All-electric-spin control in interference single electron transistors

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Single particle interference lies at the heart of quantum mechanics. The archetypal double-slit experiment\(^1\) has been repeated with electrons in vacuum\(^2,3\) up to the more massive \(C_{60}\) molecules\(^4\). Mesoscopic rings threaded by a magnetic flux provide the solid-state analogous\(^5,6\). Intra-molecular interference has been recently discussed in molecular junctions\(^7,8,9,10,11\). Here we propose to exploit interference to achieve all-electrical control of a single electron spin in quantum dots, a highly desirable property for spintronics\(^12,13,14\) and spin-qubit applications\(^15,16,17,18,19\). The device consists of an interference single electron transistor (ISET\(^10,11\)), where destructive interference between orbitally degenerate electronic states produces current blocking at specific bias voltages. We show that in the presence of parallel polarized ferromagnetic leads the interplay between interference and the exchange coupling on the system generates an effective energy renormalization yielding different blocking biases for majority and minority spins. Hence, by tuning the bias voltage full control over the spin of the trapped electron is achieved.

The all-electrical solutions to the challenge of single spin control that have been proposed\(^15,16,17,18\) and realized\(^19,20\) are based either on spin orbit-coupling\(^15,16,17,18,19\) or on tunneling-induced spin splitting in the Kondo regime\(^20\). Our proposal relies on the current blocking occurring in an ISET due to interference between degenerate states. The conditions for interference blocking are very generic\(^11\) and admit several different realizations. We consider here for clarity a benzene and a triple-dot ISET, Fig. 1. Both are described by the Hamiltonian:

\[
H = H_{\text{sys}} + H_{\text{leads}} + H_T, \tag{1}
\]

where \(H_{\text{sys}}\) represents the central system and also contains the energy shift operated by a capacitively coupled gate electrode at the potential \(V_g\). The Hamiltonian \(H_{\text{sys}}\) is in both cases invariant with respect to a discrete set of rotations around the vertical axis passing through the center of the system. This fact allows a classification of its eigenstates in terms of the \(z\) component of the angular momentum \(\ell\) and also ensures the existence of degenerate states with different \(\ell\). Then, a generic eigenstate is represented by the ket \(|N\ell\sigma E\rangle\) where \(N\) is the number of electrons on the system, \(\sigma\) is the spin and \(E\) the energy of the state. When degenerate states participate to transport they interfere since, like the two paths of the double-slit experiment, they are occupied simultaneously by the travelling electron, but in different superpositions under diverse transport conditions. \(H_{\text{lead}}\) describes the ferromagnetic leads with equal (for simplicity) parallel polarization \(P\) and with a difference \(eV_b\) between their electrochemical potentials. Finally, \(H_T\) accounts for the weak tunnelling coupling between the system and the leads, characteristic of SETs, and we consider the tunnelling events restricted to the atoms or to the dots closest to the corresponding lead (Fig. 1). We explicitly consider the Coulomb interaction only in the central part of the device (see the supplementary material\(^1\)) due to the strong confinement experienced there by the electrons while, apart from the polarization assumption, we assume a non interacting approximation for the leads.

In the weak coupling regime the current essentially consists of sequential tunnelling events at the source and drain lead that increase or decrease by one the number of electrons on the system. The different panels of Figs. 2 and 3 show the current through the benzene and triple dot ISET, respectively, as a function of bias and gate voltage. As in all SETs at low bias so called Coulomb diamonds, where transport is energetically forbidden, occur. Within the diamonds the particle number is fixed as indicated in the figures. Only exceptions are the charge degenerate points.

FIG. 1: Two examples of interference single electron transistors (ISETs): a benzene molecular junction contacted in the meta configuration (A) and a triple quantum dot artificial molecule (B). The source and drain are parallel polarized ferromagnetic leads.
where two diamonds meet. Here the energy difference of two ground states with consecutive particle numbers is equal to the equilibrium chemical potential of the leads. At finite bias the incoming electrons have enough energy to overcome the level spacing and the Coulomb repulsion and the current flows. As a signature of the new states that enter the bias window, by increasing the voltage the current typically increases steplike.

