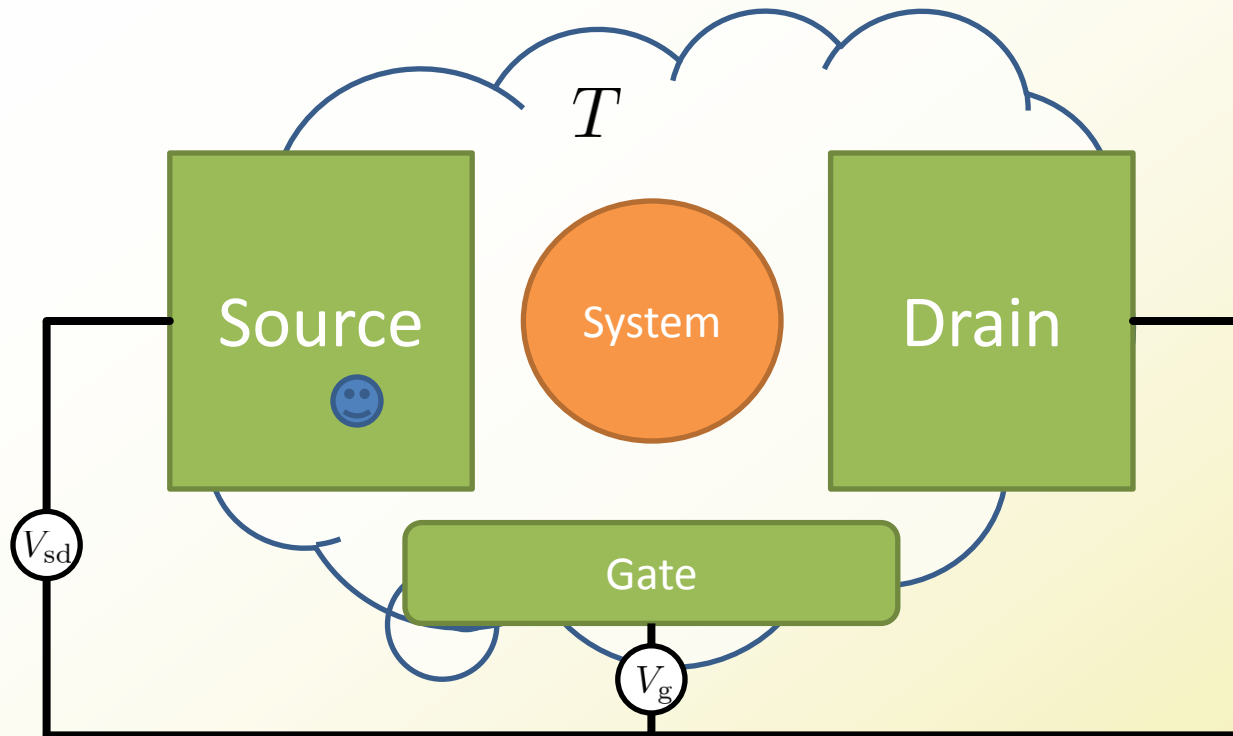


Coulomb blockade and single electron tunnelling

Andrea Donarini

Institute of theoretical physics, University of Regensburg

Three terminal device



Variation of the
electrostatic energy

$$\Delta E = \frac{e^2}{C}$$

Typical time scale $\delta t = R_t C$

Single electron detection

How **small** and how **cold** should the system be such that a **single electron** tunneling has a **measurable** effect ?



Thermal
fluctuations

$$\Delta E \gg k_B T$$



$$e^2 / C \gg k_B T$$

$$d = 100\text{nm} \quad \longleftrightarrow$$

$$C = 1\text{pF}$$

$$T \ll 1\text{K}$$



Quantum
uncertainty

$$\Delta E \delta t \gg \delta E \delta t \geq h$$



$$R_t \gg h/e^2$$

$$R_t \gg 13\text{k}\Omega$$



W. Heisenberg

First device (1987)

Aluminum- Aluminum oxide, Au-Cr gate electrode

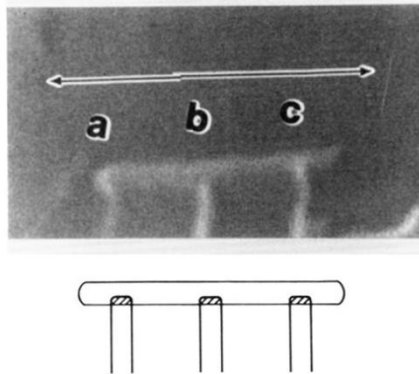


FIG. 2. A scanning-electron micrograph of a typical sample. Junctions labeled a, b, and c are formed where the vertical electrodes overlap and contact the longer horizontal central electrode. The bar is $1 \mu\text{m}$ long. The configuration is also shown in the accompanying drawing.

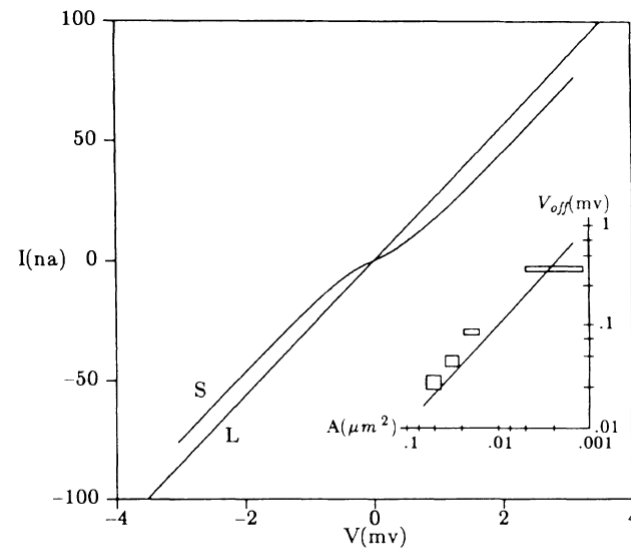


FIG. 3. I - V curves S and L for junctions corresponding to small and large C' at 1.7 K. Inset: Offset voltage vs junction areas (as determined from scanning-electron-microscopy photographs) for four different samples. The boxes represent the estimated uncertainties.

Fulton and Dolan PRL, **59** 109 (1987)

STM set-up (1992)

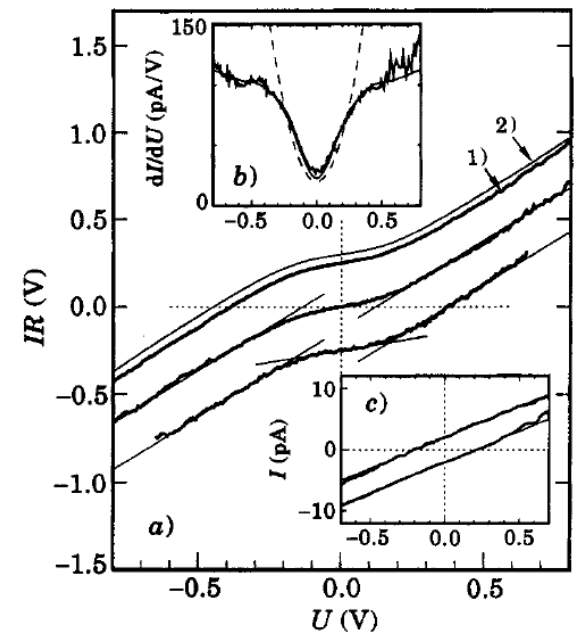
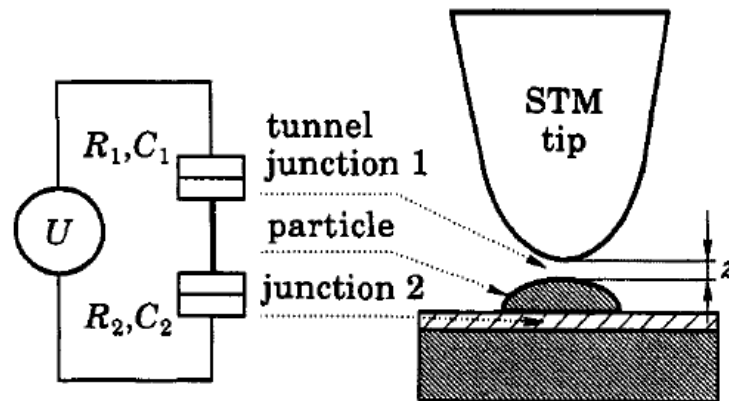
Single-Electron Tunnelling Observed at Room Temperature by Scanning-Tunnelling Microscopy.

C. SCHÖNENBERGER, H. VAN HOUTEN and H. C. DONKERSLOOT

Philips Research Laboratories - P.O.Box 80.000, 5600 JA Eindhoven, The Netherlands

(received 22 April 1992; accepted in final form 30 July 1992)

Abstract. - Ultrasmall (≤ 5 nm in lateral diameter) double-barrier tunnel junctions have been realized using a scanning tunnelling microscope, and an optimized metal particle-oxide-metallic substrate system. Three electrical transport effects, all in good agreement with the semi-classical theory of single-electron tunnelling, have been found at room temperature: the Coulomb gap, the Coulomb staircase and zero-bias conductance oscillations as a function of tip-particle distance.

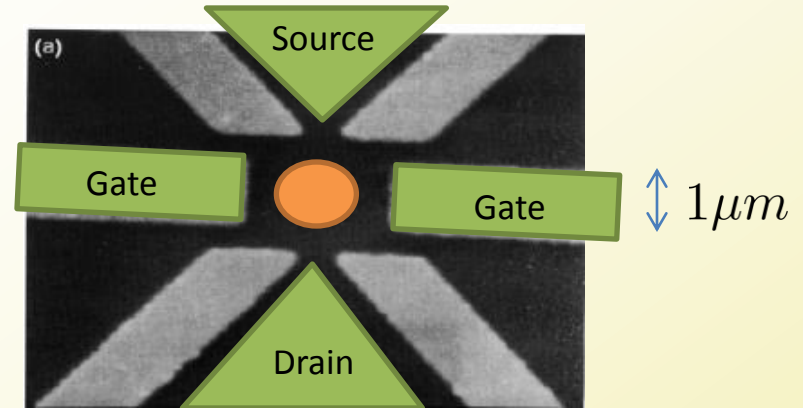
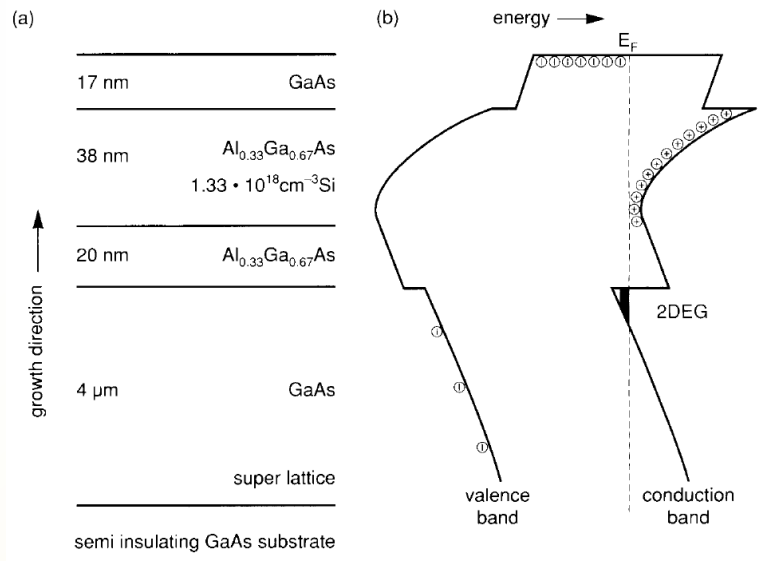


Schönenberger, van Houten and Donkersloot, EPL **20**, 249 (1992)

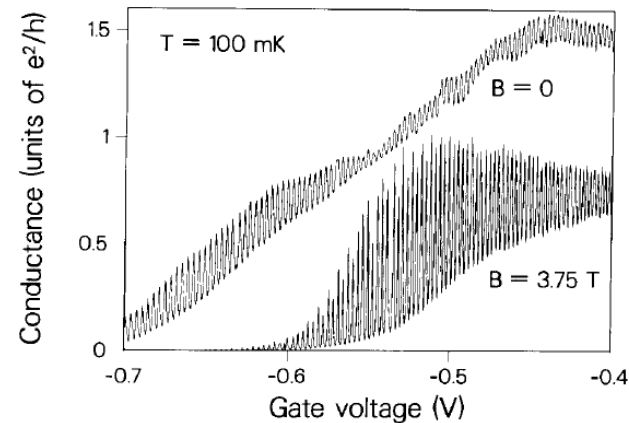
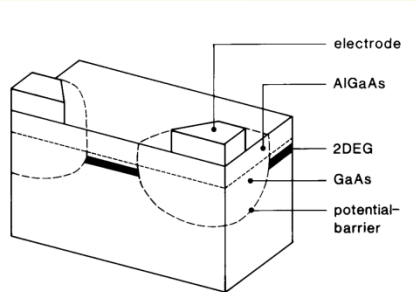
Regensburg - 19.07.2013

Single electron transistor in 2DEG structures

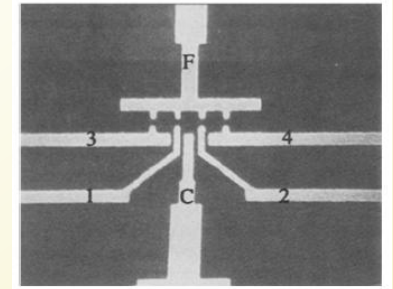
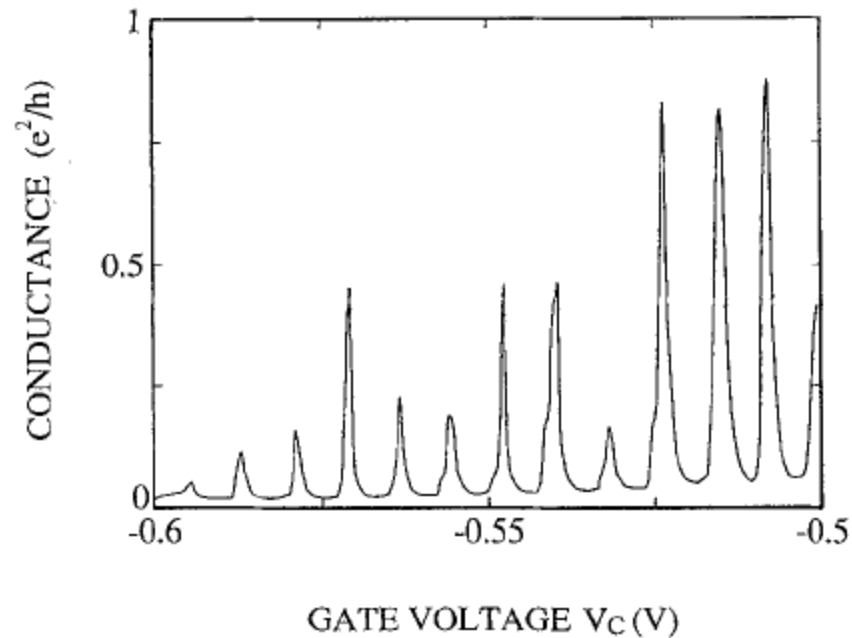
2 dimensional electron gas (2DEG)



