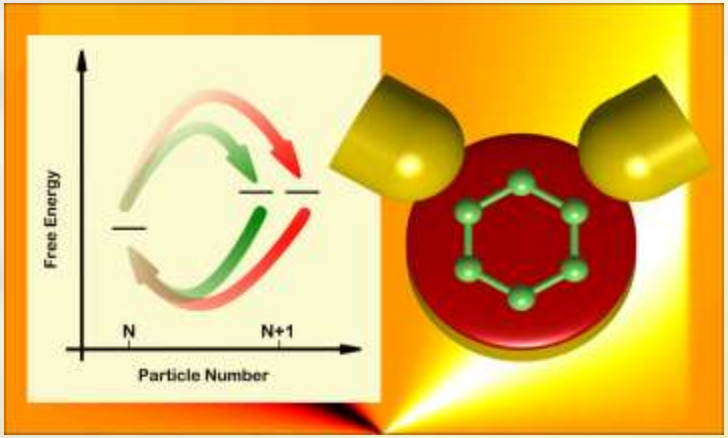
Interference and interaction in symmetric nanojunctions

Andrea Donarini



Regensburg - 31.01.2011

physikalisches Kolloquium

Double slit experiment: (London, 1801)

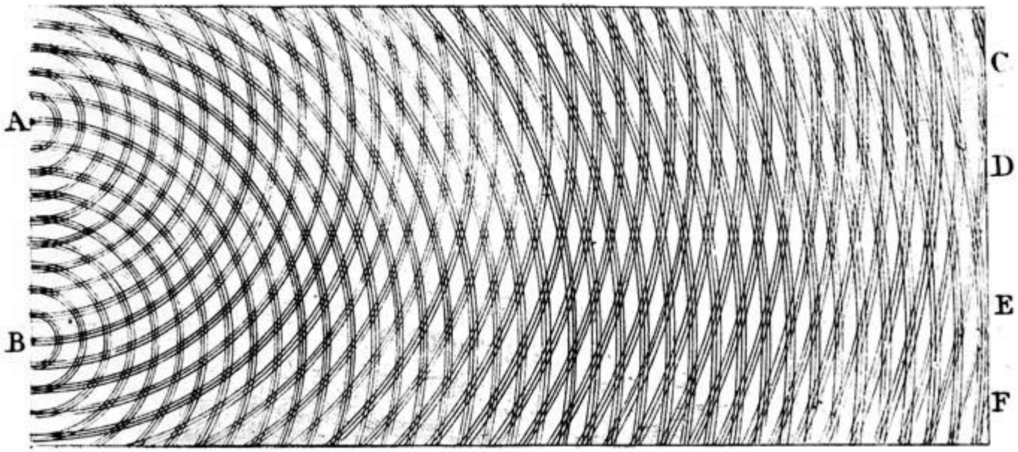
PHILOSOPHICAL

TRANSACTIONS.

I. *The* Bakerian *Lecture. Experiments and Calculations relative to physical Optics. By* Thomas Young, *M. D. F. R. S.*

Read November 24, 1803.

I. EXPERIMENTAL DEMONSTRATION OF THE GENERAL LAW OF THE INTERFERENCE OF LIGHT.



Phil. Trans. R. Soc. Lon., **94,** 12 (1804)

Double slit with electrons: (Tübingen, 1961)

Aus dem Institut für Angewandte Physik der Universität Tübingen

Elektroneninterferenzen an mehreren künstlich hergestellten Feinspalten

Von

Claus Jönsson

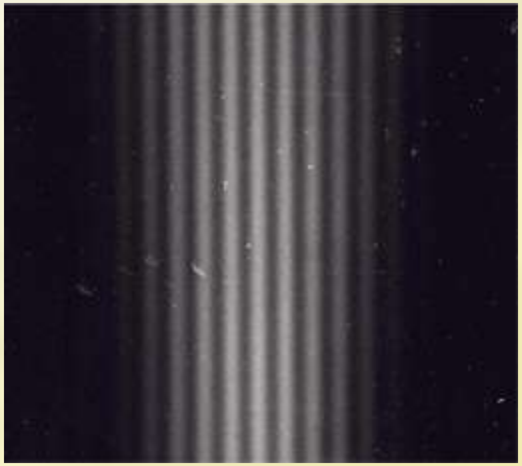
Mit 14 Figuren im Text

(Eingegangen am 17. Oktober 1960)

A glass plate covered with an evaporated silver film of about 200 Å thickness is irradiated by a line-shaped electron-probe in a vacuum of 10^{-4} Torr. A hydrocarbon polymerisation film of very low electrical conductivity is formed at places subjected to high electron current density. An electrolytically deposited copper film leaves these places free from copper. When the copper film is stripped a grating with slits free of any material is obtained. 50 μ long and 0·3 μ wide slits with a grating constant of 1 μ are obtained. The maximum number of slits is five.
The electron diffraction pattern obtained using these slits in an arrangement analogous to Young's light optical interference experiment in the Fraunhofer plane and Fresnel region shows an effect corresponding to the well-known interference phenomena in light optics.

Zeitschrift für Physik, **161**, 454 (1961)

C.Jönsson



Single electron interference (Bologna, 1974)

On the statistical aspect of electron interference phenomena

P. G. Merli
CNR–LAMEL, Bologna, Italy

G. F. Missiroli and G. Pozzi
CNR–GNSM, Istituto di Fisica, Laboratorio Microscopia Elettronica, Bologna, Italy
(Received 29 May 1974; revised 17 October 1974)

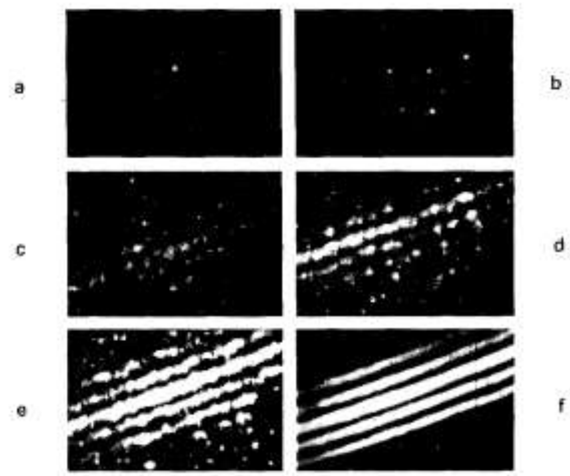


Fig. 1. (a–f) Electron interference fringe patterns filmed from a TV monitor at increasing current densities.

Am. J. Phys., **44**, 306 (1976)

Single electron interference (Tokyo, 1987)

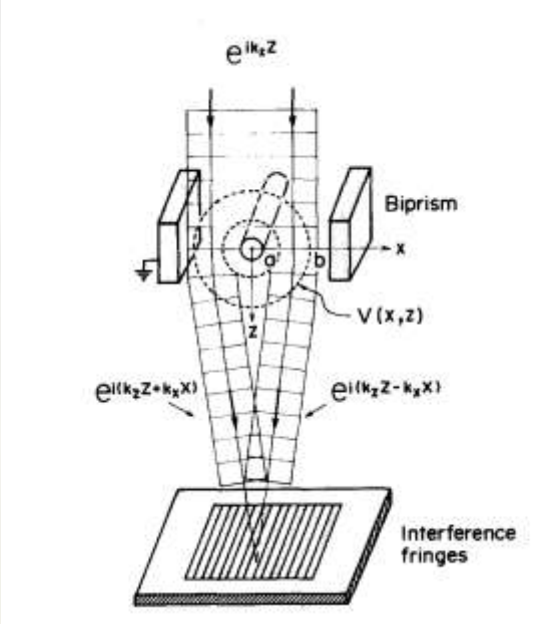
Demonstration of single-electron buildup of an interference pattern

A. Tonomura, J. Endo, T. Matsuda, and T. Kawasaki
Advanced Research Laboratory, Hitachi, Ltd., Kokubunji, Tokyo 185, Japan
H. Ezawa
Department of Physics, Gakushuin University, Mejiro, Tokyo 171, Japan

(Received 17 December 1987; accepted for publication 22 March 1988)

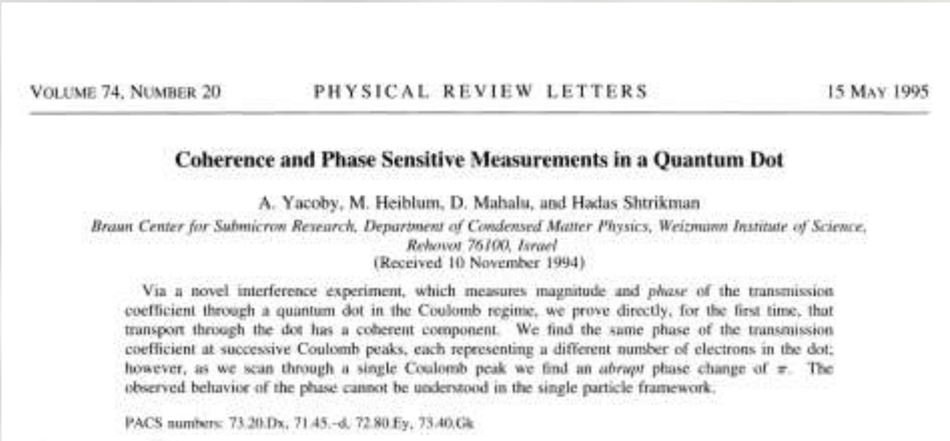
The wave–particle duality of electrons was demonstrated in a kind of two-slit interference experiment using an electron microscope equipped with an electron biprism and a position-sensitive electron-counting system. Such an experiment has been regarded as a pure thought experiment that can never be realized. This article reports an experiment that successfully recorded the actual buildup process of the interference pattern with a series of incoming single electrons in the form of a movie.

A. Tonomura



Am. J. Phys., **57,** 117 (1989)

In mesoscopic rings (Rehovot, 1995)



VOLUME 74, NUMBER 20 PHYSICAL REVIEW LETTERS 15 MAY 1995

Coherence and Phase Sensitive Measurements in a Quantum Dot

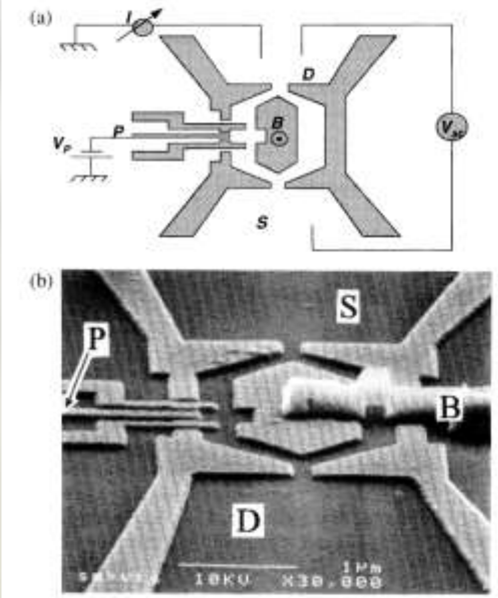
A. Yacoby, M. Heiblum, D. Mahalu, and Hadas Shtrikman

Braun Center for Submicron Research, Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot 76100, Israel

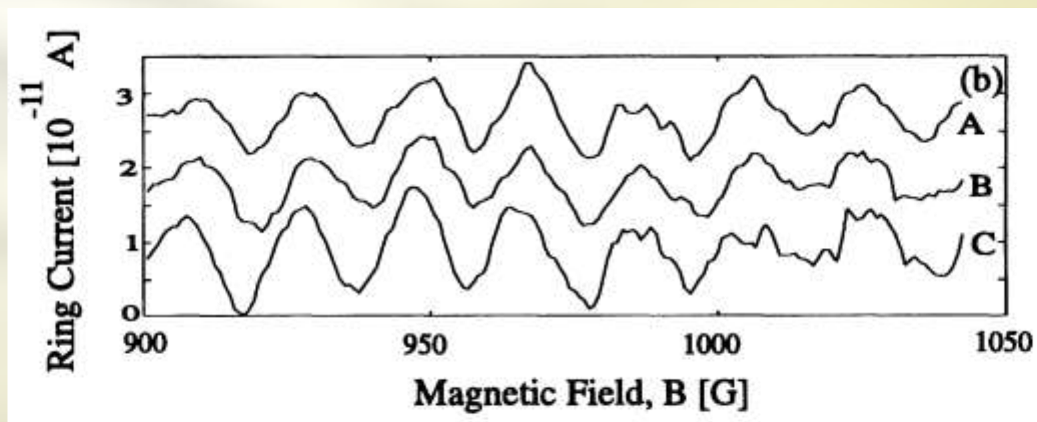
(Received 10 November 1994)

Via a novel interference experiment, which measures magnitude and *phase* of the transmission coefficient through a quantum dot in the Coulomb regime, we prove directly, for the first time, that transport through the dot has a coherent component. We find the same phase of the transmission coefficient at successive Coulomb peaks, each representing a different number of electrons in the dot; however, as we scan through a single Coulomb peak we find an *abrupt* phase change of π. The observed behavior of the phase cannot be understood in the single particle framework.