In ISETs an exception to this picture is represented by the interference blockade where the current decreases for increasing bias generating negative differential conductance (NDC) and eventually vanishes (see green lines in the panels B and C of Fig. 2 and 3). Panels B in the same figures indicate moreover that, for a given gate voltage and in absence of polarization in the leads, the current is blocked only at one specific bias voltage. For parallel polarized leads, however, at a given gate voltage, the current is blocked at two specific bias voltages, one for each spin configuration (panels C). As demonstrated below, the blocking of the minority electrons occurs for the smaller bias voltages. As such full control of the spin configuration in the ISET can be electrically achieved. The interference blockade and its spin selectivity is also demonstrated in panels A and B of Fig. 4 Along the dotted (dashed) line a majority (minority) spin electron is trapped into the molecule. The molecular spin state can thus be manipulated simply by adjusting the bias across the ISET. In the following we discuss the physics of the spin-selective interference blocking and present the necessary ingredients for its occurrence.

This novel blocking is explained by the presence of an $N$-particle non-degenerate state and two degenerate $N + 1$-particle states that simultaneously contribute to transport. It also requires that the ratio between the transition amplitudes $\gamma_{\alpha i} (i = 1, 2, \quad \alpha = L, R)$ between those $N$- and $N + 1$-particle states is different for tunneling at the left ($L$) and at the right ($R$) lead.

\[
\frac{\gamma_{L1}}{\gamma_{L2}} \neq \frac{\gamma_{R1}}{\gamma_{R2}}. \tag{2}
\]

This condition is fulfilled in both cases presented in Fig. 1 due to the geometrical configuration of the left and right lead. Due to condition (2), the degenerate states interfere among themselves such to form pairs of blocking and non-blocking states. The blocking state is only coupled to the source lead (panels C and D of Fig. 1) while the non-blocking one to both source and drain. An electron that populates the blocking state can neither leave towards the drain nor, at high enough bias, return to the source since all energetically available states are there filled. The non-blocking state is thus excluded from the dynamics and the current vanishes.

As such we would conclude that the interference blocking is a threshold effect and the current remains blocked until a new excited state participates to the transport. However, as shown in Fig. 2 and 3 the current is blocked only at specific values of the bias voltage. The explanation of this phenomenon relies on two observations: i) The blocking state (Fig. 4) must be antisymmetric with respect to the plane perpendicular to the system and passing through its center and the atom (quantum dot) closest to the drain; this state is thus also an eigenstate of the projection of the angular momentum in the direction of the drain lead. At positive (negative) bias voltages we call this state the $R(L)$-antisymmetric state $|\psi_{R(L), a}\rangle$. ii) The coupling between the system and the leads not only generates the tunneling dynamics described so far, but also contributes to an internal dynamics of the system that leaves unchanged its particle number. In fact the equation of motion for the reduced density matrix $\rho$ of the system can be cast, to lowest non vanishing order in the coupling to the leads, in the form:

\[
\dot{\rho} = -\frac{i}{\hbar} [H_{\text{sys}}, \rho] - \frac{i}{\hbar} [H_{\text{eff}}, \rho] + \mathcal{L}_{\text{tun}} \rho. \tag{3}
\]

