Topographical fingerprints of many-body interference blocking in STM junctions on thin insulating films

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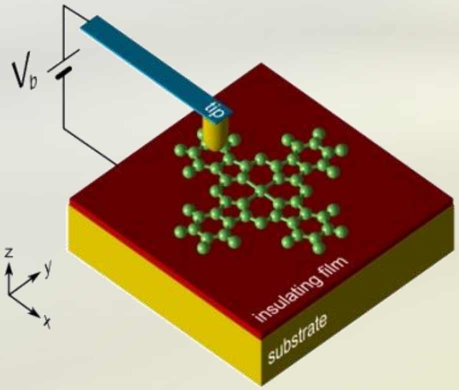

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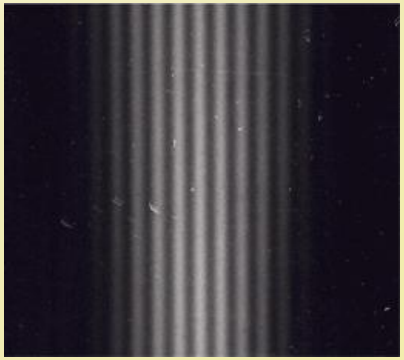
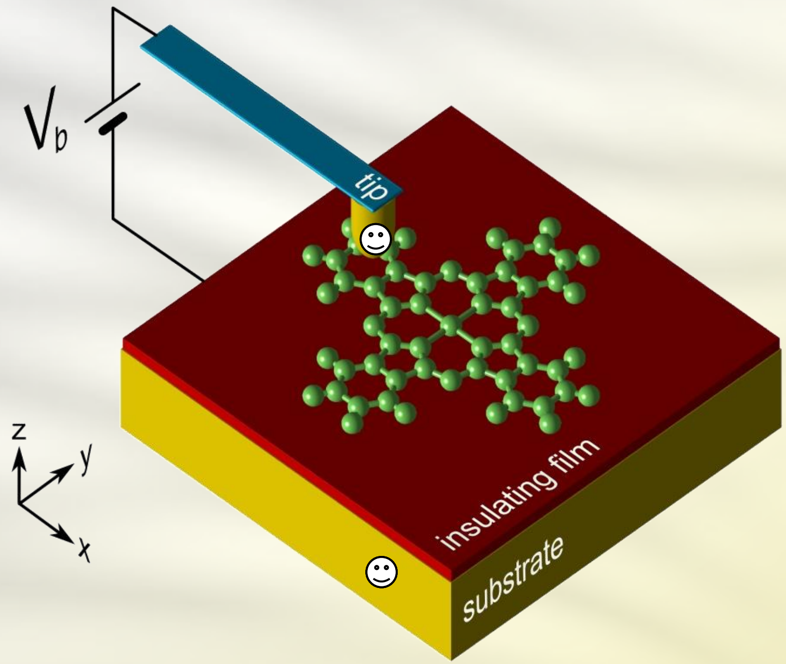


STM on thin insulating films



Weak tip-molecule **tunnelling coupling**
Low molecule-substrate **hybridization**

➡ **sequential tunnelling**

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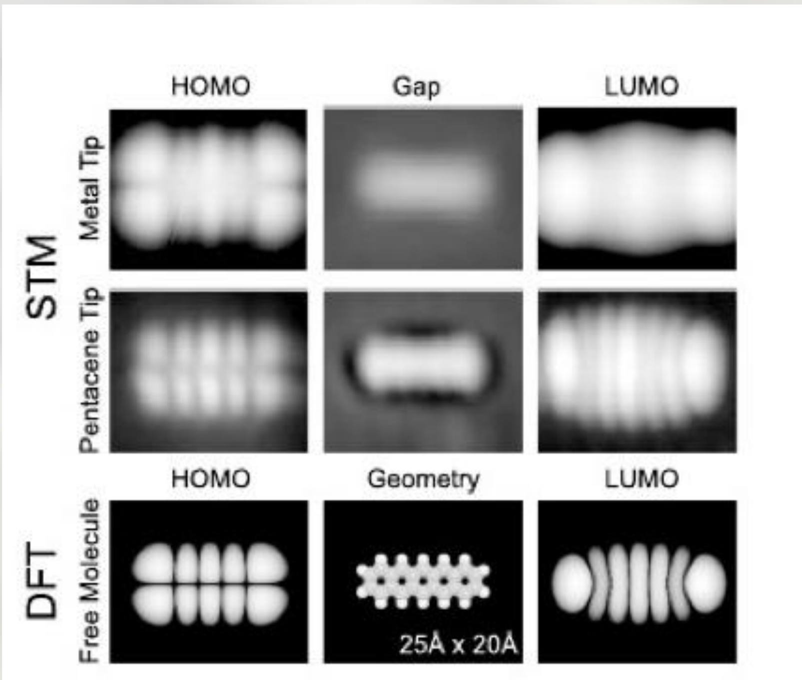
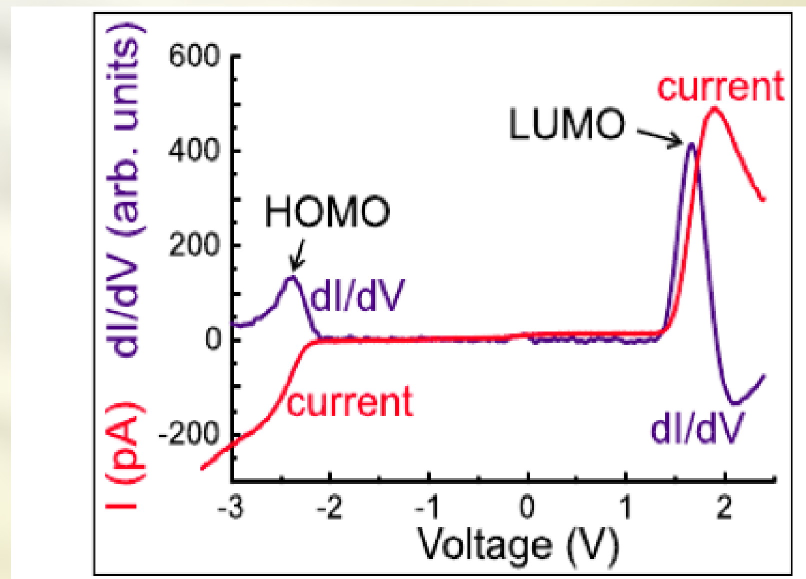
Visualization of molecular orbitals

