta)/I_{01}(0)$ and $I_{30}(\theta)/I_{30}(0)$ for different values of $V_{\text{gate}}$.

Table 1. Parameters used for the plots (if not specified differently).

| Parameter                      | Value      |
|-------------------------------|------------|
| Polarization                  | $P = 0.6$ |
| Reflection                    | $R/(\Phi P D) = 0.1 \Phi_0$ |
| Tube length                   | $L = 580 \text{ nm}$ |
| Temperature                   | $T = 20 \text{ mK}$ |
| Thermal energy                | $k_B T \approx 1.73 \mu\text{eV}$ |
| SWCNT charging energy         | $E_c = 9.49 \text{ meV}$ |
| SWCNT level spacing           | $\epsilon_0 = 2.89 \text{ meV}$ |

Besides for the mirror-symmetry with respect to $V_{\text{gate}} = 0$, there is only one more difference between (43a) and (45a): the argument of $\Psi$ is $\mu_{\Delta=2} = E_c - \mu_{\Delta=1}$ for the $\Delta = 0 \leftrightarrow \Delta = 1$ regime, but at $\Delta = 3 \leftrightarrow \Delta = 0$, additionally the level spacing comes in: $\mu_{\Delta=1} = E_c + \epsilon_0 - \mu_{\Delta=1}$. The resulting deviations, however, are washed out by the squares of the trigonometric functions and the polarization in our formula and thus are not noticeable in figures 7 and 8. As already mentioned, the dominating gate voltage dependence of the Fermi function by which $\Psi$ is divided regulates many features of the plotted curves. For instance, it determines the roaming of the peak maxima towards $V_{\text{gate}} = 0$ for $\theta \notin \{0, \pi\}$. In the case of collinear magnetizations ($\theta \in \{0, \pi\}$),...
Figure 9. In the case of symmetric tunnelling contacts, the dip width of the normalized current \( I_{01}(\theta)/I_{01}(0) \) shrinks with growing \( R \).

the maxima positions, which are identical, can be calculated easily by differentiating (43b). They lie a bit off-resonance, at

\[
e\alpha V_{\text{gate}} = \begin{cases} -k_B T \ln 2 \approx -1.2 \, \mu\text{eV}, & \text{for } \Delta = 0 \leftrightarrow \Delta = 1, \\ +k_B T \ln 2 \approx +1.2 \, \mu\text{eV}, & \text{for } \Delta = 3 \leftrightarrow \Delta = 0. \end{cases}
\]

Plugging the currents into equation (44), the evolution of the TMR can be explained. For \( \theta = \pi \), it is constant at the value \( 1 - \mathcal{P}^2 \). For non-collinear contact magnetizations, again the Fermi function in question is the decisive factor: the larger the inverse of its square becomes, the closer \( I_{\Delta\Delta+1}(\theta) \) gets to \( I_{\Delta\Delta+1}(0) \) and the TMR vanishes. In a physical sense, we can imagine the following picture: below the \( \Delta = 0 \leftrightarrow \Delta = 1 \) resonance, a spin-polarized electron which is transferred across the quantum dot will not stay inside the SWCNT too long and thus will not have time to equilibrate its spin, which hinders the transport, yielding a nonzero TMR. Above the resonance, the tube is mostly populated with \( 4N + 1 \) electrons, such that for all but exactly AP configurations, the spin of the excess electron will have equilibrated before it tunnels out, and consequently the non-collinear TMR decreases around the resonance with growing gate voltage. In analogue, the mirror-symmetric behaviour of the \( \Delta = 3 \leftrightarrow \Delta = 0 \) regime can be understood, when thinking in terms of holes rather than electrons.

This also explains the dependence of the normalized currents on \( \theta \) (figure 8). Transport is blocked by an AP polarization of the contacts, and that is why the plots exhibit a dip at \( \theta = \pi \). The more likely a spin-equilibration, the closer \( I_{\Delta\Delta+1}(\theta) \) is to \( I_{\Delta\Delta+1}(0) \), i.e. the narrower the curves get. So for \( \Delta = 0 \), the width of the dip must shrink when raising the gate voltage, while for \( \Delta = 3 \) it is just the other way round.

We see from equation (42) that \( \mathcal{P} \) grows (and hence the dips widths would shrink) with \( R \): the reflection processes fortify the equilibration of spins. As a result, the width of the normalized current curves \( I_{01}(\theta)/I_{01}(0) \) must shrink with increasing \( R \), which can clearly be seen in figure 9.