The commutator with $H_{\text{sys}}$ in Eq. 3 represents the coherent evolution of the system in absence of the leads. The operator $\mathcal{L}_{\text{tun}}$ describes instead the sequential tunnelling processes and is defined in terms of the transition amplitudes $\gamma_{\alpha i}$ between the $N$ and $N + 1$ particle states like the ones introduced in equation (2). Eventually $H_{\text{eff}}$ renormalizes the coherent dynamics associated to the system Hamiltonian. It reads:

\[
H_{\text{eff}} = \sum_{\alpha \sigma} \omega_{\alpha \sigma} L_{\alpha}, \tag{4}
\]

where $L_{\alpha}$ is the projection of the angular momentum in the direction of the lead $\alpha$ and, for paramagnetic systems, it does not depend on the spin degree of freedom $\sigma$. Moreover, $\omega_{\alpha \sigma}$ is the frequency renormalization given to the states of spin $\sigma$ by their coupling to the $\alpha$ lead. Equation 3 is an example of Bloch-Redfield equation describing the dynamics of a system coupled to thermal baths. A more detailed version of (3) is presented in the supplementary material.

For sake of simplicity we give in the following the explicit form of the transition amplitudes $\gamma_{\alpha i}$, of the operator $L_{\alpha}$ and of the associated frequency $\omega_{\alpha \sigma}$ only for the benzene ISET and for the ground state transition $6_g \rightarrow 7_g$ that is characterized by interference blocking. The argumentation is nevertheless very general and can be repeated for all the systems exhibiting rotational symmetry. The transition amplitudes read:
\[ \gamma_{\alpha \ell} = \langle 6_g00|d_M \sigma|7_g \ell \sigma \rangle e^{-i\phi_\alpha}. \] (5)

where \( |7_g \ell \sigma \rangle \) are the orbitally degenerate 7 particle ground states, \( \ell = \pm 2 \) the \( z \) projection of the angular momentum in units of \( \hbar \) and \( d_M \sigma \) destroys an electron of spin \( \sigma \) in a reference carbon atom \( M \) placed in the middle between the two contact atoms. Moreover, \( \phi_\alpha \) is the angle of which we have to rotate the molecule to bring the reference atom \( M \) into the position of the contact atom \( \alpha \). The present choice of the reference atom implies that \( \phi_L = -\phi_R = \frac{\pi}{3} \). In the Hilbert space generated by the two-fold orbitally degenerate \( |7_g \ell \sigma \rangle \) the operator \( L_\alpha \) reads:

\[ L_\alpha = \frac{\hbar}{2} \left( \frac{1}{e^{-2|\ell|\phi_\alpha}} e^{i2|\ell|\phi_\alpha} \right). \] (6)

For a derivation of (6) see the supplementary material 3. The frequency \( \omega_{\alpha \sigma} \) is defined in terms of transition amplitudes to all the states of neighbour particle numbers:

\[ \omega_{\alpha \sigma} = \frac{1}{\pi} \sum_{\sigma' \prime(E)} \Gamma_{\alpha \sigma \prime}^0, \]

\[ = \left[ \langle 7_g \ell \sigma | d_{M \sigma} | 8 \{ E \} \rangle \langle 8 \{ E \} | d_{M \sigma}^\dagger \sigma m \sigma \rangle p_\alpha | E - E_{7_g} \rangle + \langle 7_g \ell \sigma | d_{M \sigma} | 6 \{ E \} \rangle \langle 6 \{ E \} | d_{M \sigma}^\dagger \sigma m \sigma \rangle p_\alpha | E_{7_g} - E \rangle \right], \] (7)

where the compact notation \( |N \{ E \}\rangle \) indicates all possible states with particle number \( N \) and energy \( E \), \( p_\alpha (x) = -\text{Re} \left[ \frac{1}{2} + \frac{i\beta}{2\pi} (x - \mu_\alpha) \right] \) where \( \beta = 1/k_B T \), \( T \) is the temperature and \( \psi \) is the digamma function. Moreover \( \Gamma_{\alpha \sigma \prime}^0 = \)