Depleting top metal gates



Coulomb oscillations

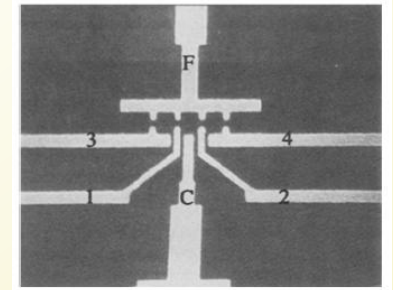
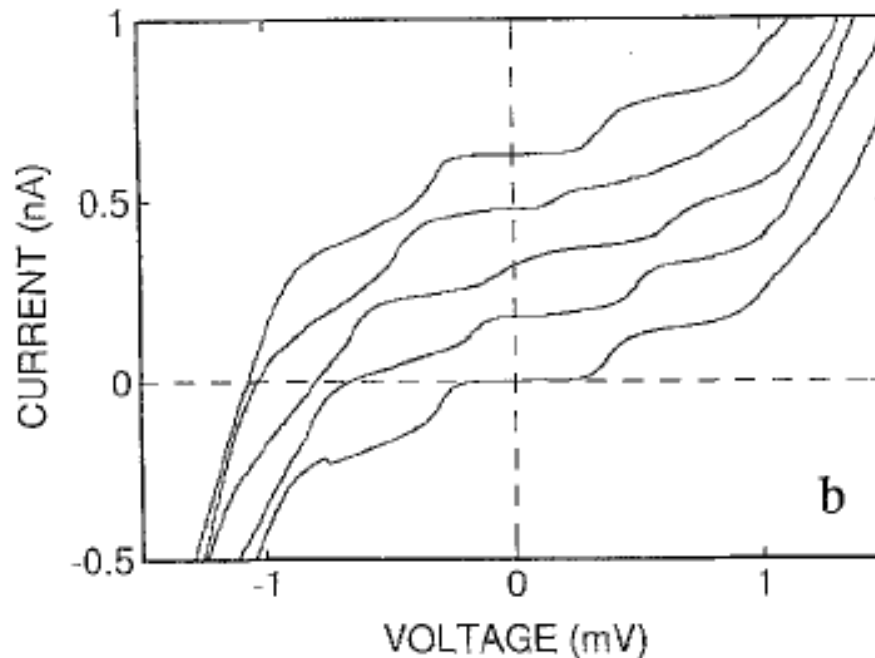


$d = 600$ nm
 $C = 0.28$ fF
 $\Delta E = 0.6$ meV
 $T = 10$ mK

Kouwenhoven *et al.*, Z. Phys. B Condensed Matter **85**, 367 (1991)

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Coulomb staircase

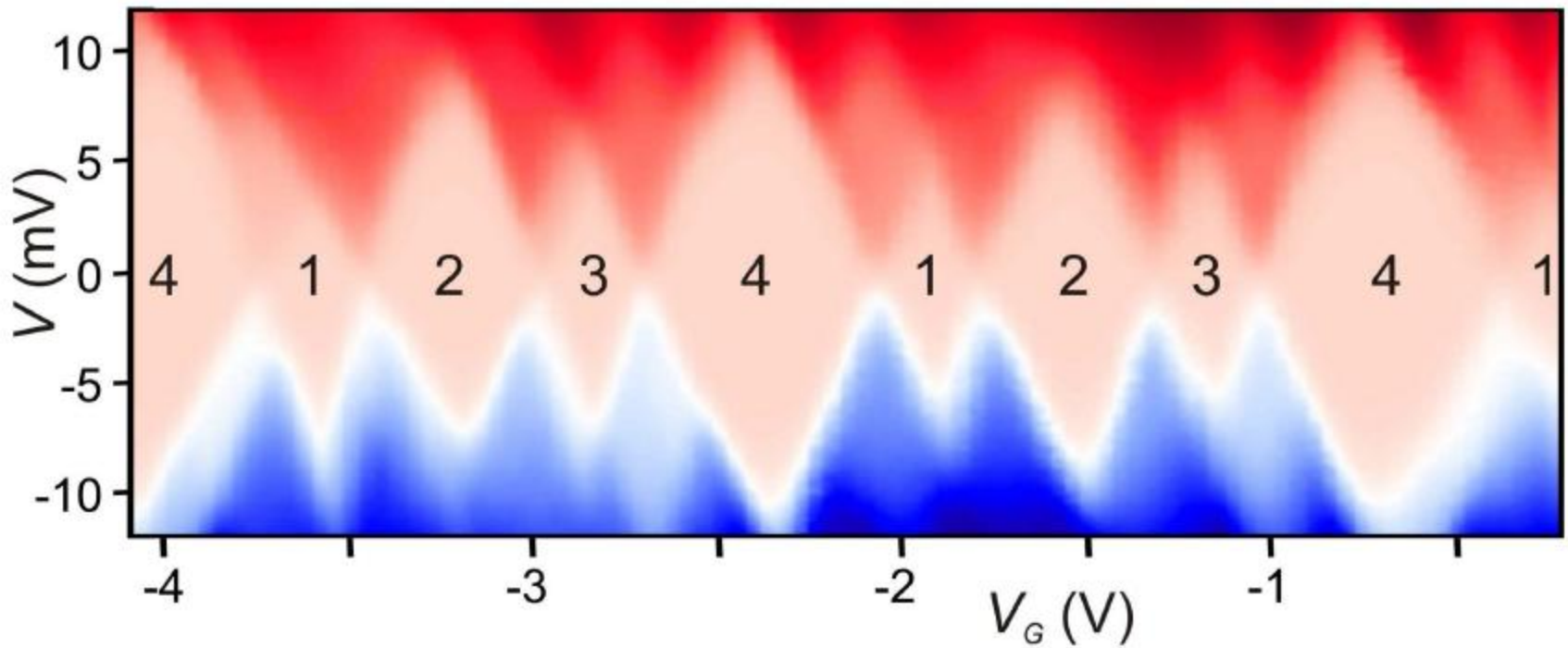
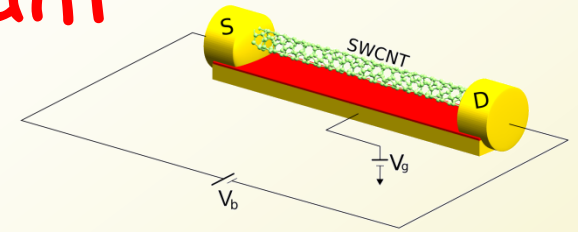


$d = 600 \text{ nm}$
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Kouwenhoven *et al.*, Z. Phys. B Condensed Matter **85**, 367 (1991)

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Stability diagram



H. van der Zant, unpublished

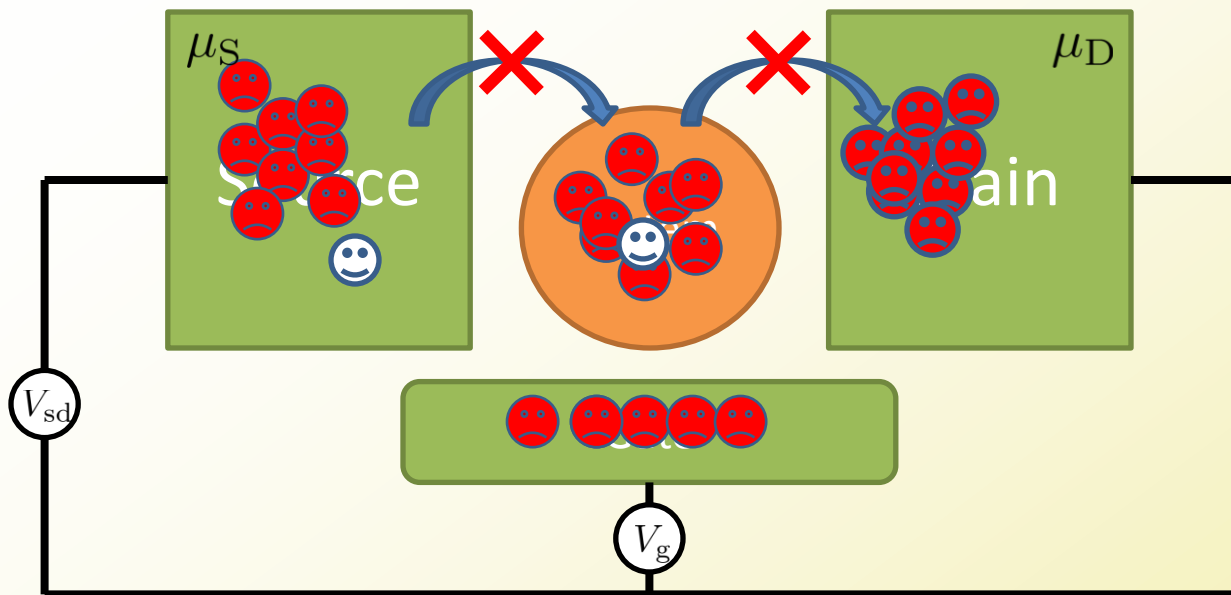
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GRK

Tutorial

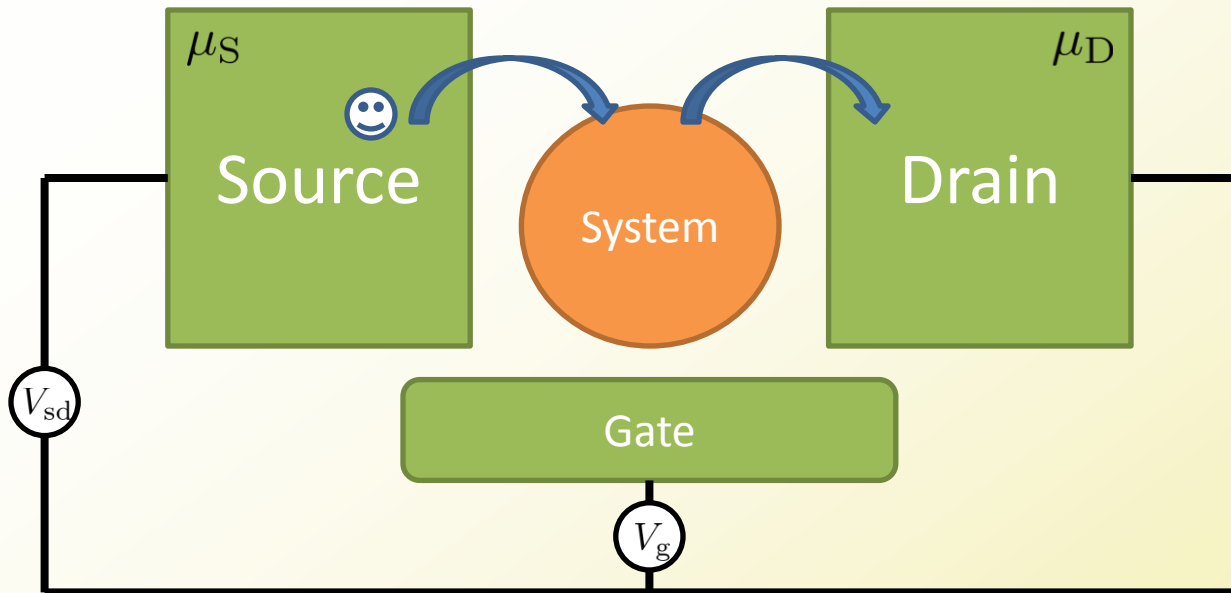
Coulomb blockade

$$\mu_S > \mu_D$$



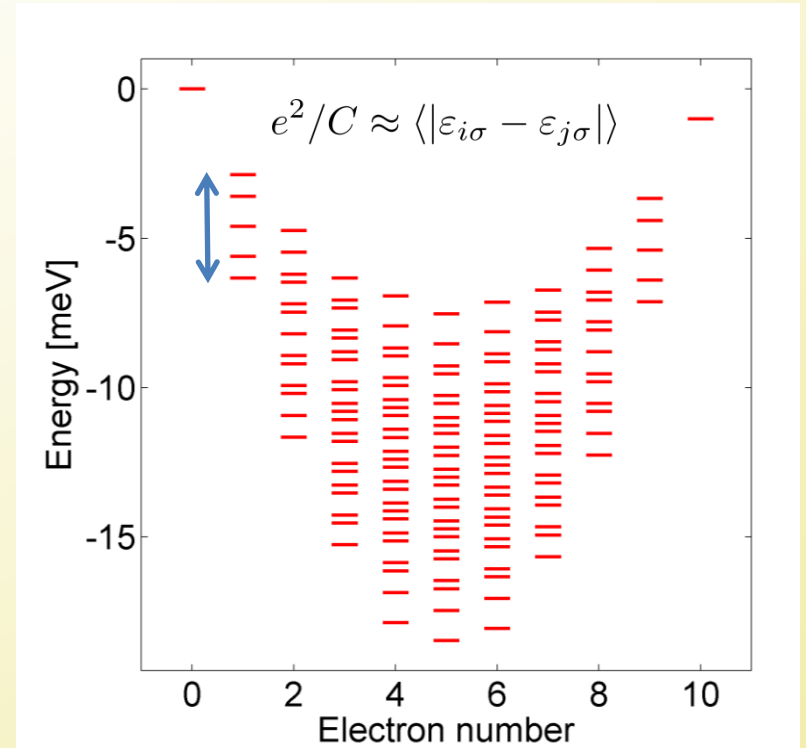
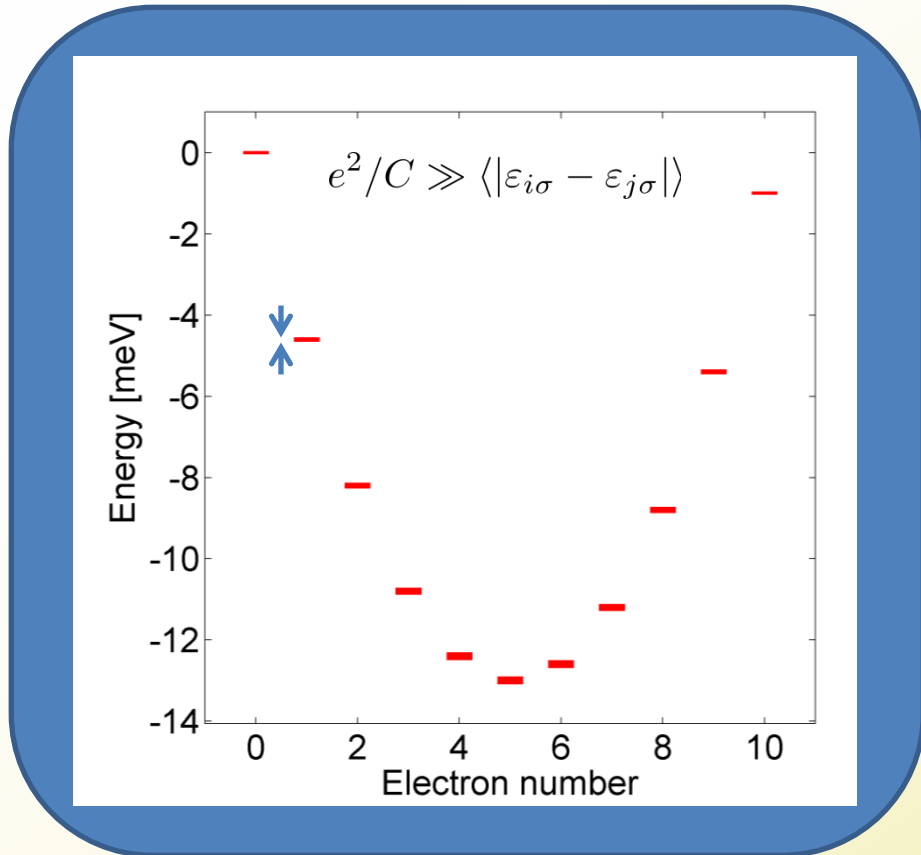
Single electron tunnelling

$$\mu_S > \mu_D$$



Discrete vs. Continuum

$$\hat{H}_{\text{sys}} = \sum_{i\sigma} \varepsilon_{i\sigma} \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} + \frac{e^2}{C} \frac{\hat{N}(\hat{N} - 1)}{2} - \hat{N} e V_g \quad \hat{N} = \sum_{i\sigma} \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma}$$



