Charge and spin transport through Interference Single Electron Transistors

Andrea Donarini

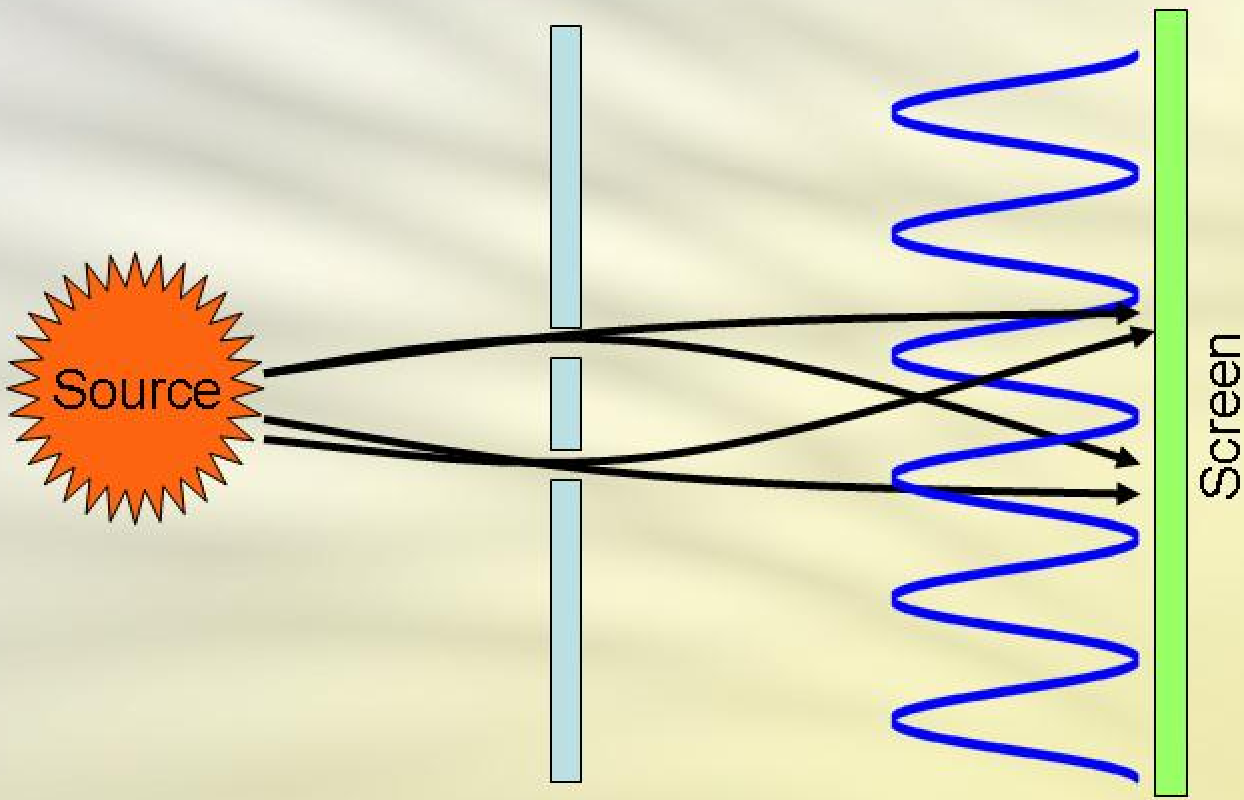
Georg Begemann, Dana Darau, Milena Grifoni

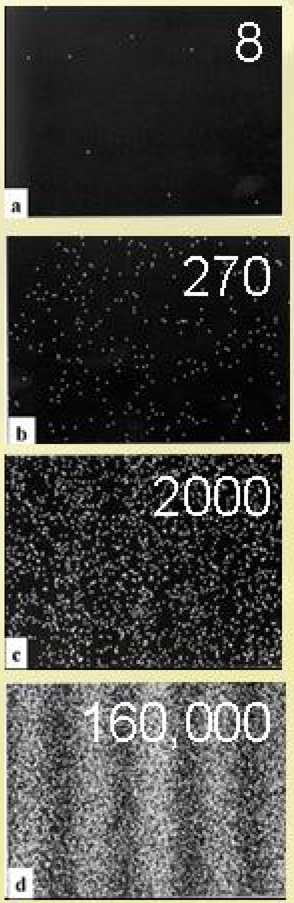
Universität Regensburg

ITP Seminar - Ulm - 27.05.2009

Macroscopic interference

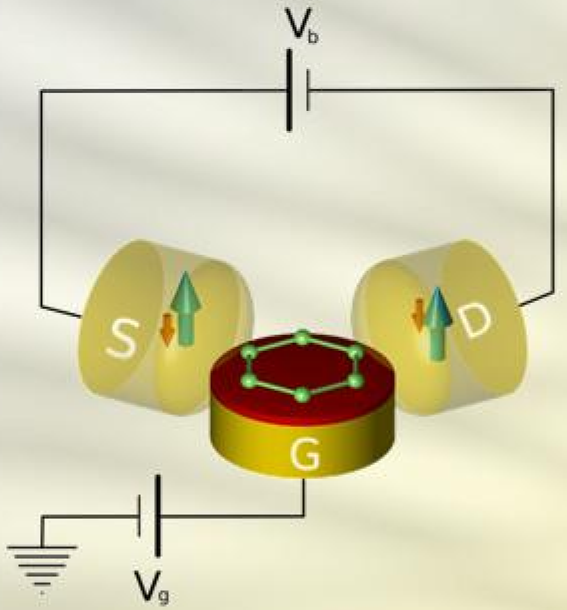
Young's light-interference experiment (1801)

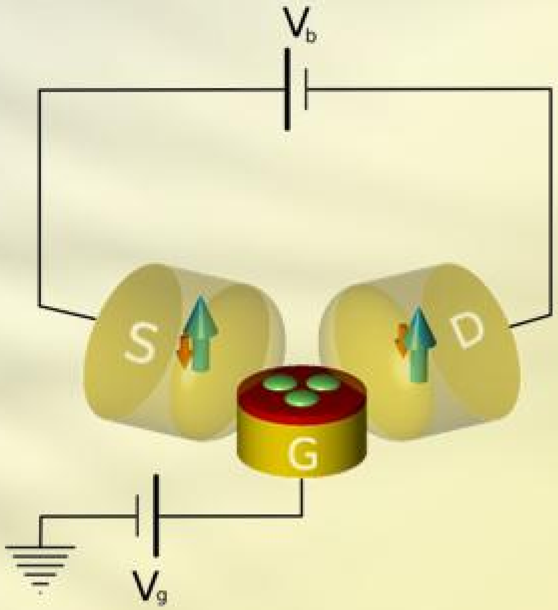




Double-slit experiment with interference
of single electrons (1961)

