Transport through a double-quantum-dot system with noncollinearly polarized leads

I. INTRODUCTION

Spin-polarized transport through nanostructures is attracting increasing interest due to its potential application in spintronics\textsuperscript{1,2} as well as in quantum computing.\textsuperscript{3} Down-scaling magnetoelectronic devices to the nanoscale implies that Coulomb interaction effects become increasingly important.\textsuperscript{4,5} In particular, the interplay between spin-polarization and Coulomb blockade can give rise to a complex transport behavior in which both the spin and the charge of the “information carrying” electron play a role. This has been widely demonstrated by many experimental studies on single-electron transistors (SETs) with ferromagnetic leads, with central element being either a ferromagnetic particle,\textsuperscript{6-8} normal metal particles,\textsuperscript{9,10} a two-level artificial molecule,\textsuperscript{11} a C\textsubscript{60} molecule,\textsuperscript{12} or a carbon nanotube,\textsuperscript{13} showing the increasing complexity and variety of the investigated systems. Initially, the theoretical work was mainly focused on the difference in the transport properties for parallel or antiparallel magnetizations in generic spin-valve SETs.\textsuperscript{14-24} More recently, the interplay between spin and interaction effects for noncollinear magnetization configurations attracted quite some interest both in systems with a continuous energy spectrum,\textsuperscript{25-28} as well as in single-level quantum dots,\textsuperscript{29-36} many-level nanomagnets,\textsuperscript{37} and in carbon nanotube quantum dots.\textsuperscript{38} In the noncollinear case, a much richer physics is expected than in the collinear one. For example, two separate exchange effects have to be taken into account. On the one hand, there is the nonlocal interface exchange, in scattering theory for noninteracting systems described by the imaginary part of the spin-mixing conductance,\textsuperscript{29} which in the context of current-induced magnetization dynamics acts as an effective field.\textsuperscript{30} Such an effective field has been experimentally found to strongly affect the transport dynamics in spin valves with MgO tunnel junctions.\textsuperscript{41} This effect has also been recently involved to explain negative tunneling magnetoresistance (TMR) effects in carbon nanotube spin valves\textsuperscript{13} and called spin-dependent interface phase shifts.\textsuperscript{22,42} The second exchange term is an interaction-dependent exchange effect due to virtual tunneling processes that is absent in noninteracting systems.\textsuperscript{25,28,30,38} This latter exchange effect is potentially attractive for quantum information processing since it allows to switch on and off magnetic fields in arbitrary directions just by a gate electric potential.

Recently, there has been increasing interest in double-quantum-dot systems (that can be realized, e.g., in semiconductor structures\textsuperscript{43} or carbon nanotubes\textsuperscript{44}) as tunable systems attractive for studying fundamental spin correlations. In fact, the exchange Coulomb interaction induces a singlet-triplet splitting, which can be used to perform logic gates.\textsuperscript{45} Moreover, Coulomb interaction together with the Pauli principle can be used to induce spin-blockade when the two electrons have triplet correlations.\textsuperscript{46-49} The Pauli spin-blockade effect can be used to obtain a spin-polarized current even in the absence of spin-polarized leads; it requires a strong asymmetry between the two on-site energies of the left and right dots.

So far, transport through a double-dot (DD) system with spin-polarized leads has been addressed in few theoretical\textsuperscript{23,24,50} and experimental\textsuperscript{11} works, for the case of collinearly polarized leads only. While Ref.\textsuperscript{23} addresses additional Pauli spin-blockade regimes when one lead is half-metallic and one is nonmagnetic, Ref.\textsuperscript{24} focuses on the effects of higher order processes in symmetric DD systems, which can, e.g., yield a zero bias anomaly or a negative tunneling magnetoresistance. In Ref.\textsuperscript{11}, Coulomb blockade spectroscopy is used to measure the energy difference between symmetric and antisymmetric molecular states and to determine the spin of the transferred electron.

In this work, we investigate spin-dependent transport in the so-far unexplored case of a DD system connected to leads with arbitrary polarization direction. Specifically, we focus on the low transparency regime where a weak coupling between the DD and the leads is assumed. Our model takes into account interface reflections as well as exchange effects due to the interactions and relevant for noncollinear polarization. We focus on the case of a symmetric DD, so that rectification effects induced by Pauli spin-blockade are excluded. In the linear transport regime, the conductance is calculated in closed analytic form. This yields four distinct resonant tunneling regimes, but due to the electron-hole sym-
FIG. 1. (Color online) Schematic picture of the model: a double-quantum-dot system attached to polarized leads. The significance of the on-site and intersite interactions $U$ and $V$, respectively, is depicted. The source and drain contacts are polarized and the direction of the magnetizations $\vec{m}_a$ is indicated by the arrows.

The method developed in this work to investigate charge and spin transport is based on the Liouville equation for the reduced density matrix (RDM) in lowest order in the reflection and tunneling Hamiltonians. The obtained equations of motion are fully equivalent to those that could be obtained by using the Green’s function method\textsuperscript{30,51} in the same weak-tunneling limit. The advantage of our approach is that it is, in our opinion, easier to understand and to apply for newcomers, as it is based on standard perturbation theory and does not require knowledge of the nonequilibrium Green’s function formalism.

The paper is organized as follows. In Sec. II, we introduce the model system for the ferromagnetic DD single-electron transistor. In Sec. III, the coupled equations of motion for the elements of the DD reduced density matrix are derived. Readers not interested in the derivation of the dynamical equations can directly go to Secs. IV and V, where results for charge and spin transfer in the linear and nonlinear regimes, respectively, are discussed. Finally, we present results for the transport characteristics in the presence of an external magnetic field in Sec. VI. Conclusions are drawn in Sec. VII.

II. MODEL

We consider a two-level DD, or a single molecule with two localized atomic orbitals, attached to ferromagnetic source and drain contacts and with a capacitive coupling to a lateral gate electrode. The system is described by the total Hamiltonian,

$$\hat{H} = \hat{H}_\odot + \hat{H}_s + \hat{H}_d + \hat{H}_T + \hat{H}_R,$$

accounting for the DD Hamiltonian, the source ($s$) and drain ($d$) leads, and the tunneling ($T$) and reflection ($R$) Hamiltonians. The two contacts are considered to be magnetized along an arbitrary but fixed direction determined by the magnetization vectors $\vec{m}_a$. The two magnetization axes enclose an angle $\Theta \in [0^\circ, 180^\circ]$ (see Fig. 1). The spin quantization axis $\vec{z}_a$ in lead $\alpha$ is parallel to the magnetization $\vec{m}_a$ of the lead. The majority of electrons in each contact will then be in the spin-up state. The Hamiltonians $\hat{H}_s, \hat{H}_d$ that model the source and drain contacts ($\alpha = s, d$) read as follows:

$$\hat{H}_\alpha = \sum_{k \ sacrifice} (\epsilon_{k\alpha} - \mu_\alpha) \hat{c}^\dagger_{k\alpha} \hat{c}_{k\alpha},$$

where $\hat{c}^\dagger_{k\alpha}$ and $\hat{c}_{k\alpha}$ are electronic lead operators. They create, respectively annihilate, electrons with momentum $k$ and spin $\sigma_\alpha$ in lead $\alpha$. The electrochemical potentials $\mu_\alpha = \mu_{0\alpha} + eV_\alpha$ contain the bias voltages $V_s$ and $V_d$ at the left and right lead with $V_s - V_d = V_{\text{bias}}$. There is no voltage drop within the DD. We denote in the following $\epsilon_{k\alpha} - \mu_\alpha = \epsilon_{k\alpha}$. Tunneling processes into and out of the DD are described by $\hat{H}_T$. We denote with $d^\dagger_{a\sigma\alpha}$, $d_{a\sigma\alpha}$ the creation and destruction operators in the DD. We assume that tunneling only can happen between a contact and the closest dot, so that we can use the convention that the lead indices $\alpha = s, d$ correspond to $\alpha = 1, 2$ for the DD. With $t_{\alpha \beta}$ the tunneling amplitude, we find

$$\hat{H}_T = \sum_{a \sigma} \left[ t_{s\alpha} d^\dagger_{a\sigma\alpha} c_{a\sigma\alpha} + t_{d\alpha} c^\dagger_{a\sigma\alpha} d_{a\sigma\alpha} \right].$$

The so-called reflection Hamiltonian $\hat{H}_R$ includes reflection events at the lead-molecule interface.\textsuperscript{30,38} For strongly shielded leads, the overall effect is the occurrence of a small energy shift $\Delta_R$ induced by the magnetic field in the contacts and built up during several cycles of reflections at the boundaries. It reads

$$\hat{H}_R = -\Delta_R \sum_{\alpha = s, d} \left( d_{a\sigma\alpha}^\dagger d_{a\sigma\alpha} - d_{a\sigma\alpha} d_{a\sigma\alpha}^\dagger \right).$$

Finally, the DD Hamiltonian needs to be specified. As the spin quantization axis of the DD, $\vec{z}_\odot$, we choose the direction perpendicular to the plane spanned by $\vec{z}_s$ and $\vec{z}_d$ (Ref. 30) (see Fig. 2). The two remaining basis vectors $\vec{x}_\odot$ and $\vec{y}_\odot$ are along $\vec{z}_s + \vec{z}_d$, respectively along $\vec{z}_s - \vec{z}_d$. The matrices that mathematically describe the above transformations read

$$M_{s \rightarrow \odot} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & e^{i\theta/4} & e^{-i\theta/4} \\ e^{i\theta/4} & +1 & e^{-i\theta/4} \\ e^{-i\theta/4} & e^{i\theta/4} & +1 \end{pmatrix} = M_{d \rightarrow \odot}.$$
ing of the creation operators is defined as $d_{\sigma}^\dagger d_{\sigma}^\dagger d_{\sigma}^2 |0\rangle$.