Topography



Spectroscopy



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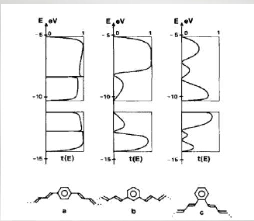
Intramolecular interference: theoretical proposals



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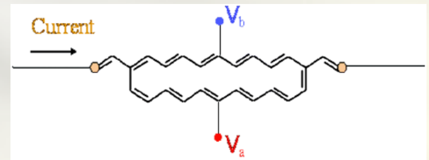

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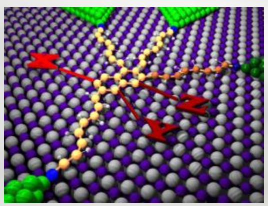

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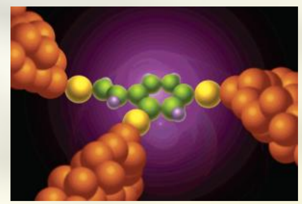

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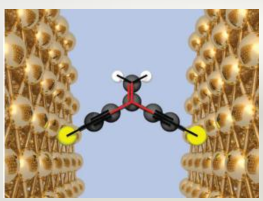

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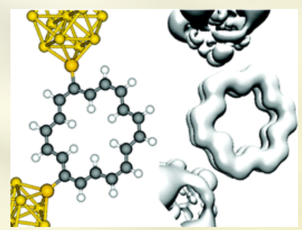

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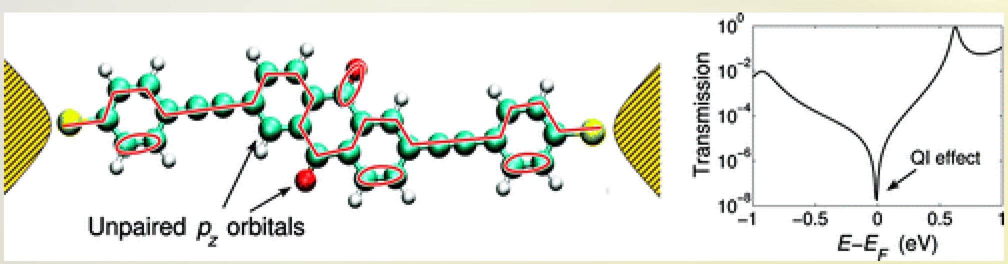

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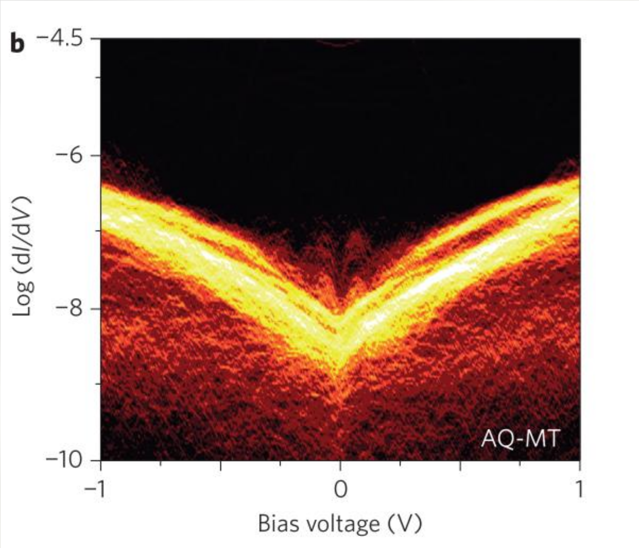
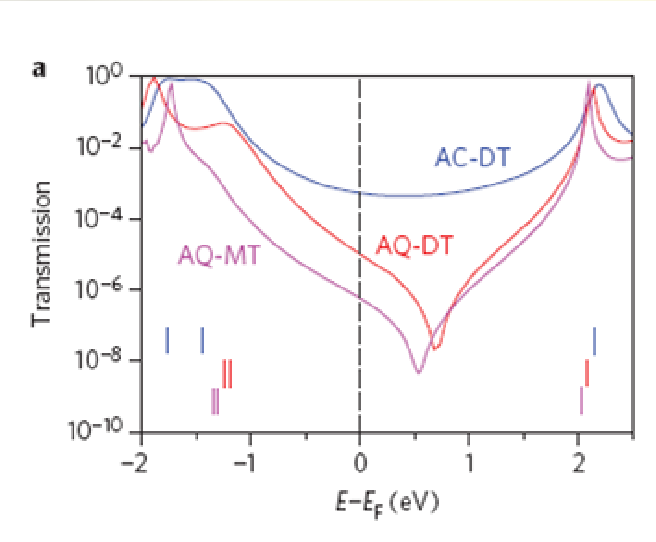
Experimental evidence