PACS numbers: 73.20.Dx, 71.45.-d, 72.80.Ey, 73.40.Gk



M. Heiblum



Phys. Rev. Lett., **74**, 4047 (1995)

…counting single electrons (Zürich, 2008)

NANO LETTERS
2008
Vol. 8, No. 8
2547-2550

Time-Resolved Detection of Single-Electron Interference

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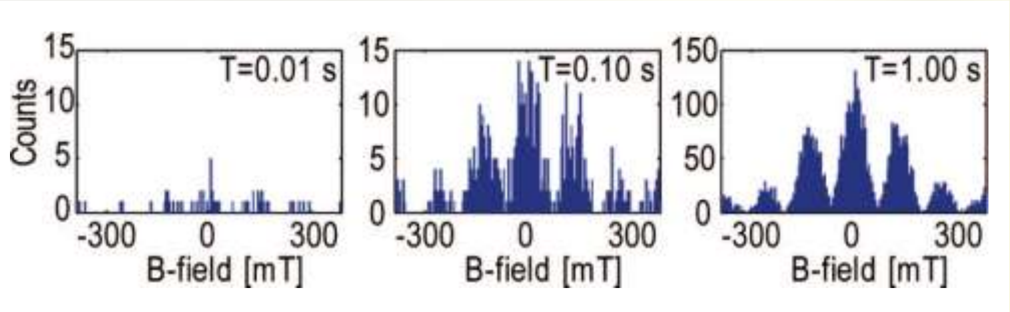
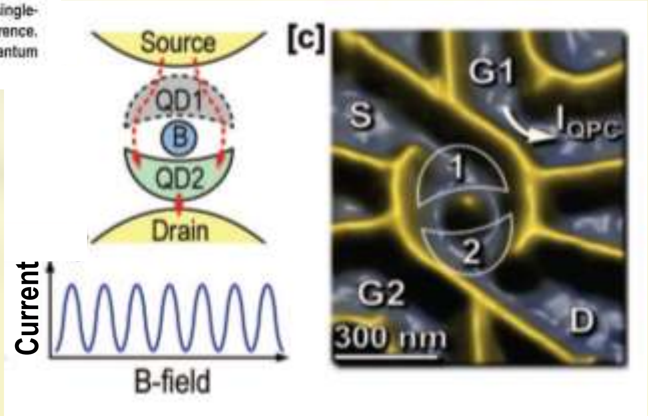
D. C. Driscoll and A. C. Gossard
Materials Departement, University of California, Santa Barbara, California 93106

Received June 13, 2008

ABSTRACT
We demonstrate real-time detection of self-interfering electrons in a double quantum dot embedded in an Aharonov−Bohm interferometer, with visibility approaching unity. We use a quantum point contact as a charge detector to perform time-resolved measurements of single-electron tunneling. With increased bias voltage, the quantum point contact exerts a back-action on the interferometer leading to decoherence. We attribute this to emission of radiation from the quantum point contact, which drives noncoherent electronic transitions in the quantum dots.

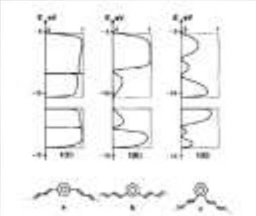
K. Ensslin



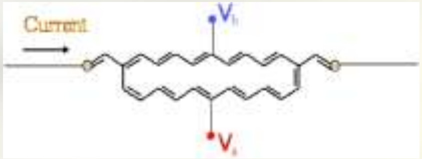


Nano Lett., **8**, 2547 (2008)

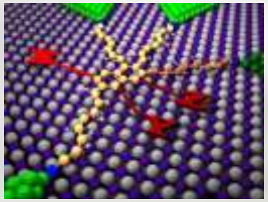
Intramolecular interference



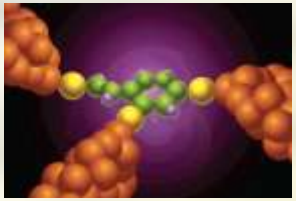
P. Sautet and C. Joachim
Chem. Phys. Lett. **153**, 511 (1988)



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

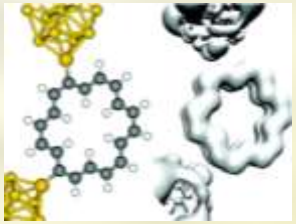


R. Stadler, et al.
Nanotechnology, **14**, 138 (2003)

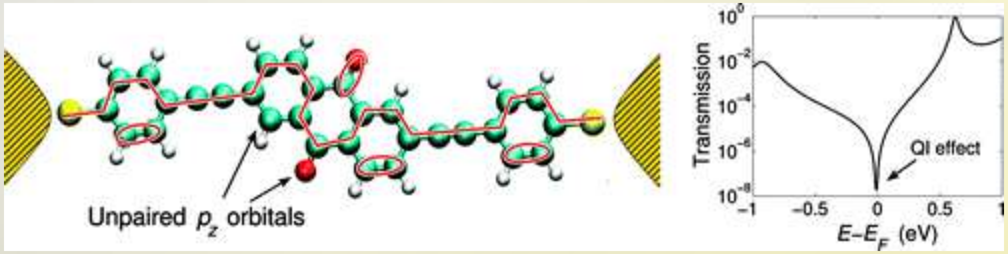


D. V. Cardamone, et al.
Nano Lett., **6**, 2422 (2006)

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

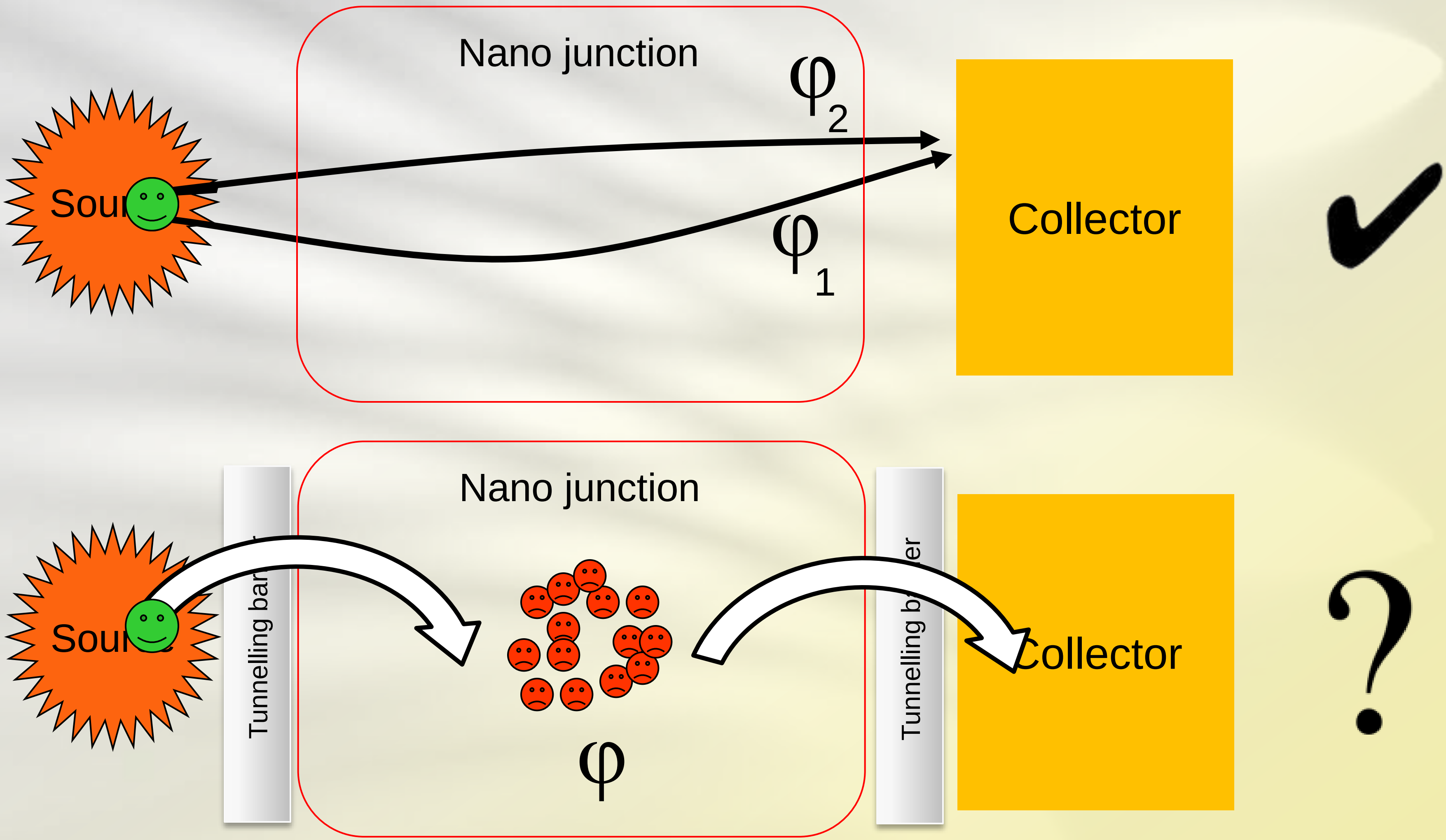


S.H. Ke, et al.
Nano Lett., **8**, 3257 (2008)

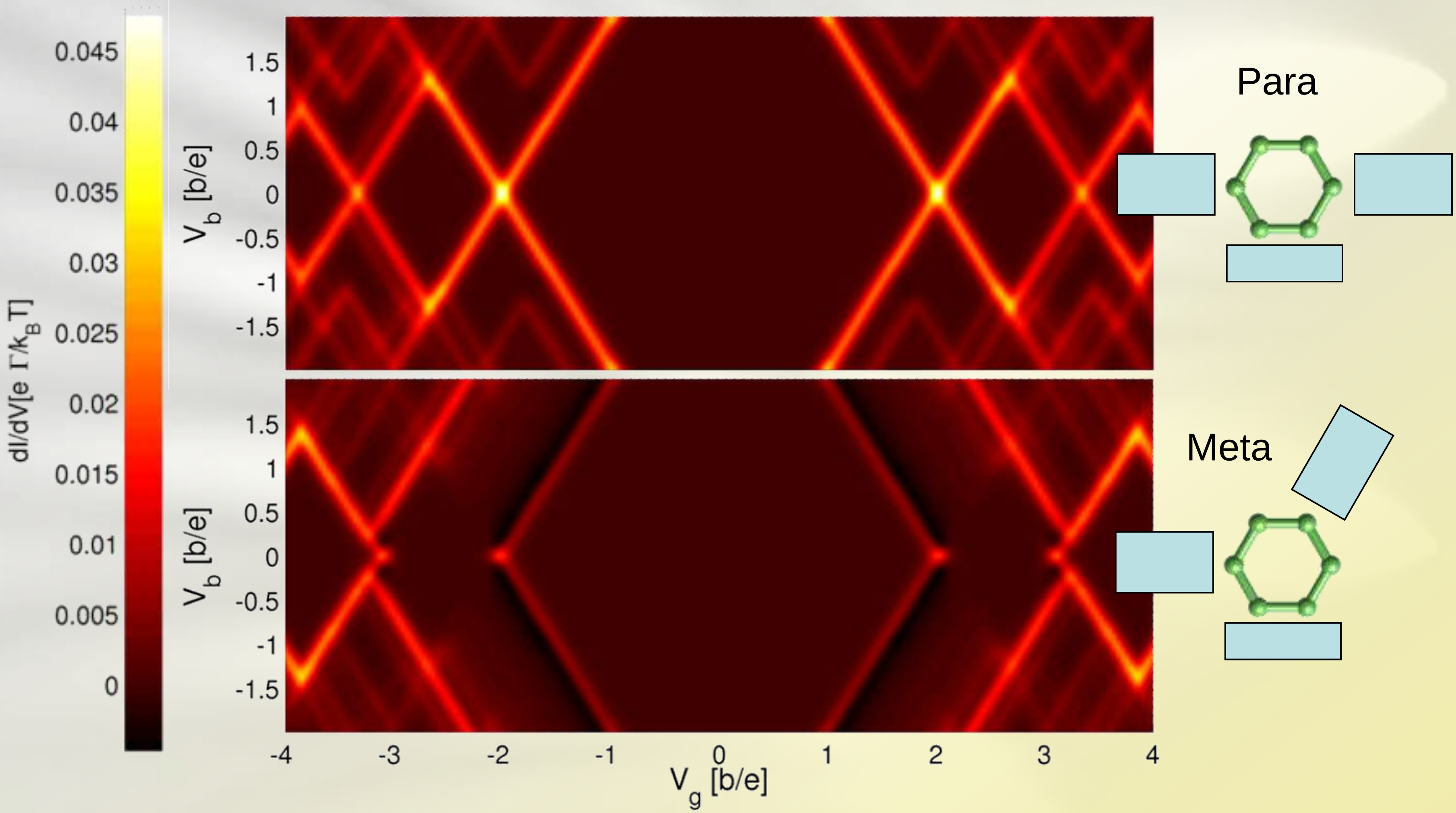


T. Markussen, et al.
Nano Lett., **10**, 4260 (2010)

Interference and dephasing

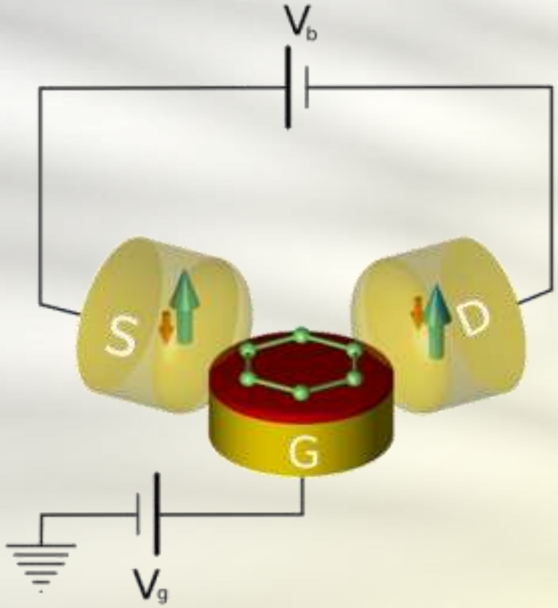


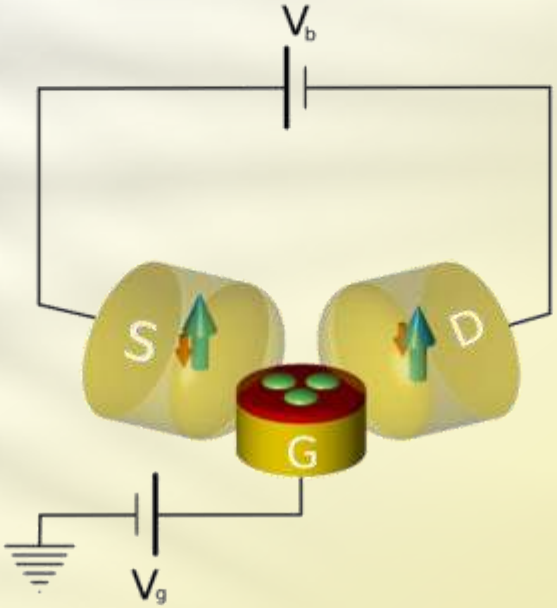
Interference in weak coupling



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

Interference
Single Electron Transistors

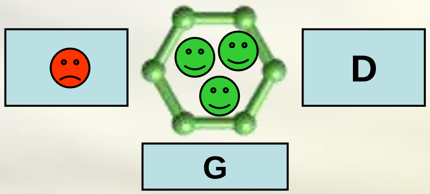




(Benzene) ISET...