ISET

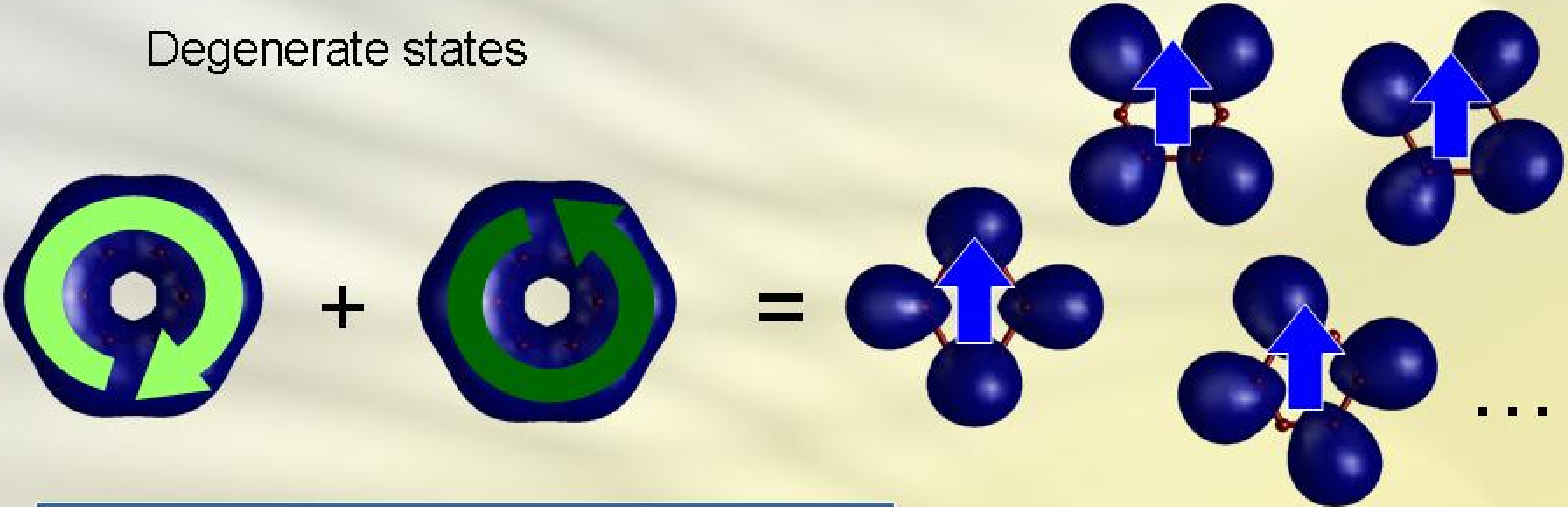


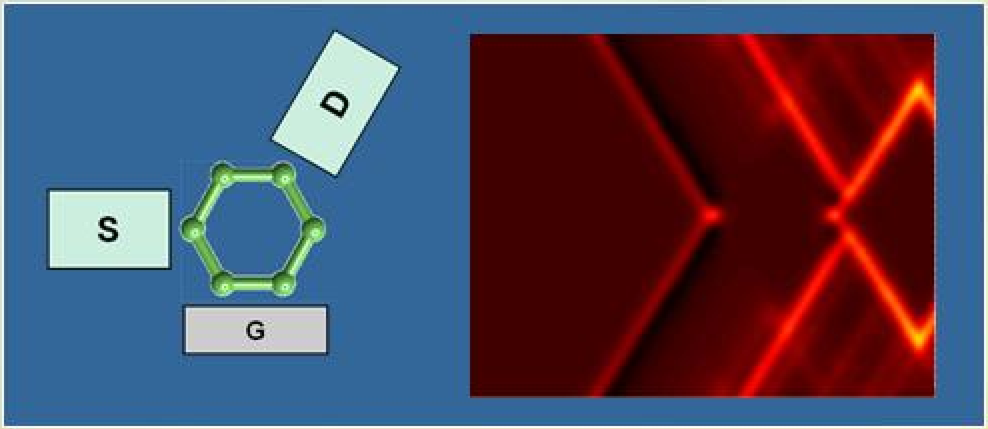


The interference occurs between transmission paths involving
orbitally (quasi-)degenerate states

Intra-molecular interference

Degenerate states





Interference visible in the current as a function of bias and gate voltages.

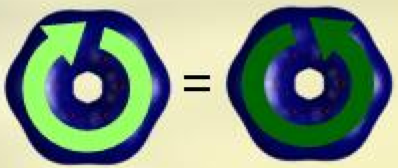
(Benzene) ISET...

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

⇒ Coulomb blockade

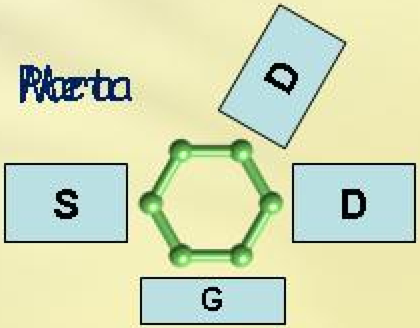
$\hbar\Gamma \ll k_{\mathrm{B}}T \ll \Delta E_{\mathrm{ex}}$

- **Rotational** symmetry ⇒ Orbitally degenerate states



$E_1 = E_2$

- Contact **geometry** ⇒ Contact symmetry breaking



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

... with a magnetic flavour

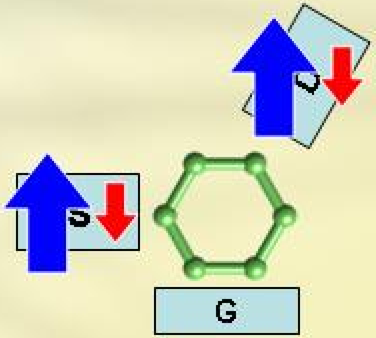
- **Coulomb** interaction
- Molecular **size**

⇒ **Exchange splitting**

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

- Parallel **ferromagnetic** leads

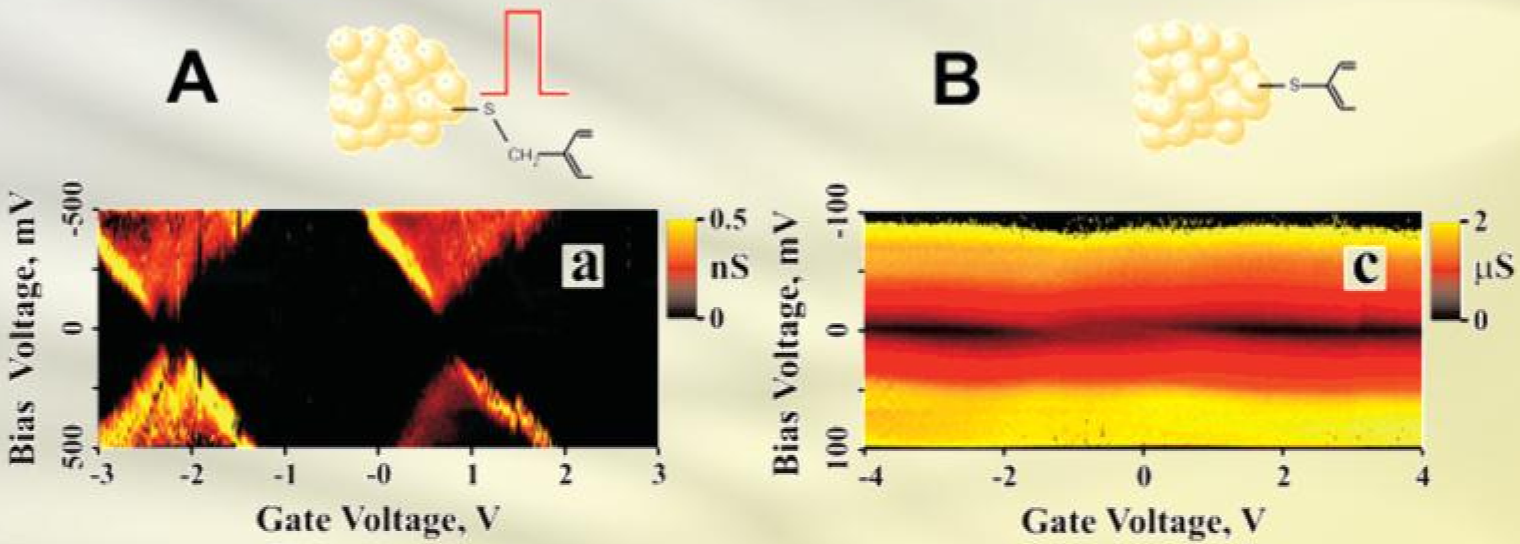
⇒ **Spin symmetry breaking**



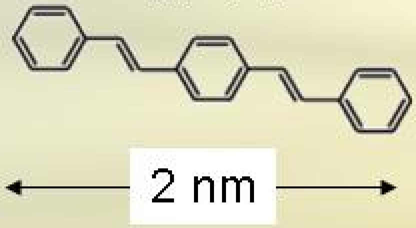
$$\Gamma_{\alpha\uparrow} \neq \Gamma_{\alpha\downarrow}$$

The interplay between orbital and spin degree of freedom allows all-electrical spin control on the molecule.

Coulomb blockade



OPV-3

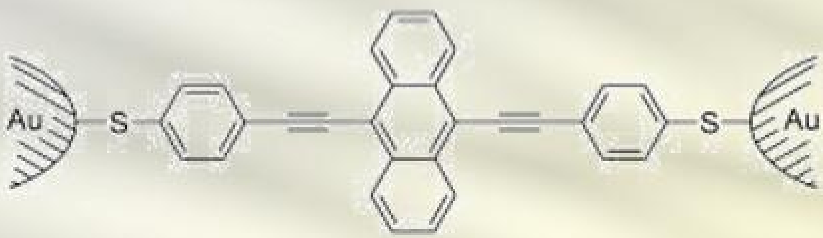


- **Gating** of 2 nm sized molecule
- **Weak coupling** realization with specific anchor groups

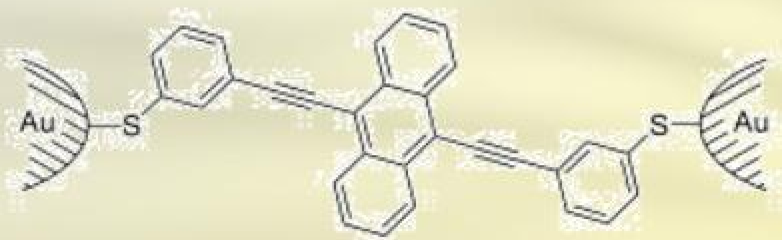
A. Danilov, S. Kubatkin, et al. Nanoletters **8**, 1 (2008)