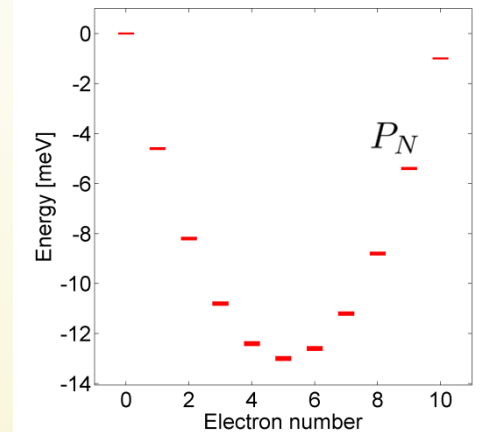
Master equation

- The only relevant quantity is the probability that the system is occupied with N electrons.

$$\dot{P}_N = -(\Gamma^{N+1,N} + \Gamma^{N-1,N})P_N + \Gamma^{N,N+1}P_{N+1} + \Gamma^{N,N-1}P_{N-1}$$

$$\Gamma^{N\pm 1,N} = \Gamma_S^{N\pm 1,N} + \Gamma_D^{N\pm 1,N}$$

- The system relaxes to “local” thermal equilibrium after each tunneling event



$$\Gamma_\alpha^{N+1,N} = \gamma_\alpha \int d\varepsilon_\alpha d\varepsilon_{\text{sys}} \underbrace{f^+(\varepsilon_\alpha - \mu_\alpha)}_{\text{lead-level occupation}} \underbrace{f^-(\varepsilon_{\text{sys}} - \mu_{\text{sys}})}_{\text{system-level occupation}} \underbrace{\delta(Ne^2/C - eV_g + \varepsilon_{\text{sys}} - \varepsilon_\alpha)}_{\text{energy conservation}}$$

$$\gamma_\alpha = \frac{2\pi}{\hbar} |t|^2 D_\alpha D_{\text{sys}}$$

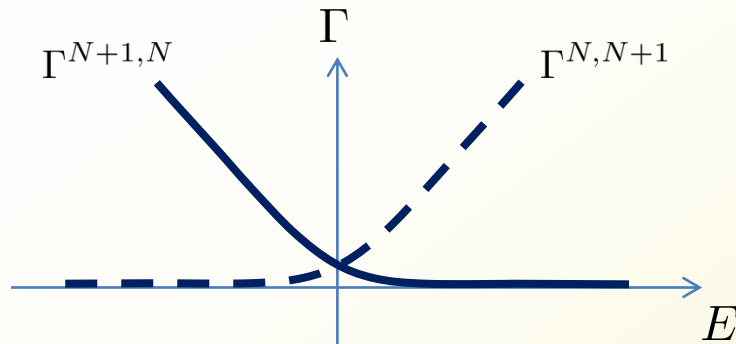
$$\Gamma_\alpha^{N+1,N} = \gamma_\alpha \frac{(Ne^2/C - eV_g + \mu_{\text{sys}} - \mu_\alpha)}{\exp[\beta(Ne^2/C - eV_g + \mu_{\text{sys}} - \mu_\alpha)] - 1}$$

Let us assume

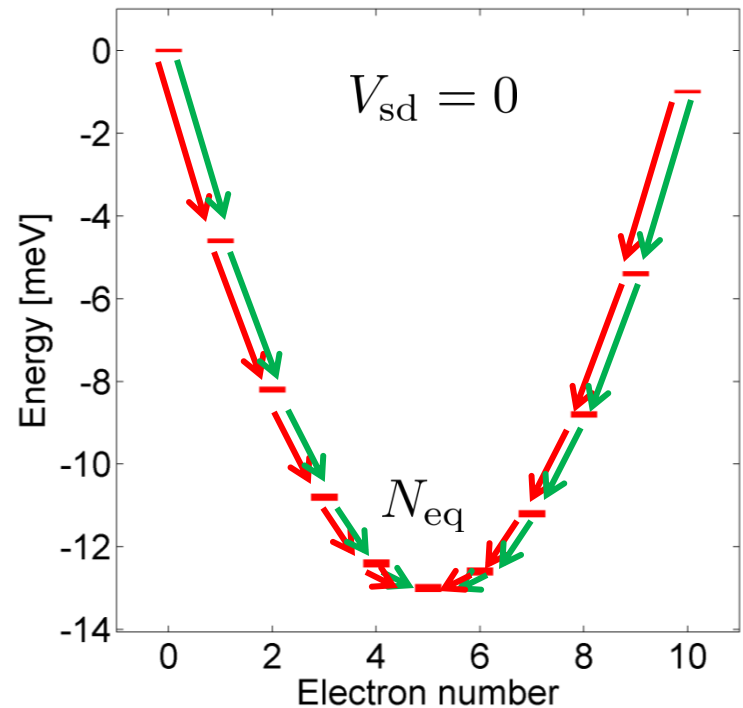
$$\mu_{S/D} = \mu_{\text{sys}} \mp \frac{eV_{\text{sd}}}{2}$$

Orthodox theory

$$\Gamma_{S/D}^{N+1,N} = \gamma_\alpha \frac{+(Ne^2/C - eV_g \pm eV_{sd}/2)}{\exp[+\beta(Ne^2/C - eV_g + \mu_{\text{sys}} - \mu_\alpha)] - 1}$$



$$\Gamma_{S/D}^{N,N+1} = \gamma_\alpha \frac{-(Ne^2/C - eV_g \pm eV_{sd}/2)}{\exp[-\beta(Ne^2/C - eV_g + \mu_{\text{sys}} - \mu_\alpha)] - 1}$$

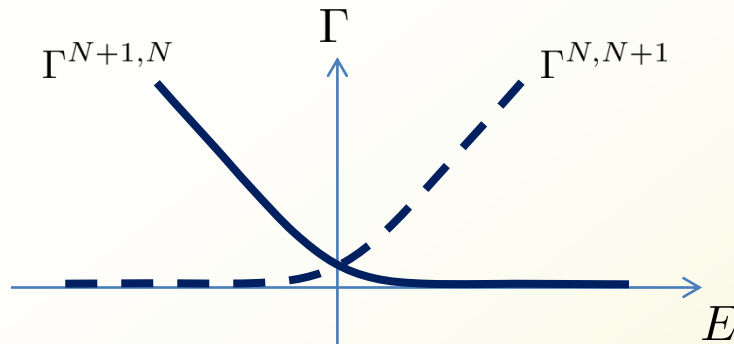


$$N_{\text{eq}} \approx V_g C / e$$

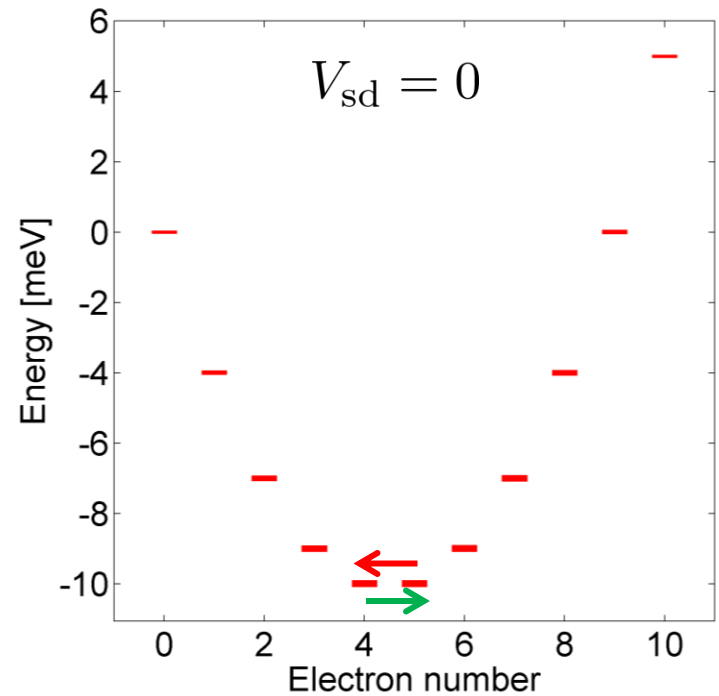
Particle number controlled by the gate

Orthodox theory

$$\Gamma_{S/D}^{N+1,N} = \gamma_\alpha \frac{+(Ne^2/C - eV_g \pm eV_{sd}/2)}{\exp[+\beta(Ne^2/C - eV_g + \mu_{sys} - \mu_\alpha)] - 1}$$



$$\Gamma_{S/D}^{N,N+1} = \gamma_\alpha \frac{-(Ne^2/C - eV_g \pm eV_{sd}/2)}{\exp[-\beta(Ne^2/C - eV_g + \mu_{sys} - \mu_\alpha)] - 1}$$



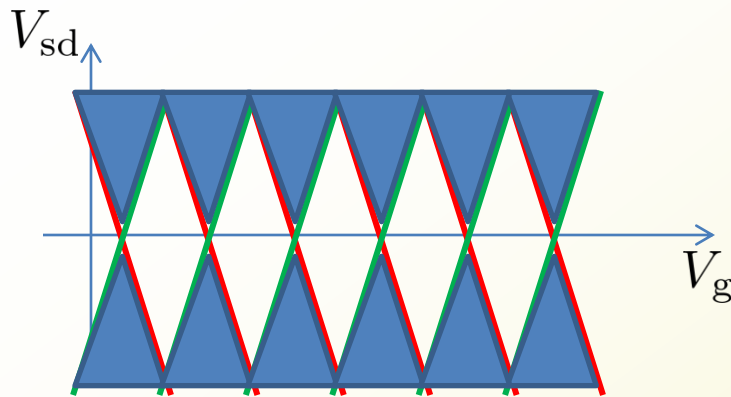
$$V_g = (N + 1/2) \frac{e}{C}$$

Conductance peak => Coulomb oscillations

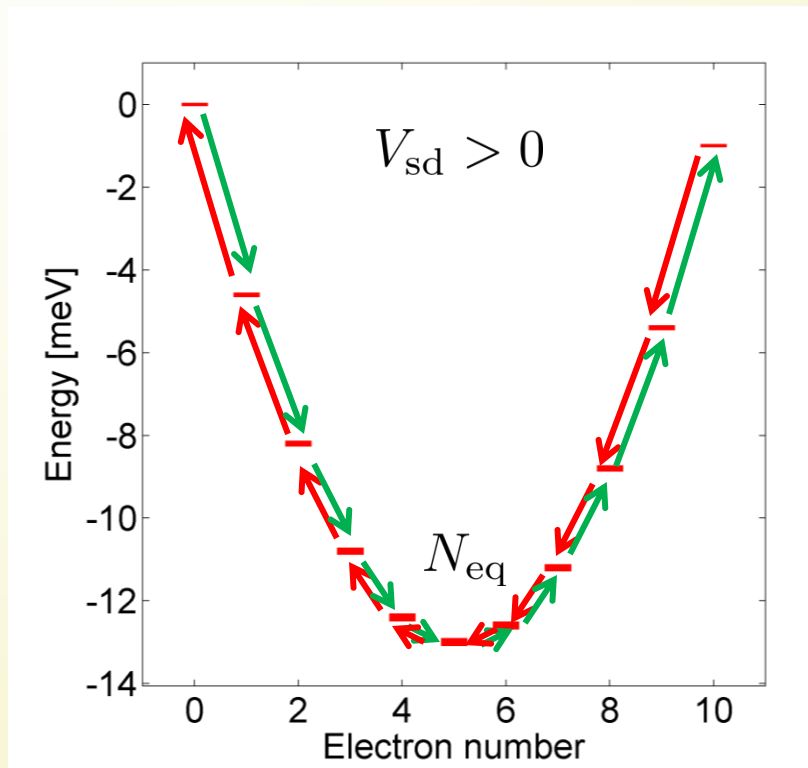
Orthodox theory

$V_{sd} > +2V_g + 2Ne/C$ Source $N \rightarrow N+1$
Resonance condition

$V_{sd} > -2V_g + 2Ne/C$ Drain $N \rightarrow N-1$
Resonance condition

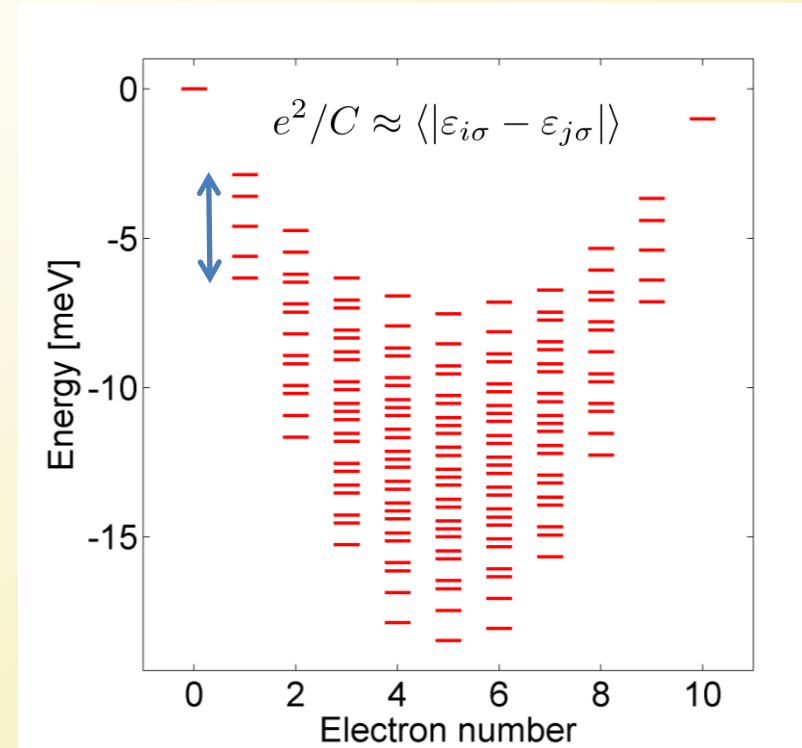
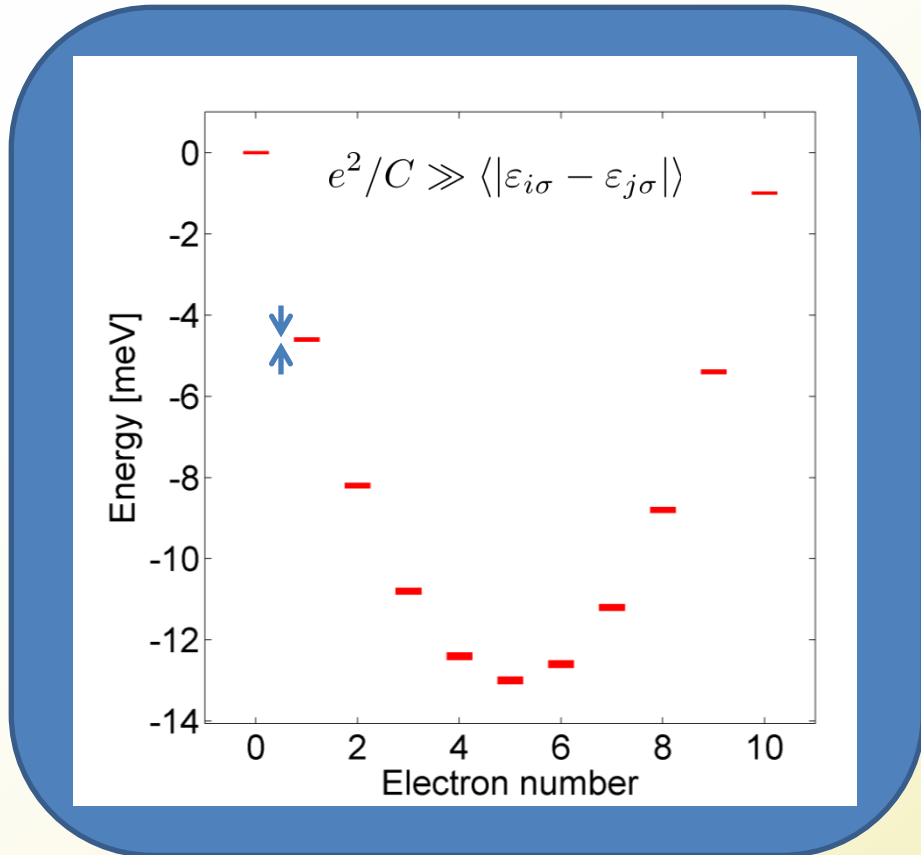