The DD Hamiltonian then reads

$$
\hat{H}_{DD} = \sum_{\sigma=\uparrow,\downarrow} e_{\sigma} d_{\sigma}^\dagger d_{\sigma} + U \sum_{\sigma=\uparrow,\downarrow} d_{\sigma}^\dagger d_{\sigma} + n_{\sigma} d_{\sigma} + \frac{1}{2} V \sum_{\sigma=\uparrow,\downarrow} n_{\sigma}^2 + V n_{\uparrow} n_{\downarrow},
$$

where the spin index $\sigma$ indicates that the operators are expressed in the spin-coordinate system of the DD. The tunneling coupling between the two sites is $U$, while $V$ and $V$ are on-site and intersite Coulomb interactions. In the following, we consider a symmetric DD with equal on-site energies $e_{\uparrow} = e_{\downarrow}$. Thus, we can incorporate the on-site energies in the parameter $\xi$ proportional to the applied gate voltage $V_{\text{gate}}$.

To understand transport properties of the two-site system in the weak-tunneling regime, we have to analyze the eigenstates of the isolated interacting system. These states, which are expressed in terms of the localized states, and the corresponding eigenvalues are listed in Table I. The eigenvalues of the total spin operator. The ground states of the DD with odd particle number are spin degenerate. In contrast, the ground states with even particle number have total spin $S=0$ and are not degenerate. In the case of the two-particle ground state, the parameters $\alpha_0$ and $\beta_0$ determine whether the electrons prefer to pair in the same dot or are delocalized over the DD structure. Since the eigenstates are normalized to one, the condition $\alpha_0^2 + \beta_0^2 = 1$ holds. The energy difference between the $S=0$ ground state and the triplet is given by the exchange energy,

$$
J = \frac{1}{2} \left( \Delta - U + V \right) = 2 |b| \left( R + \sqrt{1 + R^2} \right),
$$

where $\Delta = 4 |b| (1 + R^2)$ and $R = (V - U)/(4 |b|)$.

Besides the triplet, one observes the presence of higher two-particle excited states with total spin $S=0$. Finally, we remark that $\hat{H}_{T}$ and $\hat{H}_{R}$ contain operators of the DD, $d_{\alpha}^\dagger$ and $d_{\alpha}$, in the spin-coordinate systems of the leads, while $\hat{H}_{\otimes}$ is already expressed in terms of DD operators $d_{\alpha}^\dagger$ and $d_{\alpha}$ with spin expressed in the coordinate system of the DD.

### III. DYNAMICAL EQUATIONS FOR THE REDUCED DENSITY MATRIX

In this section, we shortly outline how to derive the equation of motion for the RDM to lowest nonvanishing order in the tunneling and reflection Hamiltonians. The method is based on the well known Liouville equation for the total density matrix in lowest order in the tunneling and reflection Hamiltonians. Equations of motion for the reduced density matrix are obtained upon performing the trace over the lead degrees of freedom, yielding, after standard approximations, Eqs. (13) and (14) below. In the case of spin-polarized leads, however, it is convenient to express the equations of motion for the RDM in the basis that diagonalizes the isolated system’s Hamiltonian and in the system’s spin quantization axis. After rotation from the leads’ quantization axis to the DD one we obtain [Eq. (21)], which forms the basis of all the subsequent analysis.

Let us start from the Liouville equation for the total density matrix $\hat{\rho}(t)$ in the interaction picture,

$$
\frac{d\hat{\rho}(t)}{dt} = [\hat{H}_{T}(t), \hat{\rho}(t)],
$$

with $\hat{H}_{T}$ and $\hat{H}_{R}$ transformed into the interaction picture by $\hat{H}_{T,R}(t) = e^{-i H_{T,R} t/\hbar} \hat{H}_{T,R}(0) e^{i H_{T,R} t/\hbar}$, where $t_0$ indicates the time at which the perturbation is switched on. Integrating Eq. (7) over time and inserting the obtained expression in the right-hand side of Eq. (7) one equivalently finds

$$
\hat{\rho}(t) = \frac{i}{\hbar} \left[ \hat{H}_{T}(0), \hat{\rho}_{\text{eq}}(0) \right] - \frac{i}{\hbar} \left[ \hat{H}_{T}(t), \hat{\rho}(t_0) \right] - \frac{1}{\hbar^2} \int_{t_0}^{t} dt' [\hat{H}_{T}(t')] \frac{d\hat{\rho}'(t')}{dt'} + \hat{H}_{R}(t) \left[ \hat{H}_{T}^\dagger(t'), \hat{\rho}'(t') \right],
$$

where $\hat{\rho}_{\text{eq}}(0) = i \hbar [\hat{H}_{T}(0), \hat{\rho}(0)]$ and $\hat{\rho}(t) = 1/2 \left( \hat{\rho}(t) + \hat{\rho}^\dagger(t) \right)$.
The time evolution of the RDM,
\[ \dot{\hat{\rho}}_C(t) := \text{Tr}_{\text{leads}}[\hat{\rho}^I(t)], \]
(9)
is now formally obtained from Eq. (8) by tracing out the lead degrees of freedom. To proceed, we make the following standard approximations.

(i) The leads are considered as reservoirs of noninteracting electrons that stay in thermal equilibrium at all times. In fact, we only consider weak tunneling and, therefore, the influence of the DD on the leads is marginal. Hence, we can approximate factorize the density matrix of the total system as
\[ \dot{\hat{\rho}}(t) \approx \dot{\hat{\rho}}_C(t) \hat{\rho}_d, \]
(10)
where \( \hat{\rho}_d \) and \( \hat{\rho}_C \) are time independent and given by the usual thermal equilibrium expression for the contacts \( \hat{\rho}_d = \frac{e^{-\beta E_d}}{Z_{s/d}}, \) where \( \beta \) is the inverse temperature and \( Z_{s/d} \) are the partition sums over all states of lead \( s/d. \)

(ii) We consider the lowest nonvanishing order in \( \hat{H}_{T/R}. \)

(iii) We apply the Markov approximation, i.e., in the integral in Eq. (8), we replace \( \hat{\rho}_C^{-1}(t') \) with \( \hat{\rho}_C(t). \) In other words, it is assumed that the system loses all memory of its past due to the interaction with the lead electrons.

Furthermore, being interested in the long term behavior of the system only, we send \( t_0 \rightarrow -\infty. \) We finally obtain the generalized master equation (GME) for the reduced density matrix,
\[ \dot{\hat{\rho}}_C(t) = -\frac{i}{\hbar} \text{Tr}_{\text{leads}}[\hat{H}_t^R(t),\hat{\rho}_C(t)\hat{\rho}_d] \\
- \frac{1}{\hbar^2} \int_0^\infty dt' \text{Tr}_{\text{leads}}([\hat{H}_t^R(t),[\hat{H}_t^R(t-t'),\hat{\rho}_C(t)\hat{\rho}_d]]). \]
(11)

A. Contribution from the tunneling Hamiltonian

In the following, we derive the explicit expression for the GME in the basis of the isolated DD. For simplicity, we omit the contribution of the reflection Hamiltonian in a first instance. When we shall have obtained the final form of the GME due to the tunneling term, we will see that it is easy to insert the contribution from the reflection Hamiltonian. Let us then start from the tunneling Hamiltonian in the interaction picture,
\[ \hat{H}_t^I(t) = \sum_{\alpha k \sigma} \sum_{i,j} t_{\alpha k} \langle \sigma | \hat{d}^\dagger_{\alpha i} \hat{d}_{\alpha j} \rangle \langle i | \exp[i(\varepsilon_l - \varepsilon_j + \varepsilon_{\alpha k \sigma})] t | j \rangle \]
+ H.c.,
(12)
where \( \langle \sigma | \hat{d}^\dagger_{\alpha i} \hat{d}_{\alpha j} \rangle \) and \( \langle \sigma | \hat{d}^\dagger_{\alpha i} \hat{d}_{\alpha j} \rangle \) are the electron annihilation and creation operators in the spin-quantization axis of lead \( \alpha \) expressed in the basis of the energy eigenstates of the quantum dot system. To simplify Eq. (11), standard approximations are invoked.

(i) The first one is the secular approximation: fast oscillations in time average out in the stationary limit we are interested in and thus can be neglected. Together with the relation \( \text{Tr}_{\text{leads}}(\hat{\rho}_d \hat{\rho}_C \hat{\rho}_d \hat{\rho}_C) = \delta_{kk'} \delta_{\sigma \sigma'} \hat{\delta}_{\alpha \alpha'} \hat{f}_d(\varepsilon_{\alpha k \sigma}), \) where \( \hat{f}_d(\varepsilon_{\alpha k \sigma}) \) is the Fermi function, and the cyclic properties of the trace we get
\[ \dot{\hat{\rho}}_C(t) = -\frac{i}{\hbar} \text{Tr}_{\text{leads}}[\hat{H}_t^R(t),\hat{\rho}_C(t)\hat{\rho}_d] \\
- \frac{1}{\hbar^2} \int_0^\infty dt' \text{Tr}_{\text{leads}}([\hat{H}_t^R(t),[\hat{H}_t^R(t-t'),\hat{\rho}_C(t)\hat{\rho}_d]]). \]
(13)
(ii) For the second approximation, we notice that we wish to evaluate single components \( \langle n| \hat{\rho}^{E_N}_{\alpha \beta} | m \rangle \) of the RDM in the system’s energy eigenbasis. Therefore, we assume that the DD is in a pure charge state with a certain number of electrons \( N \) and energy \( E_N \). In fact, in the weak-tunneling limit, the time between two tunneling events is longer than the time where relaxation processes happen. That is, we can neglect matrix elements between states with different number of electrons and only regard elements of \( \hat{\rho}^{E_N}_{\alpha \beta} \), which connect states with same electron number \( N \) and same energy \( E_N \). So, we can divide \( \hat{\rho}^{E_N}_{\alpha \beta} \) into submatrices labeled with \( N \) and \( E_N \) and find

\[
\hat{\rho}^{E_N}_{\alpha \beta}(t) = \frac{\pi}{\hbar} \sum_{\lambda_j} \left( \sum_{\lambda} \sum_{\lambda' j} \sum_{h, h'} \text{Tr}(\hat{\rho}^{E_N}_{\alpha \beta} | \lambda \rangle \langle \lambda | \hat{\rho}^{E_N}_{\alpha \beta} | \lambda' j \rangle \langle j |) \right) | t |^2 \left\{ \right.