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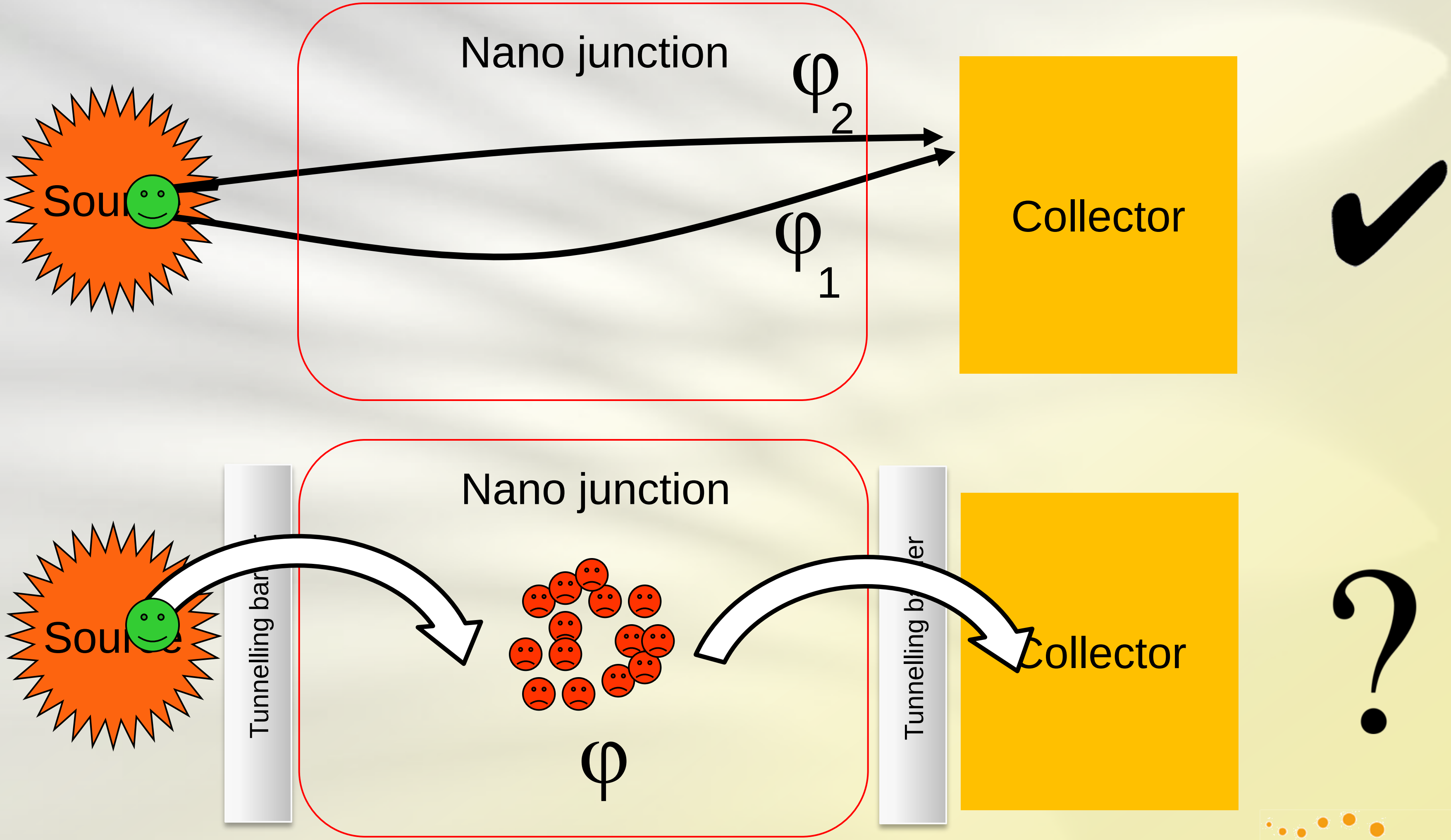
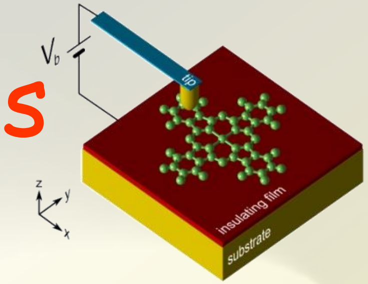
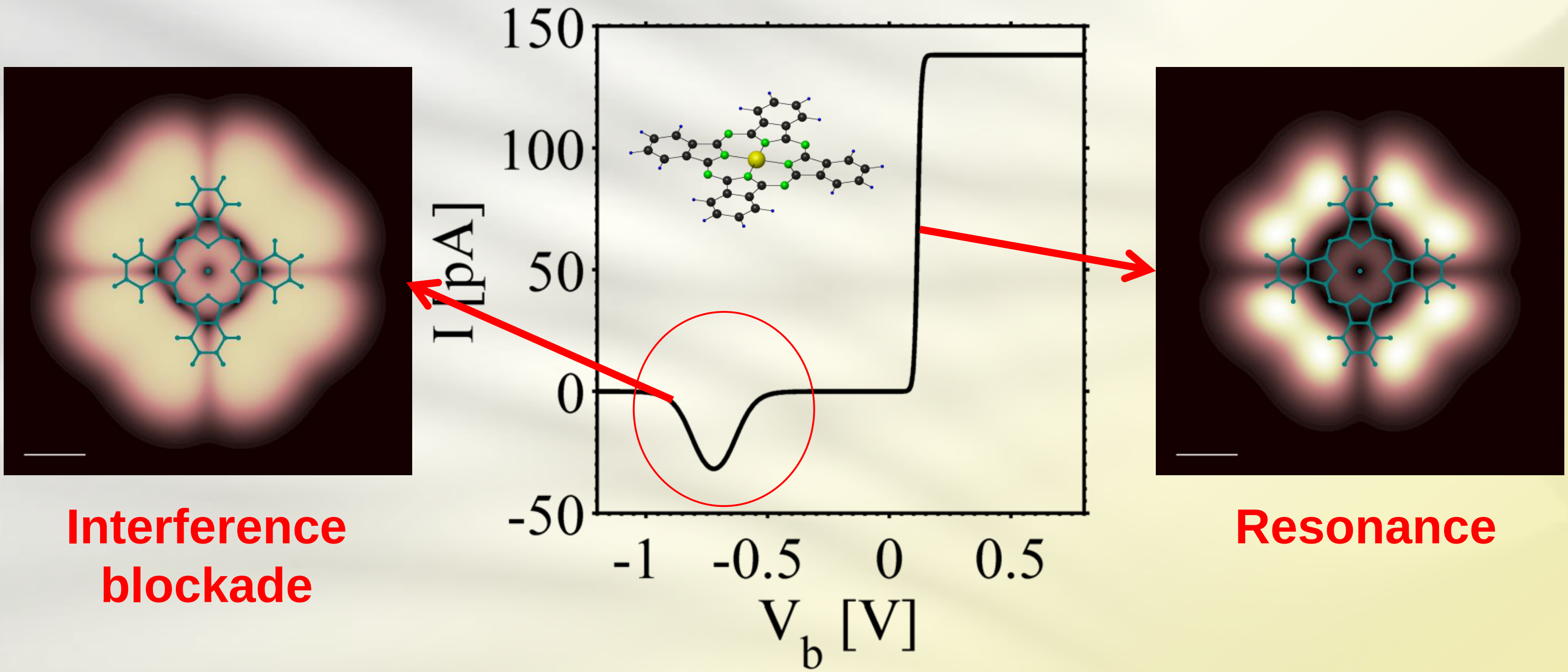
Interference and dephasing