Coulomb diamonds



Discrete vs. Continuum

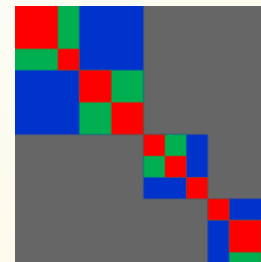
$$\hat{H}_{\text{sys}} = \sum_{i\sigma} \varepsilon_{i\sigma} \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} + \frac{e^2}{C} \frac{\hat{N}(\hat{N} - 1)}{2} - \hat{N}eV_g \quad \hat{N} = \sum_{i\sigma} \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma}$$



Generalized Master Equation

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

- We define the reduced density matrix $\sigma = \text{Tr}_{S+T}\{\rho\}$ which is **block-diagonal** in

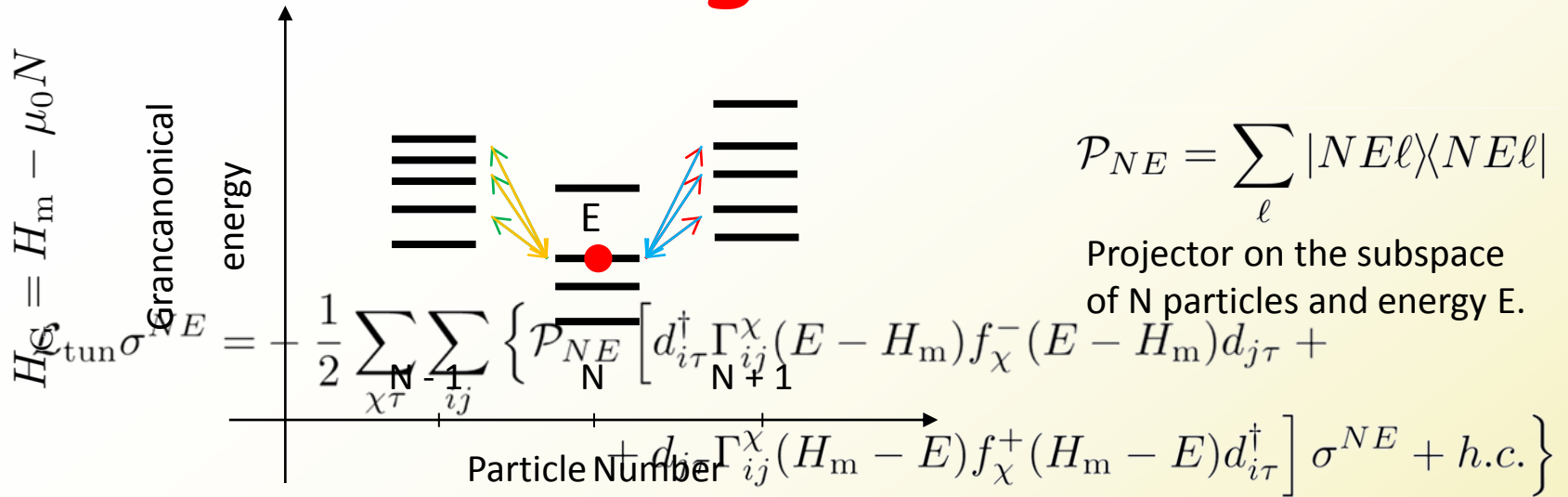


particle number
spin
energy

- We keep the coherences between **orbitally** degenerate states.
- The **Generalized Master Equation** is the equation of motion for σ :

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

Tunnelling Liouvillean



$$+ \sum_{\chi\tau} \sum_{ijE'} \mathcal{P}_{NE} \left[d_{i\tau}^\dagger \Gamma_{ij}^\chi(E - E') \sigma^{N-1E'} f_\chi^+(E - E') d_{j\tau} + d_{j\tau} \Gamma_{ij}^\chi(E' - E) \sigma^{N+1E'} f_\chi^-(E' - E) d_{i\tau}^\dagger \right] \mathcal{P}_{NE}$$

Single particle rate matrix

$$\Gamma_{ij}^{\chi}(\Delta E) = \frac{2\pi}{\hbar} \sum_k (t_{ki}^{\chi})^* t_{kj}^{\chi} \delta(\varepsilon_k^{\chi} - \Delta E)$$

$$H_{\text{eff}} = \frac{1}{2\pi} \sum_{NE} \sum_{\chi\sigma} \sum_{ij} \mathcal{P}_{NE} \left[d_{i\sigma}^{\dagger} \Gamma_{ij}^{\chi}(E - H_m) p_{\chi}(E - H_m) d_{j\sigma} \right. \\ \left. + d_{j\sigma} \Gamma_{ij}^{\chi}(H_m - E) p_{\chi}(H_m - E) d_{i\sigma}^{\dagger} \right] \mathcal{P}_{NE}$$

Effective
Hamiltonian

$$I_{\chi} = \sum_{NE\sigma ij} \mathcal{P}_{NE} \left[d_{j\sigma} \Gamma_{ij}^{\chi}(H_m - E) f_{\chi}^{+}(H_m - E) d_{i\sigma}^{\dagger} \right. \\ \left. - d_{i\sigma}^{\dagger} \Gamma_{ij}^{\chi}(E - H_m) f_{\chi}^{-}(E - H_m) d_{j\sigma} \right] \mathcal{P}_{NE}$$

Current
operator

Many-body rate matrix

The **current** is proportional to the **transition rate** between **many-body states**

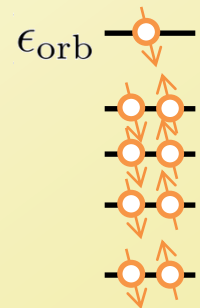
$$R_{N E_0 \rightarrow N+1 E_1}^{\chi\tau} = \sum_{ij} \langle N+1 E_1 | d_{i\tau}^\dagger | N E_0 \rangle \Gamma_{ij}^\chi(E_1 - E_0) \times \langle N E_0 | d_{j\tau} | N+1 E_1 \rangle f^+(E_1 - E_0 - \mu_\chi)$$

where

$$\Gamma_{ij}^\chi(E_1 - E_0) = \frac{2\pi}{\hbar} \sum_k (t_{ki}^\chi)^* t_{kj}^\chi \delta(\epsilon_k^\chi - E_1 + E_0)$$

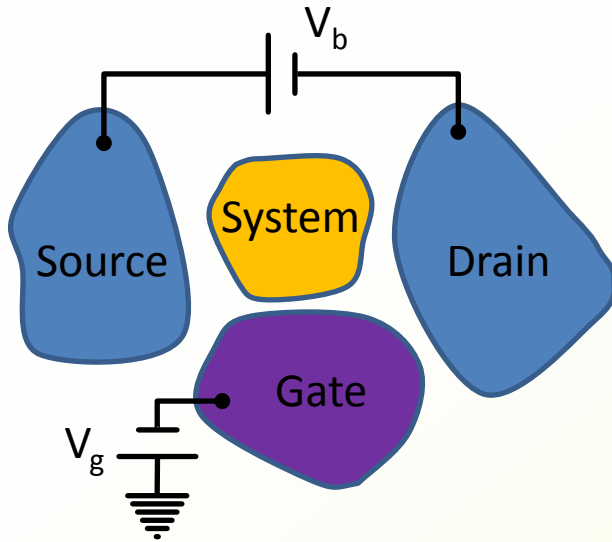
For **uncorrelated** and **non-degenerate systems** the many-body rate reduces to

$$R_{N E_0 \rightarrow N+1 E_1}^{\chi\tau} = \Gamma_{\text{orb}}^\chi(\epsilon_{\text{orb}}) f^+(\epsilon_{\text{orb}} - \mu_\chi)$$

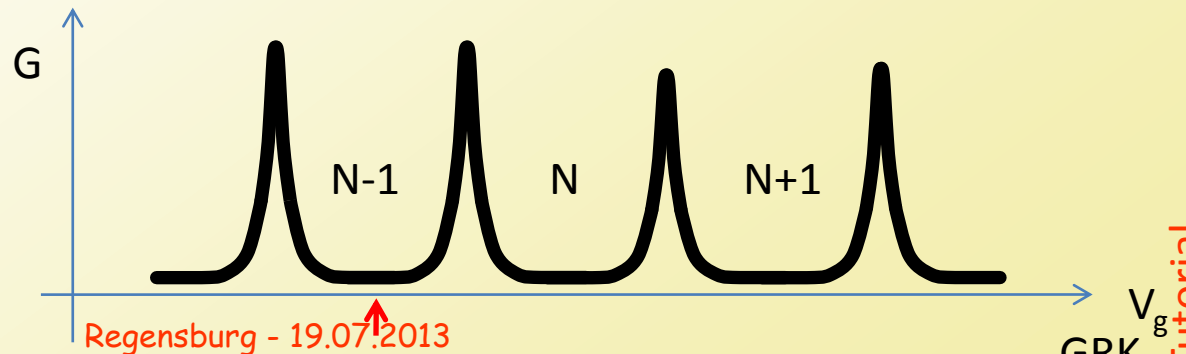
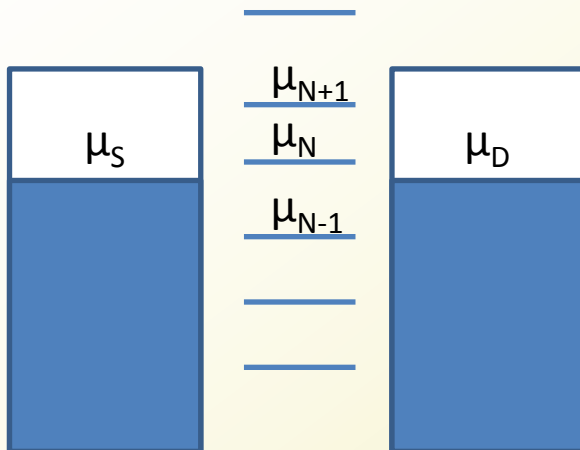
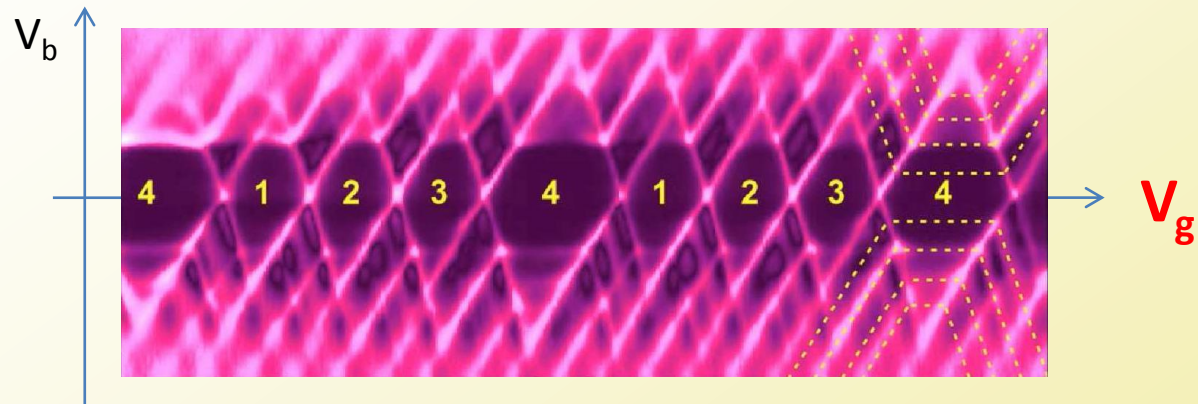