FIG. 2: Benzene ISET: polarized vs. unpolarized configuration. Panel A - Current vs. bias and gate voltage for unpolarized leads. Panel D - Current vs. bias and gate voltage for polarized leads (polarization \( P = 0.85 \)). Panels B and C - Blow up of the 6 → 7 particle transition for both configurations. The unpolarized case shows a single current blocking line and the trapped electron has either up or down polarization. The polarized case shows two current blocking lines, corresponding to the different spin of the trapped electron. The current is given in units of \( e/\Gamma \) where \( \Gamma \) is the bare average rate (supplementary material 4), and the temperature \( k_B T = 0.01b \) where \( b \) is the hopping parameter (supplementary material 1).
FIG. 3: Triple dot ISET: polarized vs. unpolarized configuration. Panel A - Current vs. bias and gate voltage for unpolarized leads. Panel D - Current vs. bias and gate voltage for polarized leads (polarization $P = 0.7$). Panels B and C - Blow up of the $6 \rightarrow 5$ particle transition for both configurations. The selective spin blocking is analogous to the one of the benzene ISET (Fig. 2).

$\frac{2 \pi \hbar}{|t|^2} D_{\alpha \sigma'}$ is the bare tunneling rate to the lead $\alpha$ of an electron of spin $\sigma'$, where $t$ is the tunnelling amplitude and $D_{\alpha \sigma'}$ is density of states for electrons of spin $\sigma'$ in the lead $\alpha$ at the corresponding chemical potential $\mu_\alpha$. Due to the particular choice of the arbitrary phase of the 7 particle ground states, $\omega_{\alpha \sigma}$ does not depend on the orbital quantum numbers $\ell$ and $m$. It depends instead on the bias and gate voltage through the energy of the 6, 7-ground and 8 particle states. In Fig. 5 the black curve depicts $\omega_{L \sigma}$ as a function of the bias in absence of polarization: the frequencies corresponding to the two spin species coincide and thus vanish at the same bias. The same condition, $\omega_{L \sigma} = 0$, also determines the bias at which the current is completely blocked. In fact, at that bias the effective Hamiltonian contains only the projection of the angular momentum in the direction of the right lead (the drain) and the density matrix corresponding to the full occupation of the 7 particle $R$-antisymmetric state ($\rho = |\psi_{R, a}\rangle \langle \psi_{R, a}|$) is the stationary solution of Eq. (5). As we leave the blocking bias the effective Hamiltonian contains also the projection of the angular momentum in the direction of the left lead and the $R$-antisymmetric state is no longer an eigenstate of $H_{\text{eff}}$. The corresponding density matrix is not a stationary solution of (5) and current flows through the system. The $L \leftrightarrow R$ symmetry of the system implies, for negative biases, the blocking condition $\omega_{R \sigma} = 0$.

All-electric-spin control is achieved, in an ISET, only in presence of ferromagnetic leads and with exchange interaction on the system. By manipulating (7) it is possible to show that the frequency splitting $\omega_{\alpha \uparrow} - \omega_{\alpha \downarrow}$ is proportional to the polarization in the $\alpha$ lead, but vanishes in the absence of exchange interaction on the system capable to lift the singlet-triplet degeneracy of the excited 6 and 8 particle states (see the supplementary material [4]). In Fig. 6 we show the frequencies $\omega_{L \sigma} = 0$ vs. bias voltage also for a finite values of the polarization $P$ calculated for the benzene ISET, where exchange splitting is ensured by the strong Coulomb interaction on the system. The interference blocking conditions $\omega_{L \sigma} = 0$ for the $L \rightarrow R$ current are satisfied at different biases for the different spin species. The dotted and dashed lines in Fig. 4 are the representation of the relations $\omega_{L \uparrow} = 0, \omega_{L \downarrow} = 0$ as a function of the bias and polarization, respectively.

In previous studies[11] we have shown that the interference current blocking does not depend on the perfect symmetry of the system but rather relies on the existence of quasi-degenerate states in which the energy splitting is smaller than...
FIG. 4: Spin control. Panel A - Current through the benzene ISET vs bias and polarization at the $6 \rightarrow 7$ electrons transition. Panel B - Population of the majority spin 7 particle state. The two zero current lines at high bias correspond to the maximum or minimum population of the 7 particle majority spin state and thus identify the spin state of the trapped electron on the molecule. Panels C and D - Schematic representation of the spin selective blocking corresponding to the dashed (C) and dotted (D) lines of the panels A and B.