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

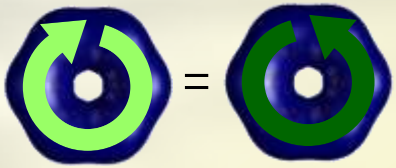
⇒ **Coulomb blockade**



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

- **Rotational** symmetry

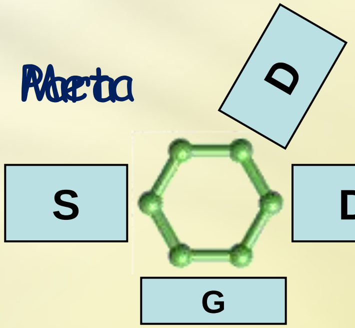
⇒ **Orbitally degenerate states**



$$E_1 = E_2$$

- Contact **geometry**

⇒ **Contact symmetry breaking**

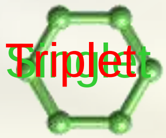


$$\frac{\gamma_{1L}}{\gamma_{2L}} \neq \frac{\gamma_{1R}}{\gamma_{2R}}$$

… with a magnetic flavour

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

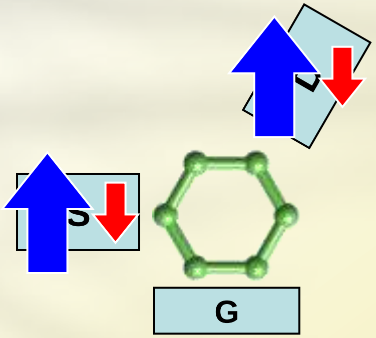
→ **Exchange splitting**



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

- Parallel **ferromagnetic** leads

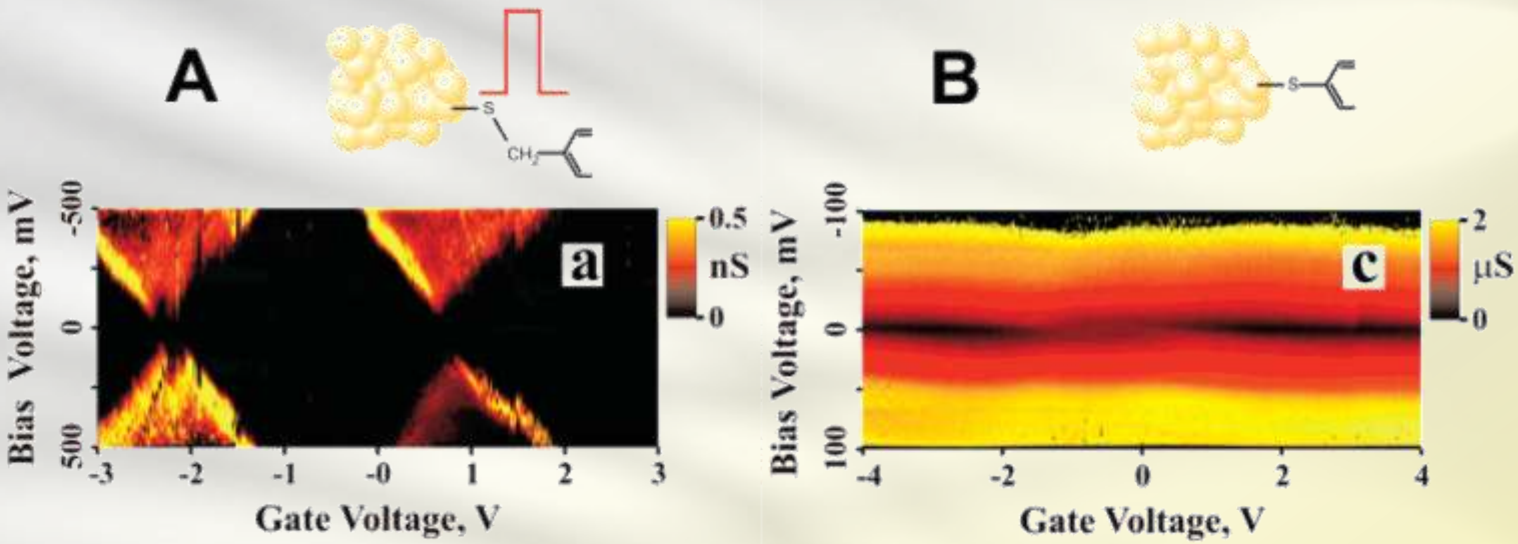
→ **Spin symmetry breaking**



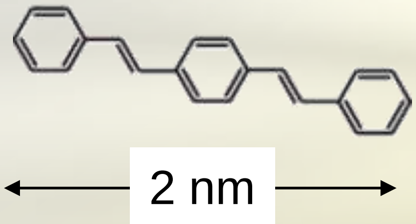
$\Gamma_{\alpha\uparrow} \neq \Gamma_{\alpha\downarrow}$

Are this conditions **achievable** in today experiments ?

Coulomb blockade



OPV-3

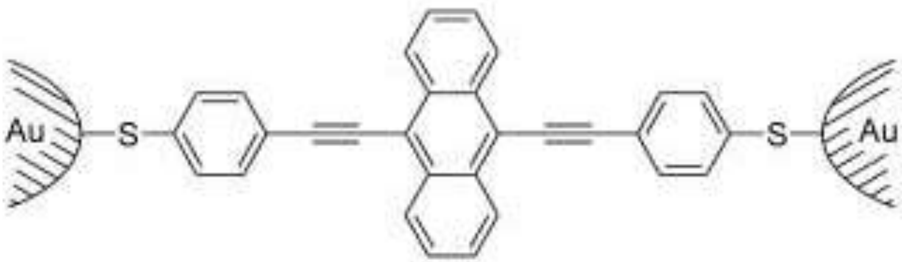


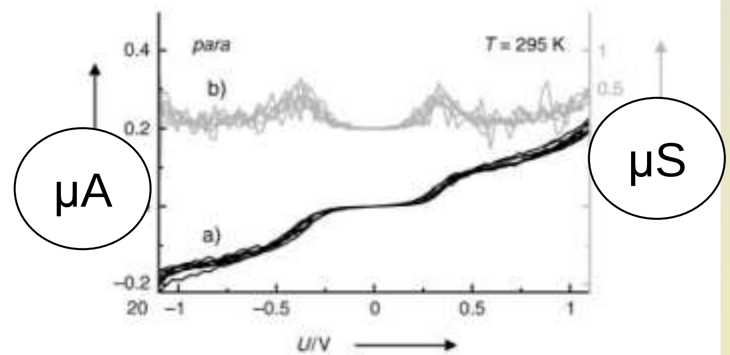
- **Gating** of 2 nm sized molecule
- **Weak coupling** realization with specific anchor groups

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

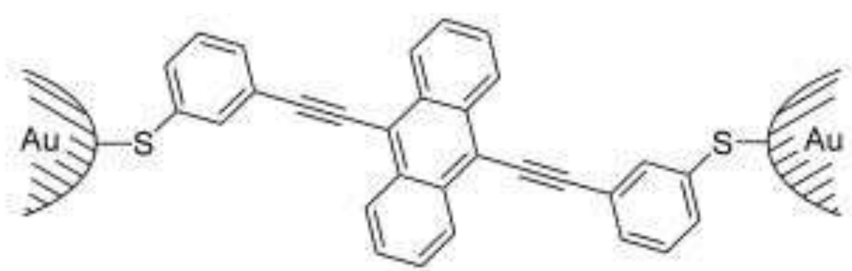
Symmetry breaking contacts

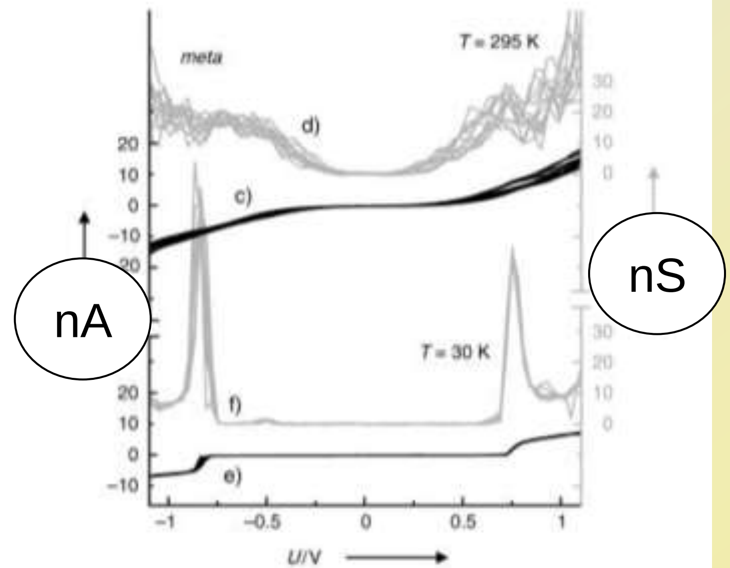
Para configuration





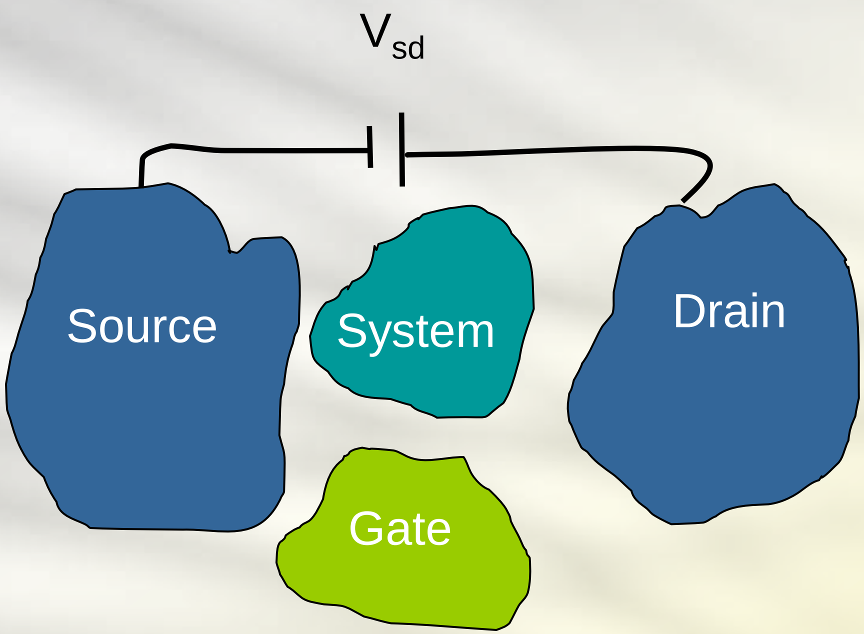
Meta configuration





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

The Hamiltonian



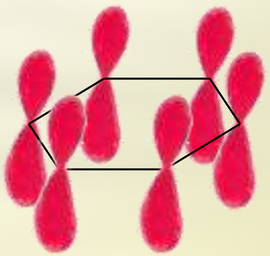
$$H = H_{\mathrm{Sys}} + H_{\mathrm{leads}} + H_{\mathrm{tun}}$$

$$\begin{cases} H_{\mathrm{Sys}} = H_{\mathrm{ben}} \;/ H_{\mathrm{TD}} \\ H_{\mathrm{leads}} = \sum_{\alpha\, k\, \sigma} \epsilon_k c^{\dagger}_{\alpha k \sigma} c_{\alpha k \sigma} \\ H_{\mathrm{tun}} = t \sum_{\alpha k \sigma} \left(d^{\dagger}_{\alpha\sigma} c_{\alpha k \sigma} + c^{\dagger}_{\alpha k \sigma} d_{\alpha\sigma} \right) \end{cases}$$