One should mention at this point that we chose the value of \( R \) (table 1) small enough to see the differences for distinct angles in figures 7 and 8. For real systems, the actual value of \( R \) might very well depend on how the different domains of the ferromagnetic contacts couple to the graphene sublattices and probably \( R \) differs from sample to sample. At the current state of the experimental art, where adjusting a well-defined angle between the lead magnetizations
is still a big challenge, we find it important to visualize the qualitative impact of the different quantities.

We can furthermore give the analytical expressions for the probabilities $P_0$ and $P_1$ to find the SWCNT populated with $N = 4\tilde{N}$ respectively, $N = 4\tilde{N} + 1$ electrons (figure 10):

$$P_0 = \frac{f(-\mu_{\Delta=1})}{1 + 3f(\mu_{\Delta=1})}, \quad P_1 = \frac{4f(\mu_{\Delta=1})}{1 + 3f(\mu_{\Delta=1})}. \quad (46)$$

Of course, the occupation probability for an additional electron grows with the gate voltage and it is worth stressing that equations (46) do not depend on anything other than $V_{\text{gate}}$. The populations become $P_0 = P_1 = \frac{1}{2}$ at $e\alpha V_{\text{gate}} = -k_B T \ln 2$, where also the maximum of the resonance is located.

The components of the average spin in the $\Delta = 1$ state follow simple dependences as well:

$$\langle S^1_x \rangle = 0,$$
$$\langle S^1_y \rangle = \frac{e}{4k_B T} V_{\text{bias}} P_1 \mathcal{P} \sin \frac{\theta}{2} \left( 1 + \mathcal{P}^2 \frac{3^2(\mu_{\Delta=1}, \mu_{\Delta=2}) \cos^2(\theta/2)}{\pi^2 f^2(-\mu_{\Delta=1})} \right)^{-1}, \quad (47)$$
$$\langle S^1_z \rangle = \mathcal{P} \frac{3^2(\mu_{\Delta=1}, \mu_{\Delta=2}) \cos(\theta/2)}{-\pi f(-\mu_{\Delta=1})} \langle S^1_y \rangle. \quad (47)$$

In figure 11, we show $\langle S^1_y \rangle$ and $\langle S^1_z \rangle$ for $V_{\text{gate}} = 0$ and $V_{\text{gate}} = \pm 5.2 \mu\text{eV}$.

The total spin grows with the polarization $\mathcal{P}$ and linearly with both the bias voltage $V_{\text{bias}}$ and the occupation probability $P_1$. Due to the choice for our coordinate system (remember figure 4), the $x_\odot$-component of the average spin is zero. The fact that $\langle S^1_y \rangle$ peaks at $\theta = \pi$ is clear, because the $y_\odot$-components of the lead magnetizations are opposite and scaling with $\sin \frac{\theta}{2}$. It is the spin precession which tilts the accumulated spin out of plane and therewith gives rise to its nonzero $z_\odot$-component; consequently $\langle S^1_z \rangle$ is proportional to $\mathcal{P}$; its sign changes with the one of the cross product in (41), at $\theta = \pi$.

What happens if the transparencies of the two contacts differ? Let us set

$$D_s = D_d = D, \quad \mathcal{P}_s = \mathcal{P}_d = \mathcal{P}, \quad \Phi_s = \varepsilon \Phi_d = \varepsilon \Phi.$$
Only for the P and the AP current, a nice analytical expression can be given:

\[ I_{01}(\theta = 0, V_{\text{gate}}, \varepsilon) = \frac{2\varepsilon}{1 + \varepsilon} I_{01}(\theta = 0, V_{\text{gate}}), \]

\[ I_{01}(\theta = \pi, V_{\text{gate}}, \varepsilon) = \frac{2\varepsilon(1 + \varepsilon)}{((1 - \mathcal{P}) + \varepsilon(1 + \mathcal{P}))(1 + \mathcal{P} + \varepsilon(1 - \mathcal{P}))} I_{01}(\theta = \pi, V_{\text{gate}}), \]

(48)

Obviously, the ratio \( \frac{I_{01}(\theta = \pi, \varepsilon)}{I_{01}(\theta = 0, \varepsilon)} \) is again constant with \( V_{\text{gate}} \) and the positions of the peak maxima are still the same as in the symmetric case.

Figure 12(a) presents the numerical results for the normalized current at two gate voltages (±5.2 µeV), for two strongly differing values of \( R \) (for simplicity, we assumed that the reflection scales with the transparency, i.e. \( R_s/\Phi_s = R_a/\Phi_a =: R/\Phi \)).