FIG. 5: Blocking condition. Renormalization frequencies $\omega_L^\sigma$ of a benzene ISET as function of the bias and for different lead polarizations. The current blocking condition $\omega_L = 0$ is fulfilled at different biases for the different spin states.

the tunnelling coupling to the source and drain leads. In the proposed structures the degeneracy is associated with the rotational symmetry and it has the advantage of a simple geometrical realization of the interference conditions [2]. Nevertheless the effect is more general and any other structure exhibiting orbital degeneracy is a good candidate for an ISET.

We acknowledge financial support by the DFG under the programs SFB689, SPP1243.

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21 The corresponding eigenvalue depends on the symmetry of the atomic (quantum dot) wave function with respect to the molecular (artificial molecule) plane: $\hbar$ or 0 for symmetric or antisymmetric wave functions respectively.
SUPPLEMENTARY MATERIAL

1. The system Hamiltonian

The Hamiltonian that describes both systems represented in Fig. 1 reads

\[ H_{\text{sys}} = \xi_0 \sum_{i\sigma} d_{i\sigma}^\dagger d_{i\sigma} + b \sum_{i\sigma} \left( d_{i\sigma}^\dagger d_{i+1\sigma} + d_{i+1\sigma}^\dagger d_{i\sigma} \right) \]

\[ + U \sum_i \left( n_{i\uparrow} - \frac{1}{2} \right) \left( n_{i\downarrow} - \frac{1}{2} \right) \]

\[ + V \sum_i \left( n_{i\uparrow} + n_{i\downarrow} - 1 \right) \left( n_{i+1\uparrow} + n_{i+1\downarrow} - 1 \right), \]

where \( d_{i\sigma}^\dagger \) creates an electron of spin \( \sigma \) in the \( p_z \) orbital of carbon \( i \) or in the ground state of the quantum dot \( i \) and \( i = 1, \ldots, 6(3) \) runs over the six carbon atoms (three quantum dots) of the system. Moreover, \( n_{i\sigma} = d_{i\sigma}^\dagger d_{i\sigma} \). The effect of the gate is included as a renormalization of the on-site energy \( \xi = \xi_0 - eV_g \) (\( V_g \) is the gate voltage) and we conventionally set \( V_g = 0 \) at the charge neutrality point. The parameters that we have used are \( b = 2.5eV, U = 9eV, V = 6eV \).

2. The generalized master equation

We describe the dynamics of the system with a generalized master equation (GME) for the reduced density matrix \( \rho \). This equation is obtained from the Liouville equation for the full density matrix as a perturbation to the lowest non-vanishing order in the coupling to the leads by tracing out the leads degrees of freedom. A generic formulation of the GME for a SET in presence of degeneracies or quasi-degeneracies can be found elsewhere e.g. \(^{11}\). We concentrate here on the range of gate and bias voltages at which the dynamics is restricted to transitions involving the \( |6g,00\rangle \) and \( |7g,\ell\sigma\rangle \) many particle states of the benzene ISET.

The seven particle states are spin and orbital degenerate. The general theory of the GME would require a priori to keep thus a full 4x4 density matrix describing the 7 particle subspace. In presence of parallel polarized leads, though, the coherences between different spin degrees of freedom can be neglected since spin is always conserved by the electrons while travelling through the device. The GME can thus be written in terms of the nine variables collected in the 1x1 matrix \( \rho^{6g} \) and the two 2x2 matrices \( \rho^{7g,\sigma} \) with \( \sigma = \uparrow, \downarrow \). Due to the rotational symmetry of the system it is more convenient to refer to another set of variables, namely to describe the dynamics in terms of the occupation probabilities \( W_6 \), \( W_7\sigma \) and the expectation values of the different projections of the angular momentum for the system. The new set of variables is:

\[ W_6 = \rho^{6g}, \]

\[ W_7\sigma = \text{Tr}\{\rho^{7g,\sigma}\}, \]

\[ L_{z\sigma} = \text{Tr}\{L_z \rho^{7g,\sigma}\}, \]

\[ L_{z\sigma} = \text{Tr}\{L_z \rho^{7g,\sigma}\}. \]

