All-electrical spin control in a benzene interference single electron transistor

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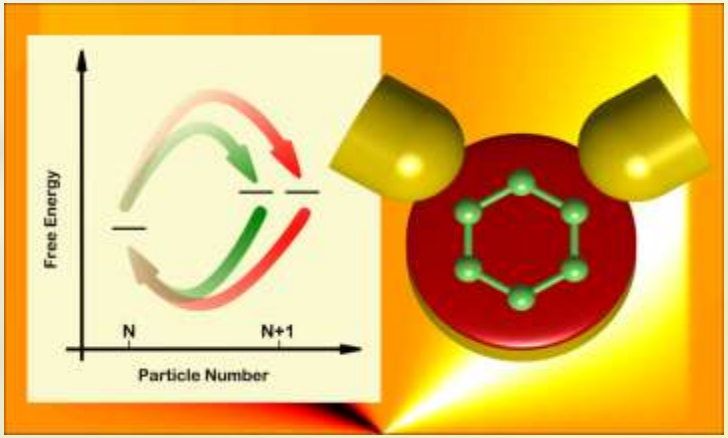
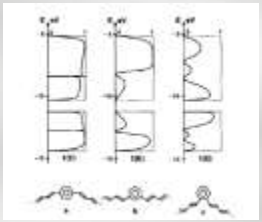
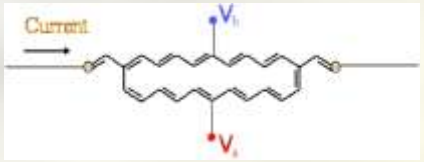


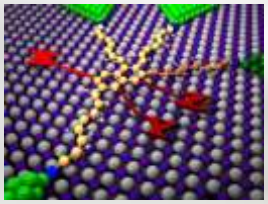
Intramolecular interference



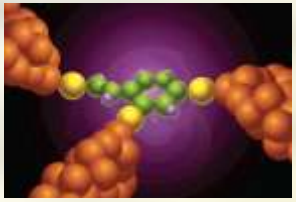
P. Sautet and C. Joachim
Chem. Phys. Lett. **153**, 511 (1988)