(a) + \left[ f_\alpha(e_j - e_i) D_{\alpha \alpha'}(e_j - e_i) \right] \frac{i}{\pi} \int dE_k \frac{f_\alpha(e_k) D_{\alpha \alpha'}(e_k)}{E_k - e_j + e_i} \left( d_{\alpha \alpha'}^{\alpha}(e_k) \tilde{\rho}^{E_N}_{j \alpha \beta}(t) \right) 

(b) + \left[ (1 - f_\alpha(e_j - e_i)) D_{\alpha \alpha'}(e_j - e_i) \right] \frac{i}{\pi} \int dE_k \frac{(1 - f_\alpha(e_k)) D_{\alpha \alpha'}(e_k)}{E_k - e_j + e_i} \left( d_{\alpha \alpha'}^{\alpha}(e_k) \tilde{\rho}^{E_N}_{j \alpha \beta}(t) \right) 

(c) + \left[ f_\alpha(e_j - e_i) D_{\alpha \alpha'}(e_j - e_i) \right] \frac{i}{\pi} \int dE_k \frac{f_\alpha(e_k) D_{\alpha \alpha'}(e_k)}{E_k - e_j + e_i} \left( E_{ij}^{E_N}(t)(d_{\alpha \alpha'}^{\alpha}(e_k) \tilde{\rho}^{E_N}_{j \alpha \beta}(t)) \right) 

(d) + \left[ (1 - f_\alpha(e_j - e_i)) D_{\alpha \alpha'}(e_j - e_i) \right] \frac{i}{\pi} \int dE_k \frac{(1 - f_\alpha(e_k)) D_{\alpha \alpha'}(e_k)}{E_k - e_j + e_i} \left( E_{ij}^{E_N}(t)(d_{\alpha \alpha'}^{\alpha}(e_k) \tilde{\rho}^{E_N}_{j \alpha \beta}(t)) \right) 

(e) - 2(1 - f_\alpha(e_j - e_i)) D_{\alpha \alpha'}(e_j - e_i) \left( d_{\alpha \alpha'}^{\alpha}(e_j - e_i) \tilde{\rho}^{E_N}_{j \alpha \beta}(t) \right) 

(f) - 2f_\alpha(e_j - e_i) D_{\alpha \alpha'}(e_j - e_i) \left( d_{\alpha \alpha'}^{\alpha}(e_j - e_i) \tilde{\rho}^{E_N}_{j \alpha \beta}(t) \right) \right\}. 

(14)

In Eq. (14), we used the notation \( \tilde{\rho}^{E_N}_{j \alpha \beta} := \langle n| \hat{\rho}^{E_N}_{\alpha \beta} | m \rangle \).

By convention, \( \{ \Sigma_{i, j}, \Sigma_j, \Sigma_{h, h'} \} \) means that in each line [(a)–(f)], we sum over the indices occurring in this line only. Notice that the sum over \( j \) is restricted to states of energy \( E_j = E_N + E_{j \alpha} = E_{\alpha} \). For the states with \( N \pm 1 \) electrons, we have to sum up to all energies; therefore, we indexed the density matrix with \( E_j = E_{\alpha} \) in line (e) and \( E_j = E_{\alpha} \) in line (f). Further, we replaced the sum over \( k \) by an integral, \( \Sigma_k \rightarrow \int dE e^{\alpha}(e_{\alpha}(e_{\alpha} - E)) \), where \( D_{\alpha \beta}(e_{\alpha}) \) denotes the density of states in lead \( \alpha \) for the spin direction \( \sigma \alpha \) and applied the following useful formula:

\[
\int dE e^{\alpha}(e_{\alpha}(e_{\alpha} - E)) = \frac{e^{\alpha}(e_{\alpha}(E))}{\pi \hbar} + \frac{i}{\hbar} \int dE \frac{G(e_{\alpha}(e_{\alpha} - E))}{\hbar}.
\]

(15)

where the prime at the integral denotes Cauchy’s principal-part integration. In our case, \( G(e_{\alpha}) \equiv D_{\alpha \beta}(e_{\alpha}) f_{\alpha}(e_{\alpha}) \), with \( f_{\alpha} \equiv f_{\alpha} \) and \( f_{\alpha} \equiv 1 - f_{\alpha} \). In order to simplify the notations, we replaced \( e_{\alpha \sigma \alpha} \) by \( e_{\alpha} \) in Eq. (14).

B. Transformation into the spin-coordinate system of the double dot

In Sec. II, we introduced the transformation rules for changing from the lead spin coordinates \( \sigma \alpha \) into the DD spin coordinates \( \sigma \alpha \). These rules give

\[
\left( \begin{array}{c}
\frac{d_{ \alpha \alpha}^{\alpha \alpha}}{d_{ \alpha \alpha}^{\alpha \alpha}}
\end{array} \right) = \frac{1}{\sqrt{2}} \left( \begin{array}{cc}
1 & e^{-i \Theta_j / 2} + e^{i \Theta_j / 2}
\end{array} \right) \left( \begin{array}{c}
\frac{d_{ \alpha \alpha}^{\alpha \alpha}}{d_{ \alpha \alpha}^{\alpha \alpha}}
\end{array} \right),
\]

(16)

with \( \Theta_j := 0 \), \( \Theta_j := + \theta \).

Thus, Eq. (14) can be easily expressed in the DD spin quantization axis. For example, it holds

\[
\sum_{\sigma \alpha} D_{\alpha \sigma \alpha}^0 d_{ \alpha \sigma \alpha}^0 d_{ \alpha \sigma \alpha} = \frac{1}{2} (D_{ \alpha \sigma \alpha}^0 + D_{ \alpha \sigma \alpha}^0) \sum_{\sigma \alpha} \Phi_{ \sigma \sigma \alpha \sigma \alpha} d_{ \alpha \sigma \alpha}^0 d_{ \alpha \sigma \alpha}^0 + \frac{1}{2} (D_{ \alpha \sigma \alpha}^0 - D_{ \alpha \sigma \alpha}^0) \sum_{\sigma \alpha} \Phi_{ \sigma \sigma \alpha \sigma \alpha} d_{ \alpha \sigma \alpha}^0 d_{ \alpha \sigma \alpha}^0 ,
\]

(17)

where we introduced
For later convenience, we also define
\[
F_{\alpha\sigma,\alpha'\sigma'} = \frac{1}{2} \left( D_{\alpha\sigma,\alpha'\sigma'}(E) + D_{\alpha'\sigma',\alpha\sigma}(E) \right), \quad \sigma \neq \sigma',
\]
and its related principal-part integral,
\[
P_{\alpha\sigma,\alpha'\sigma'}(E) = \int_0^E \text{d}e F_{\alpha\sigma,\alpha'\sigma'}(e)(e - E)^{-1}.
\]

C. Contribution from the reflection Hamiltonian

In order to give the full expression for the GME in the system’s eigenbasis, we need to compute the contribution from the reflection Hamiltonian in Eq. (11). In analogy to what we did to evaluate the contribution from the tunneling Hamiltonian, we must first transform \(H_R\) into the interaction picture and then perform the secular approximation to get rid of the time dependence. To start, we express \(H_R\) in the DD spin quantization basis,

\[
\Phi_{\alpha\sigma,\alpha'\sigma'} := \begin{cases} 
1 & \sigma = \sigma' \\
e^{i\theta_{\alpha}} & \sigma = \uparrow, \\
e^{-i\theta_{\alpha}} & \sigma = \downarrow. 
\end{cases}
\]

The commutator is easily evaluated to be
\[
-\frac{i}{\hbar} \text{Tr}_{\text{lead}}[\hat{H}_R, \rho^i(t)\rho_j]\rho_j = -\frac{i}{\hbar} \sum_{j=|N\rangle} \sum_{i=|N-1\rangle} \sum_{\alpha,\sigma} \Phi_{\alpha\sigma,\alpha'\sigma'}^* [d_{\alpha\sigma}^i, d_{\alpha'\sigma'}^j] \times (j|\rho^i(t) - \rho_j(t)\rho^j(t)|j)].
\]

In order to include this commutator in the master equation [Eq. (14)], let us introduce the following abbreviation:
\[
R_{\alpha\sigma,\alpha'\sigma'} = \frac{1}{|\alpha\sigma|^2} \Delta_{\alpha\sigma} \delta_{\alpha\sigma' \downarrow} + \delta_{\alpha\sigma' \uparrow}. \tag{20}
\]

Now, we can add \(R_{\alpha\sigma,\alpha'\sigma'}\) in Eq. (14) in lines (b) and (d) to find the final form of the complete master equation in the DD spin-coordinate system. It reads

\[
\rho_{nm}^{E_N}(t) = -\Delta_{\alpha} \sum_{\alpha} \sum_{j=|N\rangle} \sum_{i=|N-1\rangle} \Phi_{\alpha\sigma,\alpha'\sigma'}^* [d_{\alpha\sigma}^i, d_{\alpha'\sigma'}^j] |j\rangle\langle j|.
\]