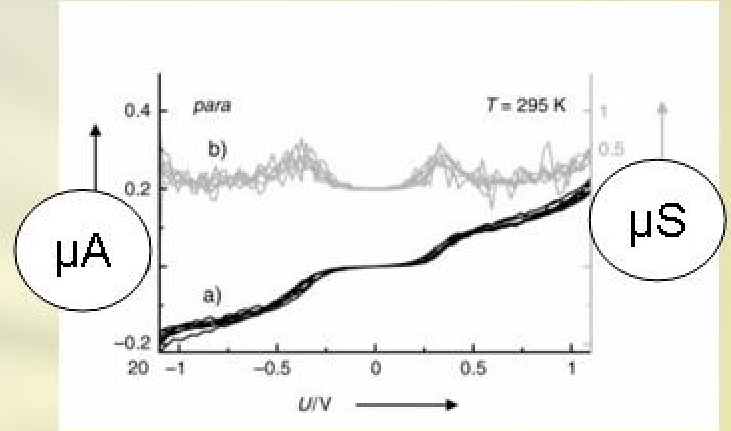
Symmetry breaking contacts

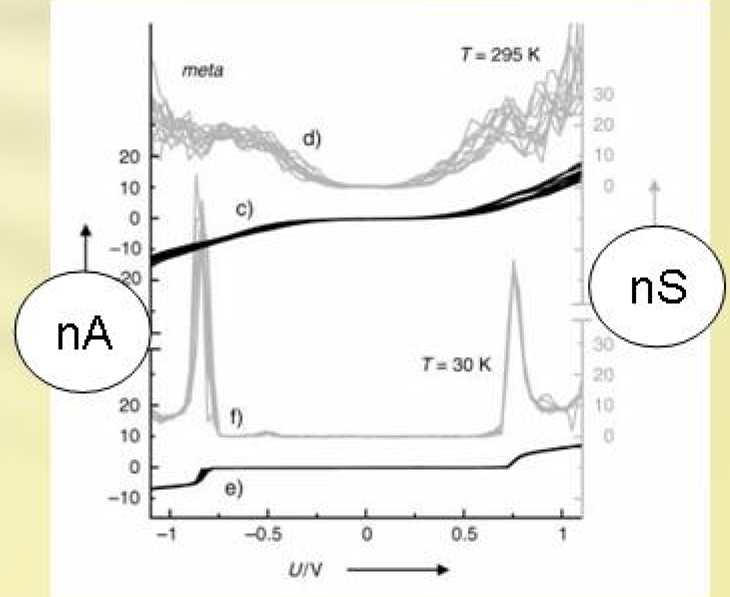
Para configuration



Meta configuration

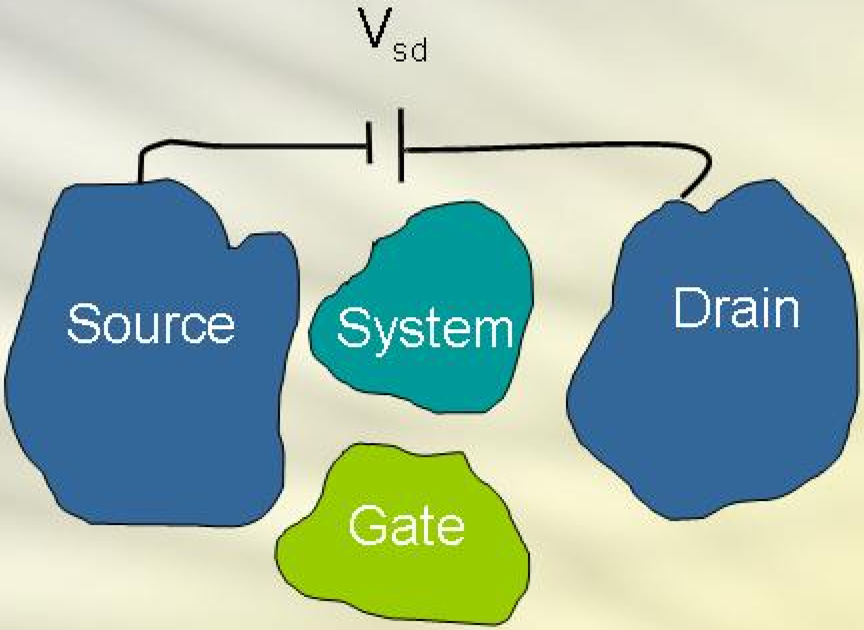






M. Mayor, H. Weber, et al. Angew. Chem. Int. Ed. **42** 5843 (2003)

The Hamiltonian

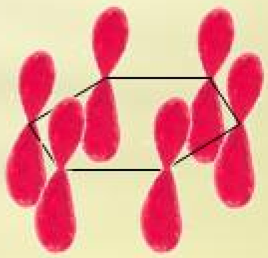


$$H = {\color{red}H_{\text{Sys}}} + H_{\text{leads}} + H_{\text{tun}} \quad \begin{cases} {\color{red}H_{\text{Sys}} = H_{\text{ben}} \;/ H_{\text{TD}}} \\ H_{\text{leads}} = \sum_{\alpha\,k\,\sigma} (\epsilon_k - \mu_\alpha) c^\dagger_{\alpha k \sigma} c_{\alpha k \sigma} \\ H_{\text{tun}} = t \sum_{\alpha k \sigma} \left(d^\dagger_{\alpha\sigma} c_{\alpha k \sigma} + c^\dagger_{\alpha k \sigma} d_{\alpha\sigma} \right) \end{cases}$$

Interacting isolated benzene

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

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



- 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

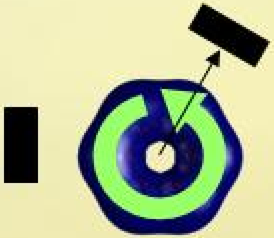
N	Degeneracy	GS energy[eV] (at $\xi = 0$)	GS symmetry representation
0	1	0	A_{1g}
1	2	-22	A_{2u}
2	1	-42.25	A_{1g}
3	4	-57.42	E_{1g}
4	3	-68.875	A_{2g}
5	4	-76.675	E_{1g}
6	1	-81.725	A_{1g}
7	4	-76.675	E_{2u}
8	3	-68.875	A_{2g}
9	4	-57.42	E_{2u}
10	1	-42.25	A_{1g}
11	2	-22	B_{2g}
12	1	0	A_{1g}

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
- $\mathcal{R}_\phi|7_g\,\ell\rangle = e^{-i\ell\phi}|7_g\,\ell\rangle$ $\ell = \pm 2$

$\ell = +2$



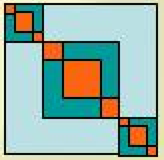
$\exp\left(+i\frac{2\pi}{3}\right)$

$\ell = -2$

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

Generalized Master Equation

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



- We 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:

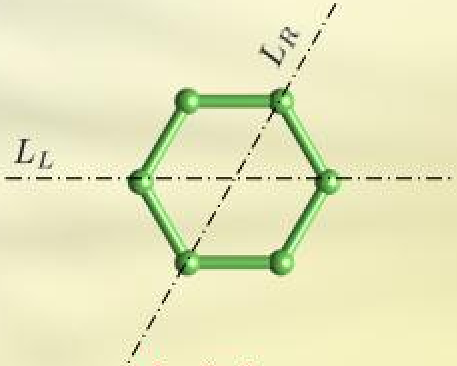
$$\dot{\sigma} = \underbrace{-\frac{i}{\hbar}[H_{\text{sys}}, \sigma]}_{\text{Coherent dynamics}} \underbrace{- \frac{i}{\hbar}[H_{\text{eff}}, \sigma]}_{\text{Effective internal dynamics}} + \underbrace{\mathcal{L}_{\text{tun}}\sigma}_{\text{Tunnelling dynamics}}$$