R. Baer and D. Neuhauser
JACS, **124**, 4200 (2002)

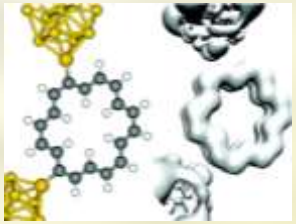


R. Stadler, et al.
Nanotechnology, **14**, 138 (2003)

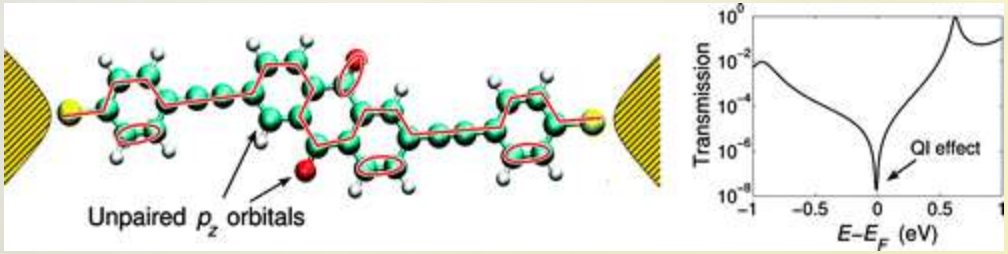


D. V. Cardamone, et al.
Nano Lett., **6**, 2422 (2006)

G. Solomon, et al.
JACS **130**, 17307 (2008)

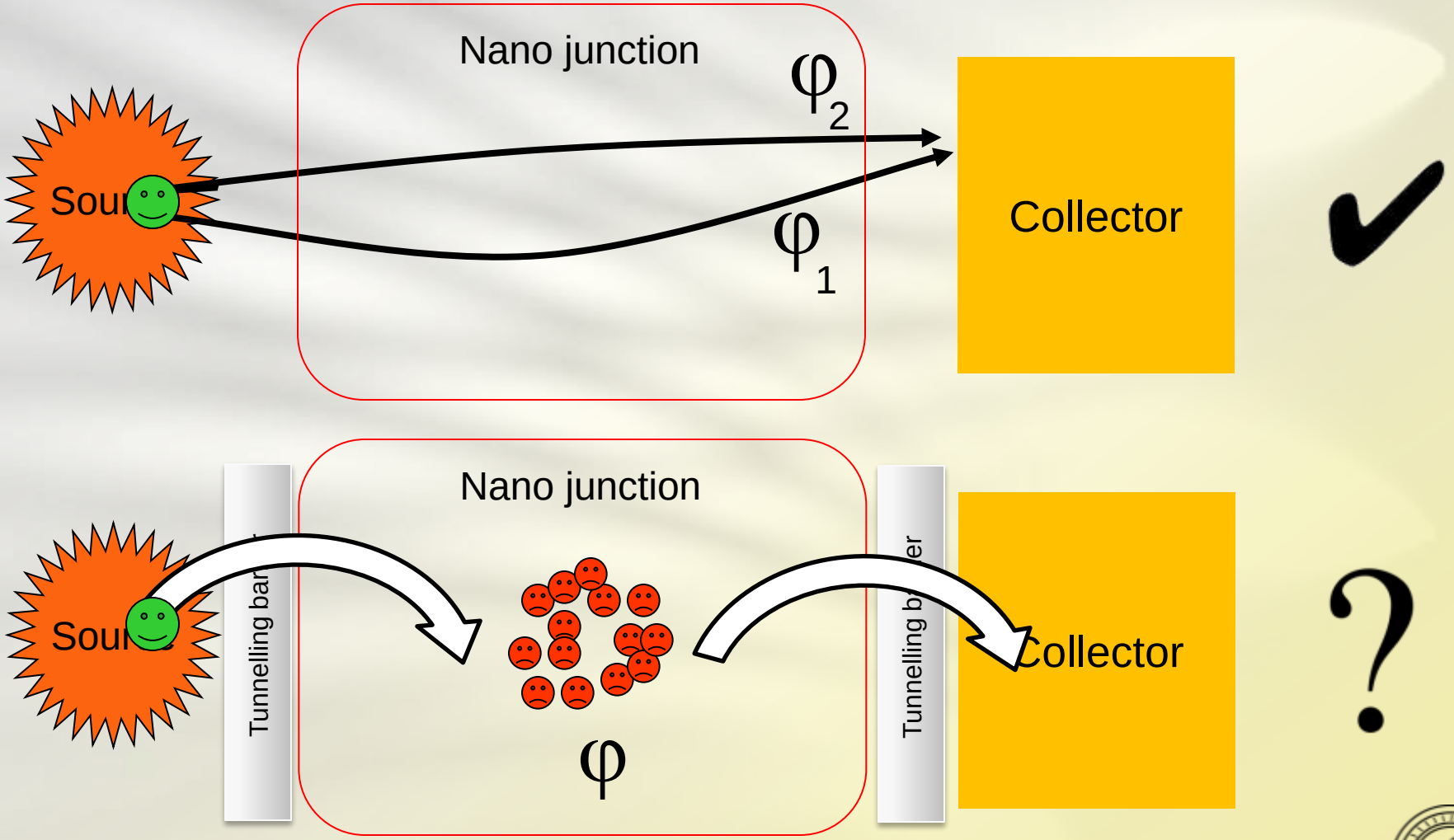


S.H. Ke, et al.
Nano Lett., **8**, 3257 (2008)

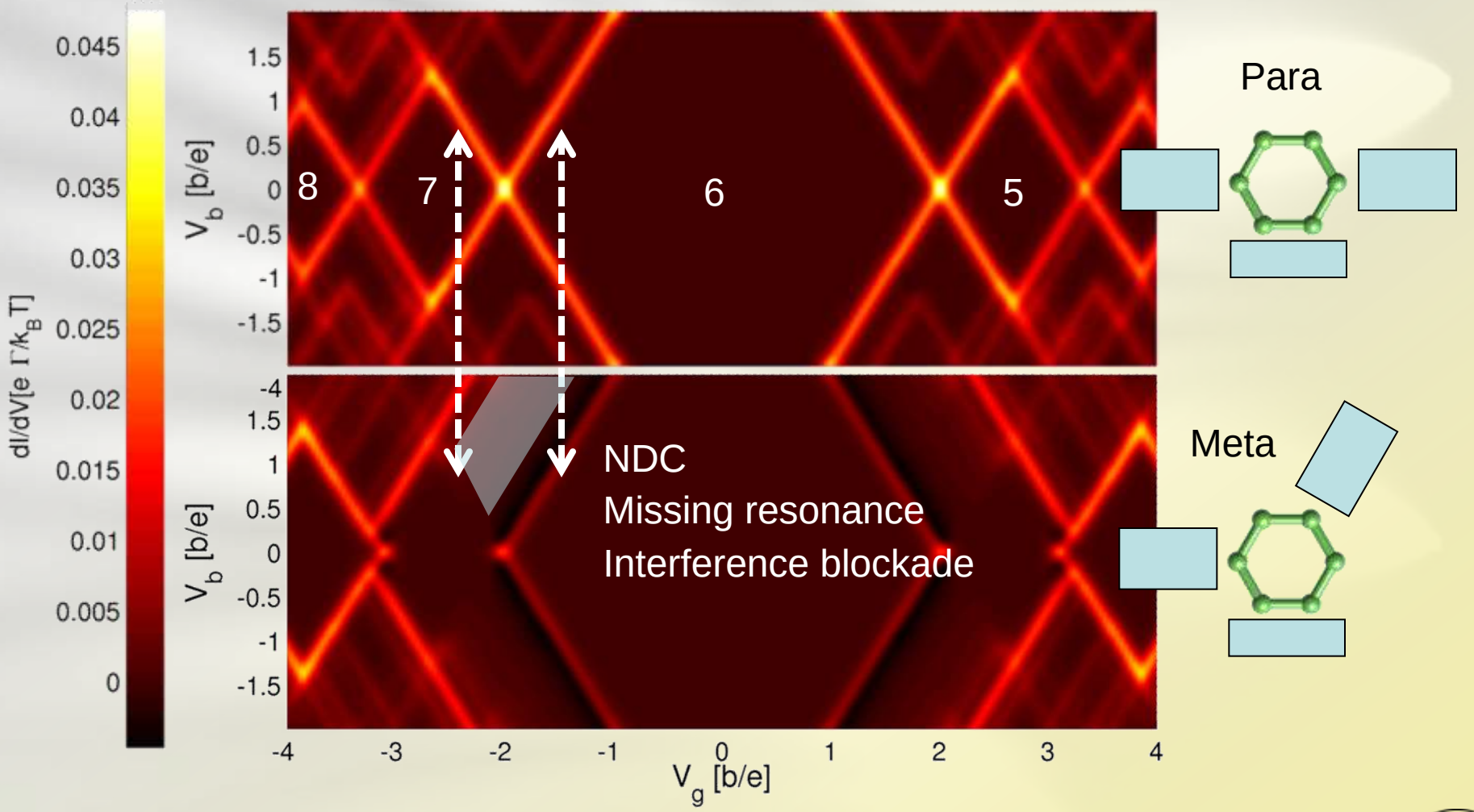


T. Markussen, et al.
Nano Lett., **10**, 4260 (2010)

Interference in weak coupling ?