We see that the four curves can hardly be distinguished from each other, and especially \( I_{01}(0^\circ, \varepsilon) \geq I_{01}(\theta, \varepsilon) \forall \theta \in [0^\circ, 360^\circ] \): there cannot appear any negative TMR. Figure 12(b) shows the analytical peaks for \( \theta = 0^\circ \) and \( 180^\circ \), along with the numerical data, which additionally gives the peak for \( \theta = 140^\circ \). \( I_{01}(0^\circ, \varepsilon) \) and \( I_{01}(180^\circ, \varepsilon) \) will, as equation (48) tells us, not differ for distinct \( R \). Also for the non-collinear magnetization, altering \( R \) over four orders of magnitude obviously has no notable effect. No negative TMR can be found throughout the full range of the gate voltage, and in particular, the dependence of the non-collinear TMR on the gate voltage is even suppressed in comparison with the symmetric coupling.

5.2. Resonant tunnelling regime \( \Delta = 1 \leftrightarrow \Delta = 2 \)

Because of the large degeneracy of the involved states, the \( \Delta = 1 \leftrightarrow \Delta = 2 \) is not accessible analytically even for the symmetric case, if \( \theta \) is arbitrary. We therefore show numerical data for \( \theta \in [0, \pi] \). The regime \( \Delta = 2 \leftrightarrow \Delta = 3 \) has again a mirror-symmetry to the one we treat here and is therefore omitted. For the P and the AP current, an analytical solution can be provided:

\[ I_{12}(\theta = 0, V_{\text{gate}}, V_{\text{bias}}) = \frac{3\pi^2e^2}{\hbar k_B T} \Phi D \frac{f(\mu_\Delta=2)f(-\mu_\Delta=2)}{2 + f(\mu_\Delta=2)} V_{\text{bias}}, \]

\[ I_{12}(\theta = \pi, V_{\text{gate}}, V_{\text{bias}}) = (1 - \mathcal{P}^2) I_{12}(\theta = 0, V_{\text{gate}}, V_{\text{bias}}), \]

(49)
in the symmetrical case.
The TMR at \( \theta = \pi \) is consequently constant, at the same value as in the \( \Delta = 0 \leftrightarrow \Delta = 1/\Delta = 3 \leftrightarrow \Delta = 0 \) regimes. The curves for \( \theta \neq \pi \), however, qualitatively differ, due to degeneracy in the \( \tilde{r} \) bands, which allows a nonzero average spin in the \( \Delta = 2 \) state. Below the resonance, there is mainly one single excess electron inside the SWCNT, with equilibrated spin. During charge transfer, an additional, spin-polarized electron enters, but it can be the first electron (the one with relaxed spin) which leaves the tube. That is why polarized non-collinear transport is easier, and hence the TMR much smaller, than below the \( \Delta = 0 \leftrightarrow \Delta = 1 \) peak. Approaching the resonance, however, the time intervals between the described process and the next charge transfer shortens so that the \( \Delta = 1 \) state does not persist long enough to allow a sufficient equilibration of the excess electron’s spin. Therefore the TMR increases. Beyond the resonance, the \( \Delta = 2 \) is stable long enough let the total spin of the two electrons equilibrate. Both excess electrons can now be involved in polarized transport and so the TMR drops to a value lower than below the resonance. Still it stays nonzero, as an outtunnelling particle always leaves a single unequilibrated spin inside the SWCNT.

Again, the current peaks are located off-resonance, at

\[
e\alpha V_{\text{gate}} = -k_B T \frac{\ln 1.5}{2} \approx -0.35 \, \mu\text{eV}
\]  

(50)

for \( \theta \in \{0, \pi\} \), as the derivative of (49) reveals. For angles in between, the peak maximum again moves slightly towards \( e\alpha V_{\text{gate}} = 0 \).

The normalized current \( I_{12}(\theta) \) in figure 13(a) looks rather familiar at \( e\alpha V_{\text{gate}} = 0 \), but nevertheless does not follow the analytical law we found for \( I_{01}(\theta) \). Off-resonance, \( I_{12}(\theta) \) obviously no longer exhibits any handsome dependence and as explained, the polarized non-collinear transport is hindered at the resonance so that the width of the dip is for \( e\alpha V_{\text{gate}} = 0 \, \mu\text{eV} \) larger than for both \( e\alpha V_{\text{gate}} = \pm 5.2 \, \mu\text{eV} \).
Figure 13. (a) $I_{12}(\theta)/I_{12}(0)$ and (b) $I_{12}(V_{\text{gate}})$ together with the related TMR around the resonance $\Delta = 1 \leftrightarrow \Delta = 2$. Notice that in contrast to the $\Delta = 0 \leftrightarrow \Delta = 1$ transition, the TMR exhibits a maximum in the vicinity of the conductance maximum.