Coherent dynamics | Effective **internal** dynamics | **Tunnelling** dynamics

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

Current operator

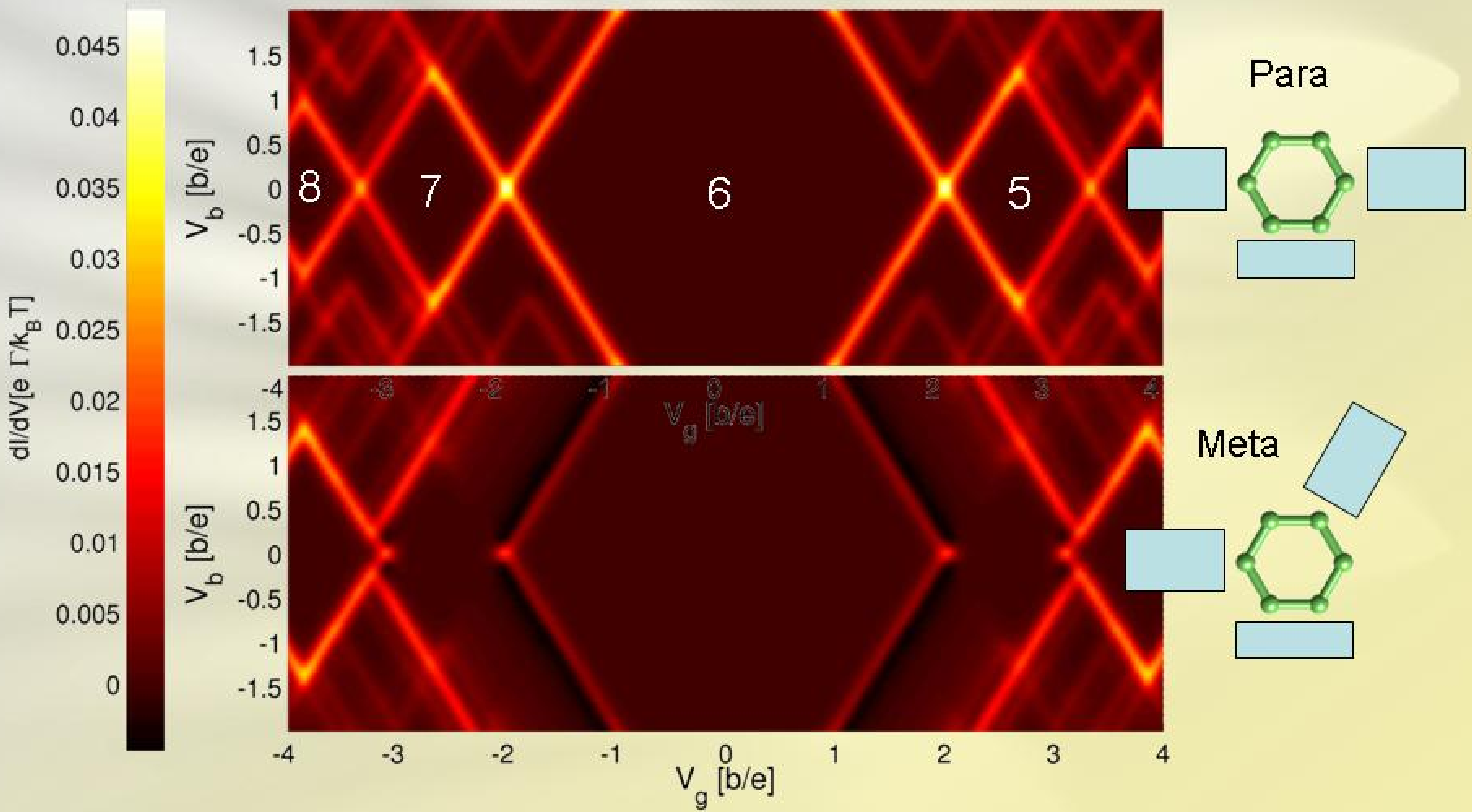
- **Current**: using the GME we find the **operator:**

$$\hat{I}_{\rm L} = \Gamma_{\rm L} \sum_{{\rm NE}\tau} \mathcal{P}_{\rm NE} \Big[d_{{\rm L}\tau} f_{\rm L}^{+}(H_{\rm ben}^{0} - E) d_{{\rm L}\tau}^{\dagger} - d_{{\rm L}\tau}^{\dagger} f_{\rm L}^{-}(E - H_{\rm ben}^{0}) d_{{\rm L}\tau} \Big] \mathcal{P}_{\rm NE'}$$

and thus calculate the **stationary current:**

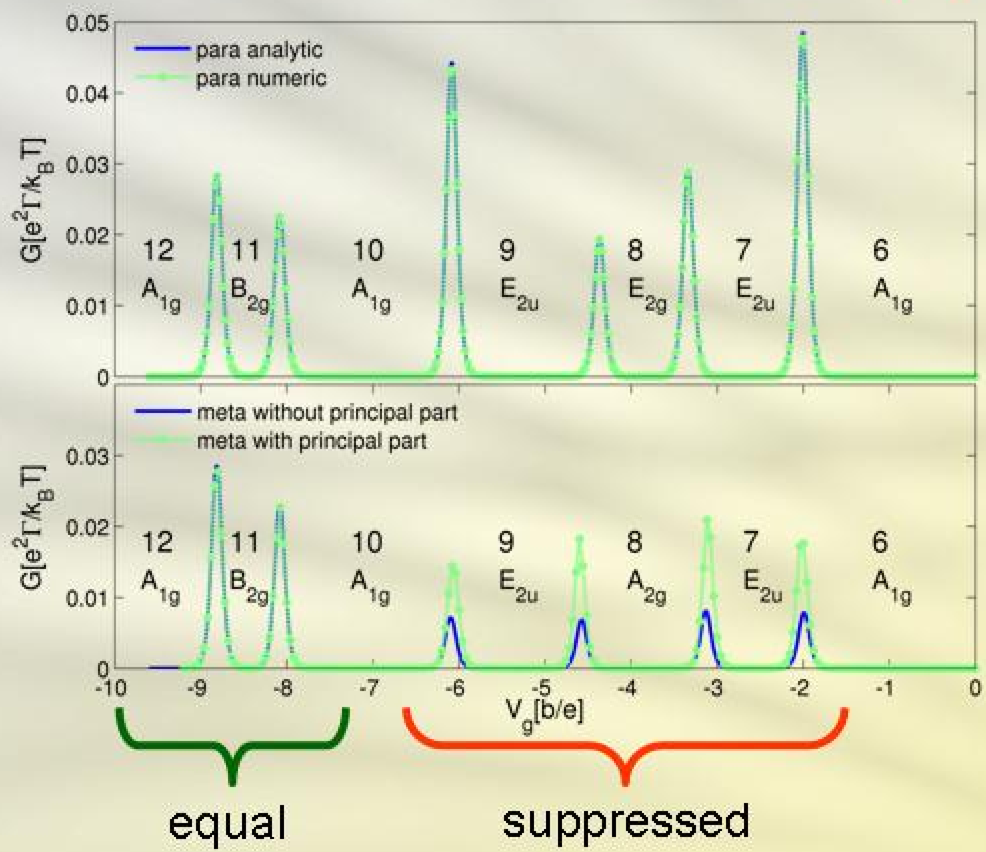
$$I_{\rm L} = {\rm Tr}\{\sigma_{\rm stat} \hat{I}_{\rm L}\} = -I_{\rm R}$$

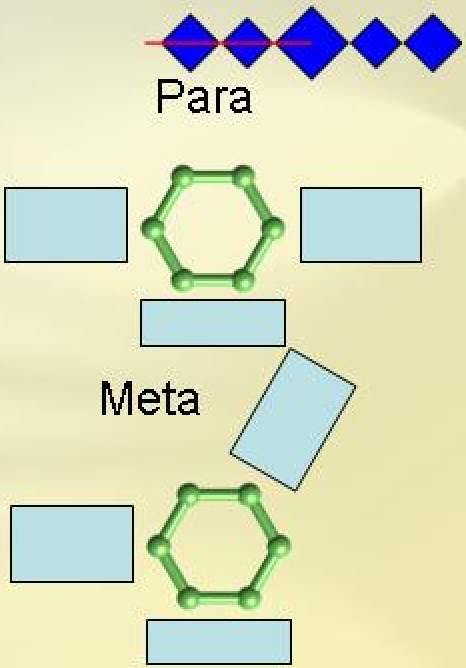
Para vs. Meta



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

Conductance suppression





A: non- degenerate ⟷ **B**: non- degenerate ⇨ Equal

A: non- degenerate ⟷ **E**: degenerate ⇨ Suppressed

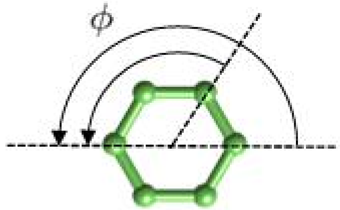
Destructive interference

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

Interference factor

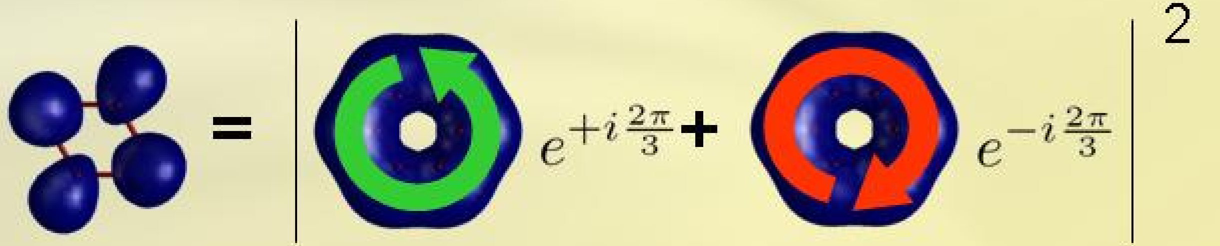
$$\Lambda = \left|\sum_{nm\tau} |\langle N,\, n| d_{\mathrm{L}\tau} |N+1,\, m\rangle|^2 e^{i\phi_{nm}}\right|^2$$

$$d^\dagger_{\mathrm{R}\tau} = \mathcal{R}^\dagger_\phi d^\dagger_{\mathrm{L}\tau} \mathcal{R}_\phi$$