D. Current formula

We now observe that Eq. (21) can be recast in the following Bloch–Redfield form:

\[
\rho_{nm}^{E_N}(t) = -\sum_{ij'lj'} R_{nmij}^{NN} \rho_{ij'}^{E_N}(t) + \sum_{hh'} R_{nmhh'}^{NN+1} \rho_{hh'}^{E_N+1}(t)
\]
\[
+ \sum_{ll'} R_{nmll'}^{NN-1} \rho_{ll'}^{E_N-1}(t).
\]
where the sums in Eq. (22) run over states with fixed particle number: \( j,j'=\{\{E_{N}N\}\}, h,h'=\{\{N+1\}\}, i,i'=\{\{N-1\}\} \). The Redfield tensors are given by \((\alpha=\sigma,d) \) (Ref. 38)

\[
R^{NN}_{\alpha ij\ell} = \sum_{a} \sum_{a'=\sigma,d} \left[ \delta_{a+a'} (\Gamma_{a,\alpha hh}^{(+)} N+1 + \Gamma_{a,\alpha hh}^{(-)} N-1) + \delta_{a+a'} (\Gamma_{a,\alpha hh}^{(+)} N+1 \Gamma_{a,\alpha hh}^{(-)} N-1) \right] \delta_{\alpha,\alpha'}, \quad (23)
\]

where the quantities \( \Gamma_{a,\alpha hh}^{(\pm)} N\pm1 \) can be easily read out from Eq. (21). They are

\[
\Gamma_{a,\alpha hh}^{(\pm)} N\pm1 = \sum_{\alpha'=\sigma,d} \left\{ \frac{\pi}{\hbar} d_{\alpha'a'} \left| I_{a}\right|^{2} \right\} F_{\sigma_a \sigma_{a'}}^{\mp} (E_{N} - E_{k}) \\
\quad \pm \frac{i}{\pi} P_{\alpha'a'}^{\mp} (E_{N} - E_{k}) \left( d_{\alpha'a'} \right) \left( d_{\alpha'a'}^{\dagger} \right) h'. \quad (24)
\]

With the stationary density matrix \( \hat{\rho}_{\sigma_{a}} \), being known, the current (through lead \( \alpha=\sigma,d \) \( \pm \)) follows from

\[
I = 2e\alpha \text{Re} \sum_{N} \sum_{N'} \left( \Gamma_{\alpha,\alpha hh}^{(N+1)} - \Gamma_{\alpha,\alpha hh}^{(N-1)} \right) p_{N,N'}^{\sigma}. \quad (25)
\]

We numerically solve Eq. (22) and use the result to evaluate the current flowing through the DD, as will be Secs. V–VII. At low-voltage voltages, however, we can make some further approximations to arrive at an analytical formula for the static dc.

### IV. LOW-BIAS REGIME

#### A. General considerations

A low-bias voltage ensures that merely one channel is involved with respect to transport properties. Here, we focus on gate voltages that align charge states \( N \) and \( N+1 \). Moreover, we can focus on density matrix elements that involve the energy ground states \( E_{N}^{(0)} \) and \( E_{N+1}^{(0)} \) only. In the following, we shall use the following compact notations:

\[
\rho_{\sigma_{a,N}}^{(N)} = \rho_{\sigma_{a,N}}^{(N)}, \quad n_{N} = \rho_{nn}^{(N)}. \quad (26)
\]

Evaluation of the current requires the knowledge of \( \hat{\rho}_{\sigma_{a}}^{(N)} \) and \( \hat{\rho}_{\sigma_{a}}^{(N+1)} \), i.e., a solution of the set of coupled equations that are obtained from Eq. (21) or, equivalently, from Eq. (22). In the low-bias regime, this task is simplified since (i) terms which try to couple states with particle numbers unlike \( N \) and \( N+1 \) can be neglected; (ii) we can reduce the sums over \( h,h' \)

in the equation for \( \hat{\rho}_{\sigma_{a}}^{(N)} \) and over \( l,l' \) in the equation for \( \hat{\rho}_{\sigma_{a}}^{(N+1)} \) to energy-ground states \( E_{N}^{(0)} \) and \( E_{N+1}^{(0)} \) because all the other transitions are exponentially suppressed by the Fermi function. Notice, however, that these two approximations are not appropriate for the principal-part terms since they are not energy conserving. The resulting equations for \( \hat{\rho}_{\sigma_{a}}^{(N)} \) and \( \hat{\rho}_{\sigma_{a}}^{(N+1)} \) [Eqs. (B1) and (B2), respectively] can be found in Appendix B. In the following, we shall apply those equations to derive an analytical expression for the conductance in the four different resonant charge state regimes possible in a DD system, i.e.,

\[
N = 0 \leftrightarrow N = 1, \quad N = 1 \leftrightarrow N = 2, \quad N = 2 \leftrightarrow N = 3, \quad N = 3 \leftrightarrow N = 4. \quad (27)
\]

In all of the four cases, we get a system of five coupled equations involving diagonal and off-diagonal elements of the RDM. The matrix elements of the dot operators between the involved states entering these equations are given in Appendix A. Before going into the details of these equations, it is instructive to analyze the structure and the physical significance of the involved RDM elements.

#### B. Elements of the reduced density matrix

**N=0.** In the case of an empty system, we have only one density matrix element in the corresponding block with fixed particle number \( N=0 \), i.e.,

\[
\rho_{\sigma_{a}}^{(0)}(t) = :W_{0}. \quad (28)
\]

describing the probability to find an empty double-dot system.

**N=1.** In this case, we have four eigenstates for the system, where the two even ones build the degenerate ground state and the two odd ones are excited states (see Table I). In the low-bias regime, we only need to take into account transitions between ground states. Therefore, we have to deal with the 2 \( \times 2 \) matrix,

\[
\left( \begin{array}{c}
\rho_{\sigma_{a},1}^{(1)}(t) \\
\rho_{\sigma_{a},1}^{(1)}(t) \\
\end{array} \right) = \left( \begin{array}{cc}
W_{1} & w_{1} e^{i\alpha_{1}} \\
w_{1} e^{-i\alpha_{1}} & W_{1} \\
\end{array} \right). \quad (29)
\]

The total occupation probability for one electron is

\[
W_{1} = W_{11} + W_{1}. \quad (30)
\]

The meaning of the off-diagonal elements, the so-called coherences, becomes clear if we regard the average spin in the system,

\[
S_{S}^{(1)} = \frac{1}{2} \text{Tr} \left( \sigma_{I} \rho_{\sigma_{a}}^{(1)}(t) \right), \quad (31)
\]

where \( i=x,y,z \) and \( \sigma_{I} \) are the Pauli spin matrices. This yields

\[
S_{S}^{(1)} = w_{1} \cos \alpha_{1}, \quad S_{S}^{(1)} = -w_{1} \sin \alpha_{1}, \quad (32)
\]

\[
S_{S}^{(1)} = \frac{1}{2} (W_{11} - W_{1}). \quad (33)
\]

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N=2. For the case N=2, we actually have six different eigenstates, but only one of them, \(|2\rangle\), is a ground state (with spin \(S=0\)), see Table I. Only this ground state must be considered in the low-bias regime, yielding
\[
P_{22}^{(2)}(t) = :W_2:.
\]
This element describes the probability to find a dot with two electrons.

N=3. In this case, we have again four eigenstates for the system, whereas the two odd ones build the degenerate ground state and the two even ones are excited. In the low-bias regime, we only need to deal with the 2×2 matrix involving the three-particle ground states,
\[
\begin{pmatrix}
\rho_{3|3\alpha}^{(3)} & \rho_{3|3\beta}^{(3)} \\
\rho_{3|3\beta}^{(3)} & \rho_{3|3\alpha}^{(3)}
\end{pmatrix} = \begin{pmatrix}
W_{3|3} & w_3 e^{i\delta_3} \\
w_3 e^{-i\delta_3} & W_{3|3}
\end{pmatrix}.
\]

The total occupation probability for three electrons is
\[
W_3 := W_{3|3} + W_{3|1}.
\]
As for the case \(N=1\), the off-diagonal elements yield information on the average spin \(S_i^{(3)} = \frac{1}{3} \text{Tr} \{ \rho_{3|3\alpha}^{(3)}(t) \} \) in the system through the following relations:
\[
S_x^{(3)} = w_3 \cos \alpha_3, \quad S_y^{(3)} = -w_3 \sin \alpha_3, \quad S_z^{(3)} = \frac{1}{2} (W_{3|3} - W_{3|1}).
\]
N=4. Finally, if the double quantum dot is completely filled with four electrons, we only have one nondegenerate state. Correspondingly, there is only one relevant RDM matrix element,
\[
\rho_{4|4}^{(4)}(t) = :W_4:
\]

C. Conductance formula

We shall exemplarily present results for the resonant transition \(N=1 \leftrightarrow N=2\). For the other transitions, similar considerations apply. The quantities of interest are \(W_1, W_2, S_i^{(1)}, S_i^{(1)}, S_i^{(1)}\), which are related through Eqs. (32) and (36) to the density matrix elements of \(\rho_{4|4}^{(1)}\). From Eqs. (B1) and (B2) and Table III, and with \(W_1 = 1 - W_2\) we finally obtain the following:
\[
\dot{S}_i^{(1)} = -\frac{\pi}{\hbar} \sum_{a=1}^{4} |t_a|^2 k_2^2 \left( 2F_{a|1}^+ (\mu_2) S_i^{(1)} - [F_{a|1}^- (\mu_2) W_2 - 4F_{a|1}^- (\mu_2)] \right)
\]
\[
-4F_{a|1}^- (\mu_2) S_i^{(1)} \cdot \vec{m}_a.
\]