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

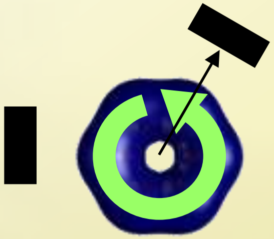
N	Degeneracy	GS energy[eV] (at $\xi = 0$)	GS symmetry representation
0	1	0	A_{1g}
1	2	-22	A_{2u}
2	1	-42.25	A_{1g}
3	4	-57.42	E_{1g}
4	3	-68.875	A_{2g}
5	4	-76.675	E_{1g}
6	1	-81.725	A_{1g}
7	4	-76.675	E_{2u}
8	3	-68.875	A_{2g}
9	4	-57.42	E_{2u}
10	1	-42.25	A_{1g}
11	2	-22	B_{2g}
12	1	0	A_{1g}

Rotation phase factors

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

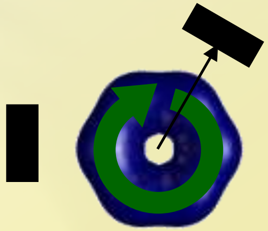
- $\mathcal{R}_\phi |6_g\rangle = |6_g\rangle$ No phase acquired
- $\mathcal{R}_\phi |7_g\,\ell\rangle = e^{-i\ell\phi} |7_g\,\ell\rangle$ $\ell = \pm 2$

$\ell = +2$



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

$\ell = -2$

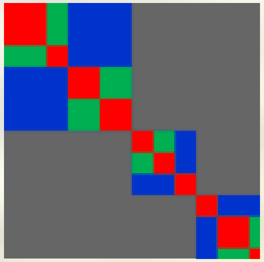


$\exp\left(-i\frac{2\pi}{3}\right)$

Generalized Master Equation

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

- We 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 :

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

Coherent dynamics

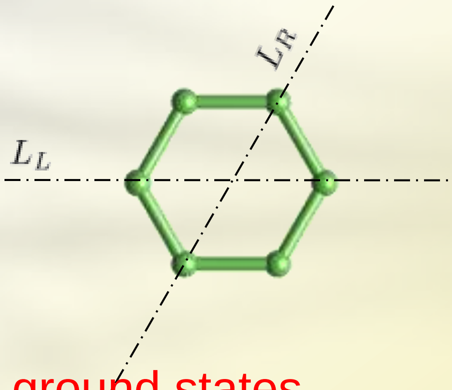
Effective **internal** dynamics

Tunnelling dynamics

The effective Hamiltonian

The effective Hamiltonian is expressed in terms of **angular momentum** operators and **renormalization frequencies:**

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



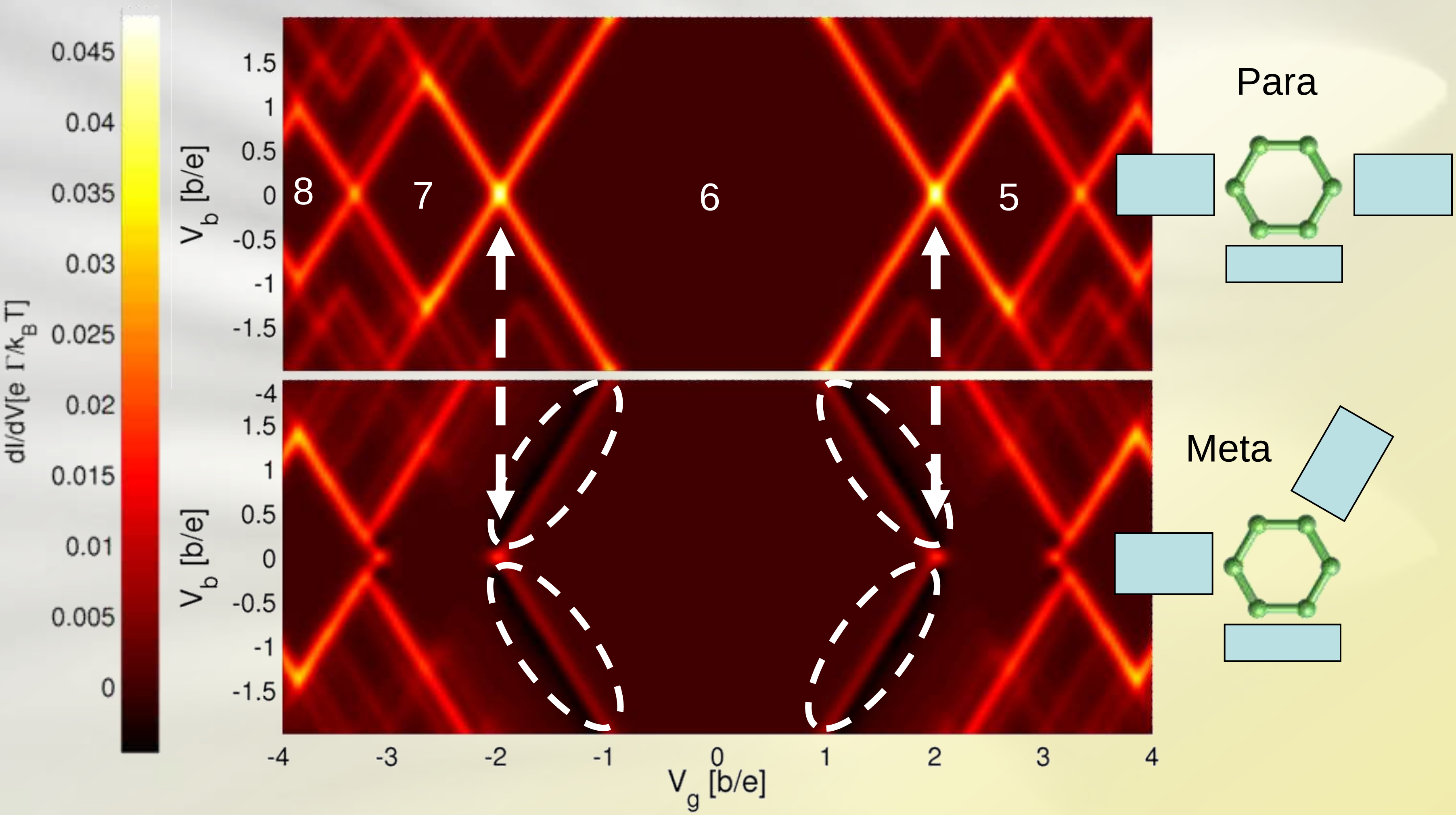
In particular in the Hilbert space of the 7 particle ground states

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

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

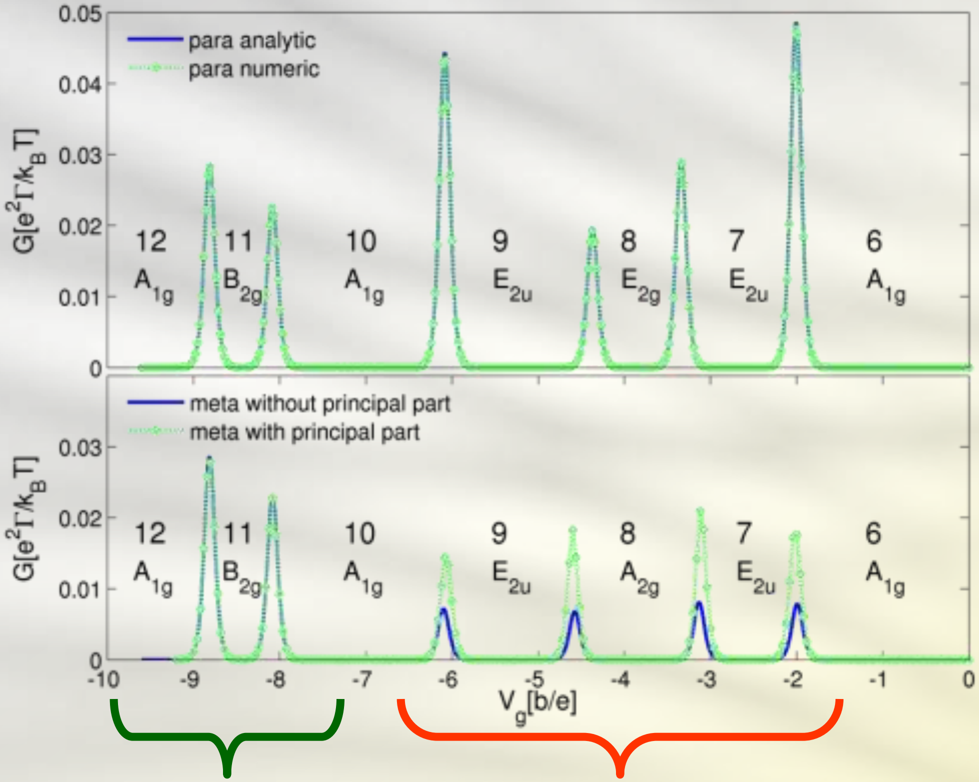
⬅ Bias and gate dependent

Para vs. Meta



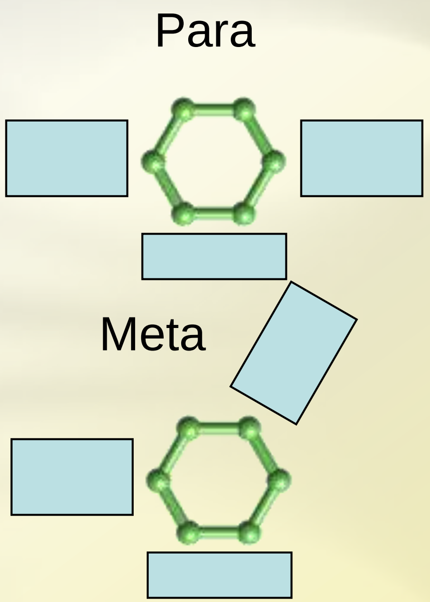
G. Begemann, D. Darau, **AD**, M. Grifoni, *Phys. Rev. B* **77**, 201406(R) (2008)

Conductance suppression



equal

suppressed



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

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

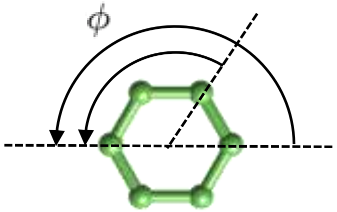
Destructive interference

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

Interference factor

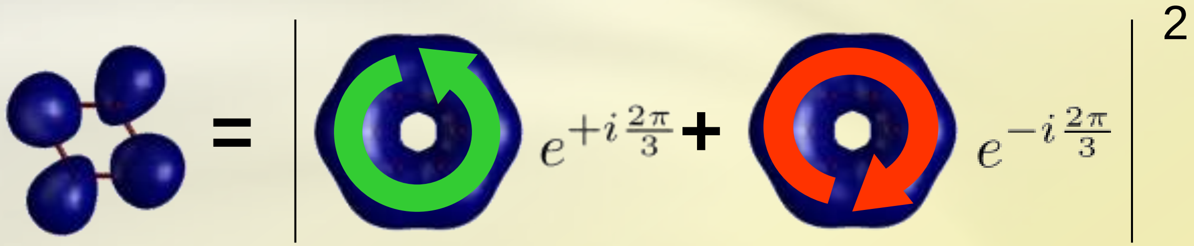
$$\Lambda = \left| \sum_{nm\tau} |\langle N,\, n | d_{\mathrm{L}\tau} | N+1,\, m \rangle|^2 e^{i\phi_{nm}} \right|^2$$

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

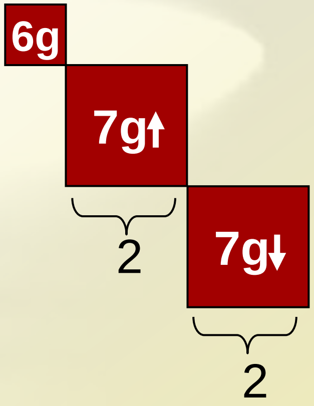


In particular for the transition **6 -7** in the **meta** configuration:

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



Negative Differential Conductance



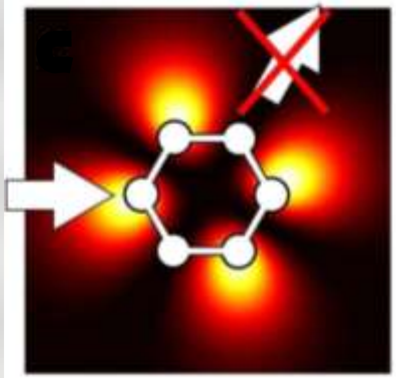
- The 7 particle ground state has spin and orbital **degeneracies**;
- **Physical basis**: the basis that diagonalizes the stationary density matrix;
- The physical basis **depends on the bias:** in whatever reference basis, **coherences** are essential for a correct description of the system;
- The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