Negative differential conductance (NDC) with symmetric contacts

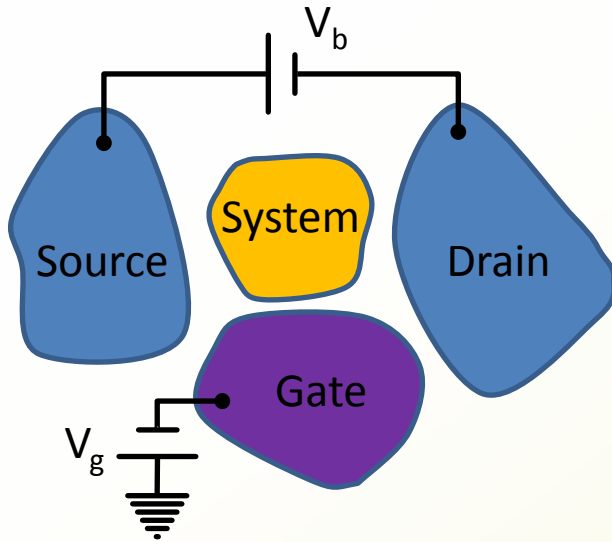
Single electron transistor



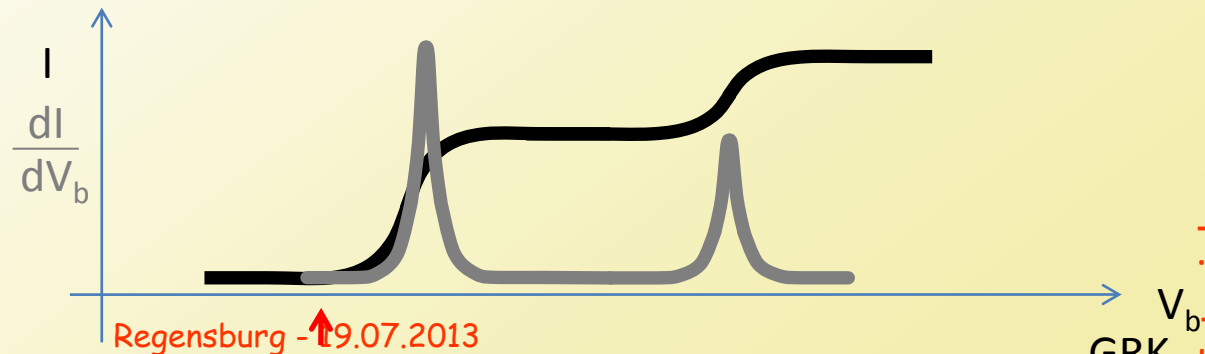
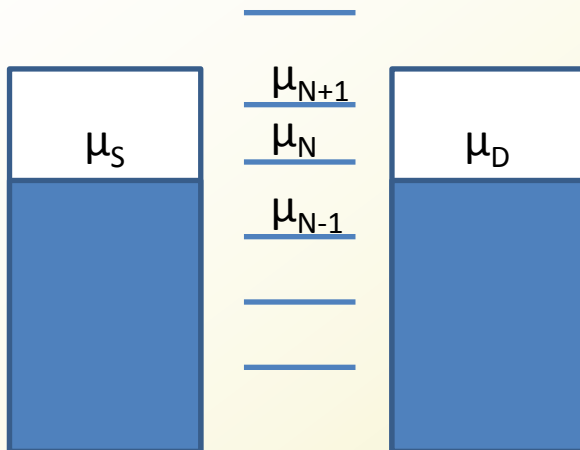
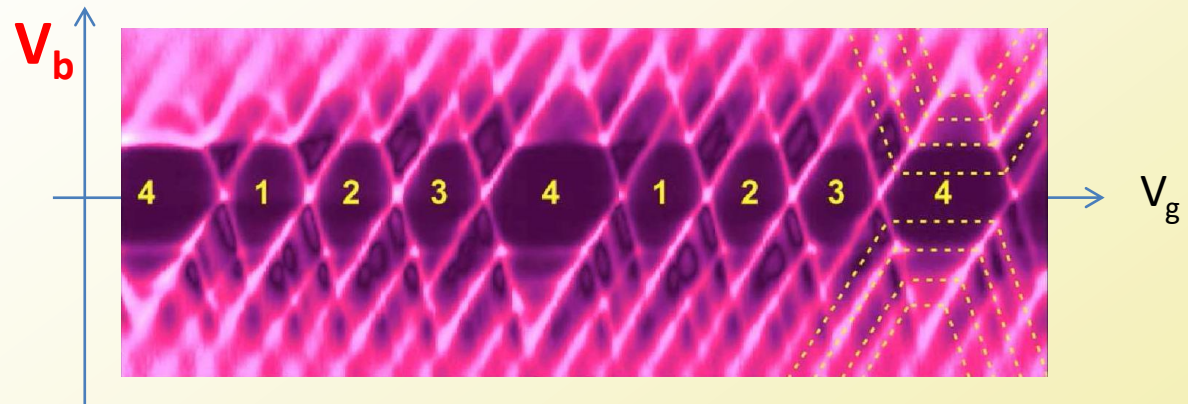
Small System size + weak System-Lead Tunnelling coupling \rightarrow Strong e-e interaction on the System \rightarrow Single electron control



Single electron transistor

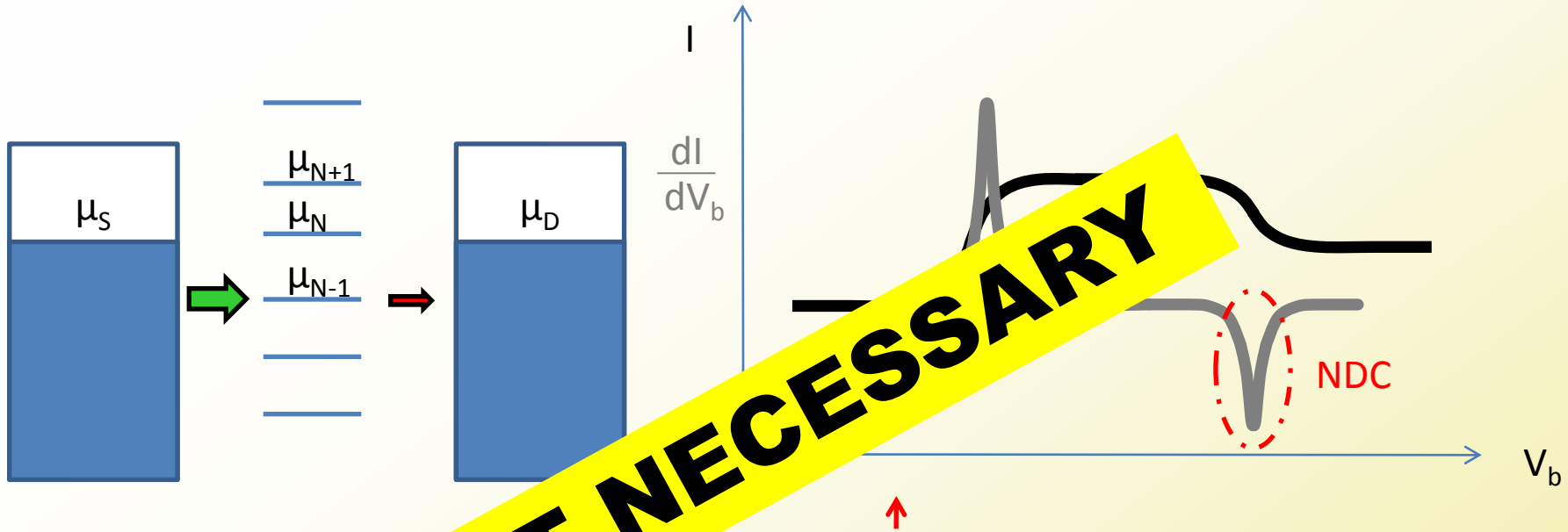


Small System size + weak System-Lead Tunnelling coupling \rightarrow Strong e-e interaction on the System \rightarrow Single electron control



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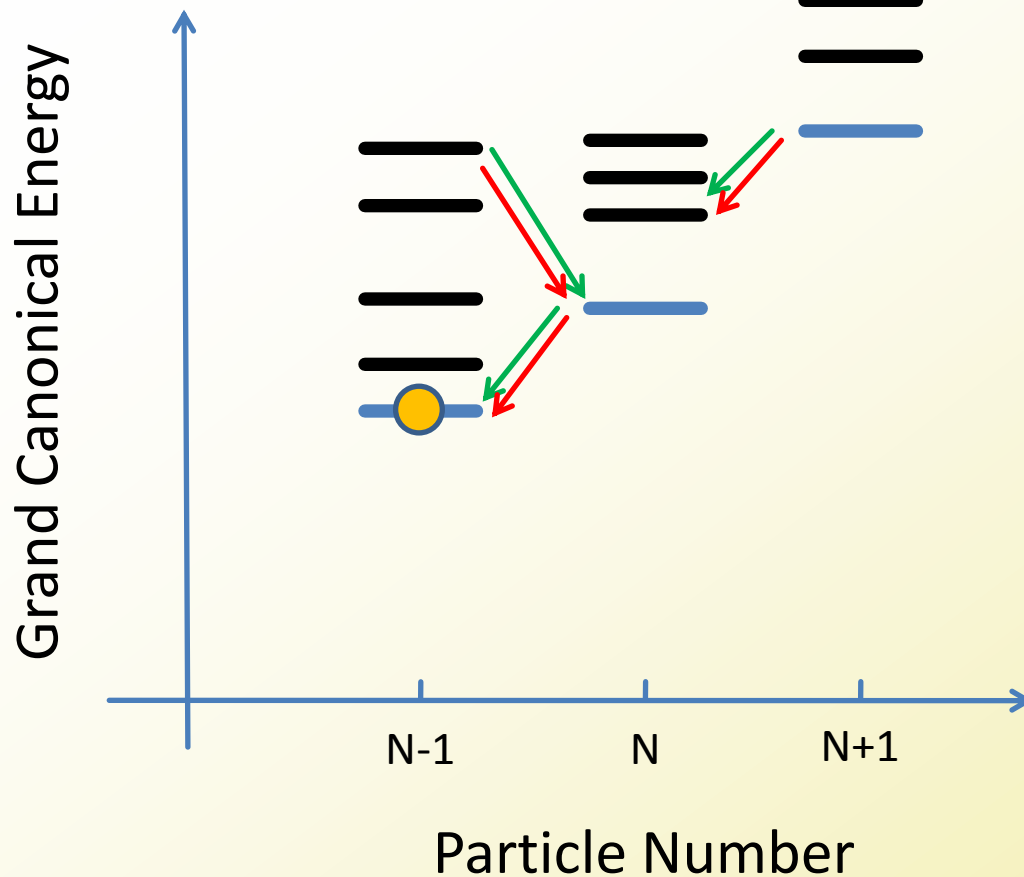
Negative Differential Conductance



NOT NECESSARY

Negative differential conductance (NDC) is usually associated with a **strong asymmetry** in the coupling to the leads

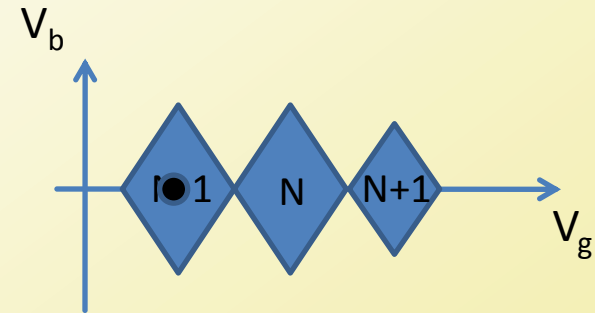
The "free energy" formulation



$$F = H - \mu_0 N \quad \text{GC energy}$$

→ Source transition

→ Drain transition



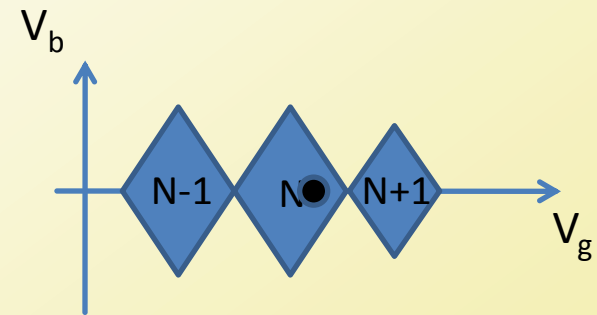
AD, G Begemann, Milena Grifoni
PRB, **82**, 125451 (2010)

The "free energy" formulation

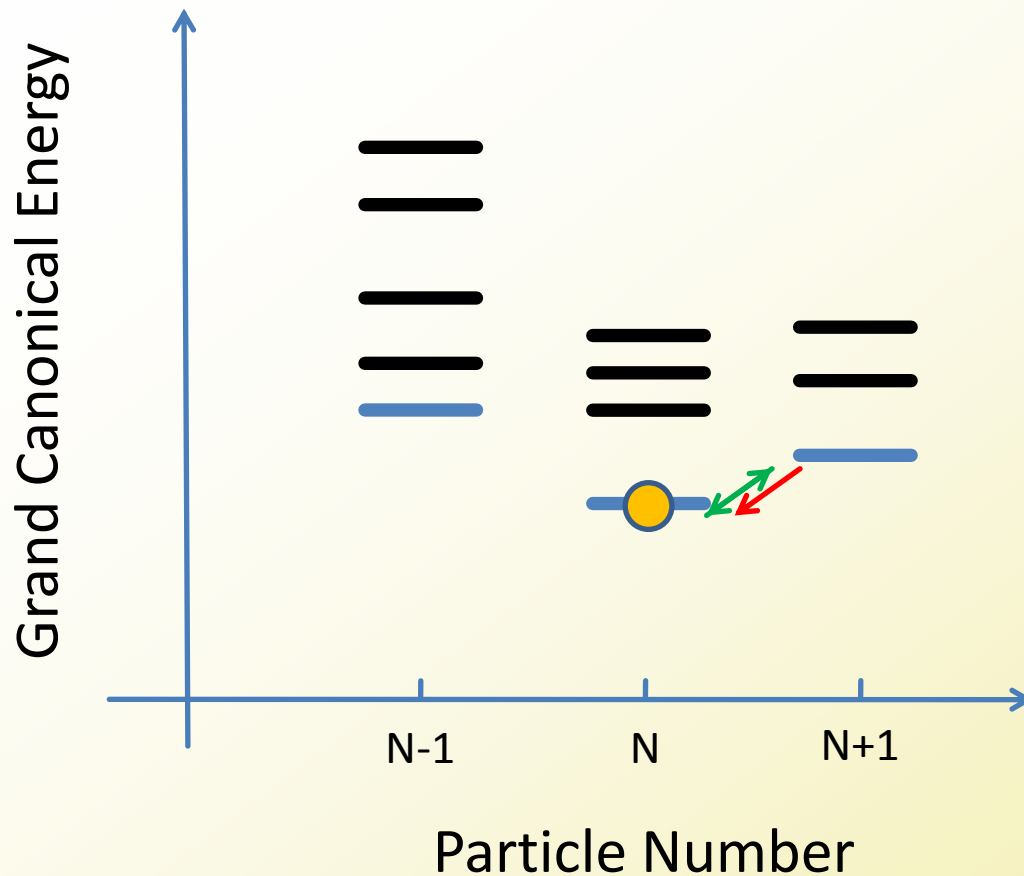
$$F = H - \mu_0 N \quad \text{GC energy}$$

→ Source transition

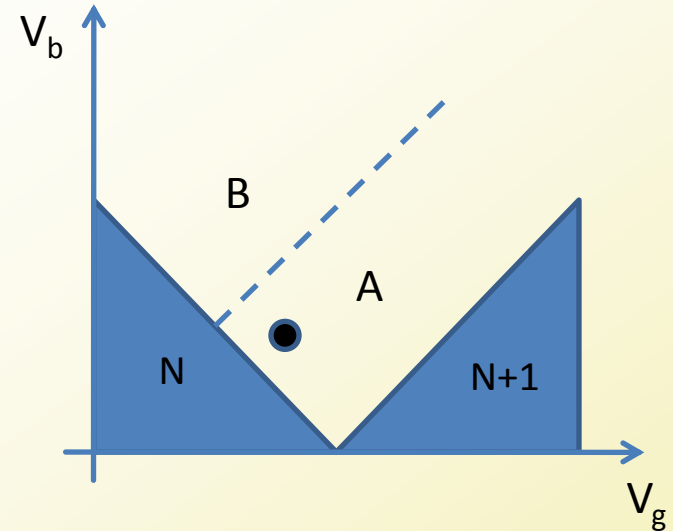
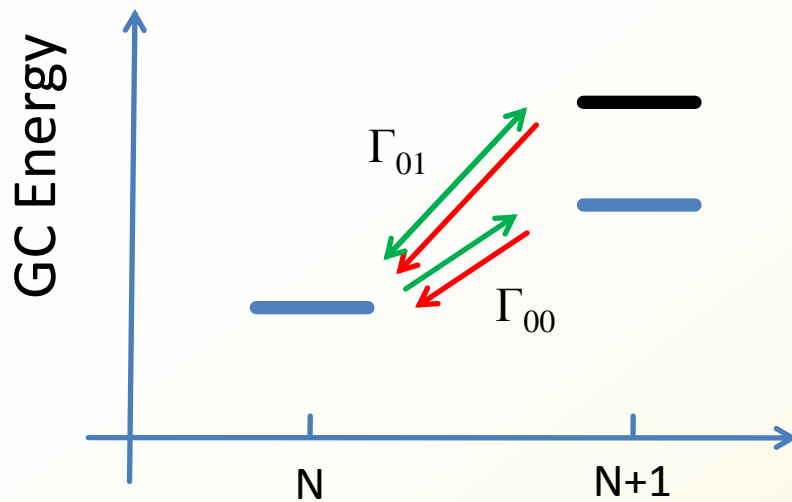
→ Drain transition