Interference fingerprints



Cu - Phthalocyanine



Interference blockade

Resonance

Constant height current maps in resonance and interference blockade

Donarini, Siegert, Sobczyk and Grifoni Phys. Rev. B 86, 155451 (2012)

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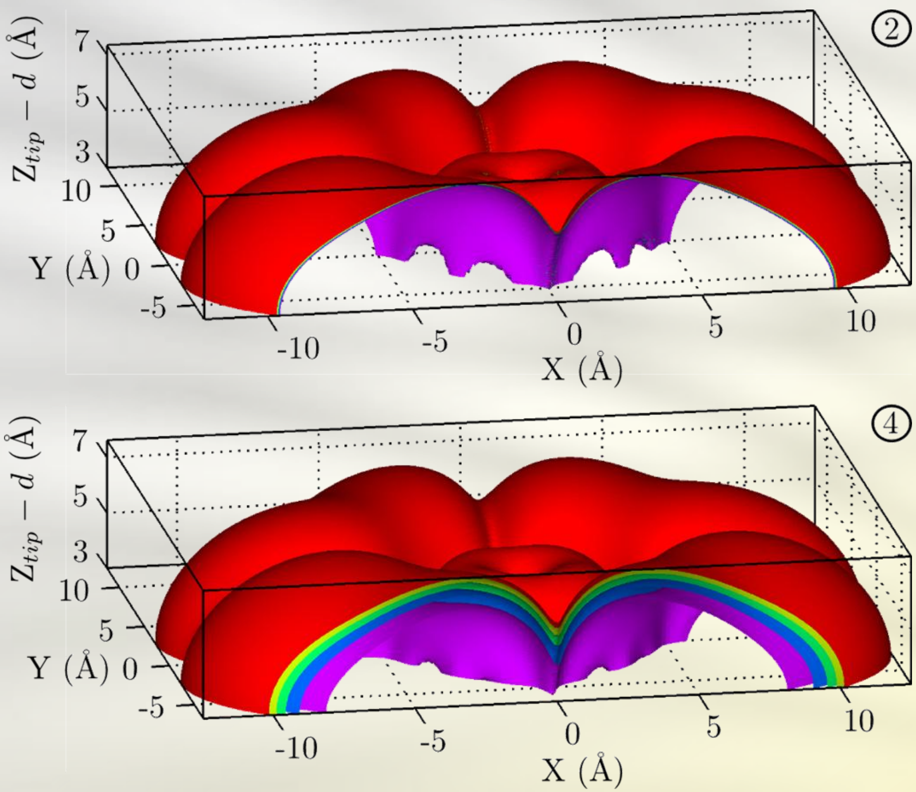
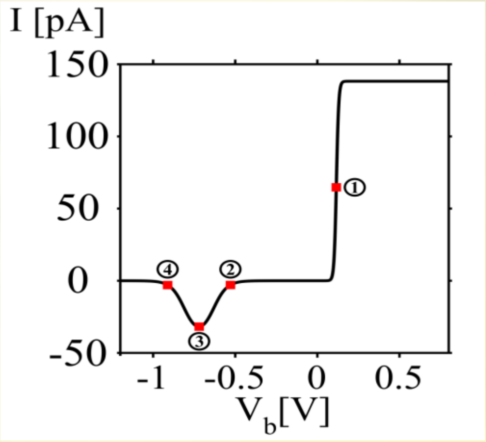
Interference fingerprints