Interference blockade



G. Begemann, D. Darau, A. Donarini, M. Grifoni, *Phys. Rev. B* **77**, 201406(R) (2008)

(Benzene) ISET...

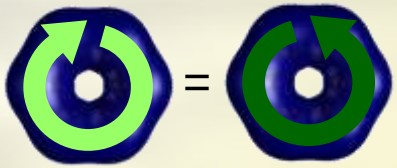
- **Weak coupling**
- **Coulomb** interaction
- Molecular **size**
- **Low** temperature

⇒ **Coulomb blockade**

$$\hbar\Gamma \ll k_B T \ll \Delta E_{ex}$$

- **Rotational** symmetry

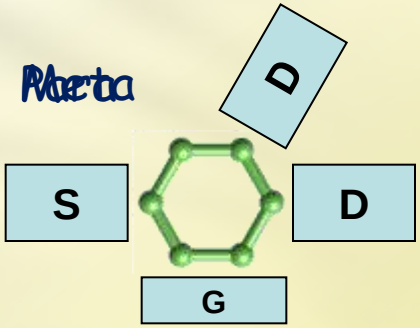
⇒ **Orbitally degenerate states**



$$E_1 = E_2$$

- Contact **geometry**

⇒ **Contact symmetry breaking**

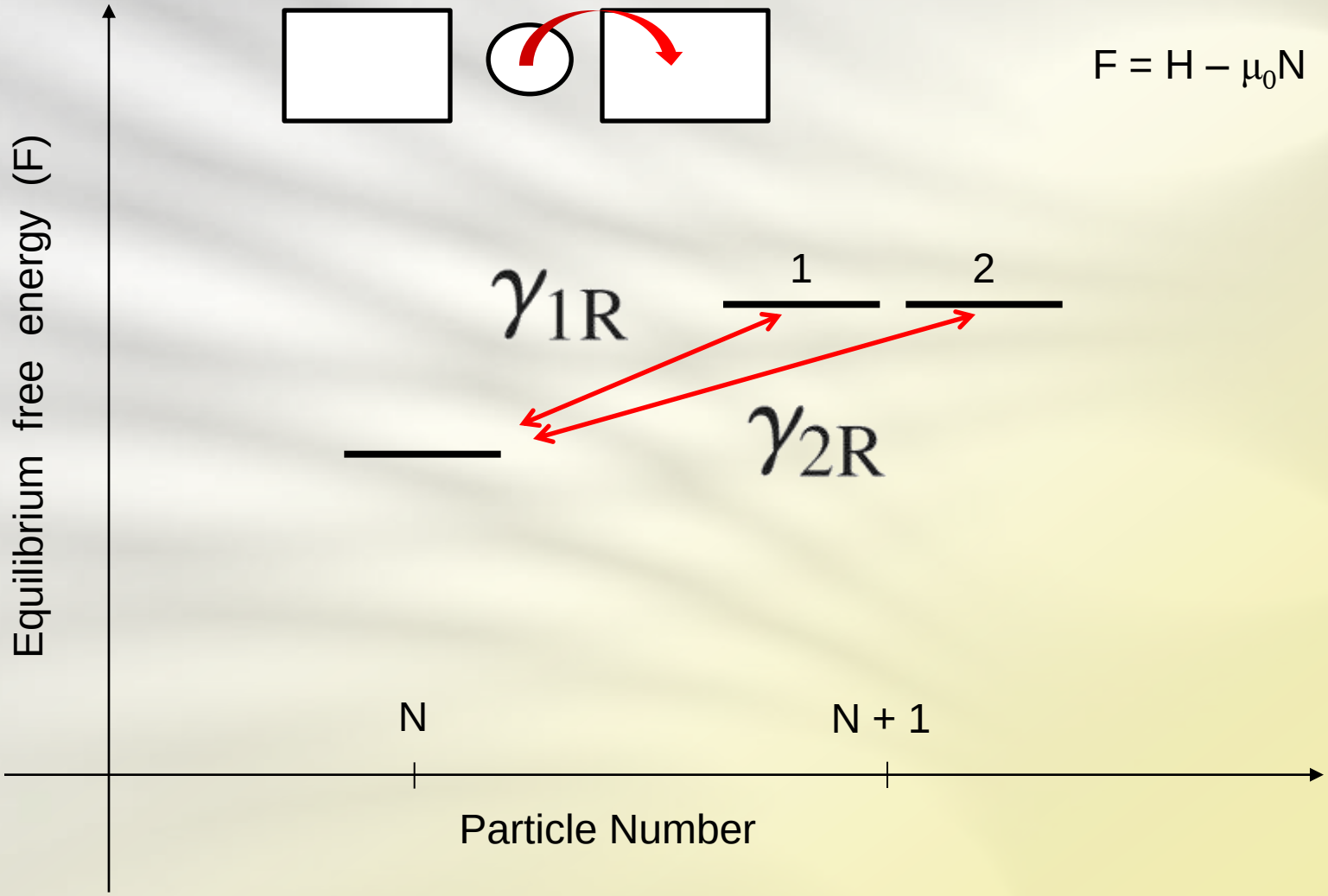


$$\frac{\gamma_{1L}}{\gamma_{2L}} \neq \frac{\gamma_{1R}}{\gamma_{2R}}$$

Many-body tunnelling amplitudes

$F = H - \mu_0 N$

Many-body tunnelling amplitudes



Contact symmetry breaking

$$|1'\rangle = a|1\rangle + b|2\rangle \quad \Rightarrow \quad \gamma_{1'L} = a\gamma_{1L} + b\gamma_{2L}$$

$$\frac{\gamma_{1L}}{\gamma_{2L}} \neq \frac{\gamma_{1R}}{\gamma_{2R}} \quad \Rightarrow \quad \exists \; |1'\rangle, |2'\rangle$$

$$\left.\begin{aligned} \gamma_{1'L} &\neq 0 \\ \gamma_{1'R} &= 0 \\ \gamma_{2'L} &\neq 0 \\ \gamma_{2'R} &\neq 0 \end{aligned}\right\} \text{Contact symmetry breaking}$$