AD, G Begemann, Milena Grifoni
PRB, **82**, 125451 (2010)



NDC with symmetric set-up



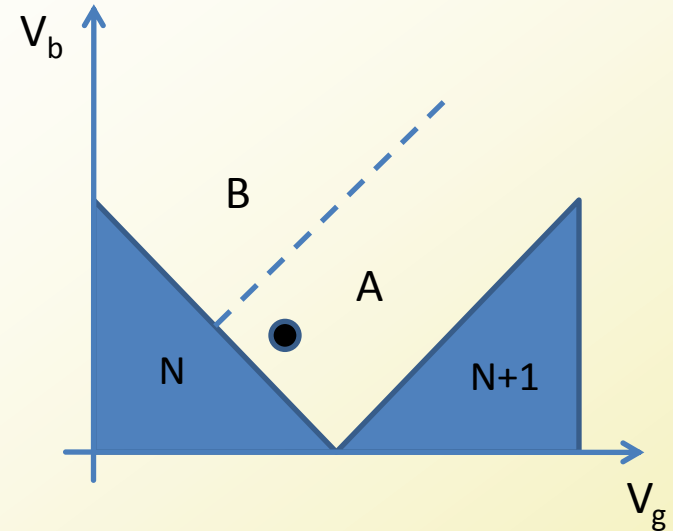
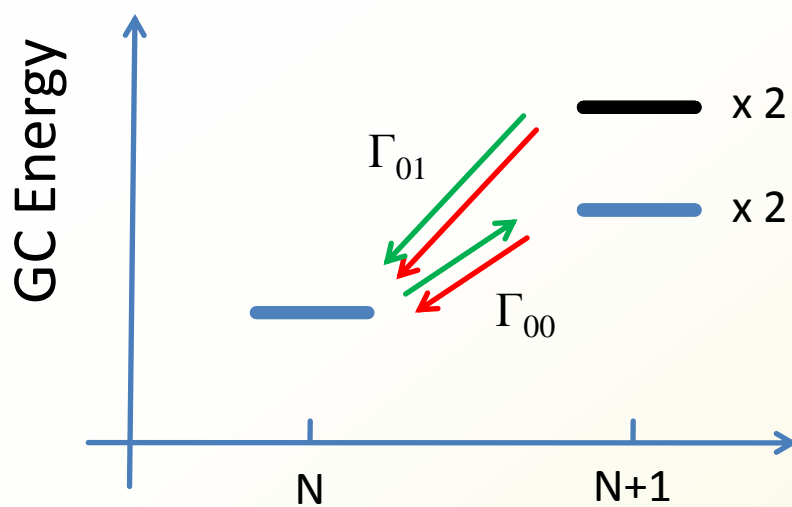
The dynamics is evaluated with the **master equation**: in the **stationary limit**

	$P_{N,0}$	$P_{N+1,0}$	$P_{N+1,1}$	Current
A	1/2	1/2	0	$\Gamma_{00}/2$
B	1/3	1/3	1/3	$1/3(\Gamma_{00} + \Gamma_{01})$



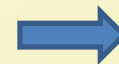
NDC if $I_B < I_A$
 $\Gamma_{01} < \Gamma_{00}/2$

Role of the degeneracies



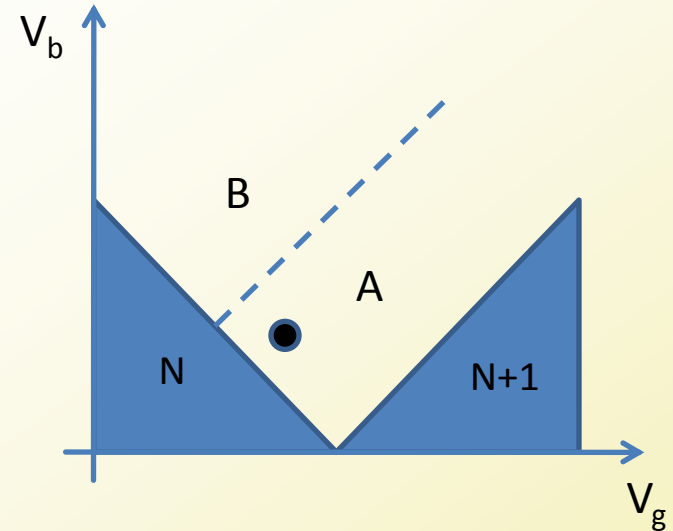
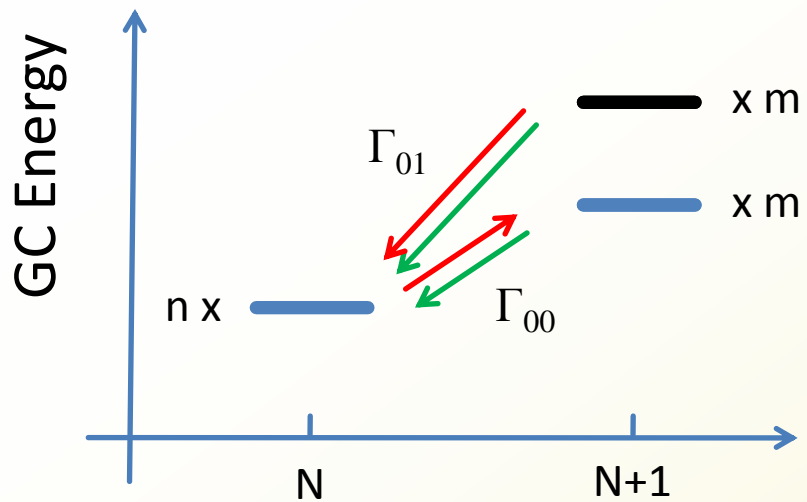
The dynamics is evaluated with the **master equation**: in the **stationary limit**

	$P_{N,0}$	$P_{N+1,0}$	$P_{N+1,1}$	Current
A	1/3	1/3	0	$2\Gamma_{00}/3$
B	1/5	1/5	1/5	$2/5(\Gamma_{00} + \Gamma_{01})$



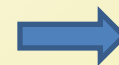
NDC if $I_B < I_A$
 $\Gamma_{01} < 2\Gamma_{00}/3$

Role of the degeneracies



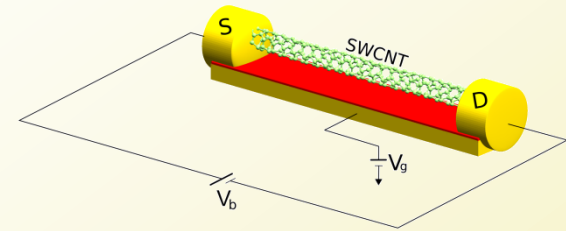
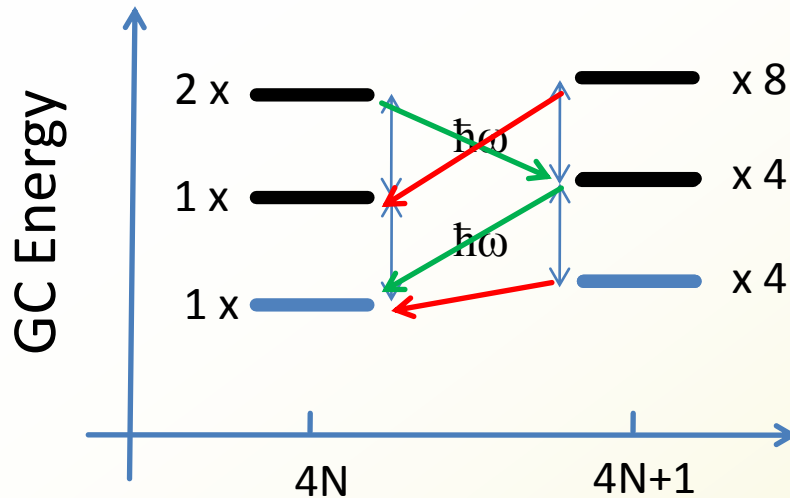
The dynamics is evaluated with the **master equation**: in the **stationary limit**

	$P_{N,0}$	$P_{N+1,0}$	$P_{N+1,1}$	Current
A	$\frac{1}{n+m}$	$\frac{1}{n+m}$	0	$\frac{nm}{n+m} \Gamma_{00}$
B	$\frac{1}{n+2m}$	$\frac{1}{n+2m}$	$\frac{1}{n+2m}$	$\frac{nm}{n+2m} (\Gamma_{00} + \Gamma_{01})$



NDC if $I_B < I_A$
 $\Gamma_{01} < \frac{m}{n+m} \Gamma_{00}$

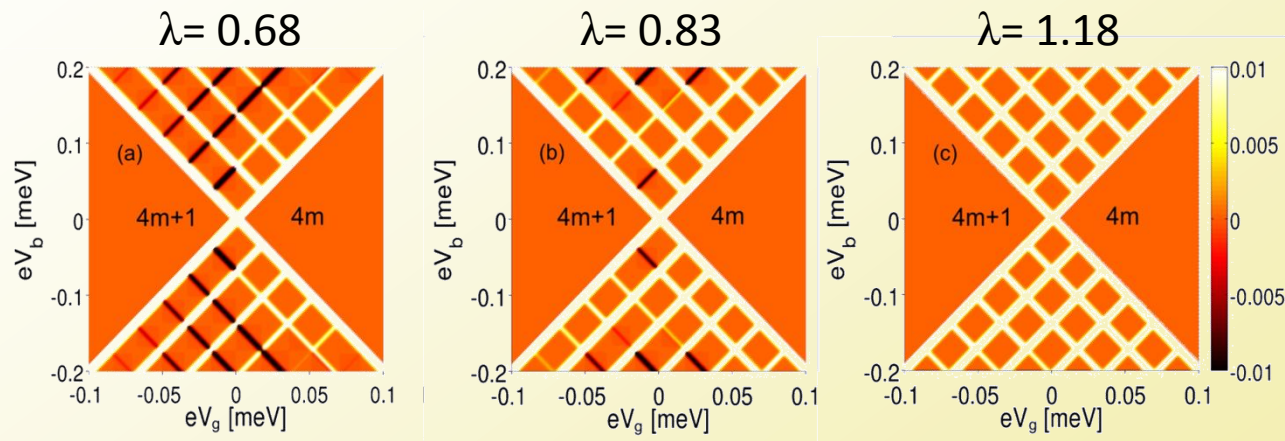
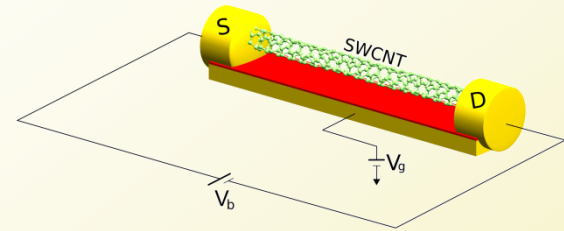
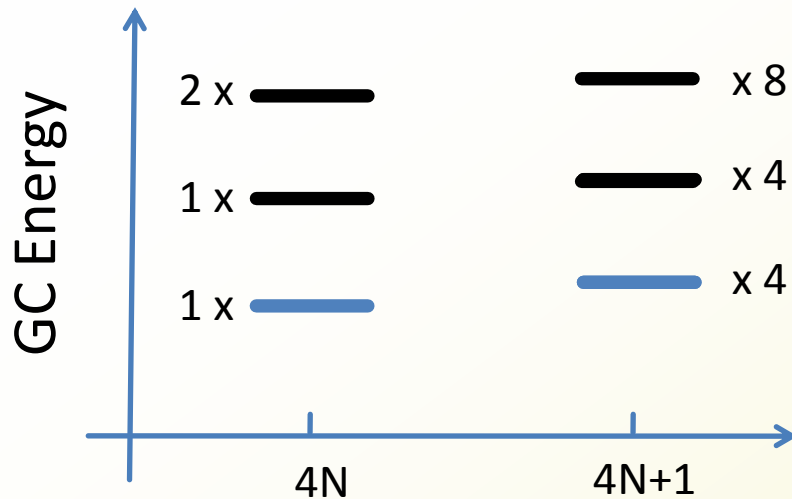
An example: a suspended CNT