For an asymmetric set-up, one finds for varying $R$ fewer deviations than in the $\Delta = 0 \leftrightarrow \Delta = 1$ regime. Again, the current in the nearly AP set-up with $\theta = 170^\circ$ hardly differs from the one for an exact AP configuration with $\theta = 180^\circ$ and the gate voltage dependence of the TMR is suppressed. Still, $I_{12}(0^\circ, \varepsilon) > I_{12}(170^\circ, \varepsilon)$ is guaranteed; the equations for the P and AP configurations can again be solved analytically and we recover our previous finding:

$$\frac{I_{\Delta\Delta+1}(\theta = \pi)}{I_{\Delta\Delta+1}(\theta = 0)} = \frac{(1 - \mathcal{P}^2)(1 + \varepsilon)^2}{((1 - \mathcal{P}) + \varepsilon(1 + \mathcal{P}))(1 + \mathcal{P} + \varepsilon(1 - \mathcal{P}))}. \quad (51)$$

So the ratio of P to AP current is independent of the gate voltage and constant even for a possible band offset.

6. Nonlinear transport

Being interested in the nonlinear transport behaviour of the ferromagnetically contacted SWCNT quantum dot, one has to take into account the various non-LEG states of the system, together with all the arising coherences. This makes an analytical calculation impossible, but the numerics delivers the relevant data respecting the influence of both real and virtual transitions with the energy $3\varepsilon_0$ of up to three (bosonic or fermionic) neutral excitations.

An inclusion of higher excited states would multiply the computational cost, but anyway, their contribution for bias voltages below the second excitation is rather small: $k_B T \ll \varepsilon_0$, so that thermal excitation is inhibited and the probability of bias voltage driven multiple subsequent excitations naturally decreases with the amount of required subsequent excitations [8].

Again, we can confine ourselves to distinguishing between the two regimes $\Delta = 0 \leftrightarrow \Delta = 1$ and $\Delta = 1 \leftrightarrow \Delta = 2$, because the symmetry arguments still hold for all states.
Figure 14. Nonlinear current $I_{01}(\theta = 0)$ below the first excitation. For the computation of this plot, transitions with a maximum energy $\epsilon_0$ were taken into account.

Figure 15. TMR vs gate and bias voltage for $\theta = 140^\circ$ and $\theta = 180^\circ$ around the $\Delta = 0 \leftrightarrow \Delta = 1$ resonance. As we have found it in the linear bias regime, figure 7, the non-collinear TMR ($\theta = 140^\circ$) drops from a constant value to zero after the resonance, following the current. For $\theta = 180^\circ$, the TMR is practically symmetric with respect to the resonant tunnelling regime, where it acquires an increased value.

6.1. Resonant tunnelling regime $\Delta = 0 \leftrightarrow \Delta = 1$

Figure 14 shows the current for bias voltages $eV_1 \ll \epsilon_0$, i.e. far below the first excitation. We can nicely see how the Coulomb blockade diamonds for $\Delta = 0$ and 1 emerge; the higher the bias, the wider the range of the gate voltage within which transport is allowed. This brings about the typical rhomb shapes.

Figures 15(a) and (b) are colormap TMR plots for the polarization angles $\theta = 140^\circ$ and $180^\circ$ in the same regime figure 14 covers. In figure 15(a), we regain what we already knew from figure 7 for the non-collinear magnetizations, namely that the TMR drops from a constant value to zero after the resonant tunnelling regime. Figure 15(b), however, shows that in the AP configuration, for higher bias voltages the TMR is not constant with the gate voltage everywhere but increases outside the Coulomb diamonds. Nevertheless, just as for the low bias, TMR ($\theta = 140^\circ$) $< \text{TMR} (\theta = 180^\circ$). Moreover, both cases lack any negative TMR and actually the TMR becomes widely constant for high-bias voltages, as the nonzero currents in between the Coulomb diamonds acquire a constant value. That is why for $eV_1 > \epsilon_0$, we only need to plot the current at one special gate voltage, e.g. $V_{\text{gate}} = 0$ (figure 16).
Nonlinear current $I_{01}(\theta)$ for $\theta \in \{0^\circ, 140^\circ, 180^\circ\}$ and bias voltages exceeding the first neutral excitation. The numerics respected a maximum transition energy of $3\epsilon_0$. For a non-collinear lead magnetization, a negative differential conductance (NDC) appears below the first excitation.