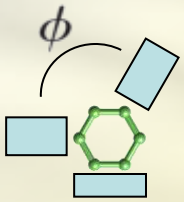
Contact symmetry breaking

$$|1'\rangle = a|1\rangle + b|2\rangle \quad \Rightarrow \quad \gamma_{1'L} = a\gamma_{1L} + b\gamma_{2L}$$

$$\frac{\gamma_{1L}}{\gamma_{2L}} = \frac{\gamma_{1R}}{\gamma_{2R}} e^{4i\phi} \quad \Rightarrow \quad \exists \; |1'\rangle, |2'\rangle$$

$$\gamma_{1'L} \neq 0 \qquad \gamma_{1'R} = 0 \qquad \gamma_{2'L} \neq 0 \qquad \gamma_{2'R} \neq 0$$

Neutral Benzene $\leftrightarrow$ Benzene Anion



POSTER

More degenerate states? See
A. Donarini , G. Begemann and M. Grifoni *Phys. Rev. B*, **82**, 125451 (2010)
for the general theory.

... with a magnetic flavour

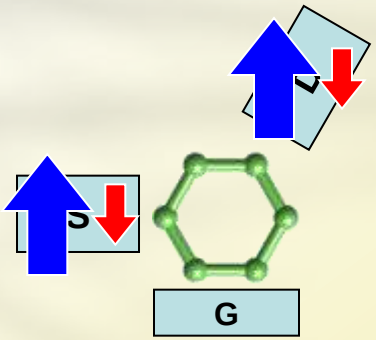
- **Coulomb** interaction
- Molecular **size**

⇒ **Exchange splitting**

$$E_{\text{triplet}} \neq E_{\text{singlet}}$$

- Parallel **ferromagnetic** leads

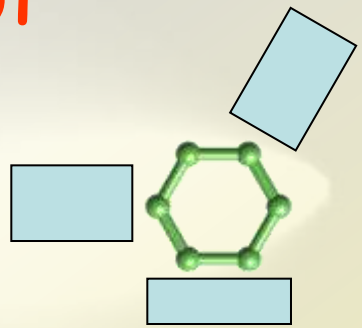
⇒ **Spin symmetry breaking**



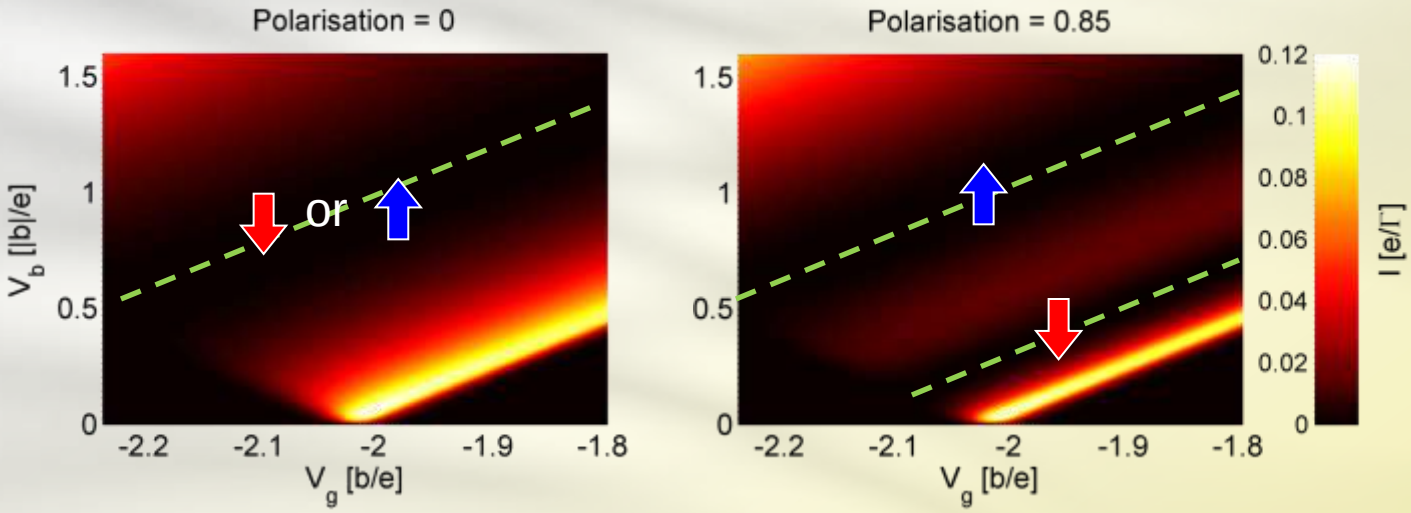
$$\Gamma_{\alpha\uparrow} \neq \Gamma_{\alpha\downarrow}$$