$\Delta I/I = 10\%$
$\Delta z = 2$Å

Constant current topographic maps calculated
at working currents: I = 3.15, 3.075, 3.0, 2.925, and 2.85 pA

Donarini, Siegert, Sobczyk and Grifoni Phys. Rev. B 86, 155451 (2012)

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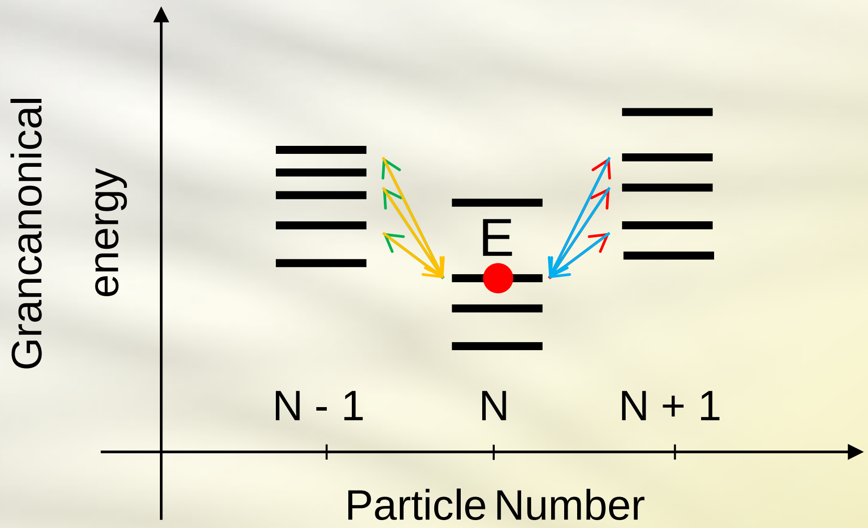
Tunnelling dynamics

$$H_{\rm G} = H_{\rm m} - \mu_0 N$$



Sobczyk, Donarini, Grifoni Phys. Rev. B 85, 205408 (2012)

Many-body rate matrix

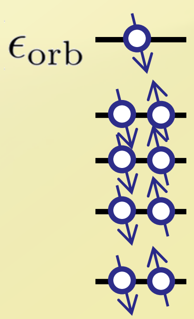
The **current** is proportional to the **transition rate** between **many-body states**

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

where

$$\Gamma^{\chi}_{ij}(E_1 - E_0) = \frac{2\pi}{\hbar} \sum_k (t^{\chi}_{ki})^* t^{\chi}_{kj} \delta(\epsilon^{\chi}_k - E_1 + E_0)$$

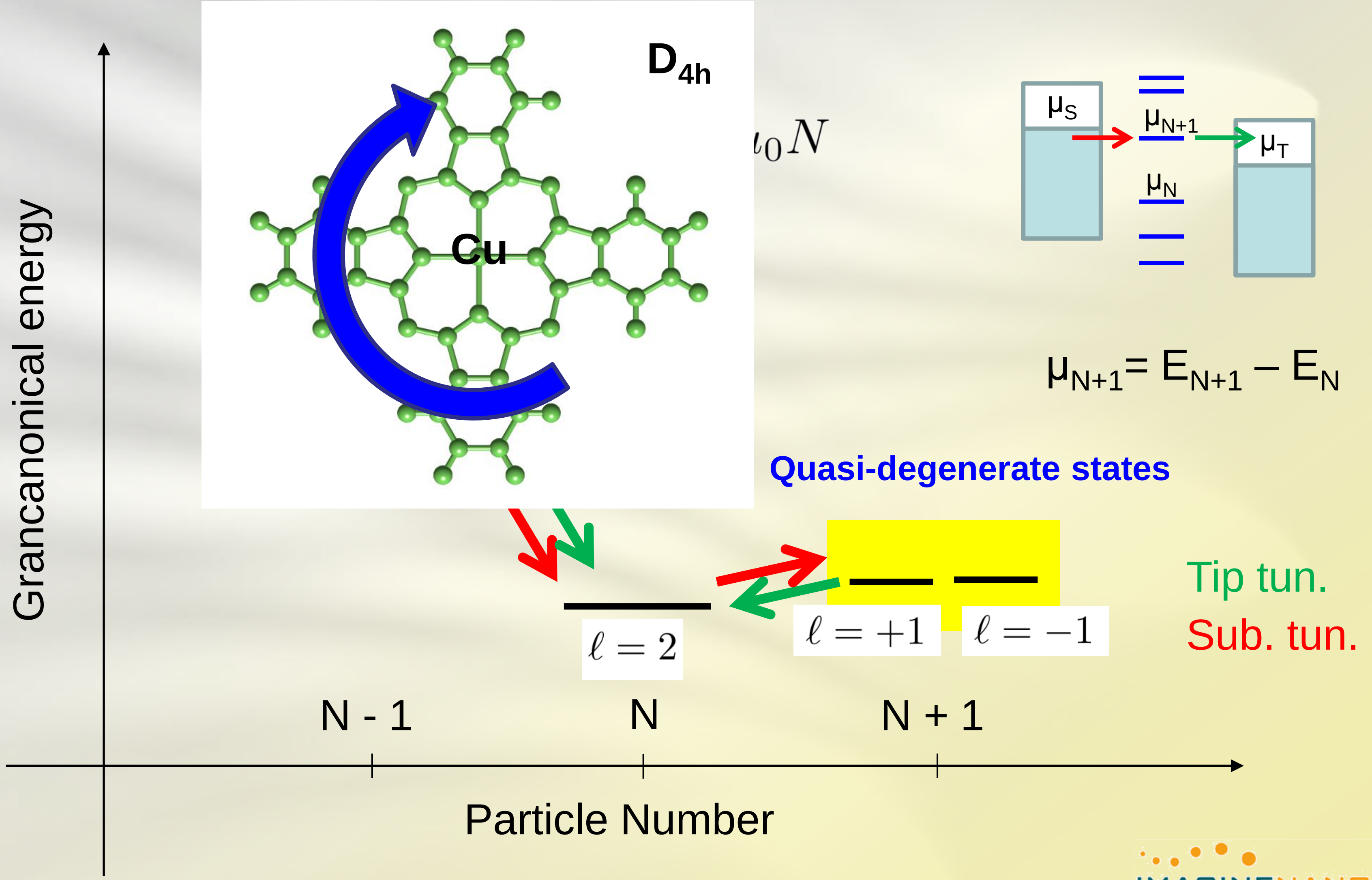
For **uncorrelated** and **non-degenerate systems** the many-body rate reduces to