Within the range of the bias voltage figure 16 shows, the curves for $\theta \in \{0^\circ, 180^\circ\}$ reach three different plateaus: below the first excitation, there is a constant current outside the Coulomb blockade diamonds. As soon as the bias voltage is high enough to excite a neutral mode, more states can contribute to transport and the current jumps to a higher value. It is important that from here on, successively by picking up the energy the bias voltage provides, multiple excited states can be generated. The probability to fall back to a less excited state, however, is always larger than the one for creating another additional excitation and therefore it is certainly valid to at an energy of $3\epsilon_0$. Indeed, another slight enlargement takes place at that value of the bias, which provides enough energy to add an additional electron to a state $|N+1, 1\rangle$; this is another allowed transition above the first neutral excitation.

The current at non-collinear magnetizations exhibits a further feature: we find a NDC below the first bosonic excitation. This behaviour becomes more evident for high polarization, but can still be seen for our passably realistic case of $\varphi = 0.6$. The explanation for the occurrence of the NDC is the decaying influence of the virtual transitions with growing bias voltage (due to the energy arguments appearing under the principal part integral, see equation (25)). We learned that the principal part terms narrow the $I_{01}(\theta)$ curve (figure 8), which means that the heavier their influence, the closer the non-collinear current for a certain magnetization angle $\theta$ comes to the maximum current $I_{01}(0^\circ)$. Thus it is clear that $I_{01}(140^\circ)$ approaches the minimal current $I_{01}(180^\circ)$ for higher bias voltages where the principal part is more and more suppressed. Notice that here $R$ has an influence, but as it does not depend on any external voltage, it tends to wipe out the effect.

6.2. Resonant tunnelling regime $\Delta = 1 \leftrightarrow \Delta = 2$

At the $\Delta = 1 \leftrightarrow \Delta = 2$ resonance, most statements from the previous subsection hold true as well. Due to the higher degeneracy of the states involved in transport, the plateau the current reaches is 50% higher than for the $\Delta = 0 \leftrightarrow \Delta = 1$ resonance [8].

For the colourmap TMR plots of figure 18, the same scale as for figure 15 was applied. Again, at $\theta = 140^\circ$ we find an evolution of the TMR we expected from the linear regime, figure 13, and for $\theta = 180^\circ$ we obtain a qualitatively similar picture as figure 15(b) for...
Figure 17. Nonlinear current $I_{12}(\theta = 0)$ below the first excitation, transitions with a maximum energy $\epsilon_0$ taken into account.

Figure 18. TMR vs gate voltage around the $\Delta = 1 \leftrightarrow \Delta = 2$ resonance. Again, the non-collinear TMR for the polarization angles $\theta = 140^\circ$ is similar to the one in the linear bias regime: before dropping to zero after the resonance, it shows a maximum near the conductance maximum. Just as the latter one, the TMR maximum widens with the bias voltage. The collinear TMR is qualitatively equivalent to the $\Delta = 0 \leftrightarrow \Delta = 1$ case of figure 15, but the increment of the TMR for $\Delta = 1 \leftrightarrow \Delta = 2$ is considerably less, namely about 10%, compared to some 40% for figure 15.

$\Delta = 0 \leftrightarrow \Delta = 1$. Figures 15(b) and 18(b), however, differ in quantity: the maximum value ($\sim 0.39$) of the TMR at $\Delta = 1 \leftrightarrow \Delta = 2$ is close to the value 0.36 for the linear bias, while at $\Delta = 0 \leftrightarrow \Delta = 1$ it still rises with the bias voltage from 0.36 to about 0.5. For the higher bias voltage regime, figure 19, we can observe the same facts described in section 6.1. It is worth mentioning that beyond the first bosonic excitation, the differences in the heights of the currents at the two distinct resonances get much smaller, because the number of involved states multiplies in both regimes.

7. Main results

Within the framework of our master equation approach, we could deduce equations for spin-dependent transport across carbon nanotube quantum dots. In the tunnelling regimes belonging to tube fillings $4N \leftrightarrow 4N + 1$ and $4N + 3 \leftrightarrow 4(N + 1)$, the system behaves equivalent to a
Figure 19. Nonlinear current $I_{12}(\theta)$ for $\theta \in \{0^\circ, 140^\circ, 180^\circ\}$ and bias voltages exceeding the first bosonic excitation. The numerics respected a maximum transition energy of $3\epsilon_0$.

8. Conclusion

We have investigated spin-dependent transport in SWCNT quantum dots. A spin-dependent equation for the RDM of a SWCNT weakly contacted to ferromagnetic leads of arbitrary magnetizations was presented. We demonstrated that the SWCNT behaves as a spin-valve single electron transistor and showed analytical and numerical results for the current flow.

