Interference blockade in symmetric nano-junctions

Andrea Donarini

Georg Begemann, Dana Darau and Milena Grifoni

University of Regensburg, Germany

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Macroscopic interference

Young's light-interference experiment (1801)

Double-slit experiment with interference of single electrons (1961)
Interference SET

The interference occurs between transmission paths involving orbitally (quasi-)degenerate states
(Benzene) ISET...

- Weak coupling
- Coulomb interaction
- Molecular size
- Low temperature

- Rotational symmetry

- Contact geometry

\[ h\Gamma \ll k_B T \ll \Delta E_{ex} \]

\[ E_1 = E_2 \]

\[ \frac{\gamma_{1L}}{\gamma_{2L}} \neq \frac{\gamma_{1R}}{\gamma_{2R}} \]
... with a magnetic flavour

- **Coulomb** interaction
- **Molecular size**

\[ E_{\text{triplet}} \neq E_{\text{singlet}} \]

- Parallel **ferromagnetic** leads

\[ \Gamma_{\alpha\uparrow} \neq \Gamma_{\alpha\downarrow} \]

**Exchange splitting**

**Spin symmetry breaking**

The interplay between orbital and spin degree of freedom allows all-electrical spin control on the junction.
The “two paths” in the ISET
Coulomb blockade

- **Gating** of 2 nm sized molecule
- **Weak coupling** realization with specific anchor groups

Symmetry breaking contacts

Para configuration

Meta configuration

The Hamiltonian

\[
H = H_{\text{Sys}} + H_{\text{leads}} + H_{\text{tun}}
\]

\[
H_{\text{Sys}} = \frac{H_{\text{ben}}}{H_{\text{TD}}}
\]

\[
H_{\text{leads}} = \sum_{\alpha k \sigma} (\epsilon_k - \mu_\alpha) c_{\alpha k \sigma}^\dagger c_{\alpha k \sigma}
\]

\[
H_{\text{tun}} = t \sum_{\alpha k \sigma} \left( d_{\alpha \sigma}^\dagger c_{\alpha k \sigma} + c_{\alpha k \sigma}^\dagger d_{\alpha \sigma} \right)
\]
Interacting isolated benzene

- The Pariser-Parr-Pople Hamiltonian for isolated benzene reads:

\[
H_{\text{ben}}^0 = \xi_0 \sum_{i\sigma} d_{i\sigma}^\dagger d_{i\sigma} + b \sum_{i\sigma} \left( d_{i\sigma}^\dagger d_{i+1\sigma} + d_{i+1\sigma}^\dagger d_{i\sigma} \right) + U \sum_i (n_{i\uparrow} - \frac{1}{2})(n_{i\downarrow} - \frac{1}{2}) + V \sum_i (n_{i\uparrow} + n_{i\downarrow} - 1)(n_{i+1\uparrow} + n_{i+1\downarrow} - 1)
\]

- The size of the Fock space for the many-body system \(4^6 = 4096\) since for each site there are 4 possibilities: \(|0\rangle, |\uparrow\rangle, |\downarrow\rangle, |\uparrow\downarrow\rangle\)

- Within this Fock space we diagonalize exactly the Hamiltonian.
Symmetry of the ground states

<table>
<thead>
<tr>
<th>N</th>
<th>Degeneracy</th>
<th>GS energy[eV] (at $\xi = 0$)</th>
<th>GS symmetry representation</th>
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<tr>
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</tbody>
</table>

Rotation phase factors

Under rotation of an angle $\phi = \frac{n\pi}{3}$

- $R_{\phi}|6_g\rangle = |6_g\rangle$ No phase acquired
- $R_{\phi}|7_g \ell\rangle = e^{-i\ell\phi}|7_g \ell\rangle$ for $\ell = \pm 2$
  
\[
\ell = +2 \quad \exp\left(\frac{i2\pi}{3}\right)
\]

\[
\ell = -2 \quad \exp\left(-\frac{i2\pi}{3}\right)
\]
Generalized Master Equation

- We start with the **Liouville** equation:
  \[ \dot{\rho} = -\frac{i}{\hbar} [\mathcal{H}, \rho] \]

- We consider a reduced density matrix **block-diagonal** in spin, energy and particle number. We keep coherencies between **orbitally** degenerate states.