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

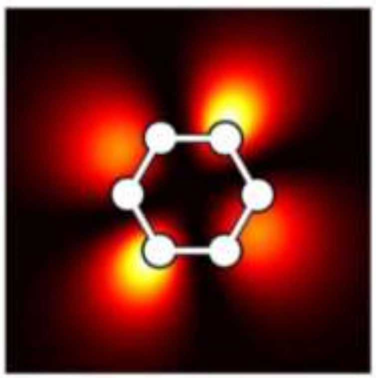
Interference blockade

Geometry

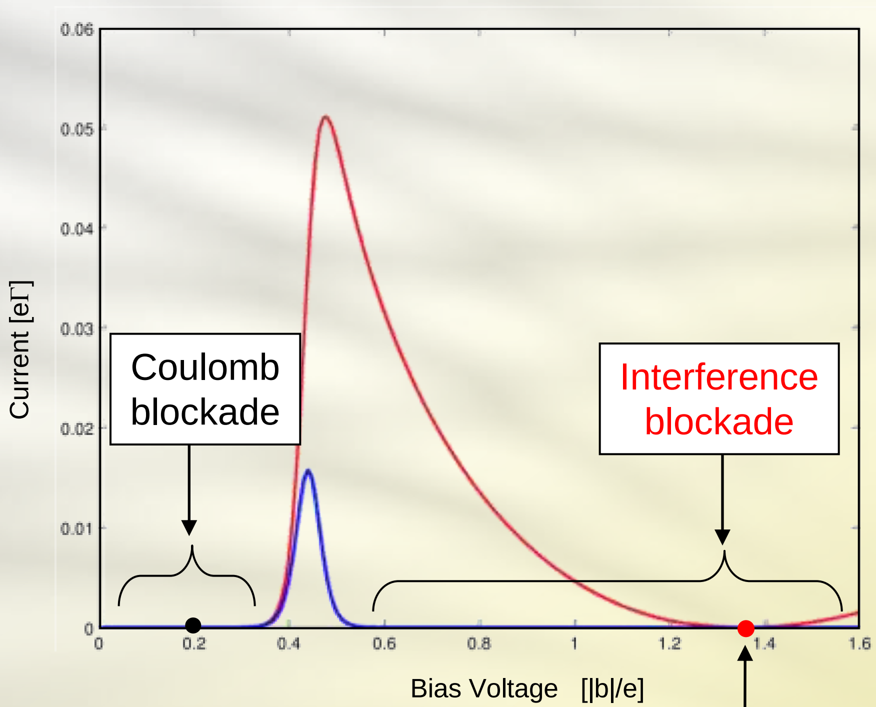
Blocking state



Non-blocking state



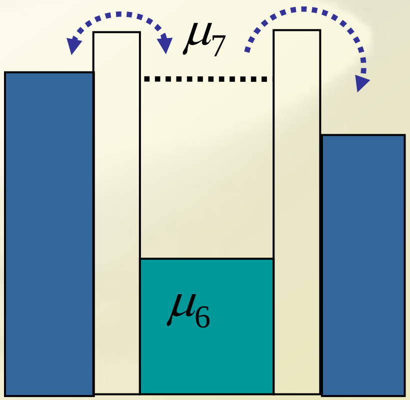
I-V for transition 6 -7



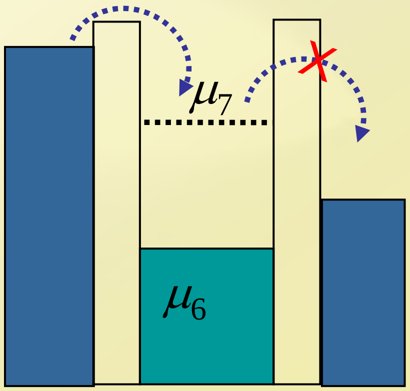
The blocking state is an eigenstate of the effective Hamiltonian

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

Energetics

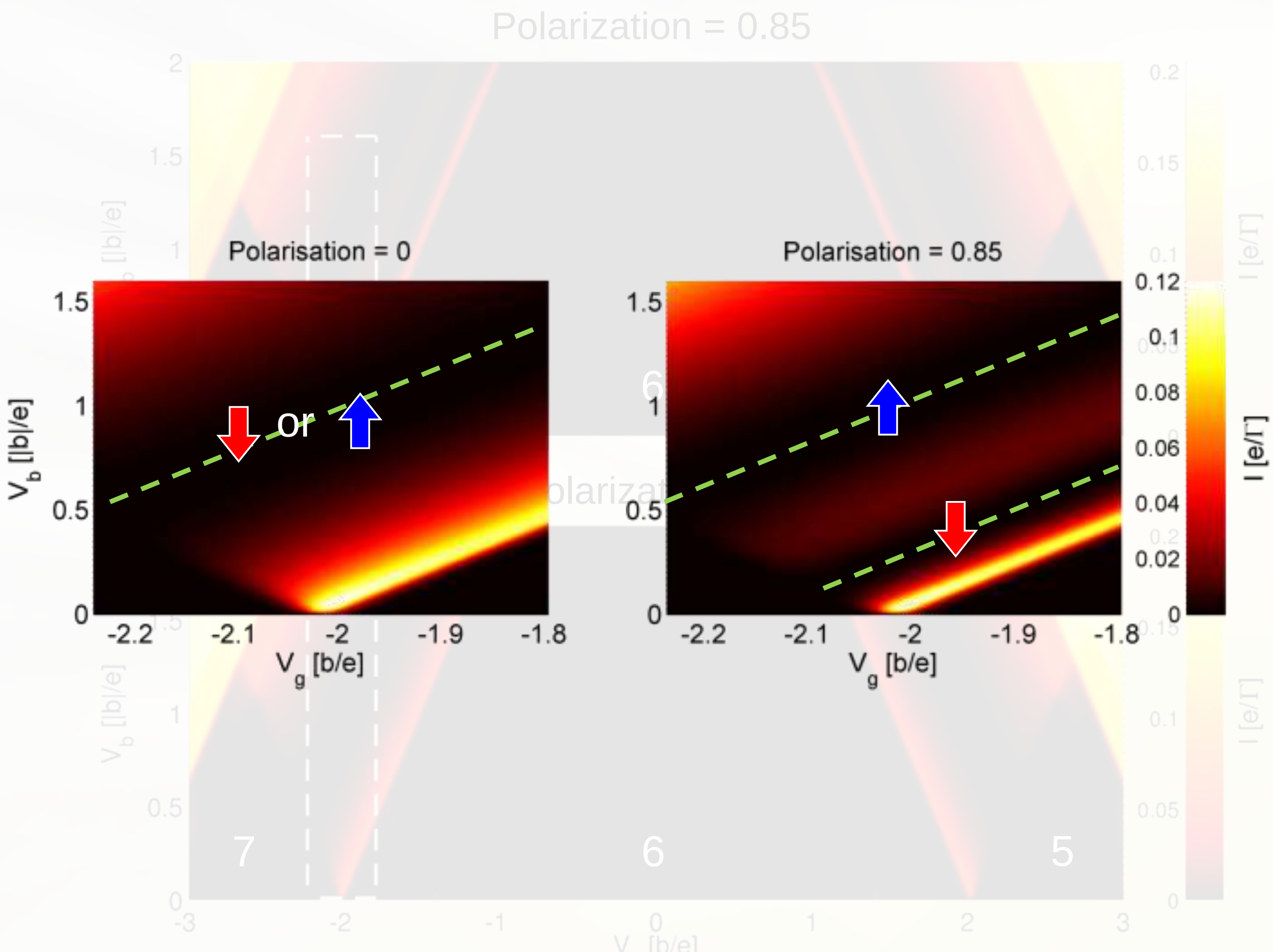


current onset

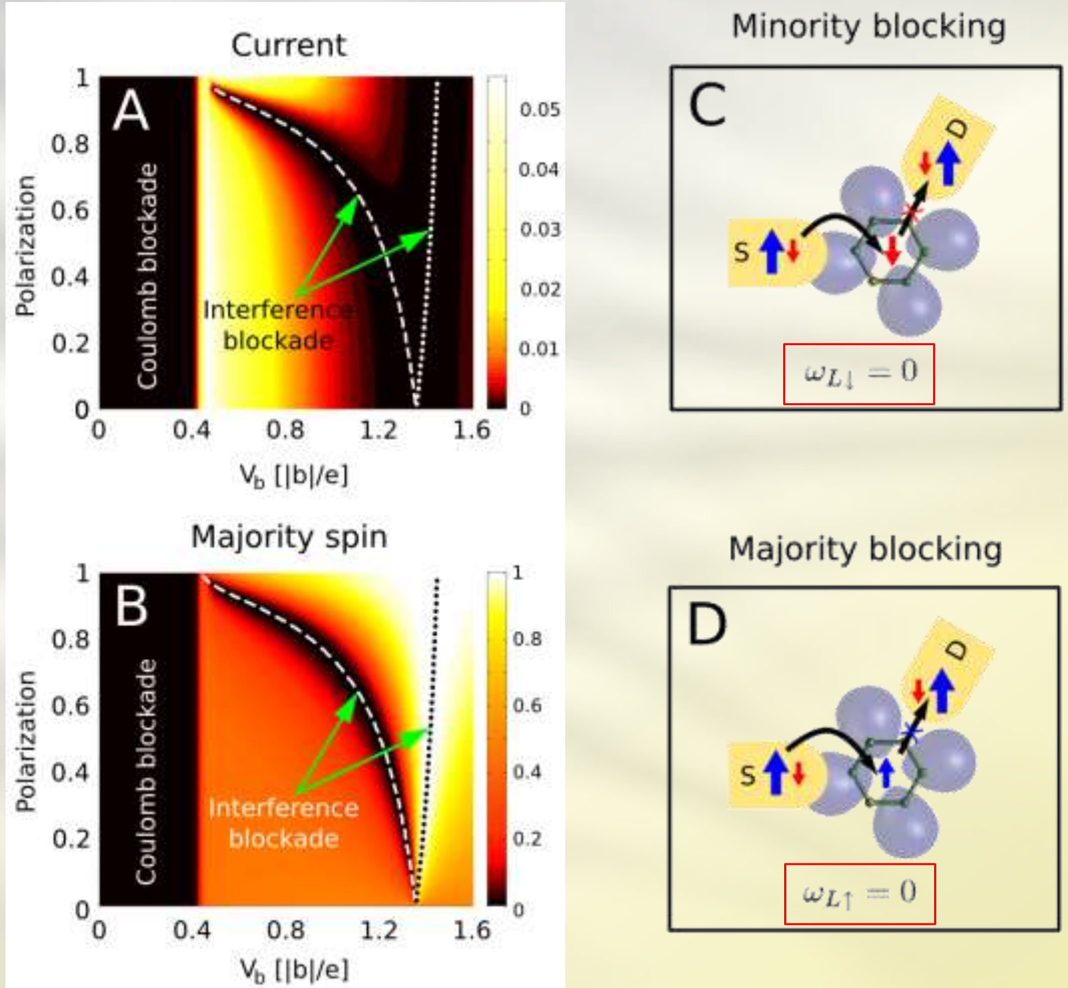


blockade

Normal vs. ferromagnetic leads



Selective Interference Blocking



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

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**:

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

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

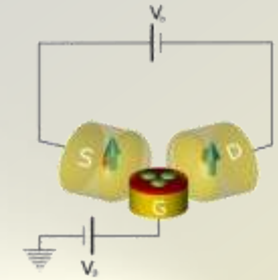
$$+ \langle 7_g \ell \uparrow | d^\dagger_{M\uparrow} | 6\{E\}\rangle\langle 6\{E\}| d_{M\uparrow} | 7_g m \uparrow\rangle p_\alpha(E_{7_g} - E)$$

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

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

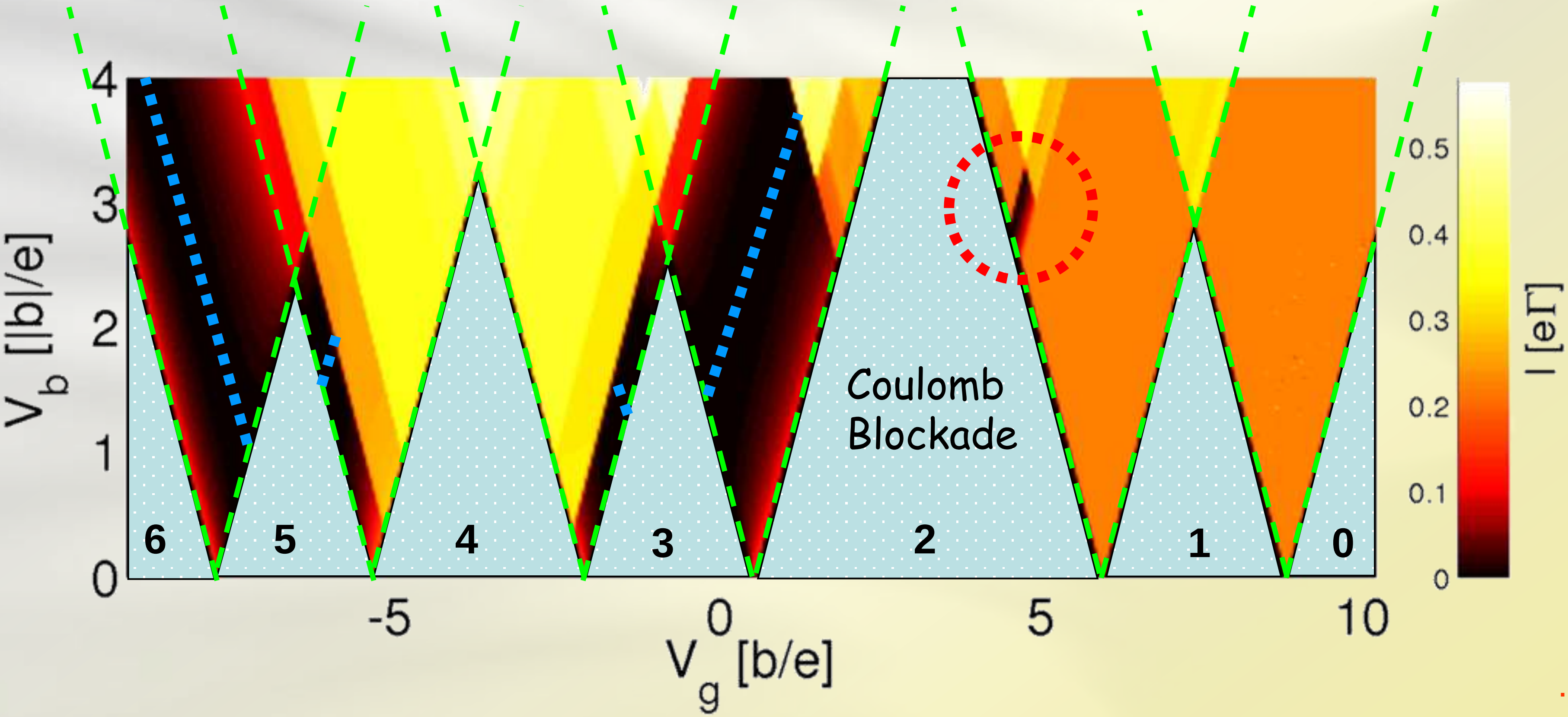
The splitting of the level renormalization depends crucially on the Coulomb interaction on the molecule and **vanishes in absence of exchange.**