Because of the fourfold periodicity for the electron number $N$, it is sufficient to discriminate between tube fillings with different values of $\Delta = N \mod 4$ and, due to mirror-symmetries in the SWCNT eigenstates, we could even restrict our examinations to the tunnelling regimes $\Delta = 0 \leftrightarrow \Delta = 1$ and $\Delta = 1 \leftrightarrow \Delta = 2$.

The analytical analysis in the case of symmetric coupling to the leads resulted for the $\Delta = 0 \leftrightarrow \Delta = 1$ resonance in a formula for the angular dependence of $I_{01}(\theta)$ equivalent to that obtained in [21] and [23], for a single level quantum dot and a metallic island respectively. The total current, due to the existence of the degenerate left and a right mover bands, is twice as large as for a single level quantum dot. The maxima of the current peaks lie slightly off-resonance; the positions for a P and AP magnetization are identical, but on the way from $\theta = 0^\circ$ to $180^\circ$ the maximum moves a bit, following a curve bent towards the resonance gate voltage (in the $I_{01}(V_{\text{gate}})$ diagram). The TMR for $\theta \neq 180^\circ$ changes around the resonance: it smoothly drops from a constant value, for gate voltages below the resonance, to zero. We additionally gave the average spin on our quantum dot SWCNT in the $\Delta = 1$ state and the two occupation probabilities $P_{\Delta=0}$ and $P_{\Delta=1}$, where we find that the latter solely depend on $V_{\text{gate}}$. For a nonzero band offset $0 < k_B T < \delta$ we can apply these results, because it makes the SWCNT at all gate voltages equivalent to a single level quantum dot.
For $\delta \ll \hbar kT (\ll \epsilon_0)$, the resonance regimes $\Delta = 1 \leftrightarrow \Delta = 2$ is more complex. The TMR around the resonance does not simply decay monotonously from one constant value to another, but shows a peak before decreasing. Nevertheless, all TMR curves are strictly positive and also for an asymmetric coupling to the leads, numerical results in the different regimes at $\theta = 140^\circ$ reveal that a non-collinear contact polarization alone cannot produce a negative TMR.

Specifically, we could deduce the general law (51) for the TMR in the case of collinear (P–AP) magnetizations. It shows that under strict lowest order perturbation treatment of both the tunnelling and the reflection parameters, the linear bias P–AP current is even for an arbitrarily asymmetric coupling independent of the gate voltage. In order to reproduce a negative TMR as observed by [17], a spin-dependent energy shift (which can be obtained from a non-perturbative treatment of $\hat{H}_R$) is necessary. Actually, exchange effects emerging from a distinguishability of inter- and intra-lattice interactions can be source of an intrinsic spin-dependent energy shift, as measurements shown in [16] on unpolarized small-diameter tubes and recent theoretical investigations exhibit [28]. The feature, however, is only present at the resonances involving $\Delta = 2$ and therefore is not responsible for the required gate-voltage independent spin-splitting.

For the nonlinear bias voltage regime of the quantum dot SWCNT, the numerical data revealed that the TMR for P–AP configurations is no longer strictly constant, but rises inside the resonant tunnelling regimes, whereas at $\Delta = 0 \leftrightarrow \Delta = 1$, the effect is much more pronounced than at $\Delta = 1 \leftrightarrow \Delta = 2$. The non-collinear TMR at $\theta = 140^\circ$ is similar to the linear bias TMR, but now it becomes obvious that the changes in value take place at the edges of the Coulomb diamonds.

Tracing the current at the resonances to bias voltages exceeding the energy of the first possible excitation, we find for both tunnelling regimes a qualitatively equivalent dependence. Besides the large jump at the first excitation, another small one can be found when the bias voltage reaches a value that provides the energy to enable transitions to states with a second additional electron. A special feature of the non-collinear polarization is that a NDC appears below the first resonance due to the decaying influence of virtual processes.

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**Appendix. Decomposition of the correlation functions**