We have introduced the notation \(4k_2^2 = (\alpha_0 \pm \beta_0)^2\). All of the nonvanishing principle-value factors \(P_{a|1}^\pm\) and the reflection parameter \(R_{a|1}\) have been merged to the following compact form:
\[
P_a (\mu_1, \mu_2, W_2) = -\frac{1}{2} [P_{a|1}^+ (\mu_1) + R_{a|1}] - k_2^2 P_{a|1}^- (\mu_2)
\]
\[
+ \frac{1}{4} P_{a|1}^+ (\mu_2) (e_{2\mu_2} - e_{1\mu_2}) - \frac{1}{4} P_{a|1}^+ (\mu_2) (e_{2\mu_2} - e_{1\mu_2})
\]
\[
- k_2^2 P_{a|1}^- (\mu_2) (e_{2\mu_2} - e_{1\mu_2}),
\]

where we introduced the chemical potential \(\mu_{N+1} = F_{N+1}^{(0)} - E_{N+1}^{(0)}\) and \(\mu_2\) denotes the fourth of two particle energies. We notice that the set of coupled equation [Eq. (39) and (40)] for the evolution of the populations and of the spin accumulation has a similar structure to that reported in Refs. 28, 30, and 38 for a single-level quantum dot, a metallic island, and a single-walled carbon nanotube, respectively. Some prefactors and the argument of the principal-part terms, however, are DD specific. In particular, as in Refs. 28 and 30, we clearly identify a spin precession term originating from the combined action of the reflection at the interface and the interaction. The associated effective exchange splitting is \(\gamma B_1\), where \(\gamma = -\mu_0\) is the gyromagnetic ratio and
\[
\tilde{B}_1 := \frac{2}{\gamma} \sum_{a} |t_a|^2 P_a \vec{m}_a
\]
is the corresponding effective exchange field. We now focus on the stationary limit. In the absence of the precession term, the spin accumulation has only a \(S_i^{(1)}\) component since, due to our particular choice of the spin quantization axis, \(S_i^{(1)} = 0\) holds. The exchange field tilts the accumulated spin out of the magnetizations' plane and gives rise to a nonzero \(S_i^{(1)}\) component proportional to \(B_1\) and \(S_i^{(1)}\).

To get further insight in the spin dynamics, we observe that since we are looking at the low-bias regime, we can linearize the Fermi function \(f_a\) in the bias voltage, i.e.,

| Table II. Matrix elements for the \(N=0 \leftrightarrow N=1\) transition induced by operators \(d_{a|1}\) and \(d_{a|1}\), \(\alpha = 1, 2\). |
|-----------------|-----------------|-----------------|-----------------|
| \(1\epsilon\)    | \(1\epsilon\)    | \(1\epsilon\)    | \(1\epsilon\)    |
| \(d_1|1\)        | \(\frac{1}{2}\)  | \(0\)           | \(\frac{1}{2}\)  |
| \(d_1|1\)        | \(0\)           | \(\frac{1}{2}\)  | \(\frac{1}{2}\)  |
| \(d_2|1\)        | \(\frac{1}{2}\)  | \(0\)           | \(-\frac{1}{2}\) |
| \(d_2|1\)        | \(0\)           | \(\frac{1}{2}\)  | \(-\frac{1}{2}\)|


By introducing the polarization of the contacts,

$$p_{d}(\xi) = \frac{D_{a\gamma}}{D_{d\gamma}(\xi) + D_{d\gamma}(\xi)} \cdot$$

we can express the $F_{\alpha\gamma\delta\epsilon}^{\pm}$ factors as

$$F_{\alpha\gamma\delta\epsilon}^{\pm}(\xi) = \frac{1}{2} D_{\alpha}(\xi) f(\mp \xi) (1 \mp f(\mp \xi) e^\phi V_d),$$

where $D_{\alpha} = D_{a\gamma} + D_{a\gamma}$. It is also sufficient for our calculations to regard the density of states as a constant quantity, $D_{\alpha}(\xi) = D_{\alpha}$. Consequently, the polarization is also constant, $p_{d}(\xi) = p_d$. Finally, we focus in the following on the symmetric case where both leads have the same properties, which, in particular, means that tunneling elements, polarizations, density of states, and reflection amplitude are equal,

$$t_1 = t_2 = \tau, \quad p_1 = p_2 = \tau p,$$

$$D_1 = D_2 = D, \quad R_{1\sigma\gamma \sigma} = R_{2\sigma\gamma \sigma} = R.$$

Upon introducing the linewidth $\Gamma = \frac{2\pi}{\hbar} |D|^2$, the conductance $G_{12} = \frac{1}{\hbar} V_{\text{bias}}$ for the resonant regime $N = 1 \rightarrow N = 2$ reads

$$G_{12}(\Theta) = \frac{e^2}{h} \left( 1 - \frac{p^2 \sin^2\left(\frac{\Theta}{2}\right)}{1 + [B_t / f(\mu_2)] \Gamma_{\text{ex}}^2 \cos^2\left(\frac{\Theta}{2}\right)} \right).$$

Similarly, we find for an arbitrary resonance $(i=0,1,2,3)$,

$$G_{ii+1}(\Theta) = \frac{e^2}{h} \left( 1 - \frac{p^2 \sin^2\left(\frac{\Theta}{2}\right)}{1 + [B_t / f(\mu_2)] \Gamma_{\text{ex}}^2 \cos^2\left(\frac{\Theta}{2}\right)} \right),$$

where $|i+1|d_\ell|\ell\rangle$ is a shortcut notation for the nonvanishing matrix elements $\langle E_{\ell}^{(0)} | i^+ | d_\ell | E_{\ell}^{(0)} \rangle$ calculated in Tables II–V. It holds $|1|d_\ell|\ell\rangle = |1|d_\ell|\ell\rangle = 1 / \sqrt{2}$ and $|2|d_\ell|\ell\rangle = |3|d_\ell|\ell\rangle = k_n$. Moreover, we gathered together the principal-part contributions and the ones coming from the reflection Hamiltonian in the effective magnetic fields,

$$\vec{B}_2 = \vec{B}_1,$$

$$\vec{B}_3 = \vec{B}_4 = \frac{2}{\gamma} \sum_{\alpha} \left| t_\alpha \right|^2 \mathcal{P}_{\alpha\gamma}(\mu_3, E_{\ell}^{(0)} - E_{\ell}^{(0)}) \hat{m}_\alpha.$$

The latter are defined in terms of the following function:

$$\mathcal{P}_{\alpha\gamma}(\mu_3, E_{\ell}^{(0)} - E_{\ell}^{(0)}) = \frac{1}{2} [P_{\alpha\gamma}(\mu_3) + R_{\alpha\gamma}(\mu_3)] - k^2 P_{\alpha\gamma}(\mu_3)$$

$$+ \left( \frac{1}{4} P_{\alpha\gamma}(\mu_3 - \mu_2) - \frac{1}{4} P_{\alpha\gamma}(\mu_3 - \mu_2) \right)$$

$$- k^2 P_{\alpha\gamma}(\mu_3 - \mu_2).$$

Moreover, a closer look to Eq. (46) shows that its angular dependence is strongly coupled to the square of the ratio $(\gamma B_t)^2 / \{h \Gamma[(\mu_2)^{-1}] \}$, which is the effective exchange splitting rescaled by the coupling and the Fermi function. The ratio occurs in the denominators, and its value depends on the gate voltage. As the change of $B_j$ under the variation in the gate voltage is comparatively small, the factor dominating the gate voltage evolution is the Fermi function. This accounts for the population of the dot: only if a nonzero spin is present (i.e., odd filling: one or three electrons) the effective magnetic field can have an influence. That is why correspondingly the renormalized effective exchange splitting vanishes for even fillings, namely, below $0 \rightarrow 1$ and $2 \rightarrow 3$, respectively, above the $1 \rightarrow 2$ and $3 \rightarrow 4$ resonances. This can nicely be seen from Fig. 3 (remember that $V_{\text{gate}} \propto - \xi$, so “below” means larger, “above” means smaller $\xi$, where the four different factors $(\gamma B_t)^2 / \{h \Gamma[(\mu_2)^{-1}] \}$ are plotted. The curves belonging to the resonances involving the half-filling do not immediately go to zero but show a more complex behavior with some small intermediate peaks due to the influence of the various excited states present for a two-electron population of the dot. As we expect, $G_{01}(\xi)$ and $G_{32}(\xi)$ [$G_{12}(\xi)$ and $G_{23}(\xi)$, respectively] are mirror symmetric with respect to each other when the gate voltage is varied. This in turn reflects the electron-hole symmetry of the DD Hamiltonian. The parameters of the figures are chosen to be as follows $(b < 0)$:

$$\kappa_b T = 4 \times 10^{-2} |b|, \quad \hbar \Gamma = 4 \times 10^{-3} |b|,$$

$$U = 6 |b|, \quad V = 1.6 |b|,$$

and $p=0.8, R=0.05D$. As expected, the peaks are mirror symmetric with respect to the half-filling gate voltage. Notice also the occurrence of different peak heights, both in the parallel and in the antiparallel case. For both polarizations, the principal-part terms entering Eq. (46) vanish, the spin accumulation is entirely in the magnetization plane, and the peak ratio is solely determined by the ratio of the ground state overlaps $2 / k_x$. For polarization angles $\Theta \neq 0, \pi$, the ra-
TABLE III. Matrix elements for the $N=1 \leftrightarrow N=2$ transition induced by $d_{11}^\dagger$ and $d_{11}'$. The notation $|2'(s_z)|$, with $s_z=0$, $\pm 1$, specifies which one of the triplet elements is addressed.

|      | $|1e^+\rangle$ | $|1e^-\rangle$ | $|1\sigma^+\rangle$ | $|1\sigma^-\rangle$ |
|------|----------------|----------------|----------------|----------------|
| $d_{11}'$: | \(\langle 2g|\) | $\frac{\alpha_0\beta_0}{2}$ | $\frac{\alpha_0\beta_0}{2}$ | $\frac{\alpha_0\beta_0}{2}$ |
| $\langle 2'(+1)|$ | $\frac{1}{2}$ | $0$ | $-\frac{1}{2}$ | $0$ |
| $\langle 2'(0)|$ | $\frac{1}{2}$ | $0$ | $0$ | $0$ |
| $\langle 2'(-1)|$ | $0$ | $0$ | $0$ | $0$ |
| $\langle 2'|\rangle$ | $\frac{1}{2}$ | $\frac{\alpha_0\beta_0}{2}$ | $0$ | $\frac{\alpha_0\beta_0}{2}$ |
| $\langle 2|\rangle$ | $\frac{1}{2}$ | $\frac{\alpha_0\beta_0}{2}$ | $0$ | $\frac{\alpha_0\beta_0}{2}$ |

$N=0 \rightarrow N=1$ and $N=3 \rightarrow N=4$

The expected mirror symmetry of $G_{01}$ and $G_{34}$ is shown in Fig. 6, where the conductance peaks are plotted for different polarization angles $\Theta$ of the contacts. Notice that the analytical expressions [Eq. (46)] (continuous lines) perfectly match the results obtained from a numerical integration of the master equation [Eq. (21)] with the current formula [Eq. (25)].