Triple dot ISET

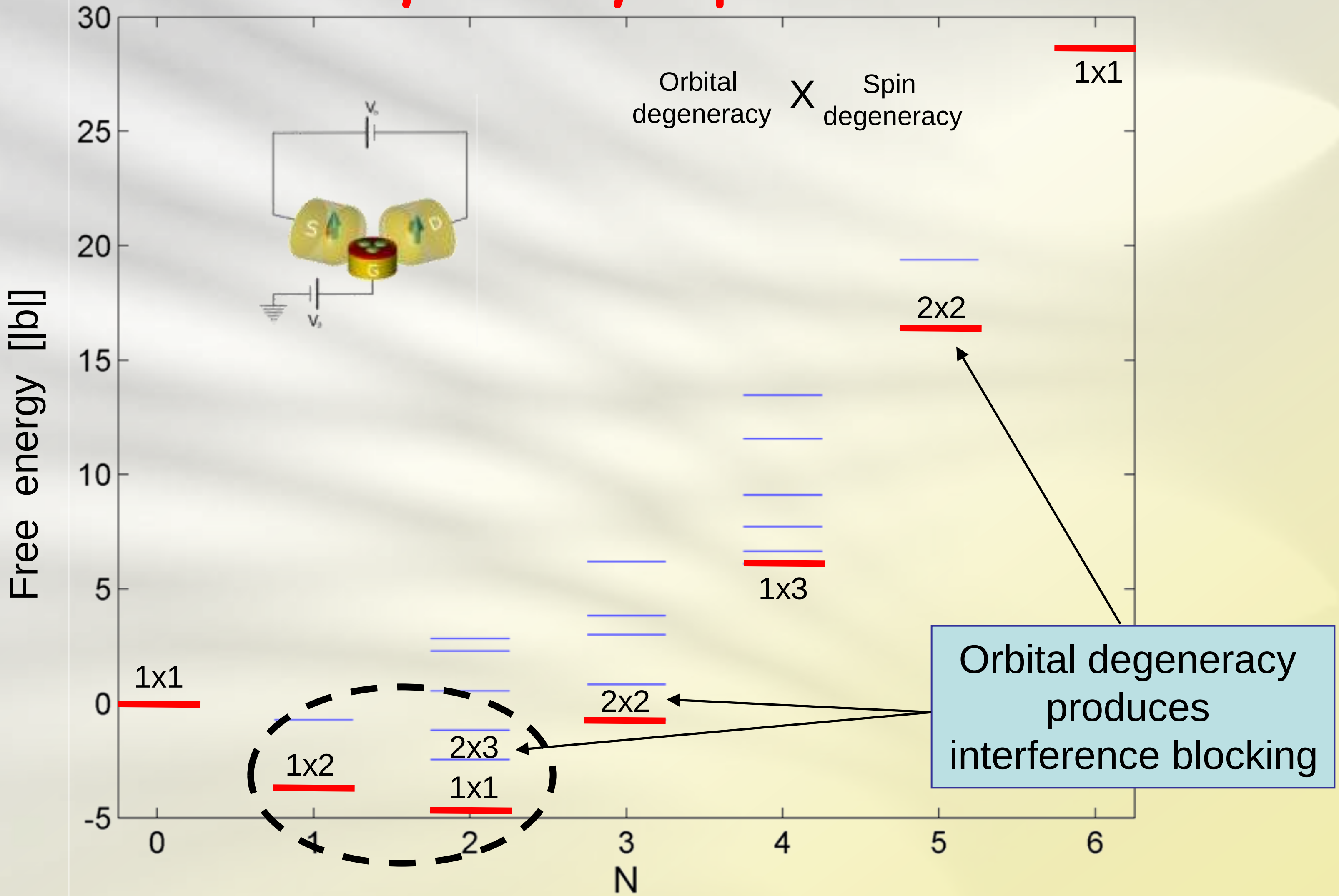


Interference Blockade
--**ground** states--

Interference Blockade
--**excited** state--



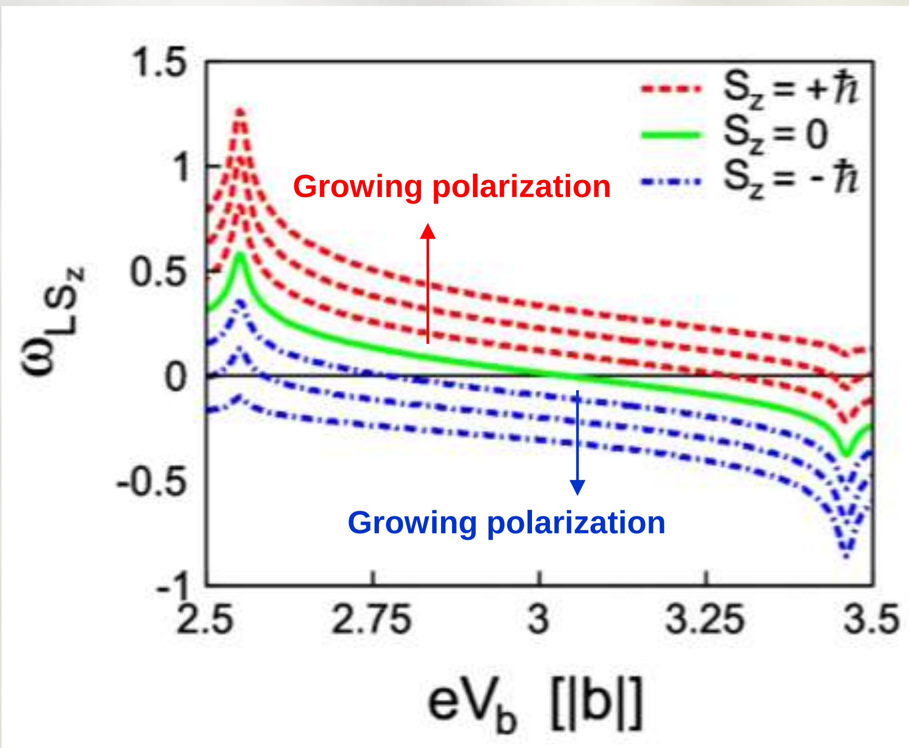
Many-body spectrum

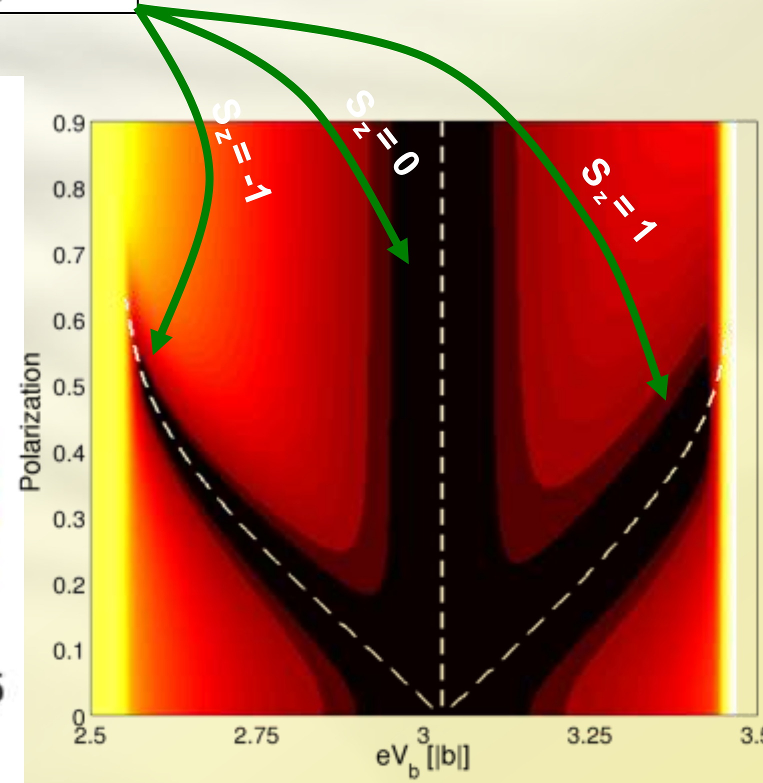


Triplet splitting

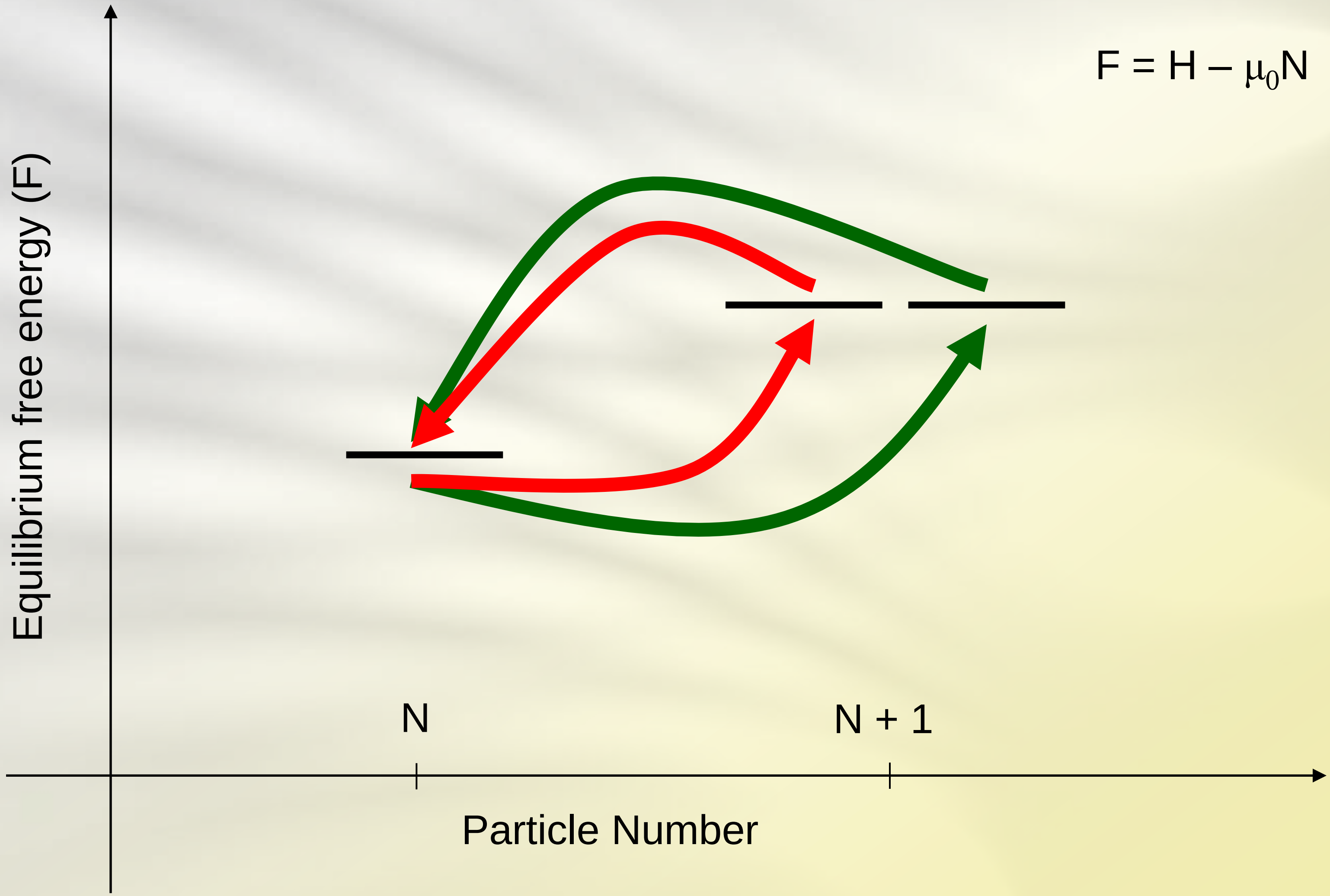
The states decoupled from the right lead are eigenstates of L_R. They are eigenstates of H_{eff} only if

$$\omega_{L\,S_z} = 0$$





The “two paths” in the ISET



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, **AD**, and M. Grifoni, *PRB,* **79**, 235404 (2009)