All-electrical spin control is achieved on the system

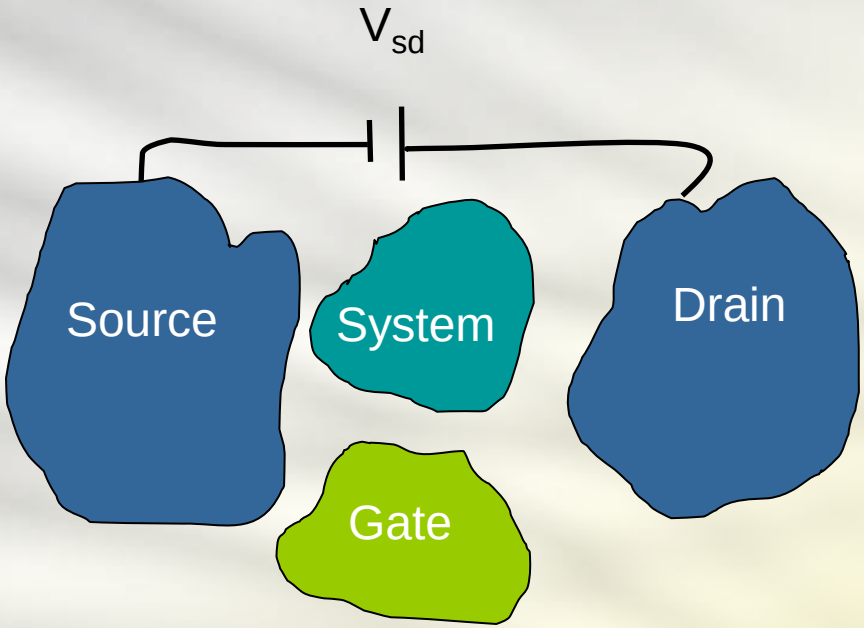
All-electrical spin control



Current maps



The Hamiltonian



$$H = H_{\mathrm{Sys}} + H_{\mathrm{leads}} + H_{\mathrm{tun}}$$

$$H_{\mathrm{Sys}} = H_{\mathrm{ben}}$$

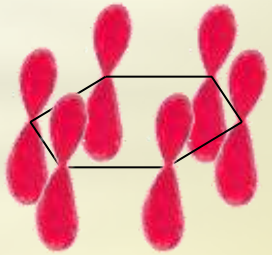
$$H_{\mathrm{leads}} = \sum_{\alpha\, k\, \sigma} \epsilon_k c^{\dagger}_{\alpha k\sigma} c_{\alpha k\sigma}$$

$$H_{\mathrm{tun}} = t \sum_{\alpha k\sigma} \left(d^{\dagger}_{\alpha\sigma} c_{\alpha k\sigma} + c^{\dagger}_{\alpha k\sigma} d_{\alpha\sigma} \right)$$

Interacting isolated benzene

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

$$H^0_{\rm ben} = \xi_0 \sum_{i\sigma} d^\dagger_{i\sigma} d_{i\sigma} + b \sum_{i\sigma} \left(d^\dagger_{i\sigma} d_{i+1\sigma} + d^\dagger_{i+1\sigma} d_{i\sigma} \right)$$
$$+U \sum_i \left(n_{i\uparrow} - \tfrac{1}{2} \right) \left(n_{i\downarrow} - \tfrac{1}{2} \right)$$
$$+V \sum_i \left(n_{i\uparrow} + n_{i\downarrow} - 1 \right) \left(n_{i+1\uparrow} + n_{i+1\downarrow} - 1 \right)$$



- 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.

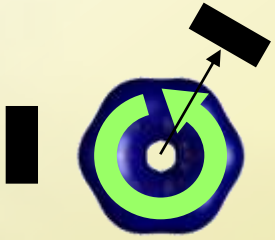
Symmetry of the ground states

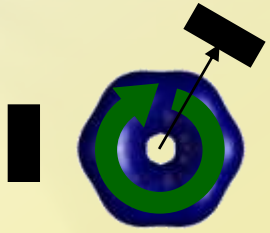
N	Degeneracy	GS energy[eV] (at $\xi = 0$)	GS symmetry representation
0	1	0	A_{1g}
1	2	-22	A_{2u}
2	1	-42.25	A_{1g}
3	4	-57.42	E_{1g}
4	3	-68.875	A_{2g}
5	4	-76.675	E_{1g}
6	1	-81.725	A_{1g}
7	4	-76.675	E_{2u}
8	3	-68.875	A_{2g}
9	4	-57.42	E_{2u}
10	1	-42.25	A_{1g}
11	2	-22	B_{2g}
12	1	0	A_{1g}

Rotation phase factors

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

- $\mathcal{R}_\phi |6_g\rangle = |6_g\rangle$ No phase acquired
- $\mathcal{R}_\phi |7_g\, \ell\rangle = e^{-i\ell\phi} |7_g\, \ell\rangle$ $\ell = \pm 2$

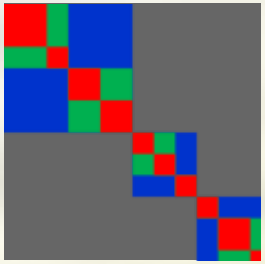
$\ell = +2$  $\exp\left(+i\frac{2\pi}{3}\right)$

$\ell = -2$  $\exp\left(-i\frac{2\pi}{3}\right)$

Generalized Master Equation

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

- We define the reduced density matrix $\sigma = \mathrm{Tr}_{\mathrm{Leads}}\{\rho\}$ which is block-diagonal in

$\sigma =$ 

particle number
spin
energy

- We keep the coherences between orbitally degenerate states.

- The **Generalized Master Equation** is the equation of motion for σ :

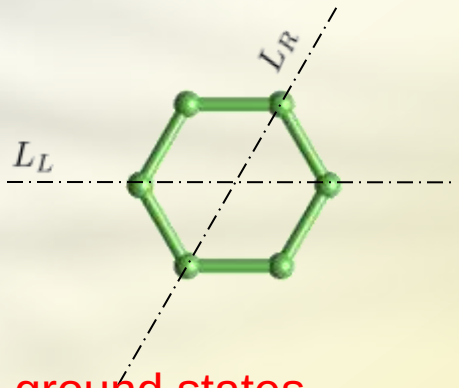
$$\dot{\sigma} = \underbrace{-\frac{i}{\hbar}[H_{\mathrm{sys}}, \sigma]}_{\text{Coherent dynamics}} \underbrace{-\frac{i}{\hbar}[H_{\mathrm{eff}}, \sigma]}_{\text{Effective internal dynamics}} + \underbrace{\mathcal{L}_{\mathrm{tun}}\sigma}_{\text{Tunnelling dynamics}}$$

- The stationary solution is calculated and the current is obtained as the average of the current operator.