The transition rates are proportional to product of Frack-Condon coefficients

$$\Gamma_{ij} = \prod_n FC^{(n)}(i, j; \lambda)$$

An example: a suspended CNT

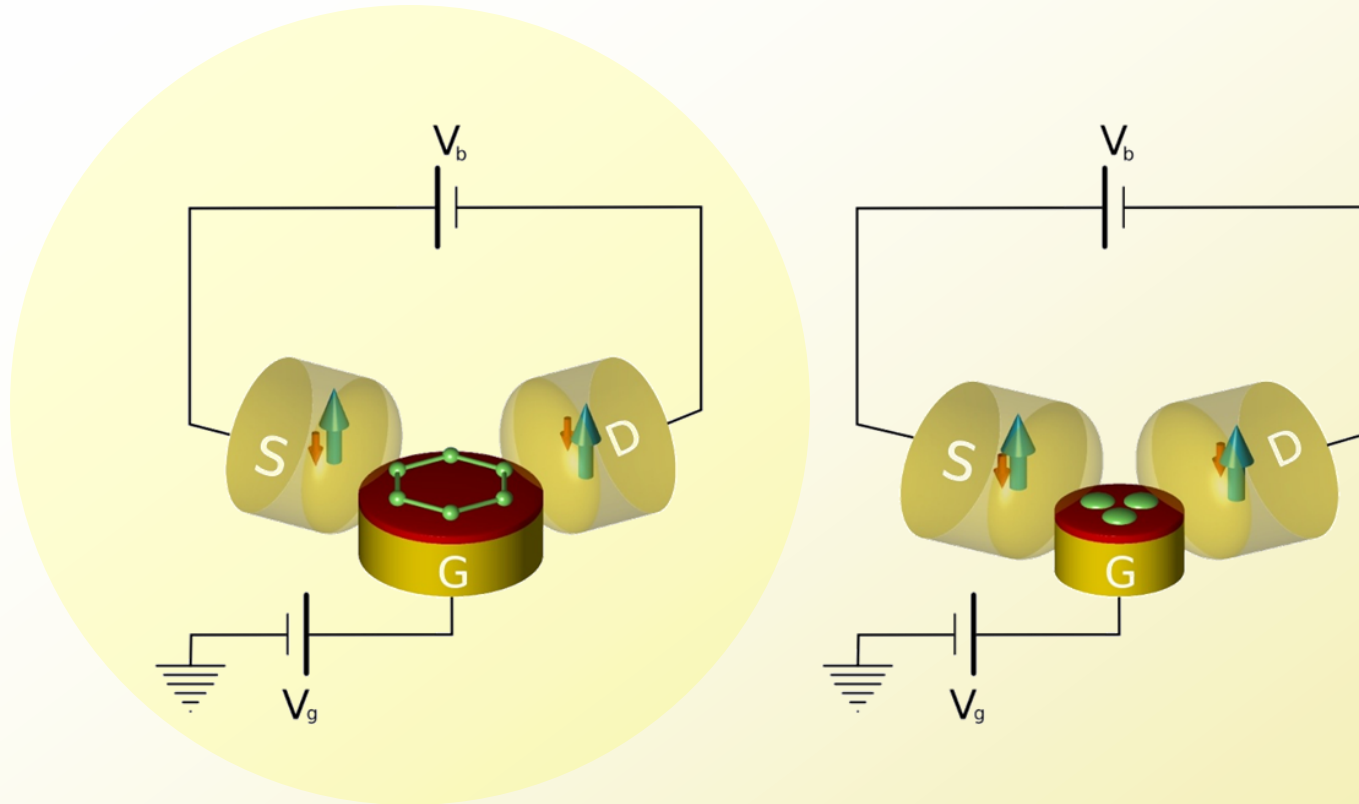


A. Yar, AD, S. Koller, and M. Grifoni, arXiv:1101.3892 (2011)

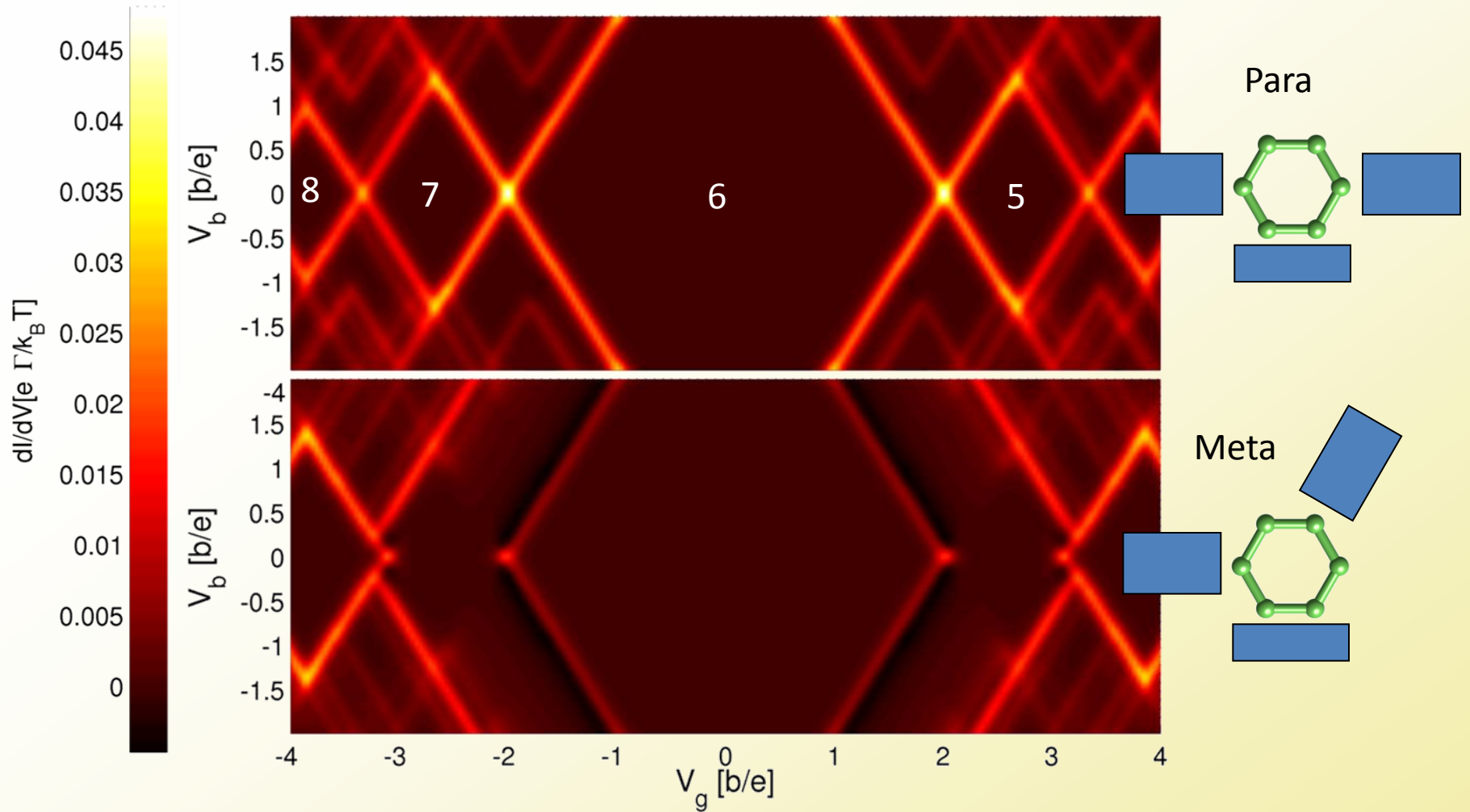
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Interference

Single Electron Transistors



Para vs. Meta



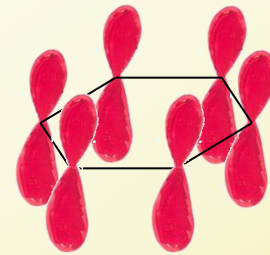
G. Begemann, D. Darau, **AD**, M. Grifoni, *Phys. Rev. B* **77**, 201406(R) (2008)

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Interacting isolated benzene

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

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



- 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

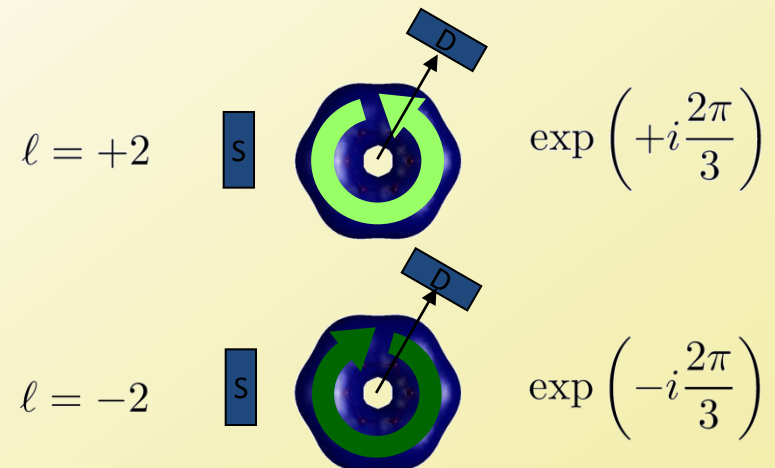
N	Degeneracy	GS energy[eV] (at $\xi = 0$)	GS symmetry representation
0	1	0	A_{1g}
1	2	-22	A_{2u}
2	1	-42.25	A_{1g}
3	4	-57.42	E_{1g}
4	3	-68.875	A_{2g}
5	4	-76.675	E_{1g}
6	1	-81.725	A_{1g}
7	4	-76.675	E_{2u}
8	3	-68.875	A_{2g}
9	4	-57.42	E_{2u}
10	1	-42.25	A_{1g}
11	2	-22	B_{2g}
12	1	0	A_{1g}

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

• $\mathcal{R}_\phi |7_g l\rangle = e^{-il\phi} |7_g l\rangle$ $l = \pm 2$



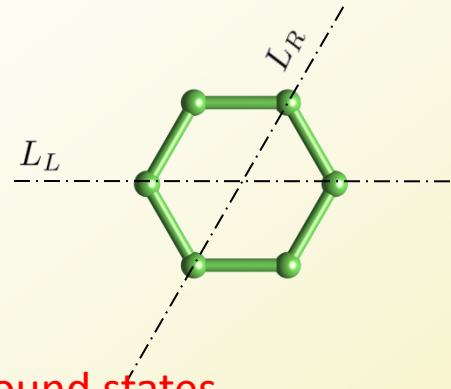
Generalized Master Equation

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

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**

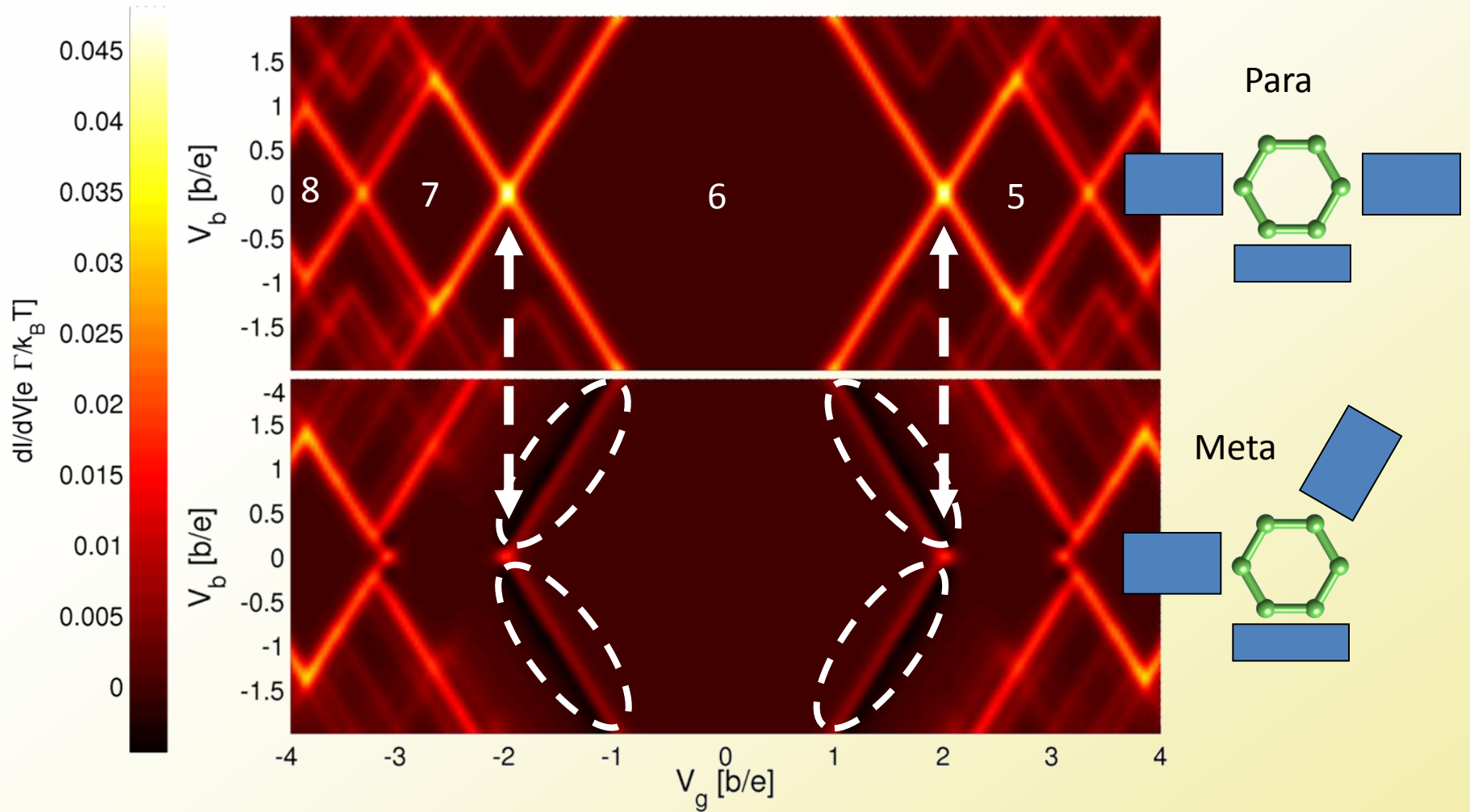
$$L_{\alpha} = \frac{\hbar}{2} \begin{pmatrix} 1 & e^{i2|\ell|\phi_{\alpha}} \\ e^{-i2|\ell|\phi_{\alpha}} & 1 \end{pmatrix}$$

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



Bias and gate dependent

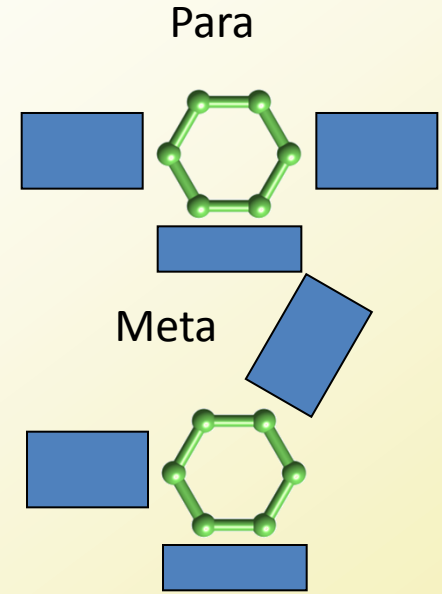
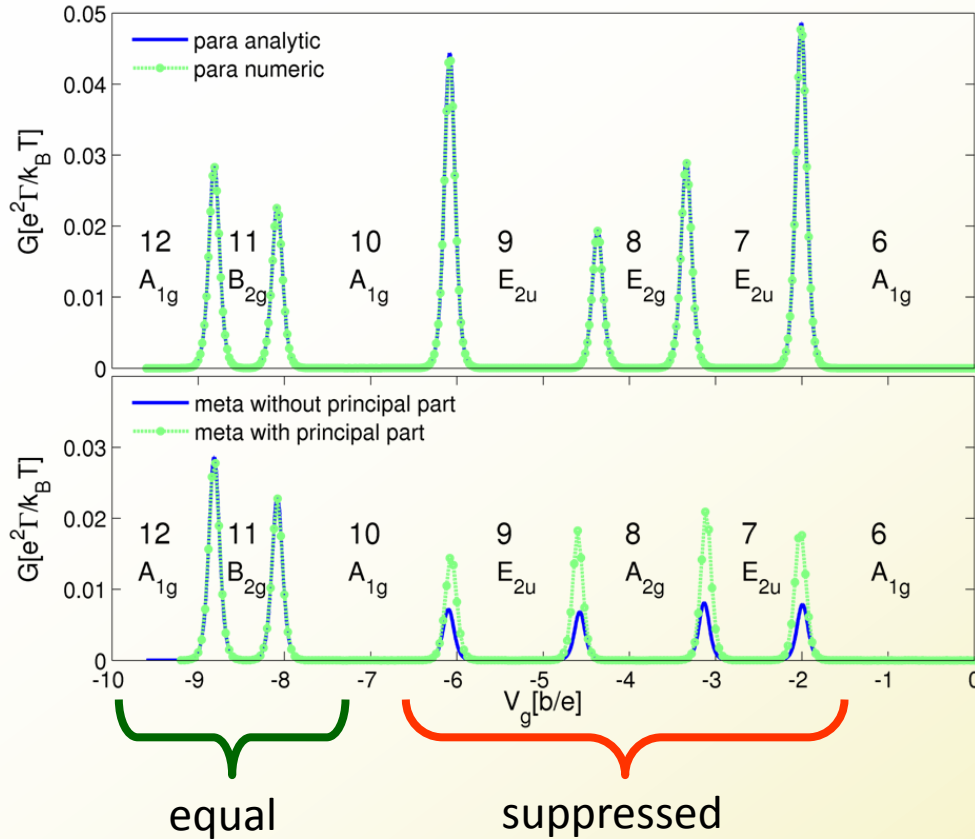
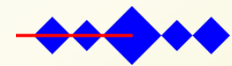
Para vs. Meta