In the following, we analyze in detail the single resonance transitions. Due to the mirror symmetry, it is convenient to investigate together the $N=0 \leftrightarrow N=1$, $N=3 \leftrightarrow N=4$ and $N=1 \leftrightarrow N=2$, $N=2 \leftrightarrow N=3$ resonances. We use the convention that for a fixed resonance, the parameter $\xi=0$ when $\mu_{N=1}$.

1. Resonant regimes $N=0 \rightarrow N=1$ and $N=3 \rightarrow N=4$

The expected mirror symmetry of $G_{01}$ and $G_{34}$ is shown in Fig. 6, where the conductance peaks are plotted for different polarization angles $\Theta$ of the contacts. Notice that the analytical expressions [Eq. (46)] (continuous lines) perfectly match the results obtained from a numerical integration of the master equation [Eq. (21)] with the current formula [Eq. (25)].

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For the $0 \leftrightarrow 1$ transition, it reads

$$
\text{TMR}_{N=0 \leftrightarrow N=1}(\Theta, \xi) = 1 - \frac{G_{N=0 \leftrightarrow N=1}(\Theta, \xi)}{G_{N=0 \leftrightarrow N=1}(0, \xi)}.
$$
$\text{TABLE IV. Matrix elements for the } N=2 \leftrightarrow N=3 \text{ transition governed by } d_{\alpha i} \text{ and } d_{\beta j}^*; \alpha = 1, 2.$

| $d_{\alpha i}$ | $|3\sigma\rangle$ | $|3\sigma^+\rangle$ | $|3\sigma^-\rangle$ | $|3\epsilon\rangle$ |
|---------------|-----------------|-----------------|-----------------|-----------------|
| $\langle 2g |$ & $\frac{\alpha_i \beta_i}{2}$ & 0 & $\frac{\alpha_i \beta_i}{2}$ & 0 |
| $\langle 2^+ |$ & 0 & 0 & 0 & 0 |
| $\langle 2^0 |$ & $\frac{1}{2}$ & 0 & $-\frac{1}{2}$ & 0 |
| $\langle 2^- |$ & 0 & $\frac{1}{2}$ & 0 & $\frac{1}{2}$ |
| $\langle 2^x |$ & $\frac{1}{2}$ & 0 & $\frac{1}{2}$ & 0 |
| $\langle 2^y |$ & $\frac{1}{2}$ & 0 & $\frac{1}{2}$ & 0 |
| $\langle 2^z |$ & $\frac{1}{2}$ & 0 & $\frac{1}{2}$ & 0 |

Hence, the TMR vanishes for $\Theta=0$ and takes the constant value,

$$\text{TMR}_{01} = \frac{p^2 \sin^2(\theta)}{1 + [B^2 f^2(-\mu_1) \Gamma^2] \cos^2(\frac{\theta}{2})},$$

at $\Theta=\pi$. For the remaining polarization angles, $\Theta \neq 0$ and $\Theta \neq \pi$, the TMR is gate voltage dependent and positive. The behavior of the TMR as a function of the gate voltage is shown in Fig. 7. To understand the gate voltage dependence of the TMR at noncollinear angles, we have to remember that the dot is depleted with increasing $\xi$. For the $0 \leftrightarrow 1$ transition, this means that at positive $\xi$, the dot is predominantly empty, so that an electron that enters the dot also leaves it fast. In this situation, the TMR is finite and its value depends in a complicated way on the amplitude of the exchange field. At negative $\xi$, the DD is predominantly occupied with an

$\text{FIG. 3. (Color online) Gate voltage dependence of the renormalized effective exchange splitting entering the conductance formula [Eq. (46)]. Notice the mirror symmetry of the } 0 \leftrightarrow 1 \text{ with the } 3 \leftrightarrow 4 \text{ curve and of the } 1 \leftrightarrow 2 \text{ with the } 2 \leftrightarrow 3 \text{ one.}$

$\text{TABLE V. Matrix elements for the } N=3 \leftrightarrow N=4 \text{ transition induced by the operators } d_{\alpha i}^d \text{ and } d_{\beta j}^*; \alpha = 1, 2.$

| $d_{\alpha i}^d$ | $|3\sigma\rangle$ | $|3\sigma^+\rangle$ | $|3\sigma^-\rangle$ | $|3\epsilon\rangle$ |
|---------------|-----------------|-----------------|-----------------|-----------------|
| $d_{\alpha i}^d; (2,2) \text{ or } d_{\beta j}^*; (2,2)$ | 0 | $\frac{1}{4}$ | 0 | $-\frac{1}{4}$ |
| $d_{\alpha i}^d; (2,2)$ | $\frac{1}{2}$ | 0 | $-\frac{1}{2}$ | 0 |
| $d_{\alpha i}^d; (2,2)$ | 0 | $-\frac{1}{2}$ | 0 | $-\frac{1}{2}$ |
| $d_{\alpha i}^d; (2,2)$ | $-\frac{1}{2}$ | 0 | $\frac{1}{2}$ | 0 |

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electron that can now interact with the exchange field, which makes the spin precess and thus eases tunneling out of the dot. Consequently, \( G_{NN+1}(\Theta, \xi) \approx G_{NN+1}(0, \xi) \) and the TMR vanishes. Finally, Fig. 8 illustrates the angular dependence of the normalized conductance for three different values of the gate voltage. We detect a common absolute minimum for the conductance \( G \), which is always smaller than that in the parallel one. An analogous result as for the 0 \( \leftrightarrow \) 2 resonance to the curves with \( |\Theta| = \pi \). Notice again the equivalence of the curves belonging to \( |\Theta| = \pi \) and \( |\Theta| = 0 \), i.e., transport is weakened in the antiparallel case. The width of the curves is dependent on the gate voltage. We detect a common absolute minimum for the conductance peaks for the 0 \( \leftrightarrow \) 3 and 3 \( \leftrightarrow \) 4 transitions vs gate voltage. The TMR is always positive and for collinear lead magnetizations, \( \Theta = 0 \) and \( \Theta = \pi \), independent of gate voltage.

\[ \text{Conductance at low bias for the parallel case } \Theta = 0 (\text{top}) \text{ and antiparallel case } \Theta = \pi (\text{bottom}). \]

\[ \text{Fig. 4. (Color online) Conductance } \frac{G}{|\mathbf{E}|^2/\hbar} \text{ at low bias for the parallel case } \Theta = 0 (\text{top}) \text{ and antiparallel case } \Theta = \pi (\text{bottom}). \]

\[ \text{Notice the different peak heights and the mirror symmetry with respect to the half-filling value } \xi = 0. \]

\[ \text{The conductance in the antiparallel configuration is always smaller than that in the parallel one.} \]

\[ \text{Fig. 5. (Color online) Conductance as a function of the polarization angle and of the gate voltage. The minimal conductance peaks occur as expected at } \Theta = \pi. \]

\[ \text{Fig. 6. (Color online) The conductance } G_{00}(\xi) \text{ (upper figure) [}G_{33}(\xi) \text{ (lower figure)] vs gate voltage for different polarization angles. The mirror symmetry of the conductance peaks for the 0 \( \leftrightarrow \) 1 and 3 \( \leftrightarrow \) 4 transitions is clearly observed.} \]

\[ \text{Notice the excellent agreement between the prediction of the analytical formula [Eq. (46)] (continuous lines) and the results of a numerical integration of Eq. (21) with Eq. (25) (symbols).} \]

V. NONLINEAR TRANSPORT

In this section, we present the numerical results deduced from the general master equation [Eq. (21)] combined with the current formula [Eq. (25)]. We show the differential conductance \( \frac{dI}{dV}(\xi, V_{\text{bias}}) \) for the three distinct angles \( \Theta = 0, \Theta = \pi \), and \( \Theta = 2\pi \).

\[ \text{Fig. 7. (Color online) TMR for the 0 \( \leftrightarrow \) 1 (upper figure) and 3 \( \leftrightarrow \) 4 (lower figure) transitions vs gate voltage. The TMR is always positive and for collinear lead magnetizations, } \Theta = 0 \text{ and } \Theta = \pi, \text{ independent of gate voltage.} \]

\[ \text{The conductance } G_{NN+1}(\xi) \text{ (upper figure) [}G_{NN+1}(\xi) \text{ (lower figure)] vs gate voltage for different polarization angles.} \]

\[ \text{The mirror symmetry of the conductance peaks for the 0 \( \leftrightarrow \) 1 and 3 \( \leftrightarrow \) 4 transitions is clearly observed.} \]

\[ \text{Notice the excellent agreement between the prediction of the analytical formula [Eq. (46)] (continuous lines) and the results of a numerical integration of Eq. (21) with Eq. (25) (symbols).} \]
...and \( \Theta = \pi \), see Fig. 10, top, middle, and bottom, respectively. The results confirm the electron-hole symmetry and the symmetry upon bias voltage inversion \( I(\xi, V_{\text{bias}}) = -I(\xi, -V_{\text{bias}}) \). In all of the three cases, we can nicely see the expected three closed and the two half-open diamonds, where the current is blocked and the electronic number of the double-dot system stays constant. At higher bias voltages, the contribution of excited states is manifested in the appearance of several excitation lines. One clearly sees that transition lines present in the parallel case are absent in the antiparallel case. Moreover, in the case of noncollinear polarization, \( \Theta = \pi/2 \), negative differential conductance (NDC) is observed.