To rewrite the rates (24),

$$\Gamma^{(\alpha)NN+1}_{l(nkk')}: = \frac{1}{\hbar^2} \sum_{\sigma_l} \int d^3r \int d^3r' \left( \hat{\Psi}^\dagger_{\sigma_l}(\vec{r}) \right)_{nk} \left( \hat{\Psi}_{\sigma_l}(\vec{r}') \right)_{k'j} \int_0^{\infty} dr_2 \mathcal{F}_{l\sigma_l}(\vec{r}, \vec{r}', t_2) e^{\alpha (i/\hbar)(E_l - E_\alpha)t_2},$$

$$\Gamma^{(\alpha)NN-1}_{l(nii')}: = \frac{1}{\hbar^2} \sum_{\sigma_l} \int d^3r \int d^3r' \left( \hat{\Psi}^\dagger_{\sigma_l}(\vec{r}) \right)_{ni} \left( \hat{\Psi}_{\sigma_l}(\vec{r}') \right)_{i'j} \int_0^{\infty} dr_2 \mathcal{E}_{l\sigma_l}(\vec{r}, \vec{r}', t_2) e^{\alpha (i/\hbar)(E_l - E_\alpha)t_2}$$

\[- \alpha \frac{i}{\hbar} \sum_{\sigma_l} \int d^3r \Delta_l(\vec{r}) \text{sgn}(\sigma_l) \left( \hat{\Psi}^\dagger_{\sigma_l}(\vec{r}) \right)_{ni} \left( \hat{\Psi}_{\sigma_l}(\vec{r}') \right)_{i'j},\]
we first have to determine the explicit form of the correlation functions (18):

\[ \langle \hat{\Psi}^{\dagger}_{l\sigma}(\vec{r}) \hat{\Psi}_{l\sigma}(\vec{r}'), -t_2 \rangle_{\text{th}} \] 

\[ \int \text{d} \epsilon \: D_{l\sigma}(E_{l\sigma}^{\text{tot}} | \epsilon) \sum_{\vec{q},l} \phi_{l\vec{q}}(\vec{r}) \phi_{l\vec{q}}^\dagger(\vec{r}') \left\{ \hat{c}_{l\sigma} \hat{c}^\dagger_{l\sigma} e^{-i(\epsilon/E_{l\sigma})} \right\} 
\]

\[ = \int \text{d} \epsilon \: D_{l\sigma}(E_{l\sigma}^{\text{tot}} | \epsilon) \sum_{\vec{q},l} \phi_{l\vec{q}}(\vec{r}) \phi_{l\vec{q}}^\dagger(\vec{r}') \left( 1 - f_i(E_{l\sigma}^{\text{tot}} | \epsilon) \right) e^{-(\epsilon/E_{l\sigma}^{\text{tot}})} t_2, \]

and in analogue

\[ \langle \hat{\Psi}^{\dagger}_{l\sigma}(\vec{r}) \hat{\Psi}_{l\sigma}(\vec{r}') \rangle_{\text{th}} = \int \text{d} \epsilon \: D_{l\sigma}(E_{l\sigma}^{\text{tot}} | \epsilon) \sum_{\vec{q},l} \phi_{l\vec{q}}(\vec{r}) \phi_{l\vec{q}}^\dagger(\vec{r}') f_i(E_{l\sigma}^{\text{tot}} | \epsilon) e^{+(\epsilon/E_{l\sigma}^{\text{tot}})} t_2. \]

Here,

\[ f_i(E_{l\sigma}^{\text{tot}} | \epsilon) = \left( 1 + \exp \frac{E_{l\sigma}^{\text{tot}} | \epsilon + eV_l - \tilde{E}_{E,l}(z)}{k_B T} \right)^{-1} \]

is the Fermi function in lead \( l \), where \( \tilde{E}_{E,l} \) is the common Fermi level for the two spin species \( \sigma_l = + \) and \( \sigma_l = - \) in contact \( l \) without any bias voltage applied.

Additionally inserting the decomposition of the SWCNT electron operator, equation (9), and introducing the quantities

\[ \Phi_{irr}(\epsilon) := \sum_{FF'} \text{sgn}(FF') \int d^3r \int d^3r' T_i(\vec{r}) T_r(\vec{r}') \sum_{\vec{q},l} \phi_{l\vec{q}}^\dagger(\vec{r}) \phi_{l\vec{q}}(\vec{r}') \psi_{[\text{sgn}(F)r]F}(\vec{r}) \psi_{[\text{sgn}(F')r']F}(\vec{r}') \]

and

\[ R_{irr}(\epsilon) := \sum_{FF'} \text{sgn}(FF') \int d^3r \Delta_i(\vec{r}) \bar{\psi}_{[\text{sgn}(F)r]F}(\vec{r}) \bar{\psi}_{[\text{sgn}(F')r']F}(\vec{r}), \]

the rates change to

\[ \Gamma_{l(nkk')j}^{(\alpha)NN+1} := \frac{L_l}{\hbar^2} \sum_{rr'} \sum_{\sigma_i} \int \text{d} \epsilon \: \Phi_{irr}(\epsilon) \left( \hat{\psi}_{\bar{r}_{\sigma}} \right)_{nk} \left( \hat{\psi}^\dagger_{\bar{r}_{\sigma}} \right)_{k'j} \]