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

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Conductance suppression



A: non-degenerate



B: non-degenerate



Equal

A: non-degenerate



E: degenerate



Suppressed

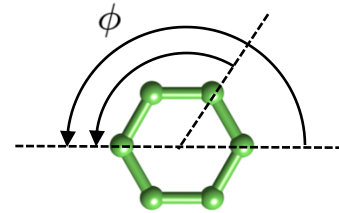
Destructive interference

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

Interference
factor

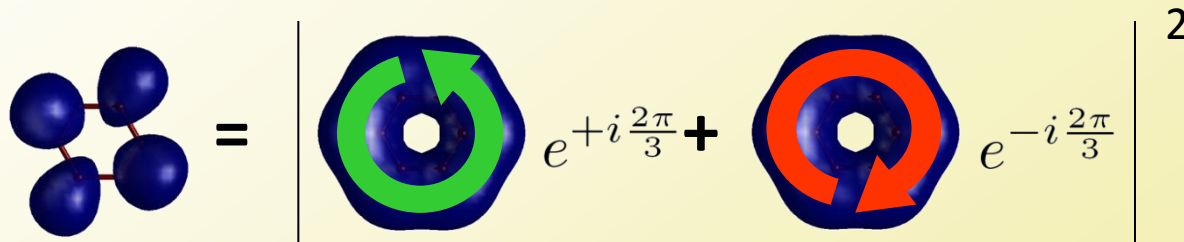
$$\Lambda = \left| \sum_{nm\tau} |\langle N, n | d_{L\tau} | N+1, m \rangle|^2 e^{i\phi_{nm}} \right|^2$$

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



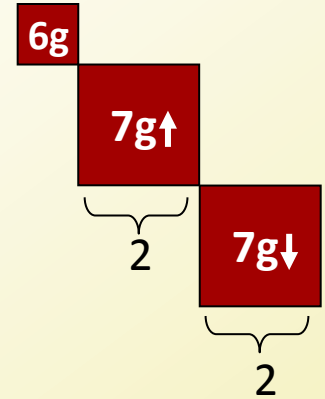
In particular for the transition **6-7** in the **meta** configuration:

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



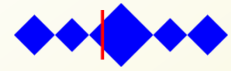
Negative Differential Conductance

- 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:



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

Interference blockade

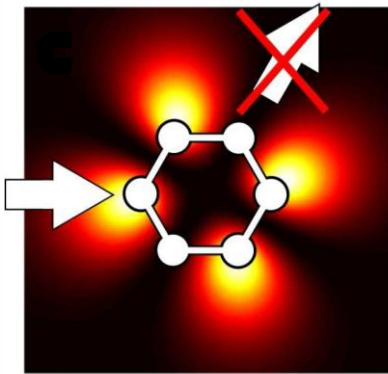


Geometry

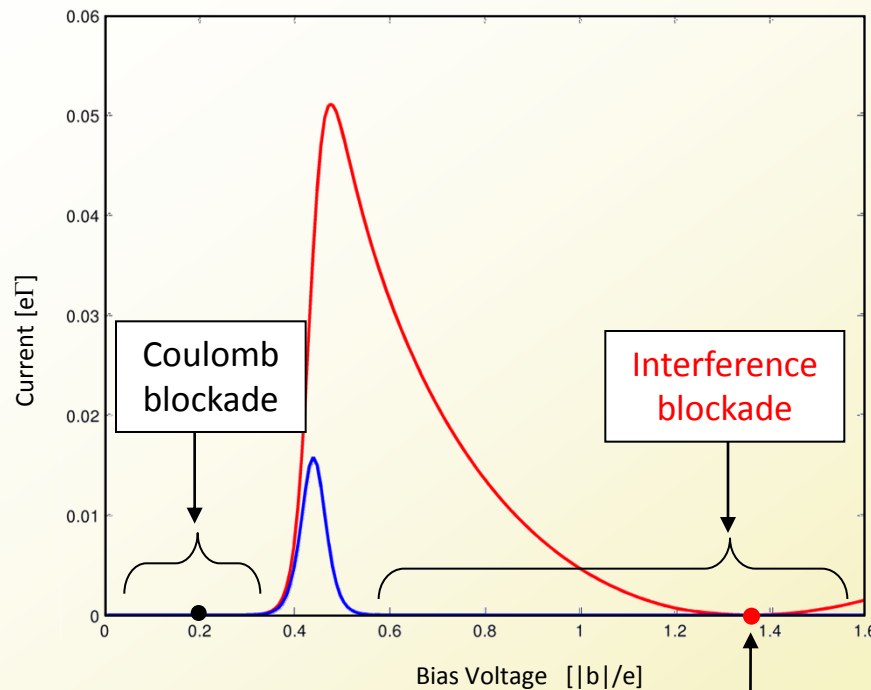
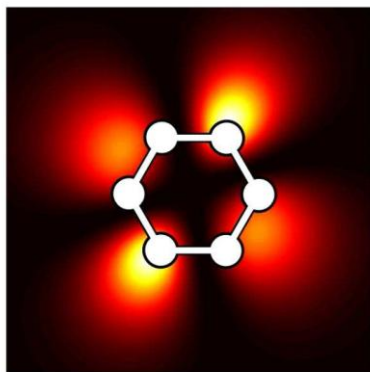
I-V for transition 6 - 7

Energetics

Blocking state

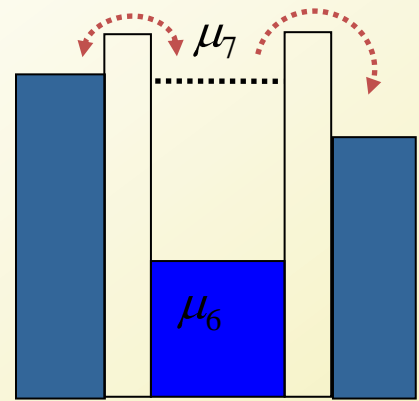


Non-blocking state

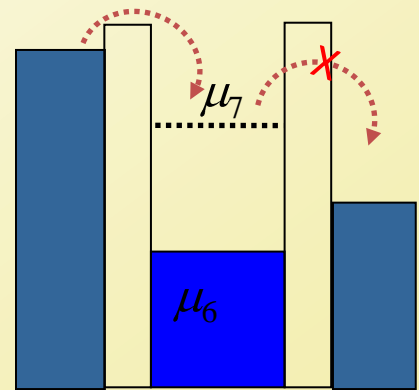


The **blocking state** is an eigenstate of the effective Hamiltonian

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

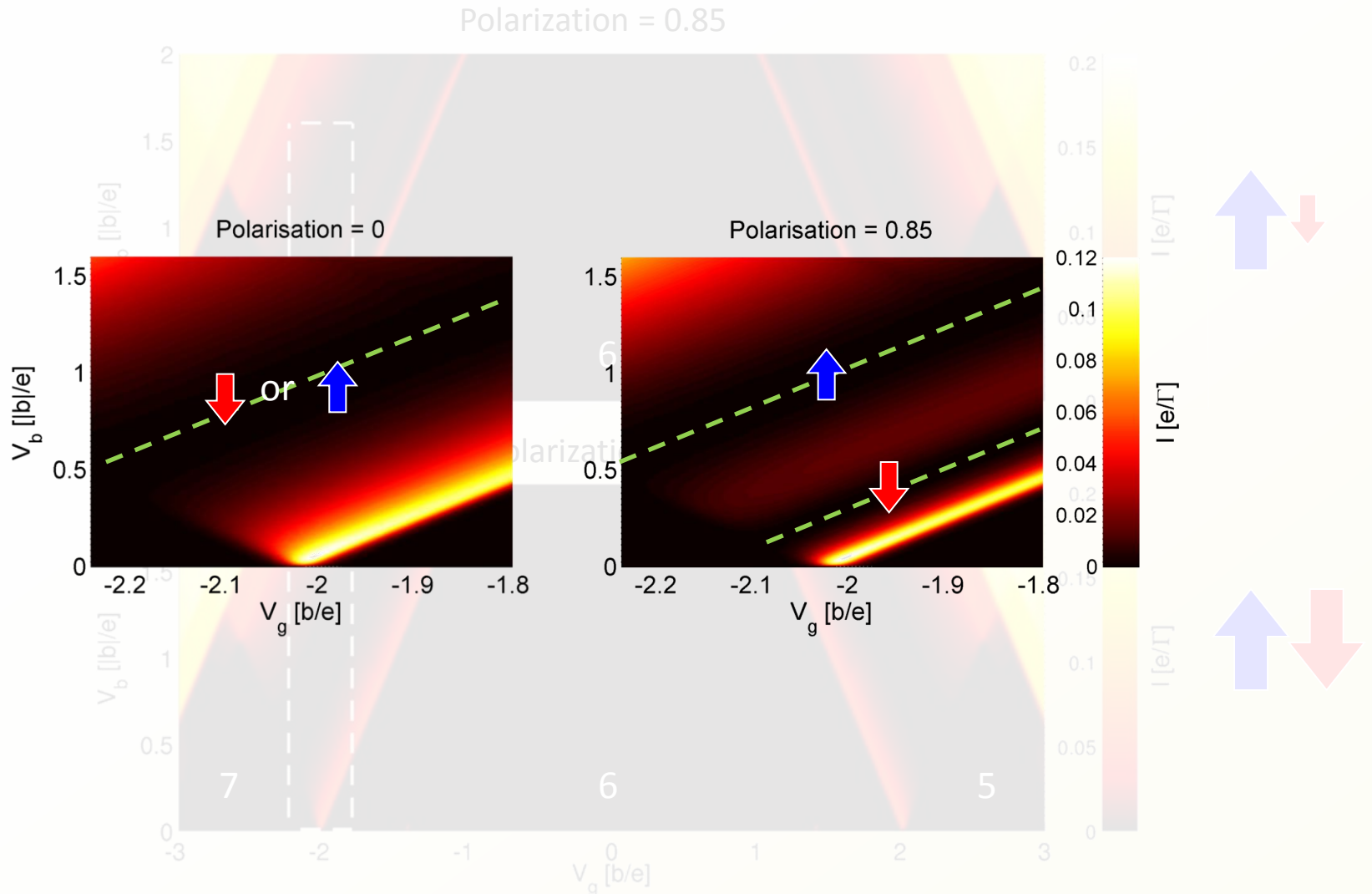


current onset

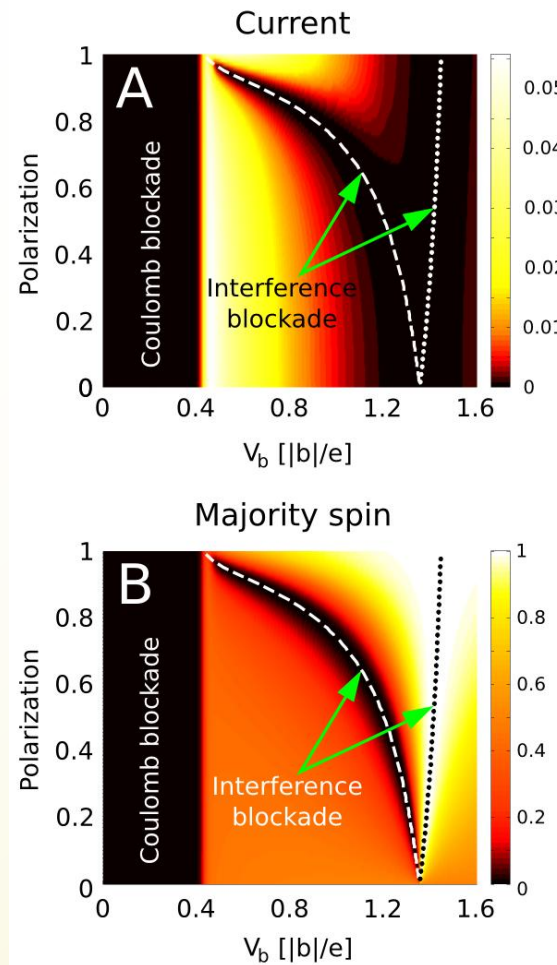


blockade

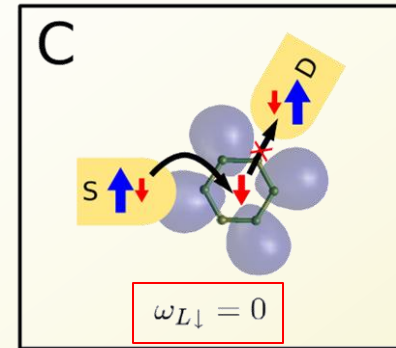
Normal vs. ferromagnetic leads



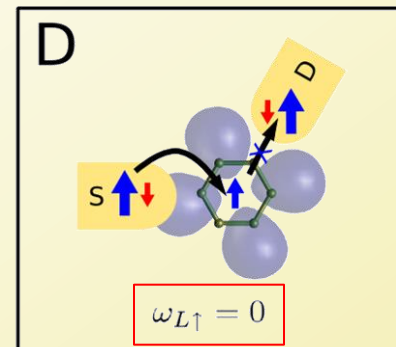
Selective Interference Blocking



Minority blocking



Majority 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**:

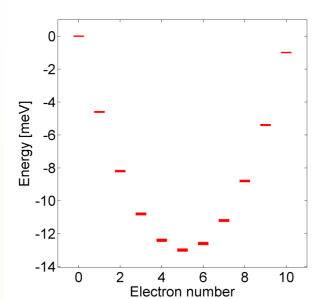
$$\omega_{\alpha\uparrow} - \omega_{\alpha\downarrow} = 2\bar{\Gamma}_\alpha^0 P_\alpha \frac{1}{\pi} \sum_{\{E\}} \left[\begin{aligned} &\langle 7_g \ell \uparrow | d_{M\uparrow} | 8\{E\} \rangle \langle 8\{E\} | d_{M\uparrow}^\dagger | 7_g m \uparrow \rangle p_\alpha(E - E_{7_g}) \\ &+ \langle 7_g \ell \uparrow | d_{M\uparrow}^\dagger | 6\{E\} \rangle \langle 6\{E\} | d_{M\uparrow} | 7_g m \uparrow \rangle p_\alpha(E_{7_g} - E) \\ &- \langle 7_g \ell \uparrow | d_{M\downarrow} | 8\{E\} \rangle \langle 8\{E\} | d_{M\downarrow}^\dagger | 7_g m \uparrow \rangle p_\alpha(E - E_{7_g}) \\ &- \langle 7_g \ell \uparrow | d_{M\downarrow}^\dagger | 6\{E\} \rangle \langle 6\{E\} | d_{M\downarrow} | 7_g m \uparrow \rangle p_\alpha(E_{7_g} - E) \end{aligned} \right]$$

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

Summary

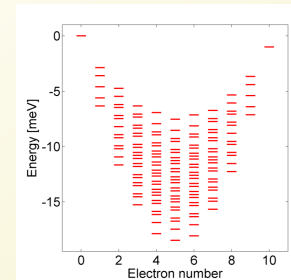
- Pedagogical introduction to the physics of the **sequential tunnelling regime** in transport through micro- and nano-junctions.
- Basic **theory** of Coulomb blockade and single electron tunneling in two flavours

Metallic island



Master equation

Quantum dot



**Generalized
Master equation**

- Application of the **GME** to different types of **quantum dot molecules**

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A. Yar



S. Kolmeder



B. Siegert



S. Pfaller

Thank you for your attention!

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