The effective Hamiltonian

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

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

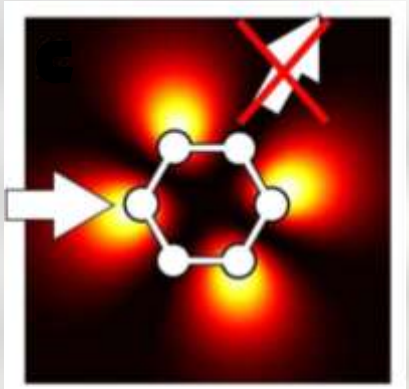
$$\omega_{\alpha\sigma} = \frac{1}{\pi} \sum_{\sigma' \{E\}} \Gamma^0_{\alpha\sigma'} \Big[\langle 7_g \ell\sigma | d_{M\sigma'} | 8\{E\} \rangle\langle 8\{E\} | d^\dagger_{M\sigma'} | 7_g m\sigma \rangle p_\alpha (E - E_{7_g}) + \langle 7_g \ell\sigma | d^\dagger_{M\sigma'} | 6\{E\} \rangle\langle 6\{E\} | d_{M\sigma'} | 7_g m\sigma \rangle p_\alpha (E_{7_g} - E) \Big]$$

Bias and gate dependent

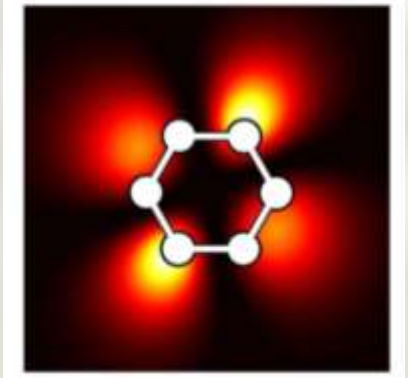
Interference blockade

Geometry

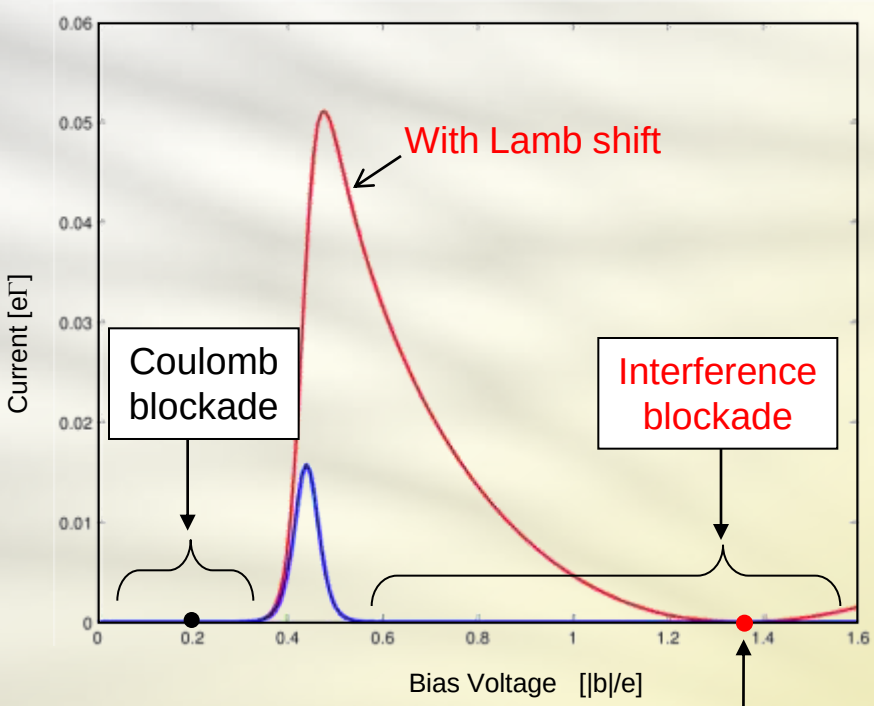
Blocking state



Non-blocking state



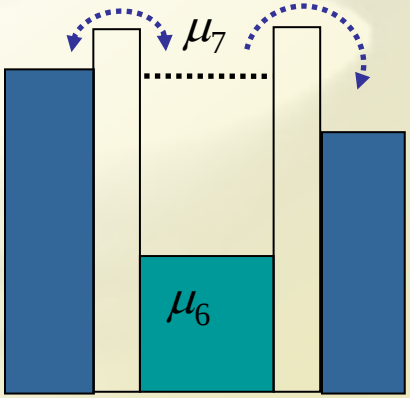
I-V for transition 6 -7



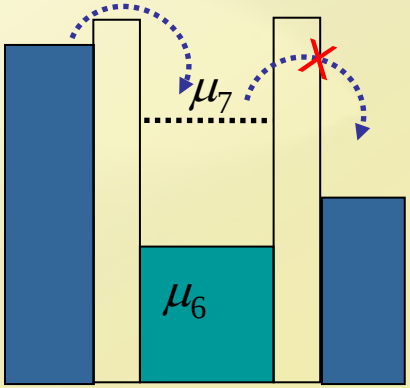
The blocking state is an eigenstate of the effective Hamiltonian