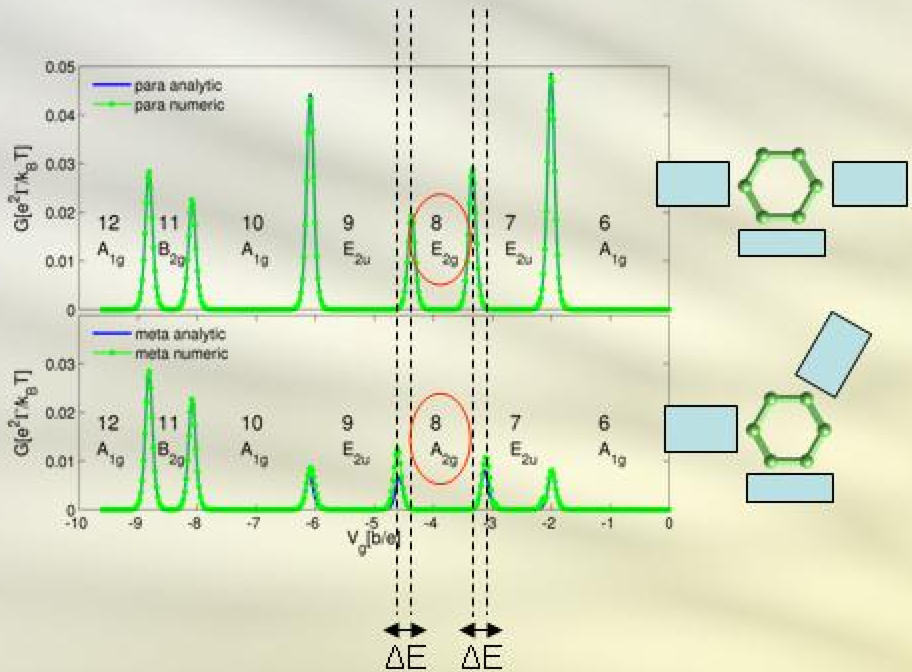


In particular for the transition **6 -7** in the **meta** configuration:

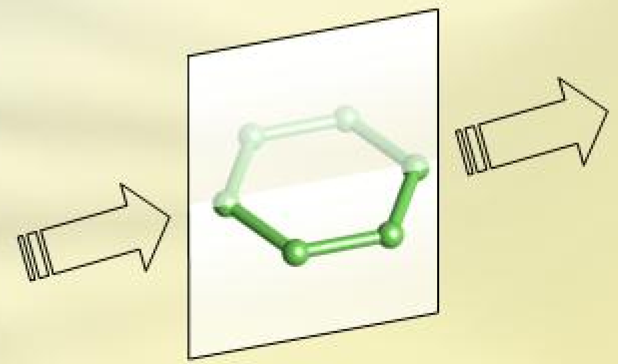
$$\Lambda = \left| |\langle 6_g | d_{\mathrm{L}\tau} | 7_g, +2, \tau\rangle|^2 e^{+i\frac{2\pi}{3}} + |\langle 6_g | d_{\mathrm{L}\tau} | 7_g, -2, \tau\rangle|^2 e^{-i\frac{2\pi}{3}} \right|^2$$



The 8 electrons "anomaly"

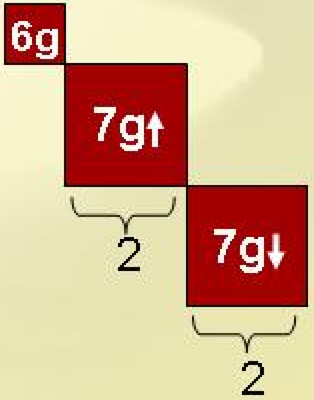


Mirror symmetry of the para-configuration



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



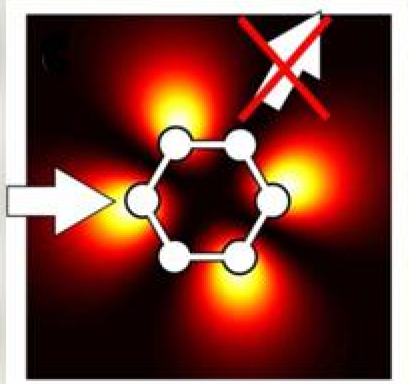
- The 7 particle ground state has spin and orbital **degeneracies**;
- **Physical basis**: the basis that diagonalizes the stationary density matrix;
- The physical basis **depends on the bias:** in whatever reference basis, **coherences** are essential for a correct description of the system;
- The **visualization tool**: **position resolved** transition probability to the physical basis:

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

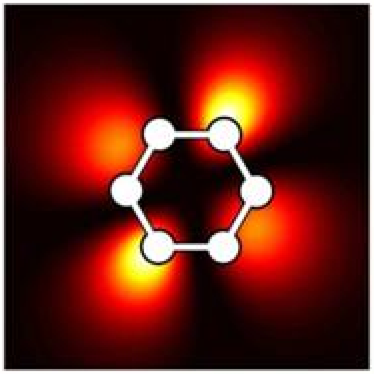
Interference blockade

Geometry

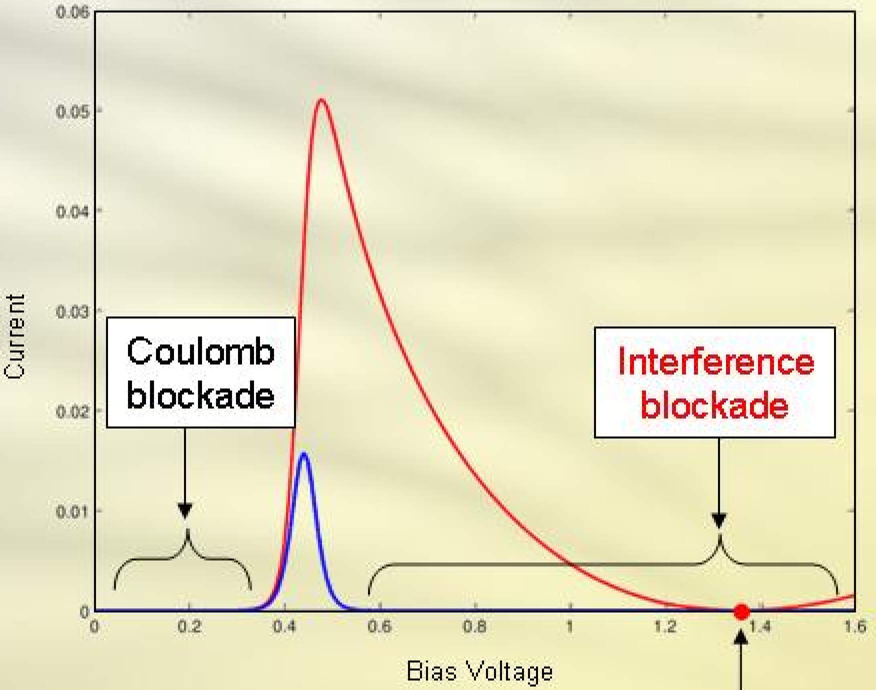
Blocking state



Non-blocking state



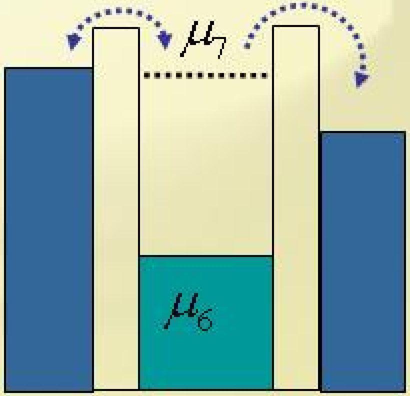
I-V for transition 6 -7



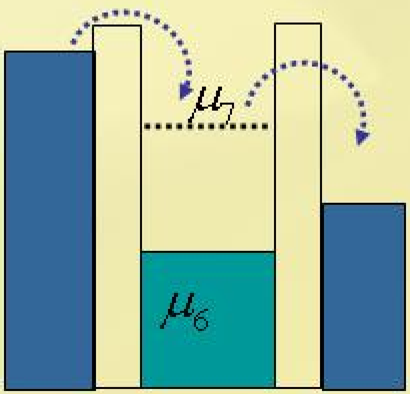
The blocking state is an eigenstate of the effective Hamiltonian