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

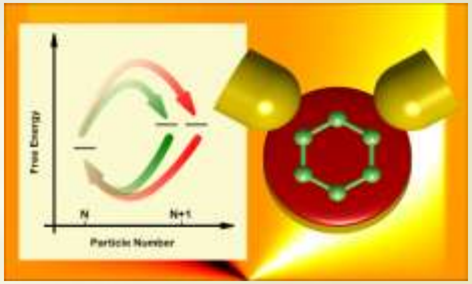
$$\mathcal{L}_{\mathrm{tun}}\sigma_{\mathrm{B}} = 0$$

- is an eigenstate of the effective Hamiltonian

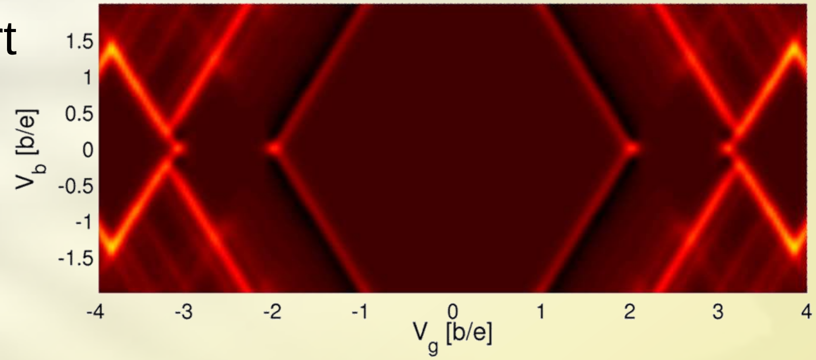
$$[H_{\mathrm{eff}}, \sigma_{\mathrm{B}}] = 0$$

Conclusions

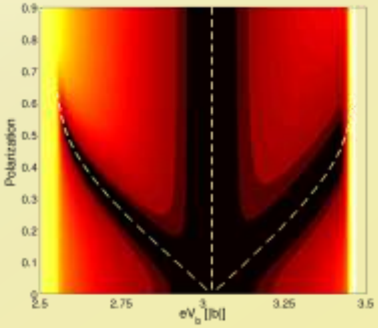
• Interference does occur even in the single-electron tunnelling regime when energetically equivalent paths involving **degenerate states** contribute to the dynamics.



• Interference effects dominates the transport characteristics of ISET both in the linear and non linear regime producing selective **suppression of the conductance** and inferference **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

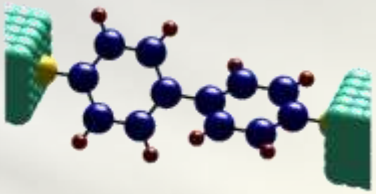
SPP 1243 Quantum Transport
at the Molecular Scale

SFB 689 Spinphänomene
in reduzierten Dimensionen

Beyond… ISET

Vibronic effects in transport through conjugated molecules

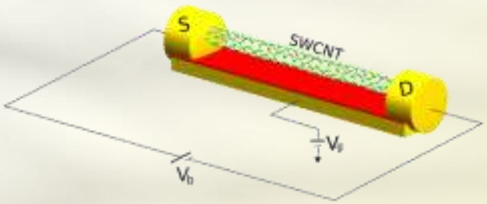
Phys. Rev. Lett., **97**, 166801 (2006)



K. Richter

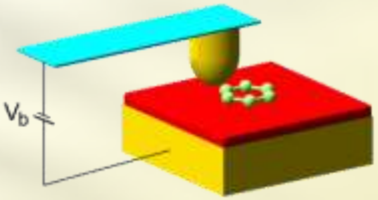
Transport through suspended single wall carbon nanotube quantum dots

arXiv:1101.3892 submitted to PRB



A. Yar

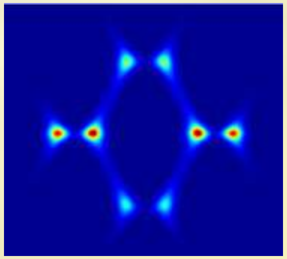
Interference effects in transport through single molecules in the STM set-up



S. Kolmeder

J. Repp

Transport through double dot structures with multiple gates



D. Preusche

A. Hüttel

Thank you for your attention!

Conclusions

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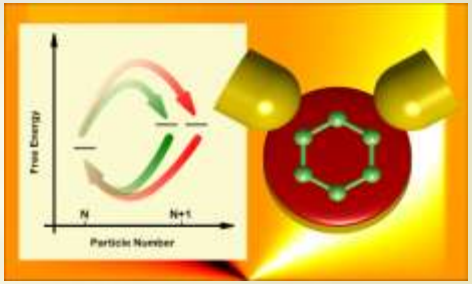

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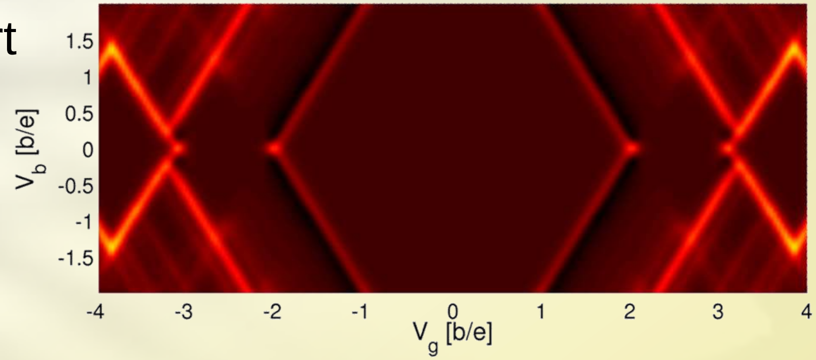

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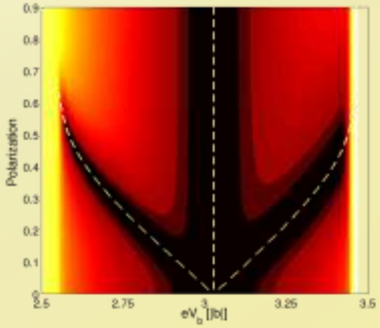
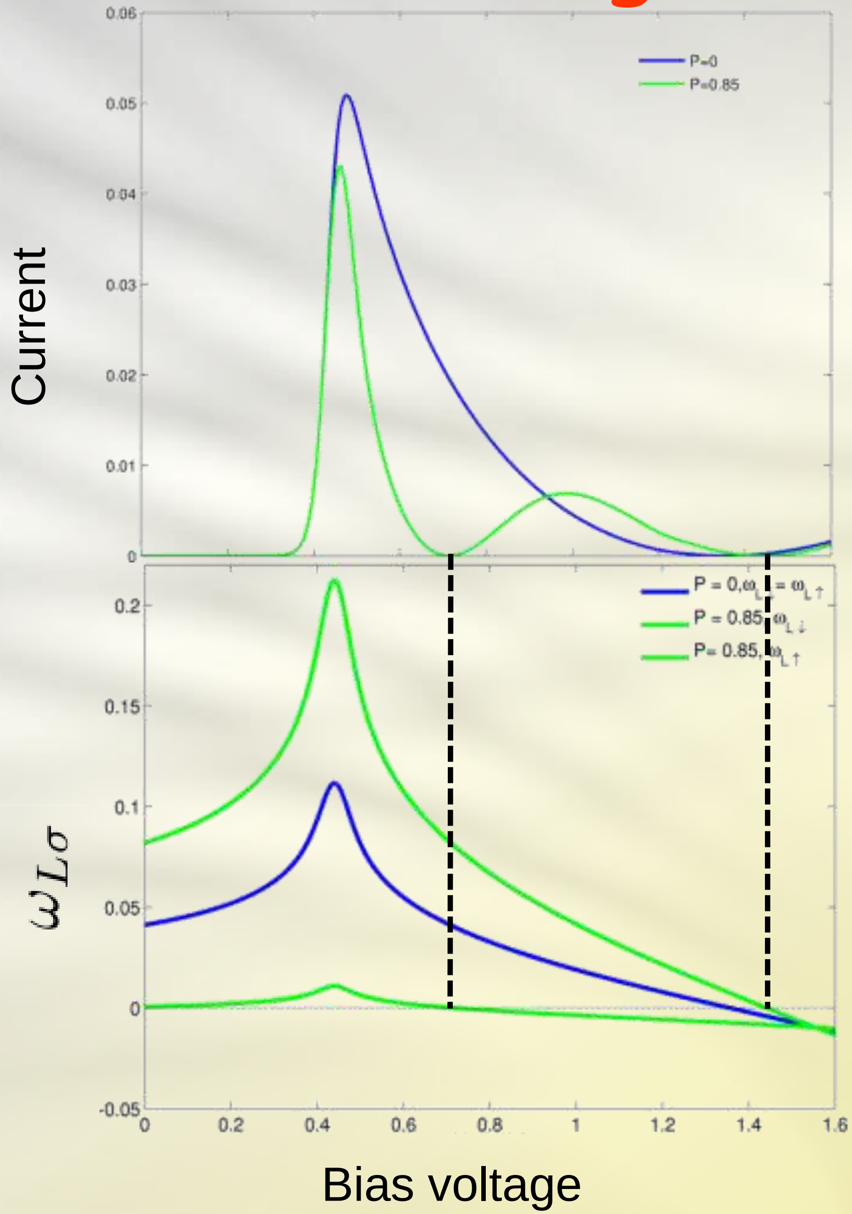
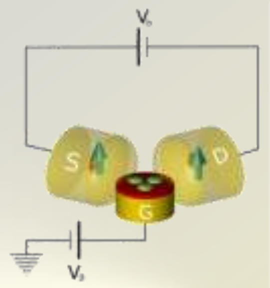


Supplementary material

Normal vs ferromagnetic leads



The triple dot ISET



$$H = H_{\rm sys} + H_{\rm leads} + H_{\rm tun}$$

$$\begin{aligned} H_{\rm sys} &= \xi_0 \sum_{i\sigma} d^\dagger_{i\sigma} d_{i\sigma} + b \sum_{i\sigma} \left(d^\dagger_{i\sigma} d_{i+1\sigma} + d^\dagger_{i+1\sigma} d_{i\sigma} \right) \\ &+U \sum_i \left(n_{i\uparrow} - \tfrac{1}{2}\right)\left(n_{i\downarrow} - \tfrac{1}{2}\right) \\ &+V \sum_i \left(n_{i\uparrow} + n_{i\downarrow} - 1\right)\left(n_{i+1\uparrow} + n_{i+1\downarrow} - 1\right) \end{aligned}$$

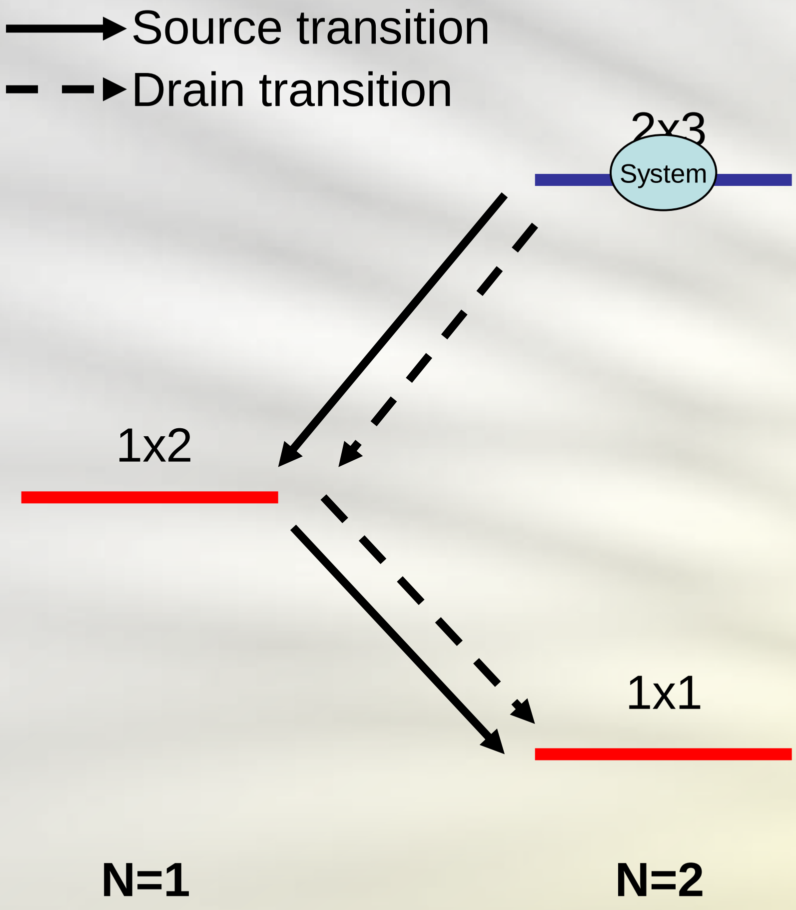
Extended Hubbard Hamiltonian with on-site and nearest neighbors **Coulomb interaction**

$$H_{\rm tun} = t \sum_{\alpha k \sigma} (c^\dagger_{\alpha k\sigma} d_{\alpha\sigma} + d^\dagger_{\alpha\sigma} c_{\alpha k\sigma})$$