In the following, we want not only to explain the origin of these two features, but alongside also give another example for spin-blockade effects, which play a decisive role in the DD physics. As a starting point, we plot in Fig. 11 (top) the current through the system for the three different angles \( \Theta = 0 \), perpendicular \( \Theta = \pi/2 \) (middle), and antiparallel \( \Theta = \pi \) (bottom) configurations. The two half diamond and three diamond regions correspond to bias and gate voltage values where transport is Coulomb blocked. The excitation lines, where excited states start to contribute to resonant transport, are clearly visible in all of the three cases. However, a negative differential conductance is observed in the perpendicular case, while some excitation lines are absent in the antiparallel configuration.

FIG. 8. (Color online) \( G_{12}(\Theta)/G_{12}(0) \) (upper figure) \( [G_{34}(\Theta)/G_{34}(0)] \) (lower figure) vs polarization angle. For all the three chosen values of the gate voltage, the curve displays an absolute minimum at \( \Theta = \pi \). Notice the overall agreement of the analytical predictions [Eq. (46)] given by the continuous curves with outcomes of a numerical solution of the master equation [Eq. (21)] together with Eq. (25) (symbols).

FIG. 9. (Color online) \( G_{12}(\Theta)/G_{12}(0) \) (upper figure) \( [G_{34}(\Theta)/G_{34}(0)] \) (lower figure) vs polarization angle. Notice the overall agreement of the analytical predictions [Eqs. (45) and (46)] (continuous lines) with the data (symbols) coming from numerical solutions of the equations for the reduced density matrix.

FIG. 10. (Color online) Differential conductance \( \frac{dI}{db} \) for the parallel \( \Theta = 0 \) (top), perpendicular \( \Theta = \pi/2 \) (middle), and antiparallel \( \Theta = \pi \) (bottom) configurations. The two half diamond and three diamond regions correspond to bias and gate voltage values where transport is Coulomb blocked. The excitation lines, where excited states start to contribute to resonant transport, are clearly visible in all of the three cases. However, a negative differential conductance is observed in the perpendicular case, while some excitation lines are absent in the antiparallel configuration.
occupation probabilities for the triplet state. We end up in a blocking state. The transition is hence forbidden.

Notice that the two distinct spin-blockade effects are different from the Pauli spin-blockade discussed in the DD literature. Moreover, the second effect, relying on the existence of degenerate triplet states, is also different from the spin-blockade found in Ref. 30 for a single-level quantum dot.

Finally, let us turn to the negative differential conductance, which occurs for noncollinearly polarized leads (see the dashed blue lines in Fig. 11) and which we find to become more evident for higher polarizations (not shown). By neglecting the exchange field, we would just expect the magnitude of the current for the noncollinear polarizations to lie somewhere in between the values for the parallel and the antiparallel current because the noncollinear polarization could, in principle, be rewritten as a linear combination of the parallel and the antiparallel configuration. Now, the effect of the exchange is to cause precession and therewith equilibration of the accumulating spin, which corresponds to shifting the balance in favor of the parallel configuration, i.e., enhancing the current. The decisive point is that the exchange field is not only gate dependent but also bias voltage dependent and reaches a minimum around $eV_{\text{bias}} \approx 8|b|$. This explains the decrease of the current up to this point. Afterward, the influence of the spin precession regains weight.

The same consideration applies for the other NDC regions observed in Fig. 10, e.g., in the gate voltage region $\xi \approx 2|b|$ involving the $N=0 \leftrightarrow N=1$ transition, as described in Ref. 30.

VI. EFFECTS OF AN EXTERNAL MAGNETIC FIELD

In this section, we wish to discuss the qualitative changes brought by an external magnetic field applied to the DD. Specifically, the magnetic field is assumed to be parallel to the magnetization direction of the drain. For simplicity, we focus on the experimental standard case of parallel and antiparallel lead polarizations and of low-bias voltages. Then, the magnetic field causes an energy shift $\tilde{E}_{\text{Zeeman}}$, depending on whether the electron spin is parallel or antiparallel, respectively, to it. For collinear polarization angles, the principal-part contributions vanish, and the equations for the RDM are easily obtained. We exemplarily report results for $0 \leftrightarrow 1$ and $1 \leftrightarrow 2$ transitions. Let us then consider the parameter regime nearby the $0 \leftrightarrow 1$ resonance and setup a system of three equations with three unknown variables, $W_0$, $W_1$, and $W_{1\downarrow}$. The first equation corresponds to the normalization condition $W_{1\uparrow} + W_1 + W_0 = 1$. The remaining equations are the equations of motion for $W_{1\downarrow}$, which can be written as

$$W_{1\downarrow} = -\frac{\pi}{\mathcal{H}} \sum_{\alpha=\uparrow,\downarrow} |t_{\alpha}|^2 [F_{\alpha}(\mu_{1\uparrow}) W_{1\uparrow} - F_{\alpha}^+(\mu_{1\downarrow}) W_0],$$  (49)
\[ W_{1i} = -\frac{\pi}{\hbar} \sum_{\alpha, \alpha'} |\rho|^2 [F_{\alpha i}(\mu_{11}) - F_{\alpha' i}(\mu_{11})] W_0, \]

where

\[ \mu_{11} = \mu_1 \pm E_{Zeeman}, \]

and \( F_{\alpha i}(E) = D_{\alpha i} f(E). \) \( D_{\alpha i} = D_{\alpha i} \alpha' \). On the other hand, \( D_{11} = D_{zz} \) and \( D_{12} = D_{dz} \) in the antiparallel case.

Upon considering symmetric contacts \( t_1 = t_2 = t, D_1 = D_2 = D \), we find the following in the parallel case:

\[
G_{00}(\Theta = 0) = \frac{\Gamma e^2}{2\beta} f(-\mu_1) f(-\mu_1) \times \frac{1 - \rho^2 [f(\mu_1) + f(\mu_1)]}{\rho [f(\mu_1) - f(\mu_1)] + f(\mu_1) + f(\mu_1)}. \tag{52}
\]

For the antiparallel case, we obtain

\[ G_{00}(\Theta = \pi) = G_{00}(\Theta = 0) \times \frac{1 - \rho^2 [f(\mu_1) + f(\mu_1)]}{\rho [f(\mu_1) - f(\mu_1)] + f(\mu_1) + f(\mu_1)}. \tag{53}\]

Analogously, we find the following for the 1 \( \leftrightarrow \) 2 transition:

\[
G_{12}(\Theta = 0) = \frac{\Gamma e^2 k^2}{2\beta} f(\mu_2) f(\mu_2) \times \frac{1 - \rho^2 [f(\mu_2) + f(\mu_2)]}{\rho [f(\mu_2) - f(\mu_2)] + f(\mu_2) + f(\mu_2)}. \tag{54}\]

\[ G_{12}(\Theta = \pi) = G_{12}(\Theta = 0) \times \frac{1 - \rho^2 [f(\mu_2) + f(\mu_2)]}{\rho [f(\mu_2) - f(\mu_2)] + f(\mu_2) + f(\mu_2)}. \tag{55}\]

The remaining resonances are analogously calculated. Figure 12 shows the four conductance resonances for the parallel and antiparallel configurations. Strikingly, the applied magnetic field breaks the symmetry between tunneling regimes 0 \( \leftrightarrow \) 1 and 3 \( \leftrightarrow \) 4, as well as between 1 \( \leftrightarrow \) 2 and 2 \( \leftrightarrow \) 3 resonances in case of parallel contact polarizations. The reason for this behavior is the following: in the low-bias regime, transitions between ground states dominate transport. In particular, the magnetic field removes the spin degeneracy of \( |1e\alpha\rangle \) and \( |3o\alpha\rangle \) states, such that states with spin aligned to the external magnetic field are energetically favored. Therefore, the transport electron in tunneling regime 0 \( \leftrightarrow \) 1 is a majority spin carrier. For the case 3 \( \leftrightarrow \) 4, however, two of the three electrons of the ground state \( |3o\uparrow\rangle \) have spin-up, such that the fourth electron that can be added to the DD has to be a minority spin carrier. Therefore, the conductance gets diminished with respect to the 0 \( \leftrightarrow \) 1 transition, and the mirror symmetry present in the zero field case is broken. Analogously, the broken symmetry in the case of the 1 \( \leftrightarrow \) 2 and 2 \( \leftrightarrow \) 3 transitions can be understood. Correspondingly, the TMR can become negative for values of the gate voltages around the 2 \( \leftrightarrow \) 3 and 3 \( \leftrightarrow \) 4 resonances. We observe that a negative TMR has recently been predicted in Ref. 42 for the case of a single impurity Anderson model with orbital and spin degeneracies (see Fig. 13). In that work, a negative TMR arises due to the assumption that multiple reflections at the interface cause spin-dependent energy shifts. In our approach, however, where the contribution from the reflection Hamiltonian is treated to the lowest order, see Eq. (19), such spin-dependent energies originate from the magnetic-field-induced Zeeman splitting.

\[ FIG. 12. \] (Color online) Conductance vs gate voltage for parallel (continuous line) and antiparallel (dashed lines) contact configurations and Zeeman splitting \( E_{Zeeman} = 0.05|b| \). The magnetic field breaks the mirror symmetry with respect to the gate voltage in the parallel configuration.

\[ FIG. 13. \] (Color online) TMR vs gate voltage in the presence of an external magnetic field. In contrast to the zero field case, the TMR can become negative in the vicinity of the 2 \( \leftrightarrow \) 3 and 3 \( \leftrightarrow \) 4 resonances.

\[ VII. CONCLUSIONS \]

In summary, we have evaluated linear and nonlinear transport through a double-quantum-dot (DD) coupled to polarized leads with arbitrary polarization directions. Due to strong Coulomb interactions, the DD operates as a single-electron transistor, a F-SET, at low enough temperatures. A detailed analysis of the current-voltage characteristics of the DD and comparison with results of previous studies on other
F-SET systems with noncollinear polarization (a single-level quantum dot,\textsuperscript{30} a metallic island,\textsuperscript{28} and a carbon nanotube\textsuperscript{38}) brings us to the identification of \textit{universal} behaviors of a F-SET, i.e., a behavior shared by \textit{any} of those F-SETs, independent of the specific kind of conductor that is considered as the central system, as well as system-specific features.