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

Energetics

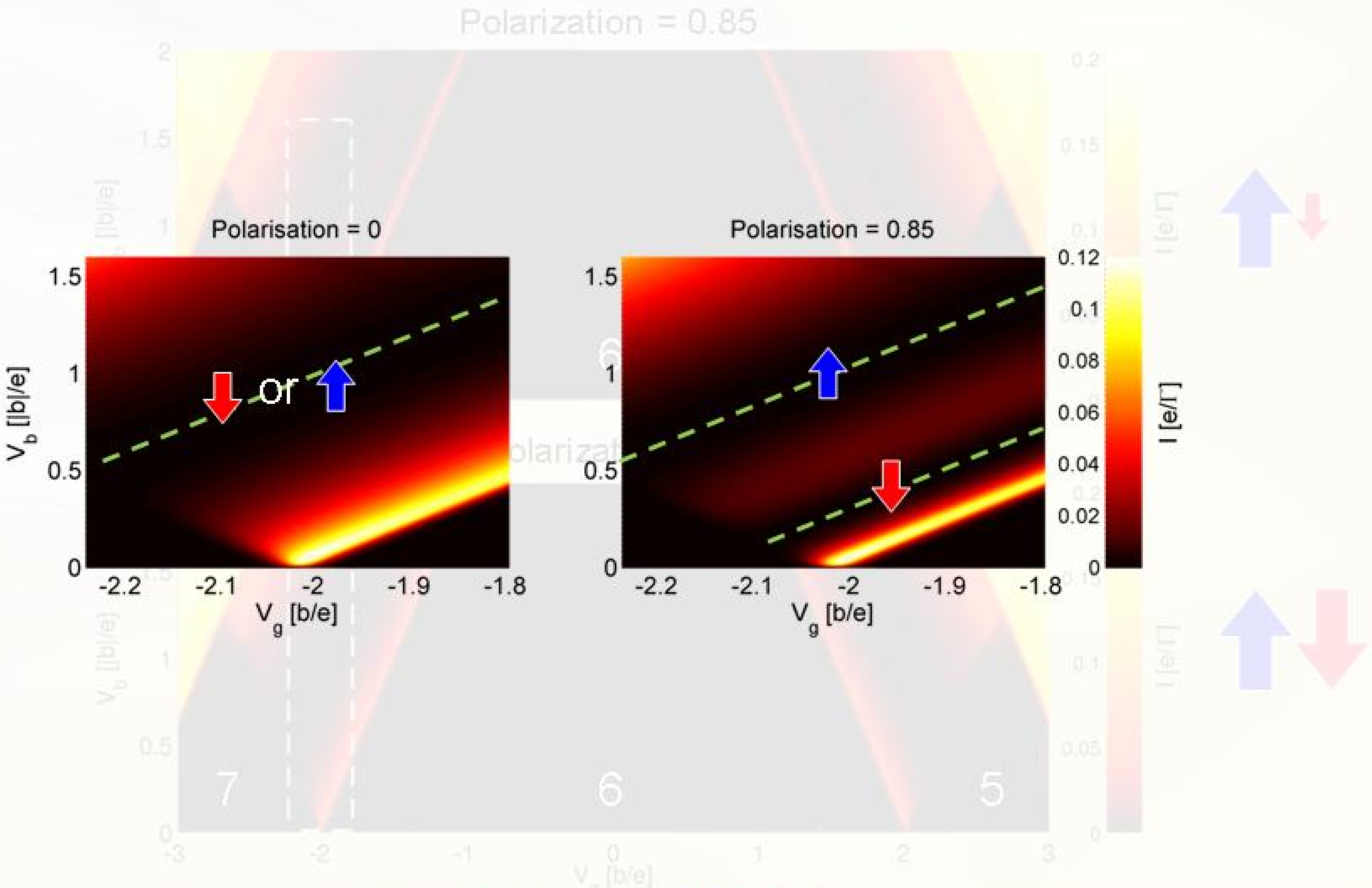


current onset

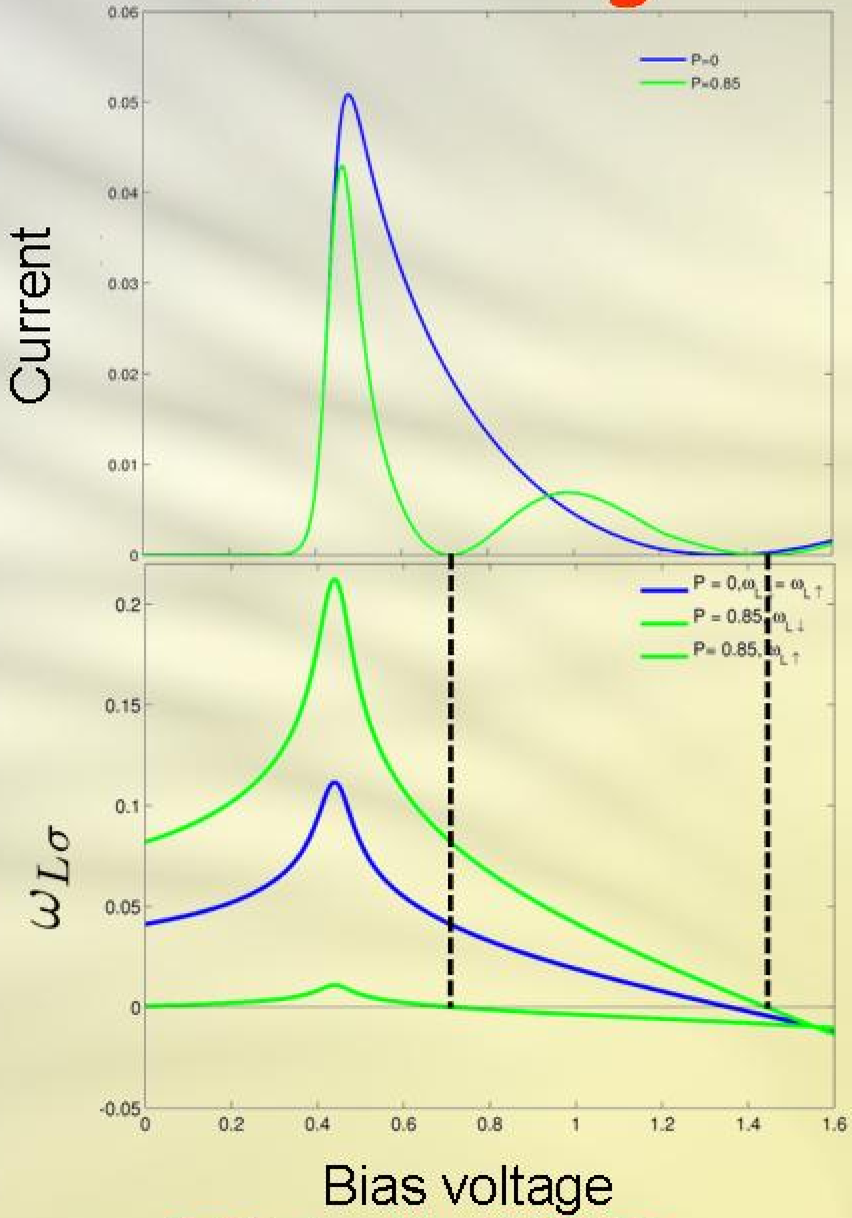


blockade

Normal vs. ferromagnetic leads



Normal vs ferromagnetic leads



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**:

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

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

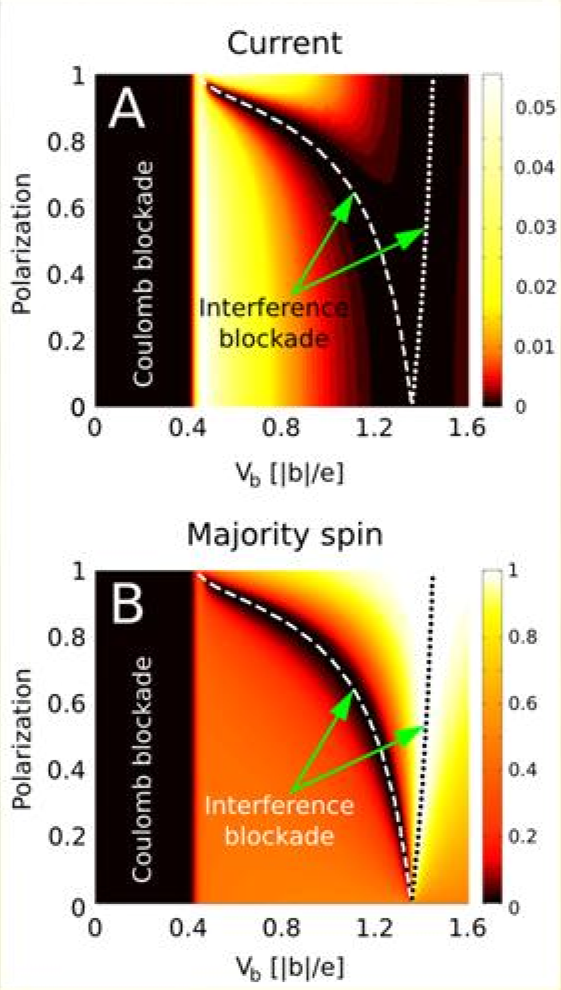
$$+\langle 7_g\ell\uparrow|d^\dagger_{M\uparrow}|6\{E\}\rangle\langle 6\{E\}|d_{M\uparrow}|7_g m\uparrow\rangle p_\alpha(E_{7_g}-E)$$

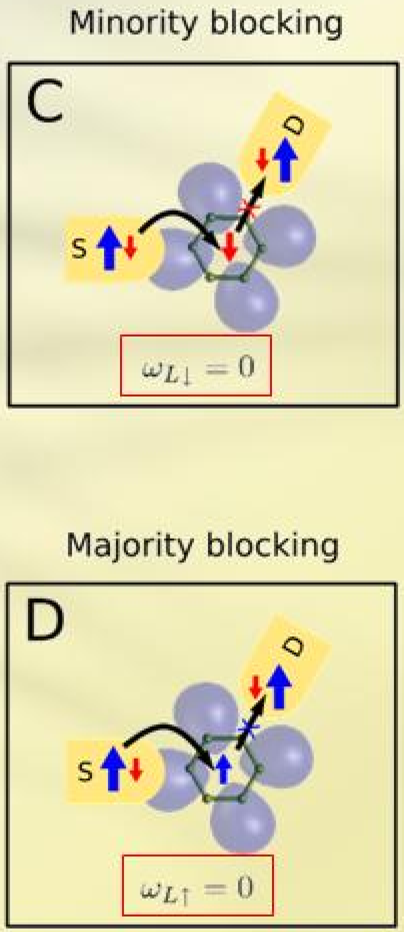
$$-\langle 7_g\ell\uparrow|d_{M\downarrow}|8\{E\}\rangle\langle 8\{E\}|d^\dagger_{M\downarrow}|7_g m\uparrow\rangle p_\alpha(E-E_{7_g})$$

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

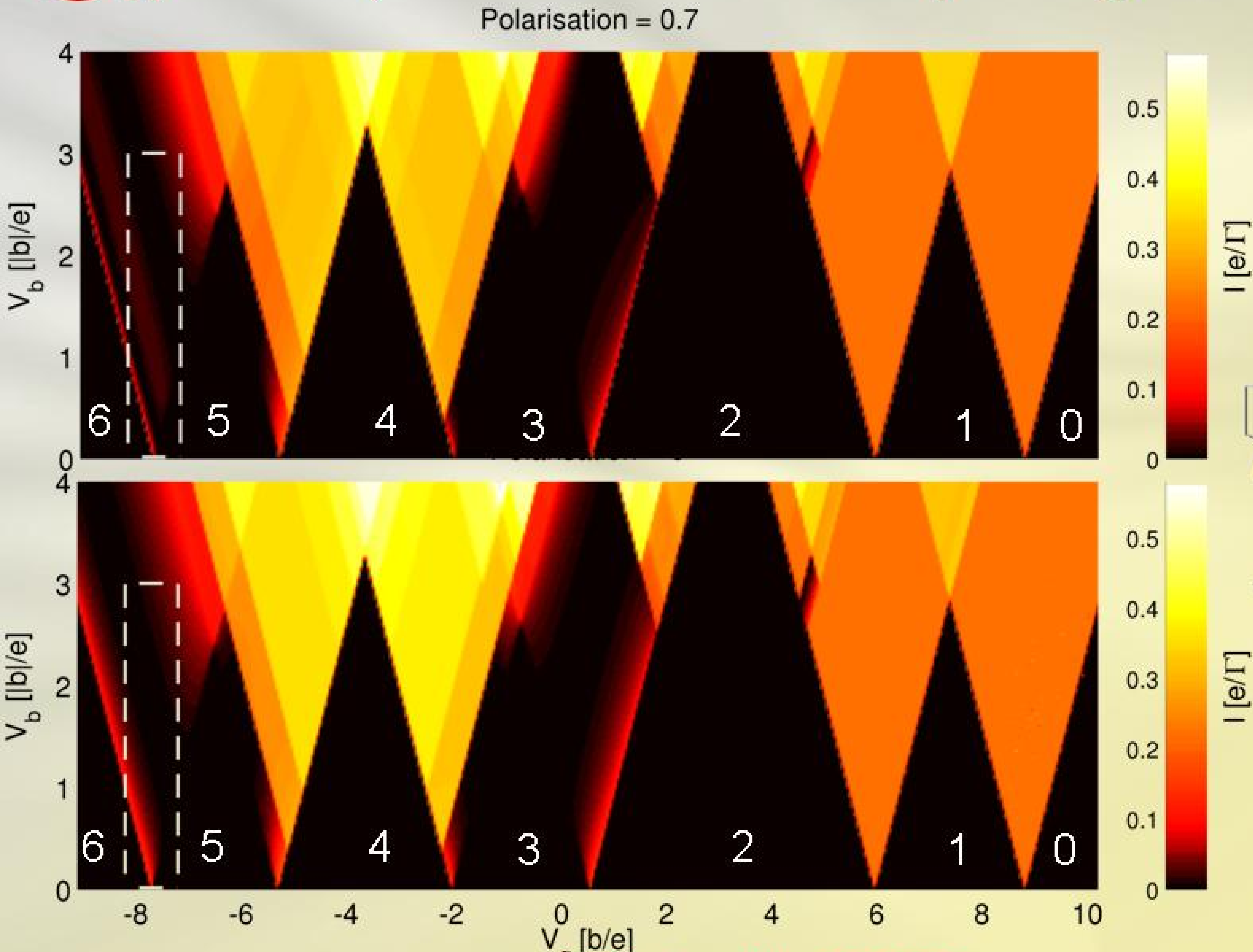
The splitting of the level renormalization depends crucially on the Coulomb interaction on the molecule and **vanishes in absence of exchange**.

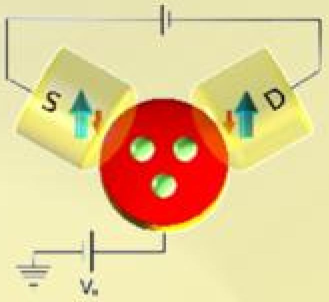
Selective Interference Blocking





Triple dot: Stability diagram





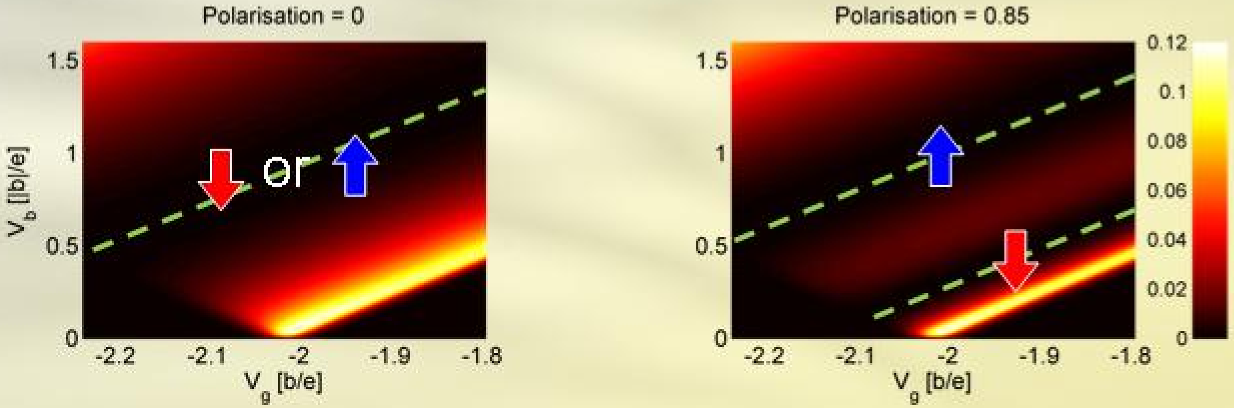
Robustness

- 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:

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

Conclusions

- The interplay between electron-electron **interaction** and orbital **symmetry** is important to understand transport through a benzene ISET;
- **Distructive 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 molecule.