$$R^{\chi\tau}_{N\,E_0\rightarrow N+1\,E_1} = \Gamma^{\chi}_{\rm orb}(\epsilon_{\rm orb}) f^{+}(\epsilon_{\rm orb} - \mu_{\chi})$$

The **constant current map** is the **isosurface** of a **specific molecular orbital**.

Dynamics in energy space



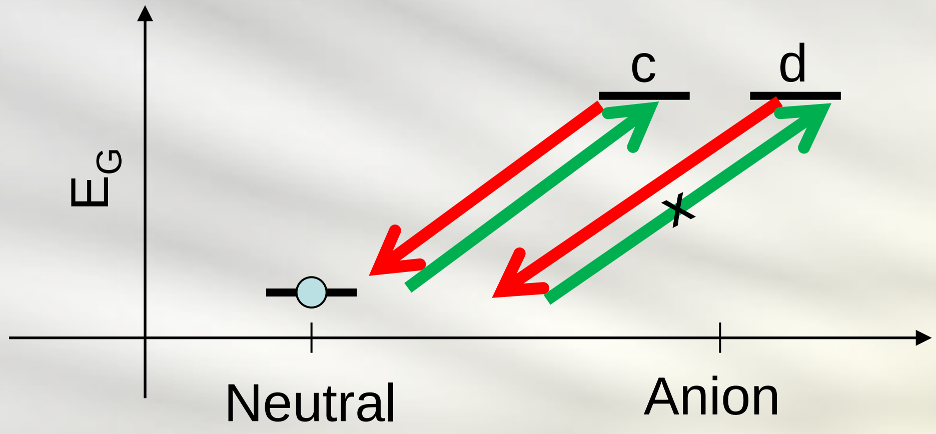
Interference: decoupling basis

Degenerate anionic ground state ⇨ **Matrix form** for the **many-body tunnelling rate** between the neutral and anionic ground states.

	Angular momentum basis	Decoupling basis
Tip	$\ell = +1$ $\quad$ $\ell = -1$ $\mathbf{R}^T = R_0^T \begin{pmatrix} 1 & e^{-2i\phi} \\ e^{+2i\phi} & 1 \end{pmatrix}$ **Mixes** angular momentum	$\tilde{\mathbf{R}}^T = R_0^T \begin{pmatrix} 2 & 0 \\ 0 & 0 \end{pmatrix}$ One of the anionic state is **decoupled from the tip**
Substrate	$\mathbf{R}^S = R_0^S \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$ **Conserves** angular momentum	$\tilde{\mathbf{R}}^S = R_0^S \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$

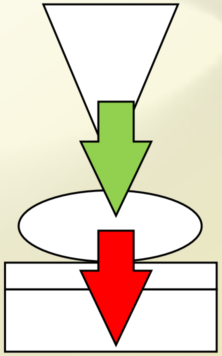
Notice that the decoupling basis **depends** on the **tip position**.