- The **Generalized Master Equation** is an equation of motion for the reduced density matrix. We calculate it in the lowest non-vanishing order in the coupling to the leads and in the Markov approximation. It reads:

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

**Coherent dynamics**  **Effective internal dynamics**  **Tunnelling dynamics**
The effective Hamiltonian is expressed in terms of angular momentum operators and renormalization frequencies:

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

In particular in the Hilbert space of the 7 particle ground states

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

\[ \omega_{\alpha \sigma} = \frac{1}{\pi} \sum_{\sigma'} \Gamma_{\alpha \sigma}^{0} \left[ \langle 7_g \ell \sigma | d_{M \sigma'}^{\dagger} | 8 \{ E \} \rangle \langle 8 \{ E \} | d_{M \sigma'}^\dagger | 7_g m \sigma \rangle p_\alpha (E - E_{7_g}) + \langle 7_g \ell \sigma | d_{M \sigma'}^{\dagger} | 6 \{ E \} \rangle \langle 6 \{ E \} | d_{M \sigma'}^{\dagger} | 7_g m \sigma \rangle p_\alpha (E_{7_g} - E) \right] \]
Current operator

- **Current**: using the GME we find the **operator**:

\[
\hat{I}_L = \Gamma_L \sum_{N E \tau} P_{N E} \left[ d_{L \tau} f_L^+ (H^0_{\text{ben}} - E) d_{L \tau}^+ - d_{L \tau}^+ f_L^- (E - H^0_{\text{ben}}) d_{L \tau} \right] P_{N E}.
\]

and thus calculate the **stationary current**:

\[
I_L = \text{Tr}\{\sigma_{\text{stat}} \hat{I}_L\} = -I_R
\]
Para vs. Meta


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Conductance suppression

A: non-degenerate  ↔  B: non-degenerate  →  Equal

A: non-degenerate  ↔  E: degenerate  →  Suppressed

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Destructive interference

\[ \Lambda = \left| \sum_{nm, \tau} \langle N, n | d_{L, \tau} | N + 1, m \rangle \langle N + 1, m | d_{R, \tau} | N, n \rangle \right|^2 \]

\[ \Lambda = \left| \sum_{nm, \tau} | \langle N, n | d_{L, \tau} | N + 1, m \rangle |^2 e^{i \phi_{nm}} \right|^2 \]

\[ d_{R, \tau} = R_{\phi}^\dagger d_{L, \tau} R_{\phi} \]

In particular for the transition 6 -7 in the meta configuration:

\[ \Lambda = \left| \langle 6_g | d_{L, \tau} | 7_g, +2, \tau \rangle \right|^2 e^{i \frac{2\pi}{3}} + \left| \langle 6_g | d_{L, \tau} | 7_g, -2, \tau \rangle \right|^2 e^{-i \frac{2\pi}{3}} \right|^2 \]

\[ \begin{array}{c}
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\end{array} \]
The 8 electrons “anomaly”

Mirror symmetry of the para-configuration

The tunnelling preserves this mirror symmetry: the lowest 8 electron state involved in transport is the mirror-symmetric (first excited) state with $E_{2g}$ symmetry.
NDC: the role of coherences

- The 7 particle ground state has spin and orbital degeneracies;

- **Physical basis**: the basis that diagonalizes the stationary density matrix;

- The physical basis depends on the bias: in whatever reference basis, coherences are essential for a correct description of the system;

- The visualization tool: position resolved transition probability to the physical basis:

\[
P(x, y; \ell \tau) = \lim_{L \to \infty} \sum_\sigma \frac{1}{2L} \int_{-L/2}^{L/2} dz |\langle 7_g \ell \tau | \psi_\sigma^\dagger(\vec{r}) | 6_g \rangle|^2
\]
Interference blockade

Geometry

Blocking state

Non-blocking state

I-V for transition 6 - 7

Energetics

Current onset

Coulomb blockade

Interference blockade

The blocking state is an eigenstate of the effective Hamiltonian

$\omega_{L\sigma} = 0$
Normal vs. ferromagnetic leads
Normal vs ferromagnetic leads

Current

$\omega L_0$

Bias voltage

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Level renormalization in presence of polarized leads

We obtain a difference in the renormalization frequencies for the 2 spin directions linear in the polarization of the leads:

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

The splitting of the level renormalization depends crucially on the Coulomb interaction on the molecule and vanishes in absence of exchange.
Selective Interference Blocking

AD, G. Begemann, and M. Grifoni *Nano Lett.* 9, 2897 (2009)
Robustness

• We have tested the robustness of the effects against:
  
  – Residual potential drop on the benzene molecule (in weak coupling to the leads the potential drop is concentrated at the contacts)
  
  – On-site energy renormalization of the contact atom due to different anchor groups
  
  – Lifting of the electronic degeneracy due to deformation (static Jahn-Teller effect)

• The minimal necessary condition is quasi-degeneracy:

\[ \delta E \ll \hbar \Gamma \]
Blocking conditions

The interference blocking state:

- is a linear combination of (quasi-)degenerate system eigenstates
- is achievable from the global minimum via a finite number of allowed transitions
- has vanishing tunnelling amplitudes for all energetically allowed outgoing transitions

\[ L_{\text{tun}} \sigma_B = 0 \]

- is an eigenstate of the effective Hamiltonian

\[ [H_{\text{eff}}, \sigma_B] = 0 \]
The triple dot ISET

\[ H = H_{\text{sys}} + H_{\text{leads}} + H_{\text{tun}} \]

\[ H_{\text{sys}} = \xi_0 \sum_{i\sigma} d_{i\sigma}^\dagger d_{i\sigma} + b \sum_{i\sigma} \left( d_{i\sigma}^\dagger d_{i+1\sigma} + d_{i+1\sigma}^\dagger d_{i\sigma} \right) \]
\[ + U \sum_i \left( n_{i\uparrow} - \frac{1}{2} \right) \left( n_{i\downarrow} - \frac{1}{2} \right) \]
\[ + V \sum_i \left( n_{i\uparrow} + n_{i\downarrow} - 1 \right) \left( n_{i+1\uparrow} + n_{i+1\downarrow} - 1 \right) \]

\[ H_{\text{tun}} = t \sum_{\alpha k \sigma} \left( c_{\alpha k \sigma}^\dagger d_{\alpha \sigma} + d_{\alpha \sigma}^\dagger c_{\alpha k \sigma} \right) \]

Extended Hubbard Hamiltonian with on-site and nearest neighbors Coulomb interaction

Tunnelling restricted to the dot closest to the corresponding lead

Ferromagnetic leads with equal parallel polarization
Stability diagram

Interference Blockade --ground states--

Interference Blockade --excited state--

Coulomb Blockade

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Polarized leads

Parallel polarized leads
No magnetic field on the system
All-electric spin control
Many-body spectrum

Orbital degeneracy
Spin degeneracy

Orbital degeneracy produces interference blocking
Excited state blocking

Source transition
Drain transition

2x3 System

Coulomb Blockade

N = 2

V_g

N = 1

N = 2
Excited state blocking

Source transition
Drain transition

2x3

1x2

1x1
System

N=1
N=2

N=2

Current

V_g

V_o

2_1 \leftrightarrow 3_0
1_0 \leftrightarrow 2_1
1_1 \leftrightarrow 2_1
1_0 \leftrightarrow 2_0

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Excited state blocking

- Source transition
- Drain transition

2x3

Interference Blockade

1x2

N=1

1x1

System

N=2

Three linear combinations of 2-particle excited states are coupled ONLY to the source.
Triplet splitting

The states decoupled from the right lead are eigenstates of $L_R$. They are eigenstates of $H_{\text{eff}}$ only if

$$\omega_L S_z = 0$$
Conclusions

- The interplay between electron-electron interaction and orbital symmetry is important to understand transport through an ISET;

- **Destructive interference** between degenerate states implies current blocking at specific bias voltages.

- In presence of parallel ferromagnetic leads the **current blocking is spin-selective**. We obtain all-electrical spin control on the junction.

- **Coherences** between degenerate states are essential to capture the interference effects in an ISET.

- Interference is **robust** against symmetry breaking.
Thanks

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References:

G. Begemann, D. Darau, AD, and M. Grifoni PRB 77, 201406(R) (2008)
D. Darau, G. Begemann, AD, and M. Grifoni PRB 79, 235404 (2009)
AD, G. Begemann, and M. Grifoni Nano Lett. 9, 2897 (2009)

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