- **Coherences** between degenerate states are essential to capture the interference effects in a benzene ISET.
- Interference is **robust** against symmetry breaking.

Thanks

Georg Begemann

Milena Grifoni

Dana Darau

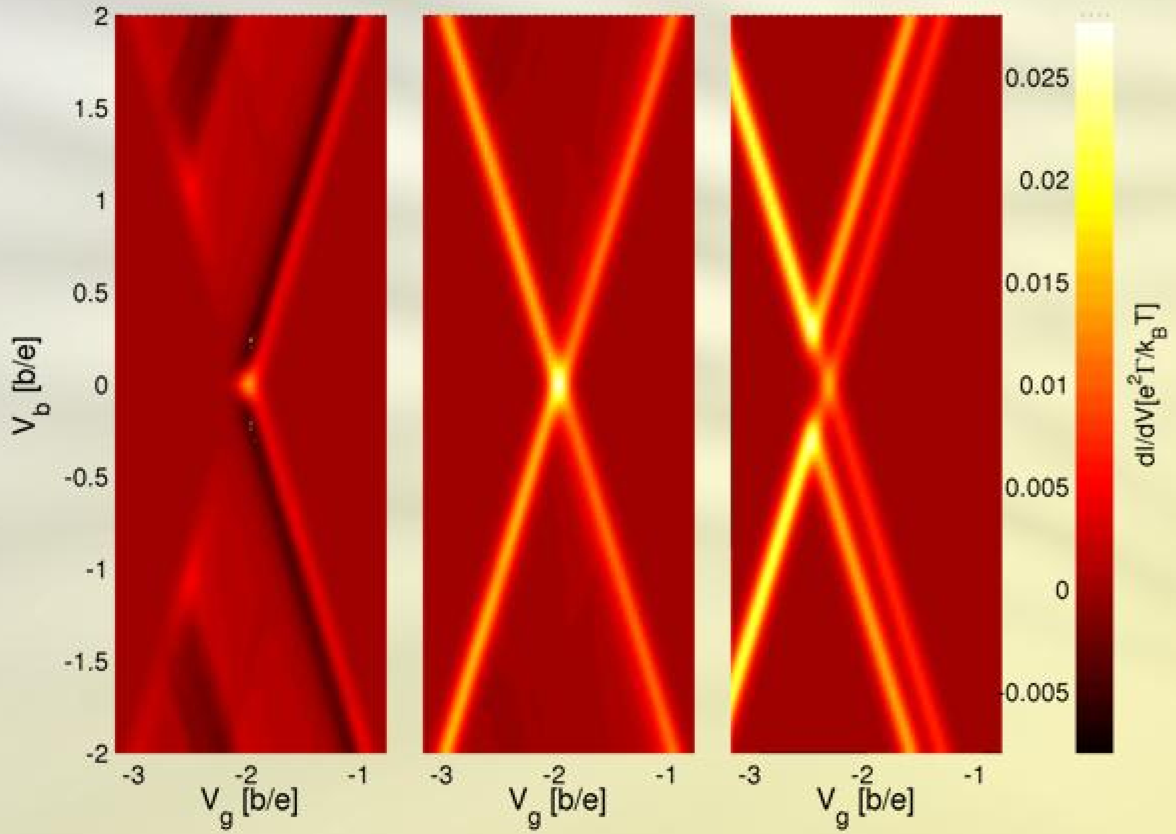
...and you for your attention!

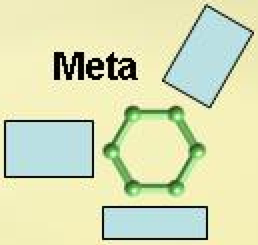
Phys. Rev. B 77, 201406(R) (2008)

arXiv:0904.0167 (Nano Lett. in press)
arXiv:0810.2461 (PRB in press)

Contact effect

$$H'_{\mathrm{ben}} := H_{\mathrm{contact}} = \xi_{\mathrm{c}} \sum_{\alpha\sigma} d^{\dagger}_{\alpha\sigma} d_{\alpha\sigma}, \quad \alpha = L, R$$

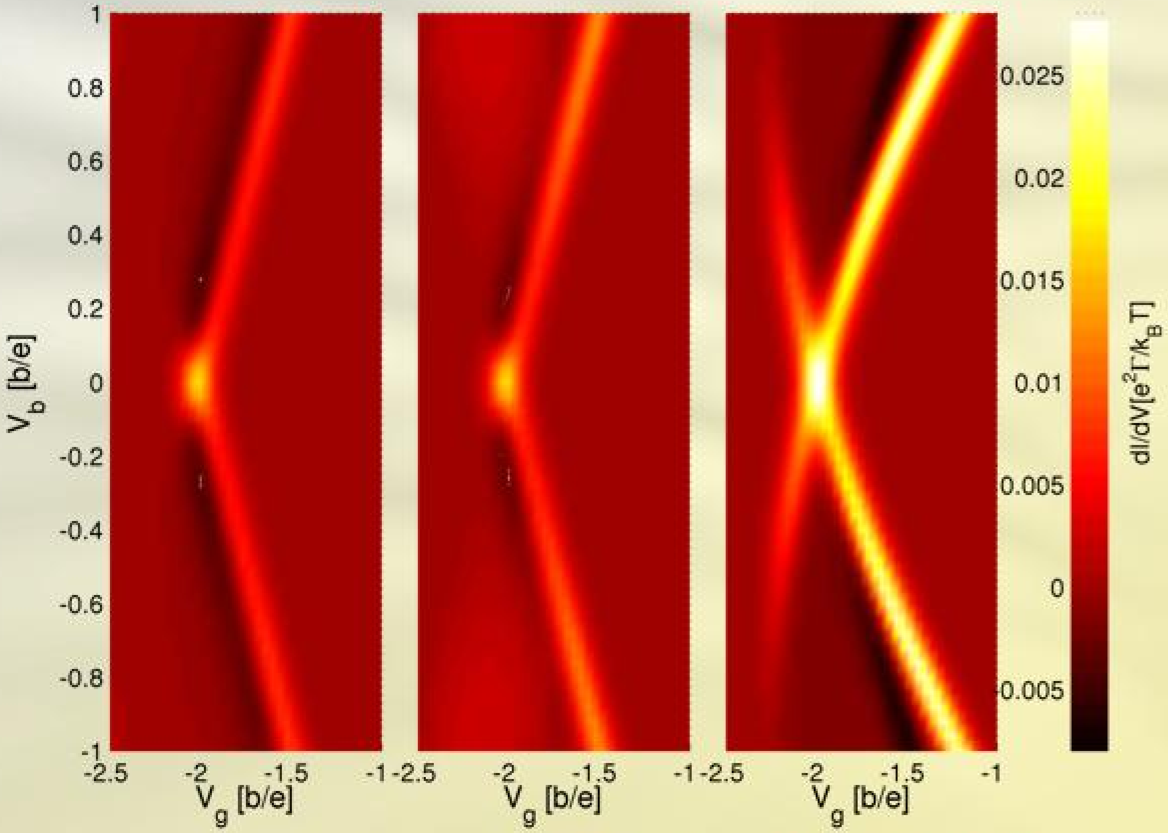


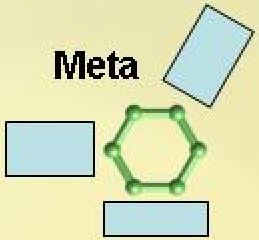


Bias effect

$$H'_{\mathrm{ben}} := H_{\mathrm{bias}} = e\sum_{i\sigma}\xi_{\mathrm{b}_i} d^{\dagger}_{i\sigma} d_{i\sigma}$$

$$\xi_{\mathrm{b}_i} = \int d\mathbf{r} p_{z_i}(\mathbf{r}) V_{\mathrm{b}}(\hat{\mathbf{r}}) p_{z_i}(\mathbf{r}).$$





Bias + Contact