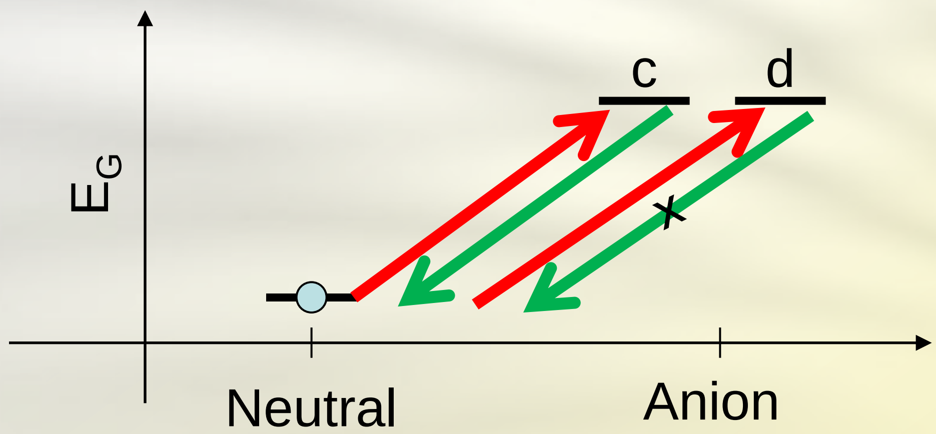
Interference: current blocking



$V_b > -\Delta E_G/ec$

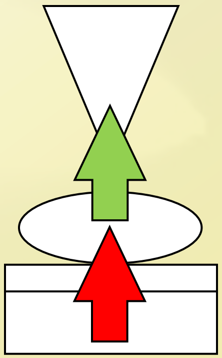
Current





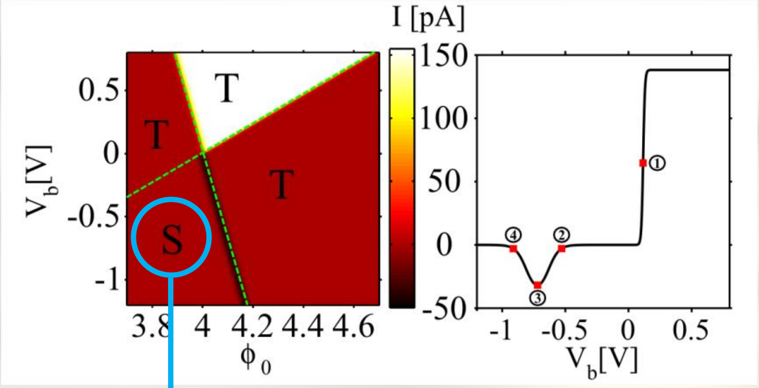
$V_b < \Delta E_G/e(1-c)$

No current

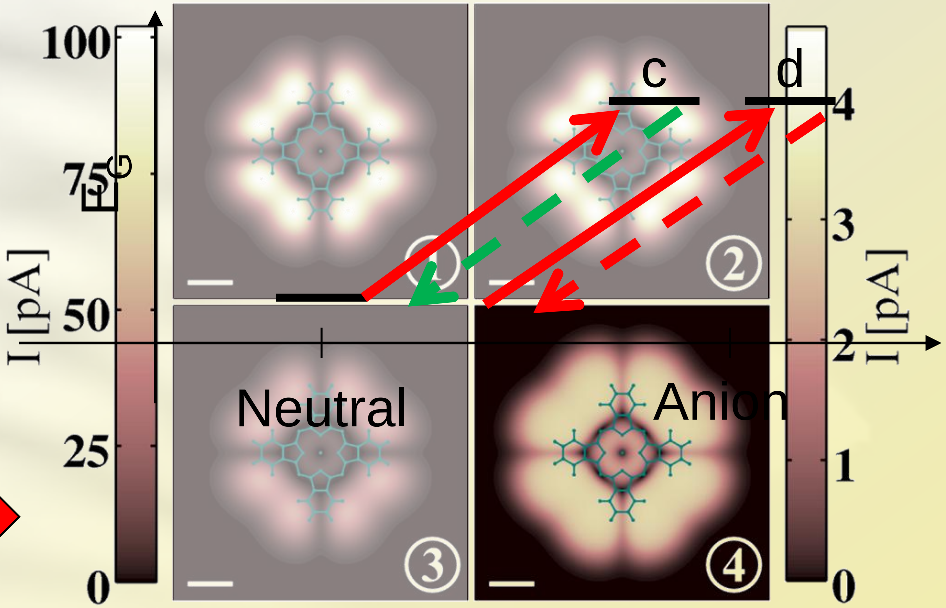


$\mu_T = \mu_0 - c\,eV_b$ $\qquad$ $\mu_S = \mu_0 + (1-c)eV_b$ $\qquad$ $c \approx 0.9$

A new bottle-neck process



$$I_{\mathrm{IB}} = e\frac{R_0^S f_S^- R_0^T f_T^-}{R_0^S f_S^- + R_0^T f_T^-}$$



The **depopulation** of the blocking state via a **substrate transition** dominates the transport.

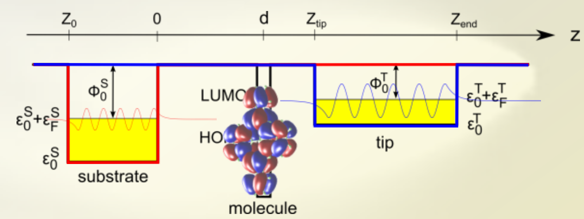
Conclusions

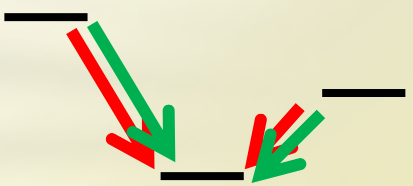
- We developed a **semi-quantitative model** for the description of double barrier STM junctions with π-conjugated molecules.