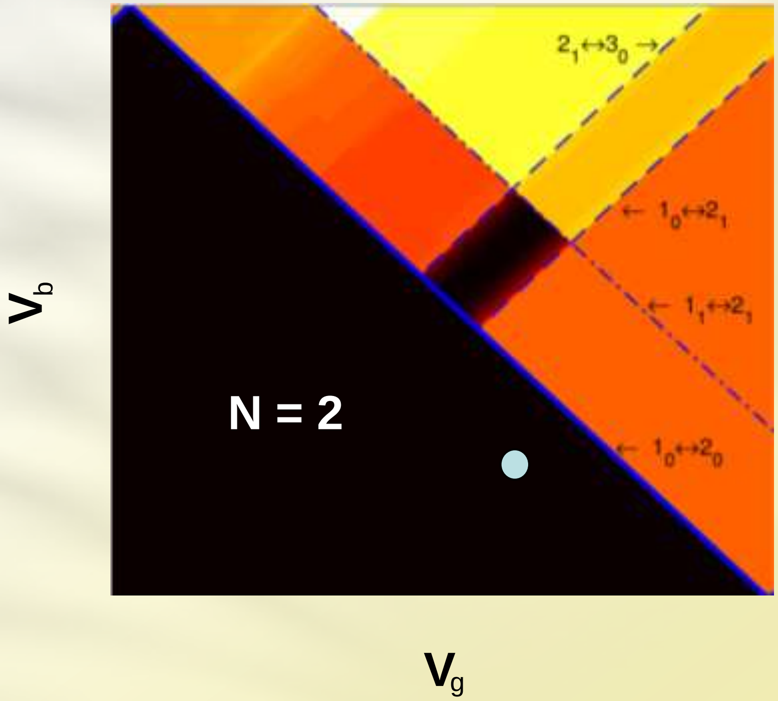
Tunnelling restricted to the dot closest to the corresponding lead

$H_{\rm leads}$ Ferromagnetic leads with equal parallel polarization

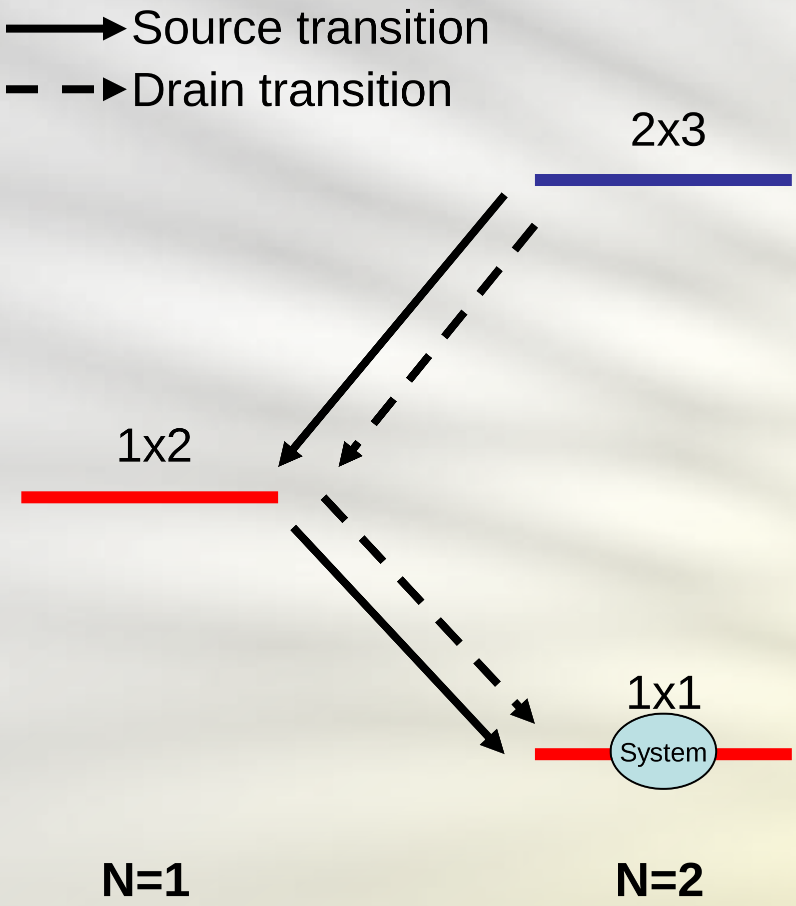
Excited state blocking

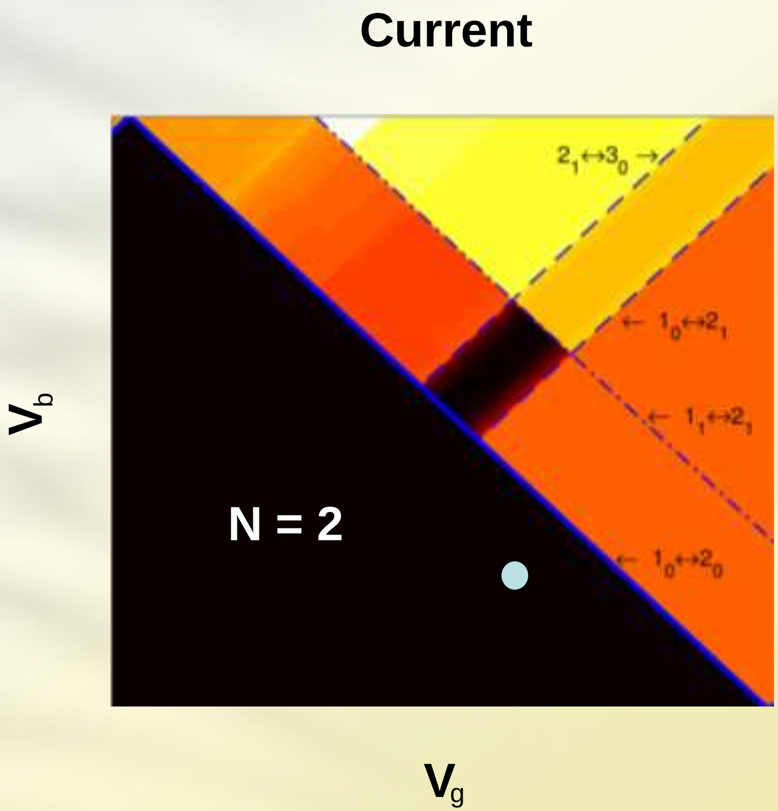


Coulomb Blockade

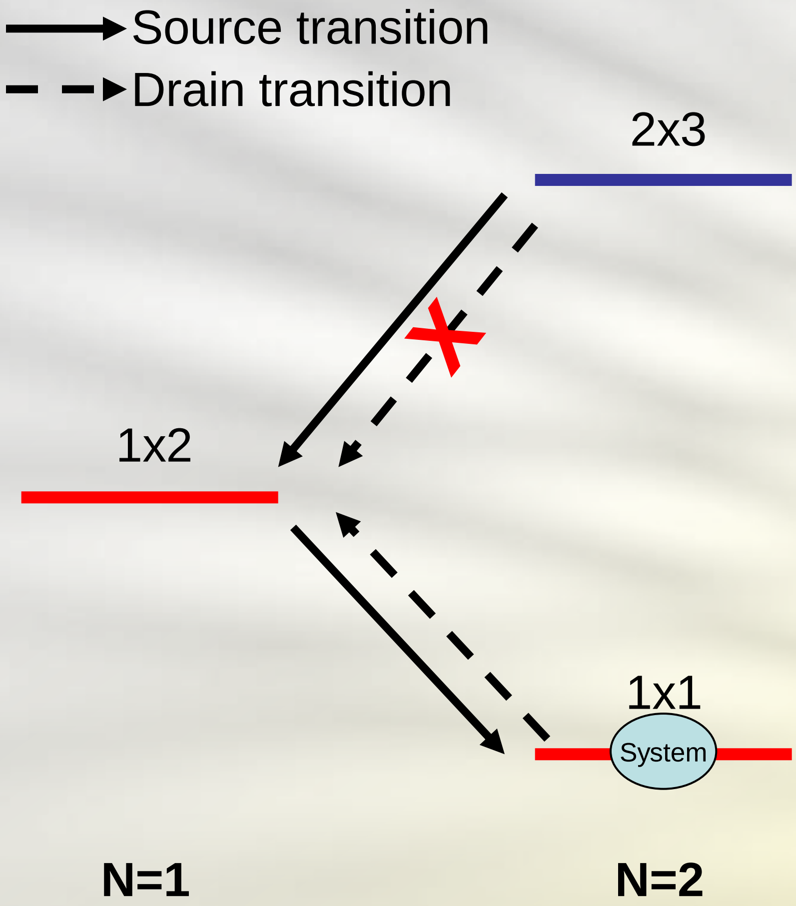


Excited state blocking

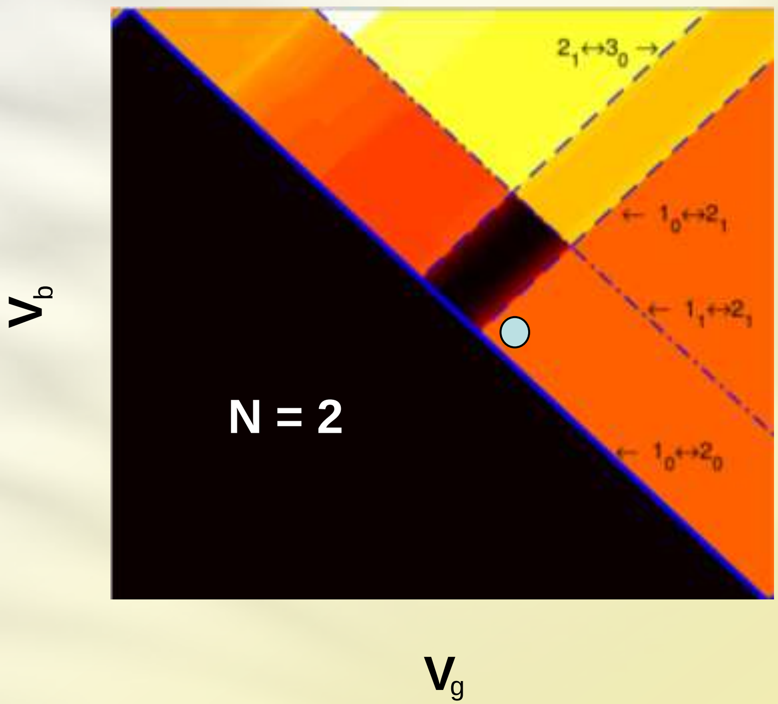




Excited state blocking

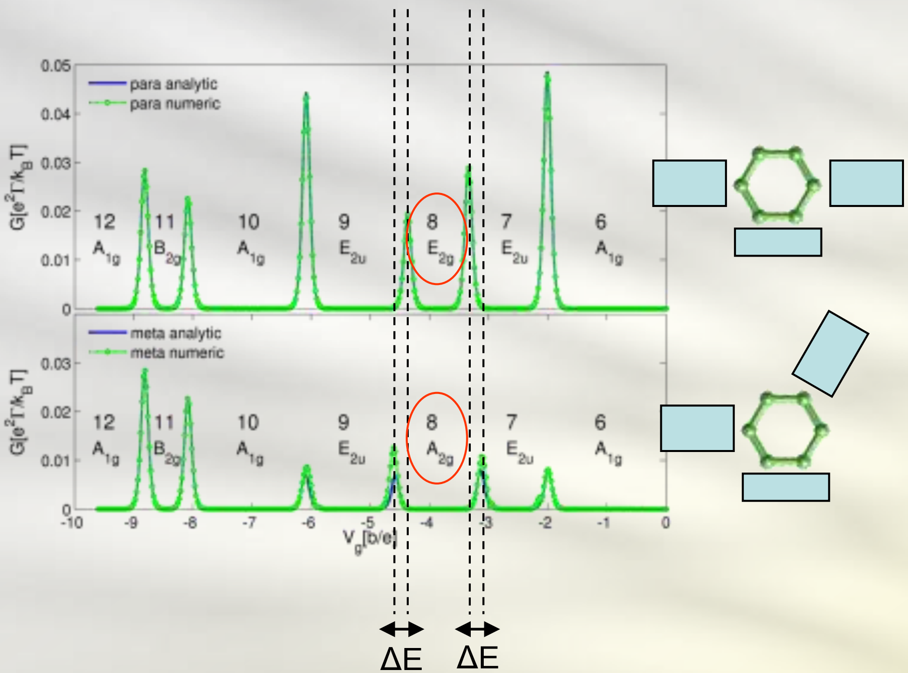


Interference Blockade

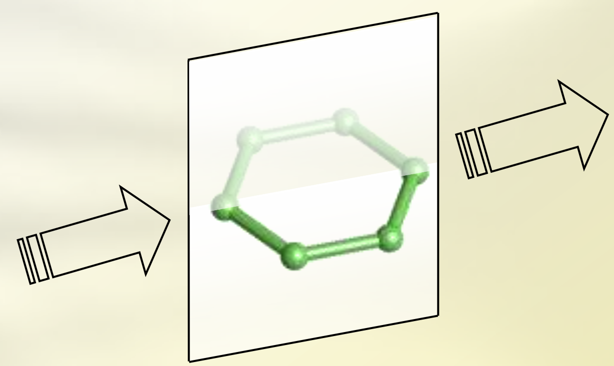


Three linear combinations of 2-particle excited states are coupled **ONLY to the source**.

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.