$$\omega_{L\sigma} = 0$$

Energetics

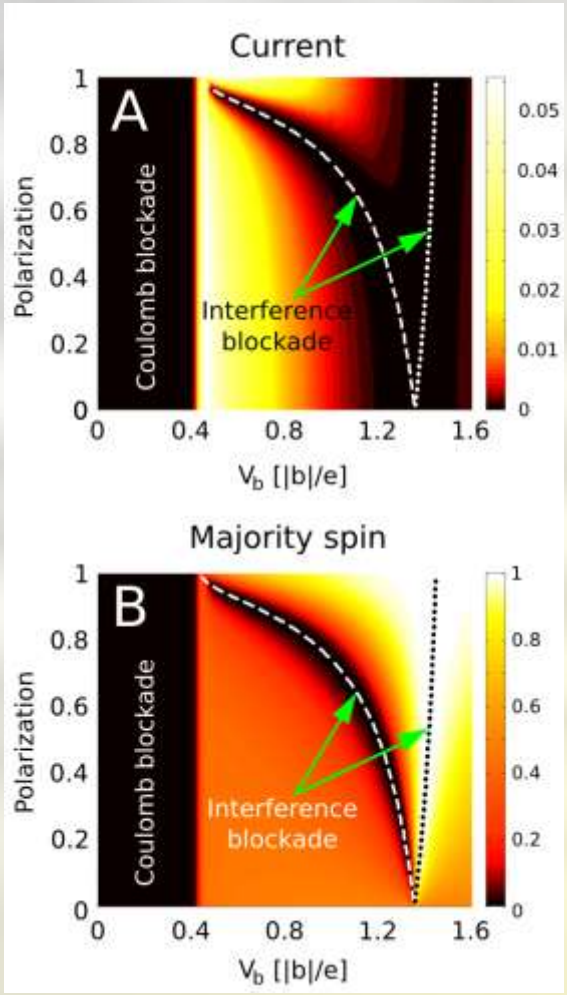


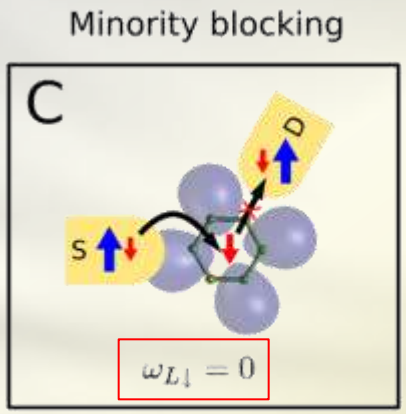
current onset

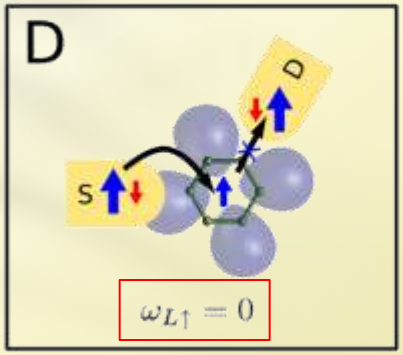


blockade

Selective Interference Blocking







A . Donarini, G. Begemann, and M. Grifoni *Nano Lett.* **9**, 2897 (2009)

Lamb shift due to polarized leads

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

$$\omega_{\alpha\uparrow}-\omega_{\alpha\downarrow}=2\bar{\Gamma}^{0}_{\alpha}P_{\alpha}\frac{1}{\pi}\sum_{\{E\}}$$

$$\Big[\langle 7_g\ell\uparrow|d_{M\uparrow}|8\{E\}\rangle\langle 8\{E\}|d^{\dagger}_{M\uparrow}|7_g m\uparrow\rangle p_{\alpha}(E-E_{7_g})$$

$$+\langle 7_g\ell\uparrow|d^{\dagger}_{M\uparrow}|6\{E\}\rangle\langle 6\{E\}|d_{M\uparrow}|7_g m\uparrow\rangle p_{\alpha}(E_{7_g}-E)$$