(10)

The operator \( L_z \) is the generator of the set of discrete rotations around the axis perpendicular to the plane of the benzene molecule that bring the molecule into itself and can be written within the 7 particle Hilbert space spanned by the vectors \( |7g,\ell\sigma\rangle \) as \( L_z = -\hbar |\ell\rangle \sigma_z \), where \( \sigma_z \) is the third Pauli matrix. The operator \( L_\alpha \) generates, in the same space, the discrete rotations around the axis in the molecular plane and passing through the center and the atom closest to the contact \( \alpha \). Finally, the dynamics for the variables introduced in Eq. (10) is given by the equations:
\[
\begin{align*}
\hat{W}_6 &= 2 \sum_{\alpha} \Gamma_{\alpha\sigma} \left[ -f_\alpha^+(\Delta E)W_6 + f_\alpha^-(\Delta E)L_{\alpha\sigma} \right], \\
\hat{W}_{7\sigma} &= 2 \sum_{\alpha} \Gamma_{\alpha\sigma} \left[ f_\alpha^+(\Delta E)W_6 - f_\alpha^-(\Delta E)L_{\alpha\sigma} \right], \\
\hat{L}_{\alpha\sigma} &= -2 \Gamma_{\alpha\sigma} f_\alpha^-(\Delta E) + 2 \left\{ \Gamma_{\alpha\sigma} f_\alpha^+(\Delta E) + \Gamma_{\alpha\sigma} f_\alpha^+(\Delta E) \cos^2|\ell|(\phi_\alpha - \phi_\beta) \right\} W_6 \\
&\quad + \Gamma_{\alpha\sigma} f_\alpha^-(\Delta E) \sin^2|\ell|(\phi_\alpha - \phi_\beta)W_{7\sigma} - \Gamma_{\alpha\sigma} f_\alpha^-(\Delta E)(L_{\alpha\sigma} + L_{\beta\sigma}) + \frac{\sin[2|\ell|(\phi_\alpha - \phi_\beta)]}{4} \omega_{\alpha\beta} L_{2\sigma}, \\
\hat{L}_{2\sigma} &= -\sum_{\alpha} \Gamma_{\alpha\sigma} f_\alpha^-(\Delta E)L_{2\sigma} - 2 \tan[|\ell|(\phi_\alpha - \phi_\beta)](\omega_{L\sigma} - \omega_{R\sigma})(W_{7\sigma} - L_{L\sigma} - L_{R\sigma}) \\
&\quad - 2 \cot[|\ell|(\phi_\alpha - \phi_\beta)](\omega_{L\sigma} + \omega_{R\sigma})(L_{L\sigma} - L_{R\sigma}),
\end{align*}
\]

where \( \Gamma_{\alpha\sigma} = \Gamma_{\alpha\sigma}^0 (|6\sigma\rangle \langle 6\sigma|)(\ell g \ell \sigma)|^2 \) is the tunnelling rate at the lead \( \alpha \) involving the ground states with 6 and 7 particles. Terms describing sequential tunnelling from and to the lead \( \alpha \) are proportional to the Fermi functions \( f_\alpha(x) := f(x - \mu_\alpha) \) and \( f_\alpha(x) := 1 - f_\alpha^+(x) \), respectively, and \( \Delta E = E_{6g} - E_{7g} + eV_g \) where \( E_{6g} \) and \( E_{7g} \) are the energies of the 6 and 7 particle ground states. Finally with \( \bar{\alpha} \) we mean the lead opposite to the lead \( \alpha \). By using the expression \(|\ell|\) (to be substituted with 2 for the 6 → 7 particle transition) we maintained the generality of the equations. The replacement \(|\ell| = 2 \rightarrow 1\) and the appropriate redefinition of \( \Delta E \) is enough to treat the 6 → 5 transition. Another important generalization concerns the position of the leads. The para \((\phi_\alpha - \phi_R = \pi)\) and ortho \((\phi_\alpha - \phi_R = \pi/3)\) configuration are also treated within the same equations. In particular one can see that all the terms containing the renormalization frequencies drop from the equations in the para configuration and that the equations for the ortho and meta configuration coincide.