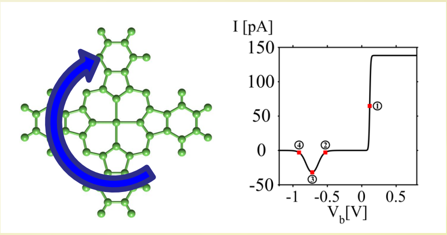
- The dynamics is described in terms of **many-body** transitions.

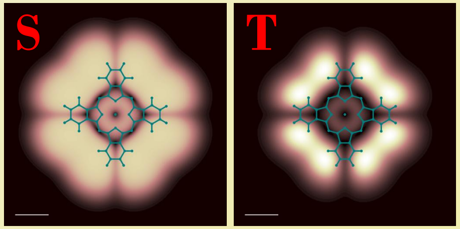
- Transport through **degenerate states** is associated to **electron interference** blockade at negative sample biases.

- Close to the interference blocking regime, **substrate** tunnelling dominates the transport and gives **flat constant height current maps**.









Interference Resonance

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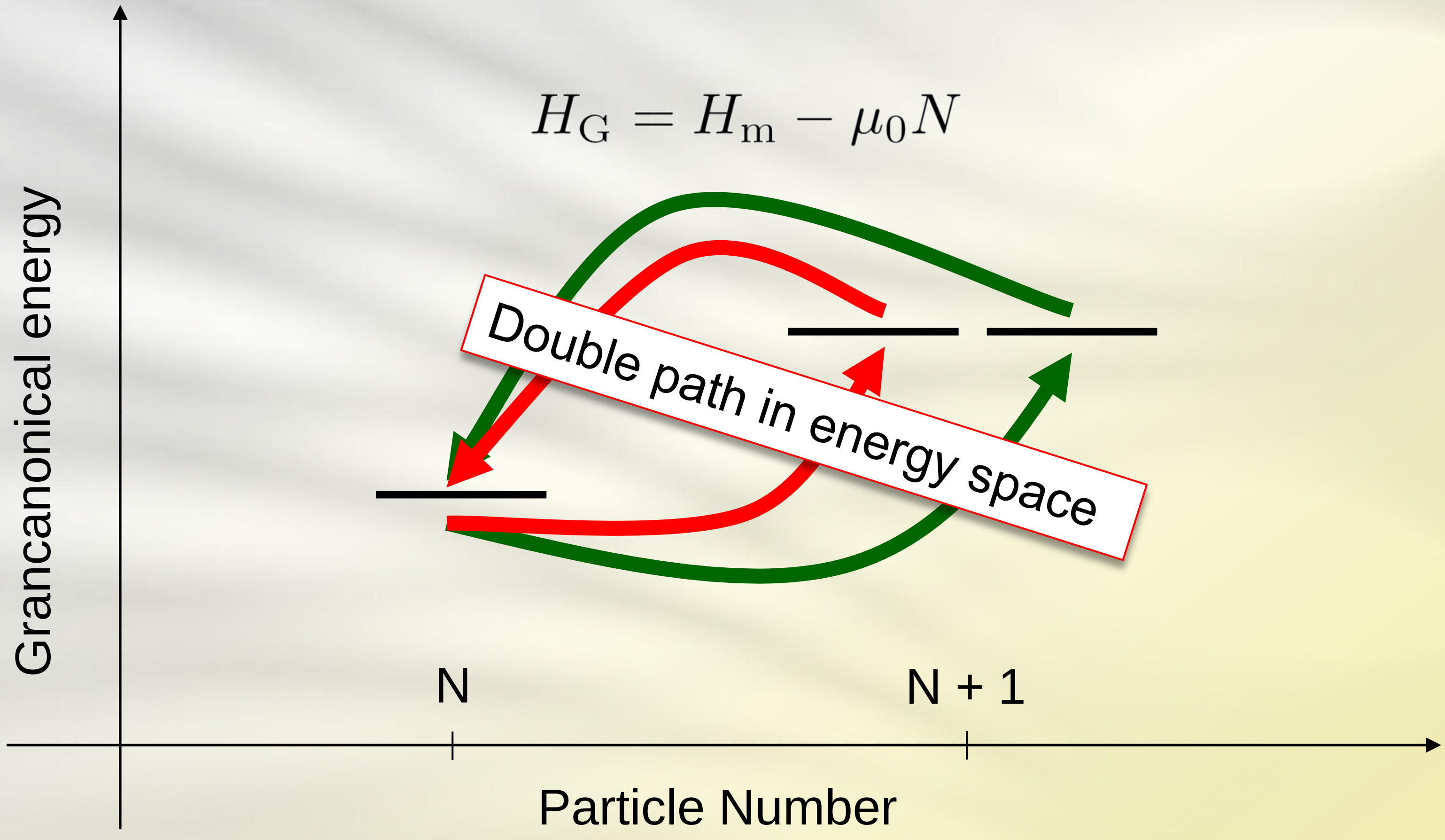
Interfernce + interaction ✔



Donarini, Begemann, and Grifoni, *Phys. Rev. B* **82,** 125451 (2010)

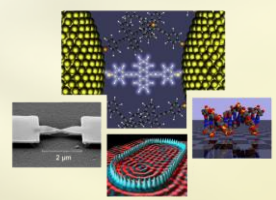
Thanks

Benjamin Siegert

Sandra Sobczyk

Milena Grifoni

DFG Deutsche Forschungsgemeinschaft



SPP 1243

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Thank you for your attention…

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