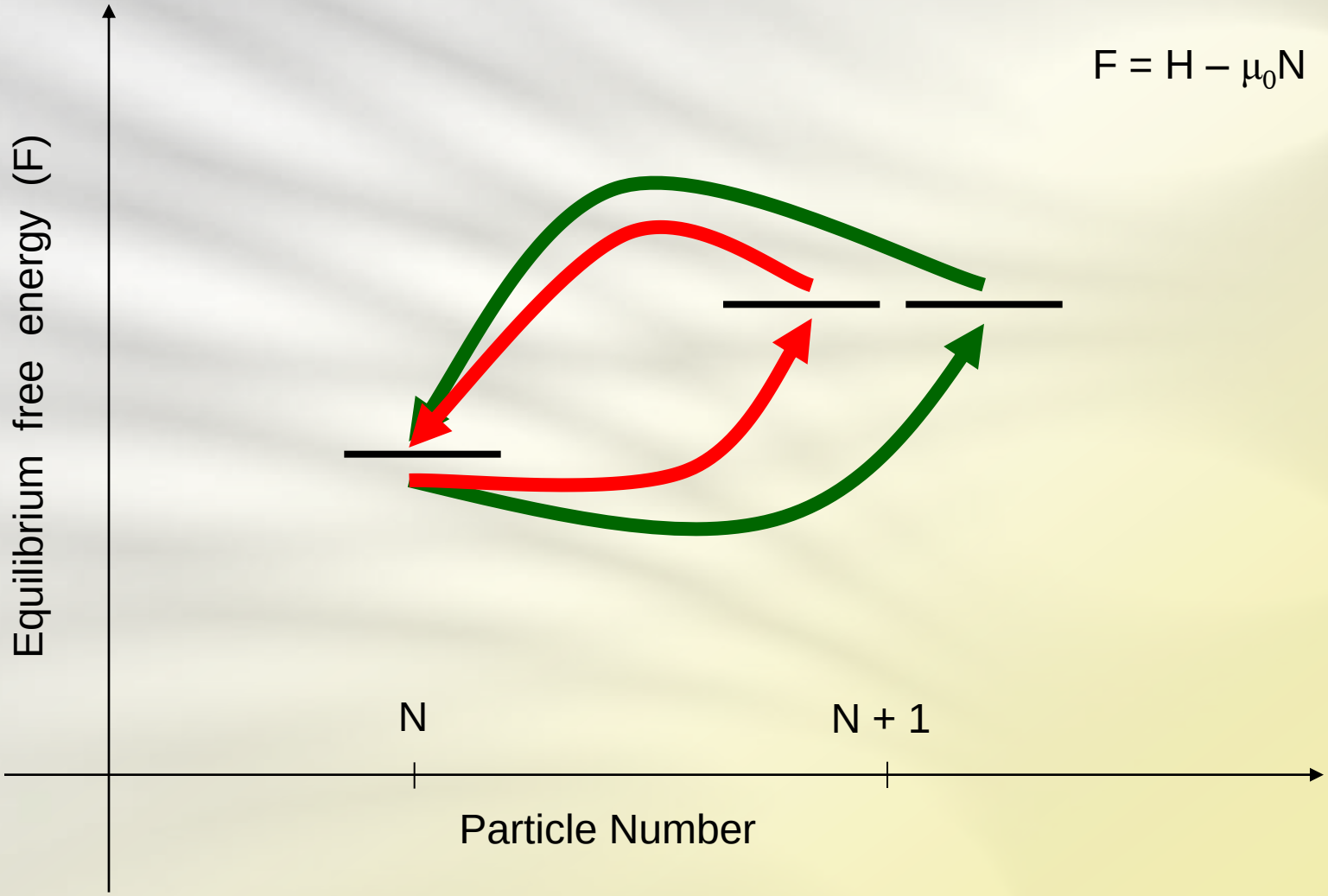
$$-\langle 7_g\ell\uparrow|d_{M\downarrow}|8\{E\}\rangle\langle 8\{E\}|d^{\dagger}_{M\downarrow}|7_g m\uparrow\rangle p_{\alpha}(E-E_{7_g})$$

$$-\langle 7_g\ell\uparrow|d^{\dagger}_{M\downarrow}|6\{E\}\rangle\langle 6\{E\}|d_{M\downarrow}|7_g m\uparrow\rangle p_{\alpha}(E_{7_g}-E)\Big]$$

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

The "two paths" in the ISET

$F = H - \mu_0 N$



Robustness

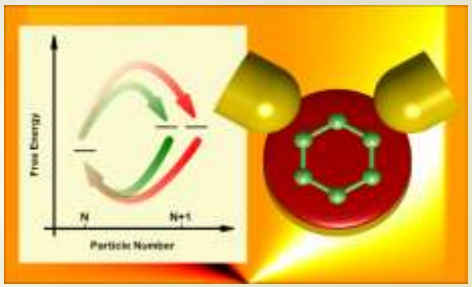
- We have tested the robustness of the effects against:
  - Residual potential drop on the (artificial) molecule (in weak coupling to the leads the potential drop is concentrated at the contacts)
  - On-site energy renormalization of the contact atom due to different anchor groups
  - Lifting of the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

$$\delta E \ll \hbar\Gamma$$

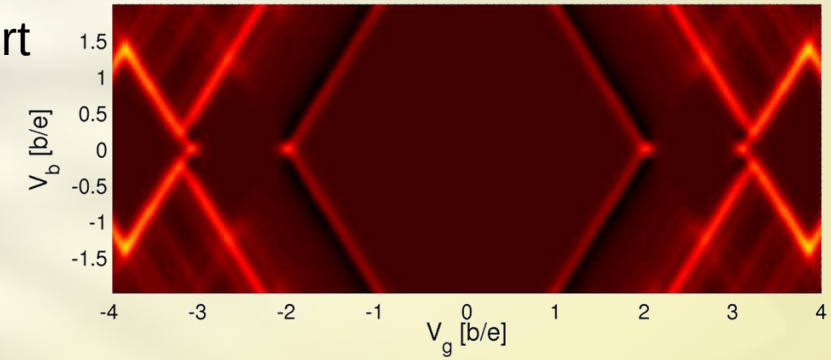
D. Darau, G. Begemann, A. Donarini , and M. Grifoni, *PRB,* **79**, 235404 (2009)

Conclusions

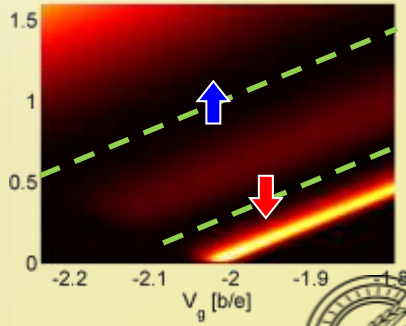
• Interference does occur even in the single-electron tunnelling regime when energetically equivalent paths involving **degenerate states** contribute to the dynamics.



• Interference effects dominates the transport characteristics of ISET. In the non linear regime it produces interference **current blocking**.



• In the presence of ferromagnetic leads, the interplay between interference and exchange on the ISET allows to achieve **all-electrical spin control** of the junction.



Thanks

Georg Begemann

Milena Grifoni

Dana Darau

Deutsche Forschungsgemeinschaft DFG

in the research programs

SPP 1243 Quantum Transport
at the Molecular Scale

SFB 689 Spinphänomene
in reduzierten Dimensionen

Thank you for your attention!