### 3. Matrix form of the operator \( L_\alpha \)

The explicit form of \( L_\alpha \) is given in Eq. \((13)\). We give here its derivation. It is convenient, for this purpose, to choose the arbitrary phases of the states \(|7g\ell\sigma\rangle\) in such a way that the rotation of \( \pi \) around the axis passing through a reference atom \( M \) and the center of the molecule transforms \(|7g\ell\sigma\rangle\) into \(-|7g - \ell\sigma\rangle\). In other terms

\[
\exp\left(i\frac{\pi L_\alpha}{\hbar}\right) = -\sigma_x,
\]

where \( \sigma_x \) is the first Pauli matrix. The relation is in fact an equation for \( L_M \) and the solution reads:

\[
L_M = \frac{\hbar}{2} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}.
\]

Eventually we obtain \( L_\alpha \) by rotation of \( L_M \) in the molecular plane, namely:

\[
L_\alpha = e^{-\frac{i}{2} \phi_\alpha L_z} L_M e^{\frac{i}{2} \phi_\alpha L_z} = \frac{\hbar}{2} \begin{pmatrix} 1 & e^{i2|\ell|\phi_\alpha} \\ e^{-i2|\ell|\phi_\alpha} & 1 \end{pmatrix},
\]

where \( \phi_\alpha \) is the angle of which we have to rotate the molecule to bring the reference atom \( M \) into the position of the contact atom \( \alpha \).
4. The spin splitting of the renormalization frequencies

The spin splitting of the renormalization frequencies is obtained from Eq. (7). By introducing the average bare rate
\[ \Gamma = \frac{\Gamma_0^\alpha + \Gamma_0^\beta}{2}, \]
for simplicity equal in both leads, and using the fact that benzene is paramagnetic we get:

\[
\omega_\alpha \uparrow - \omega_\alpha \downarrow = 2 \Gamma_0^\alpha P_\alpha \frac{1}{\pi} \sum_{\{E\}} \left[ \langle \ell \uparrow | d_{M^\dagger} | \ell^\alpha 8\{E\} \rangle \langle 8\{E\} | d_M^\dagger | \uparrow \rangle p_\alpha (E - E_{7g}) \right. \\
+ \langle \ell \uparrow | d_{M^\dagger} | \ell^\alpha 6\{E\} \rangle \langle 6\{E\} | d_M^\dagger | \uparrow \rangle p_\alpha (E_{7g} - E) \\
- \langle \ell \uparrow | d_{M^\dagger} | \ell^\alpha 8\{E\} \rangle \langle 8\{E\} | d_M^\dagger | \downarrow \rangle p_\alpha (E - E_{7g}) \\
- \langle \ell \uparrow | d_{M^\dagger} | \ell^\alpha 6\{E\} \rangle \langle 6\{E\} | d_M^\dagger | \downarrow \rangle p_\alpha (E_{7g} - E) \right]
\]

(15)

where one appreciates the linear dependence of the spin splitting on the lead polarization \( P_\alpha \). The first and the third term of the sum would cancel each other if the energy of the singlet and triplet 8 particle states would coincide. An analogous condition, but this time on the 6 particle states, concerns the second and the fourth terms. For this reason the exchange interaction on the system is a necessary condition to obtain spin splitting of the renormalization frequencies and thus the full all-electric spin control.