The presence of an interfacial exchange field together with an interaction-induced one is universal. These exchange fields can act only for noncollinear polarizations and cause a precession of the accumulated spin on the dot and therewith ease the tunneling. This effect has various implications. It determines, e.g., the gate and angular dependences, as well as the height of single conductance peaks and can yield negative differential conductance features. Another universal feature is the occurrence of a negative tunneling magnetoresistance—even in the weak-tunneling limit—if a Zeeman-splitting exists.

The following features are specific to the DD system: in the low-bias regime, the problem can be analytically solved, and for tunneling regimes $0 \rightarrow 1$ and $1 \rightarrow 2$ (2 $\rightarrow$ 3 and 3 $\rightarrow$ 4, respectively), the system behaves equivalent to a single-level quantum dot, where the Coulomb blockade peaks are found to be mirror symmetric with respect to the charge neutrality point. This mirror symmetry reflects the electron-hole symmetries of a system and is therefore typical for the DD, as well as the ratio of the peak heights. An external magnetic field lifts this symmetry and can cause a negative tunneling magnetoresistance. In the nonlinear bias regime, the presence of various excited states gives rise to interesting DD specific features. For example, a suppression of several excitation lines for an antiparallel lead configuration originates from a spin-blockade effect. It occurs because a trapping state is formed whenever a transition involves a two-electron state with total spin zero. A second spin-blockade effect we described involves the two-electron triplet state. The common mechanism of these two spin-blockades is the following: in both cases, a tunneling event can only occur if initially the dot is populated with an unpaired electron possessing the majority spin of the drain. The second step is that a majority electron of the source will enter, forming a spin-zero state.

Then, the first electron can leave the dot, causing a spin flip. If the triplet state is involved, the dot will be left in a trapping state once a second majority electron from the source enters. Otherwise, we are already in a blocking state. Finally, for noncollinear lead polarizations, negative differential conductance can be observed. All in all, due to their universality and the multiplicity of their properties, F-SETs based on DD systems seem good candidates for future magnetoelectronic devices.

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**APPENDIX A: MATRIX ELEMENTS OF THE DOT OPERATORS**

Tables II–V show all the possible matrix elements $\langle N-1|d_{\sigma \uparrow}^{\dagger}|N \rangle$ and $\langle N|d_{\sigma \downarrow}^{\dagger}|N-1 \rangle$ with $\sigma=\{1,2\}$ and $\sigma_0=\{1,\}$, which occur in the master equations [Eqs. (B1) and (B2)]. Notice that we only need to illustrate either the matrix elements $\langle N-1|d_{\sigma \uparrow}^{\dagger}|N \rangle$ or $\langle N|d_{\sigma \downarrow}^{\dagger}|N-1 \rangle$ because they are complex conjugated to each other.

**APPENDIX B: MASTER EQUATION FOR THE REDUCED DENSITY MATRIX IN THE LINEAR REGIME**

We explicitly report here the coupled equations of motion for elements $\rho_{nm}^{(N)}(t)$ and $\rho_{nm}^{(N+1)}(t)$ of the RDM to be solved in the low-bias regime. They are obtained from the generalized master equation [Eq. (21)] upon observing that (i) in the linear regime, terms that couple states with particle numbers unlike $N$ and $N+1$ can be neglected; (ii) we can reduce the sum over $h$ and $h'$ only to energy ground states. In the remaining energy nonconserving terms, the sum has to go also over excited states. With $\mu_{N+1} = E_{N+1}^{(0)} - E_{N}^{(0)}$ being the chemical potential, we finally arrive at the following two master equations:

\begin{align}
\dot{\rho}_{nm}^{(N)}(t) &= -\frac{i}{\hbar} \sum_{\sigma_\alpha, \sigma_\beta} \left\{ \sum_{l \in \{N-1\}} \sum_{j, \ell \in \{N\}} h_h \sum_{h' \in \{N+1\}} \sum_{l' \in \{N\}} \Phi_{aa' \sigma_\alpha \sigma_\beta}^{(N)}(\mu_{N+1})(d_{\sigma_\alpha}^{(l \ell)})(d_{\sigma_\beta}^{(l' \ell')})h_{\sigma_\alpha \sigma_\beta}^{(N)}(t)
+ \Phi_{aa' \sigma_\alpha \sigma_\beta}^{(N)} \sum_{l \in \{N-1\}} \sum_{j, \ell \in \{N\}} h_h \sum_{h' \in \{N+1\}} \sum_{l' \in \{N\}} \Phi_{aa' \sigma_\alpha \sigma_\beta}^{(N)}(\mu_{N+1})(d_{\sigma_\alpha}^{(l \ell)})(d_{\sigma_\beta}^{(l' \ell')})h_{\sigma_\alpha \sigma_\beta}^{(N)}(t)
+ \Phi_{aa' \sigma_\alpha \sigma_\beta}^{(N)} \sum_{l \in \{N-1\}} \sum_{j, \ell \in \{N\}} h_h \sum_{h' \in \{N+1\}} \sum_{l' \in \{N\}} \Phi_{aa' \sigma_\alpha \sigma_\beta}^{(N)}(\mu_{N+1})(d_{\sigma_\alpha}^{(l \ell)})(d_{\sigma_\beta}^{(l' \ell')})h_{\sigma_\alpha \sigma_\beta}^{(N)}(t)
+ \Phi_{aa' \sigma_\alpha \sigma_\beta}^{(N)} \sum_{l \in \{N-1\}} \sum_{j, \ell \in \{N\}} h_h \sum_{h' \in \{N+1\}} \sum_{l' \in \{N\}} \Phi_{aa' \sigma_\alpha \sigma_\beta}^{(N)}(\mu_{N+1})(d_{\sigma_\alpha}^{(l \ell)})(d_{\sigma_\beta}^{(l' \ell')})h_{\sigma_\alpha \sigma_\beta}^{(N)}(t)
\right\},
\end{align}
\begin{align}
\rho_{nm}^{(N+1)}(t) &= -\frac{i}{\hbar} \sum_{\alpha, \sigma, d} |t|^2 \sum_{\sigma_0, \sigma_1} \left\{ \sum_{i \in \{E_{k}(N)\}} \sum_{j \in \{E_{j}(N+1)\}} \left[ \Phi_{\alpha \sigma_0 \sigma_1} \left( P_{\alpha \sigma_0 \sigma_1} (\varepsilon_{k} - \varepsilon_{j}) \right) \right. \\
& \times \left( d_{\alpha \sigma_0}^{(N)} (d_{\alpha \sigma_0}^{(N)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} + F_{\alpha \sigma_0 \sigma_1}^{(N+1)} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} \right) \right\} \\
& \times \left\{ P_{\alpha \sigma_0 \sigma_1} \left( P_{\alpha \sigma_0 \sigma_1} (\varepsilon_{j} - \varepsilon_{k}) + R_{\alpha \sigma_0 \sigma_1} \right) \right\} \\
& \left. \times \left( d_{\alpha \sigma_0}^{(N)} (d_{\alpha \sigma_0}^{(N)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} + F_{\alpha \sigma_0 \sigma_1}^{(N+1)} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} \right) \right\} \\
& \times \left( d_{\alpha \sigma_0}^{(N)} (d_{\alpha \sigma_0}^{(N)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} + F_{\alpha \sigma_0 \sigma_1}^{(N+1)} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} \right) \right\} \\
& \times \left( d_{\alpha \sigma_0}^{(N)} (d_{\alpha \sigma_0}^{(N)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} + F_{\alpha \sigma_0 \sigma_1}^{(N+1)} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} \right) \right\} \\
& \times \left( d_{\alpha \sigma_0}^{(N)} (d_{\alpha \sigma_0}^{(N)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} + F_{\alpha \sigma_0 \sigma_1}^{(N+1)} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} \right) \right\} \\
& \left( d_{\alpha \sigma_0}^{(N)} (d_{\alpha \sigma_0}^{(N)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} + F_{\alpha \sigma_0 \sigma_1}^{(N+1)} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} \right) \right\} \\
& \left( d_{\alpha \sigma_0}^{(N)} (d_{\alpha \sigma_0}^{(N)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} + F_{\alpha \sigma_0 \sigma_1}^{(N+1)} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} \right) \right\} \\
& \left( d_{\alpha \sigma_0}^{(N)} (d_{\alpha \sigma_0}^{(N)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} + F_{\alpha \sigma_0 \sigma_1}^{(N+1)} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} (d_{\alpha \sigma_1}^{(N+1)})^{\dagger} \right) \right\} \right). \tag{B2}
\end{align}

Notice that we kept the sums over excited states \( n \), \( \hat{n} \) in Eq. (B1) and \( i \), \( \hat{i} \) in Eq. (B2), which are responsible for the virtual transitions.