\[ \times D_{l\sigma}(E_{l\sigma}^{\text{tot}} | \epsilon) f_i(E_{l\sigma}^{\text{tot}} | \epsilon) e^{+\alpha i/\hbar \left( E_{l\sigma}^{\text{tot}} | \epsilon - E_{l\sigma} \right)} t_2, \]

\[ \Gamma_{l(nii')j}^{(\alpha)NN-1} := \frac{L_l}{\hbar^2} \sum_{rr'} \sum_{\sigma_i} \int \text{d} \epsilon \: \Phi_{irr}^*(\epsilon) \left( \hat{\psi}^\dagger_{\bar{r}_{\sigma}} \right)_{ni} \left( \hat{\psi}_{\bar{r}_{\sigma}} \right)_{i'j} D_{l\sigma}(E_{l\sigma}^{\text{tot}} | \epsilon) \left( 1 - f_i(E_{l\sigma}^{\text{tot}} | \epsilon) \right) \]

\[ \times e^{-\alpha i/\hbar \left( E_{l\sigma}^{\text{tot}} | \epsilon - E_{l\sigma} \right)} t_2 - \frac{iL_l}{\hbar} \sum_{rr'} R_{irr'} \sum_{\sigma_i} \text{sgn}(\sigma_i) \left( \hat{\psi}_{\bar{r}_{\sigma}} \right)_{ni} \left( \hat{\psi}^\dagger_{\bar{r}_{\sigma}} \right)_{i'j}, \]

where \( E_{a} - E_{b} := E_{ab} \), and it was assumed that the tunnelling and the reflection processes take place mainly close to the leads, which justifies to drop the position dependence of the electron operators.

More details on this can be found in the appendix of [8], where it is also nicely explained how some considerations about the main contributions under the integrals in (A.1) and the fact
that the SWCNT is unpolarized with respect to the $\tilde{L}$- and $\tilde{R}$-bands allow to set $\Phi_{l_{rr}'}(\epsilon) = \delta_{rr} \Phi_l$, and actually in analogue $R_{l_{rr}'}(\epsilon) = \delta_{rr} R_l$. Then it is rather easy to carry out the integration $\int d\epsilon \int_0^\infty d\tau_2$.

Any formulary tells us for some real function $G(\epsilon)$:

$$
\text{Re} \left( \int d\epsilon \, G(\epsilon) \int_0^\infty d\tau_2 \, e^{\pm i(\hbar)(\epsilon - E)\tau_2} \right) = \pi \hbar G(E),
$$

$$
\text{Im} \left( \int d\epsilon \, G(\epsilon) \int_0^\infty d\tau_2 \, e^{\pm i(\hbar)(\epsilon - E)\tau_2} \right) = \pm \hbar \int_{-\infty}^\infty d\epsilon \, \frac{G(\epsilon)}{\epsilon - E},
$$

where $\mathcal{P}$ denotes a principal part integration.

So finally:

$$
\Gamma^{(a)NN+1}_{l_{kk'}j} := \frac{\pi L_l}{\hbar} \sum_r \sum_{\sigma_i} \Phi_l \left( \hat{\psi}_{\tilde{r} \sigma_l} \right)_{nk} \left( \hat{\psi}^\dagger_{\tilde{r} \sigma_l} \right)_{k'j} \times \left[ D_{l\sigma_l}(E_{kj}) \left(1 - f_i(E_{kj})\right) - \frac{i}{\pi} \left( \mathcal{P} \int_{-\infty}^\infty d\epsilon \left( D_{l\sigma_l}(\epsilon) f_i(\epsilon) \left(1 - f_i(\epsilon)\right) + \frac{1}{\Phi_l R_l}\right) \right) \right],
$$

$$
\Gamma^{(a)NN-1}_{l_{ii'}j} := \frac{\pi L_l}{\hbar} \sum_r \sum_{\sigma_i} \Phi_l \left( \hat{\psi}_{\tilde{r} \sigma_l} \right)_{ni} \left( \hat{\psi}^\dagger_{\tilde{r} \sigma_l} \right)_{i'j} \times \left[ D_{l\sigma_l}(E_{ji}) \left(1 - f_i(E_{ji})\right) - \frac{i}{\pi} \left( \mathcal{P} \int_{-\infty}^\infty d\epsilon \left( D_{l\sigma_l}(\epsilon) f_i(\epsilon) \left(1 - f_i(\epsilon)\right) + \frac{1}{\Phi_l R_l}\right) \right) \right].
$$

(A.2)

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