Electromechanical properties of a biphenyl transistor

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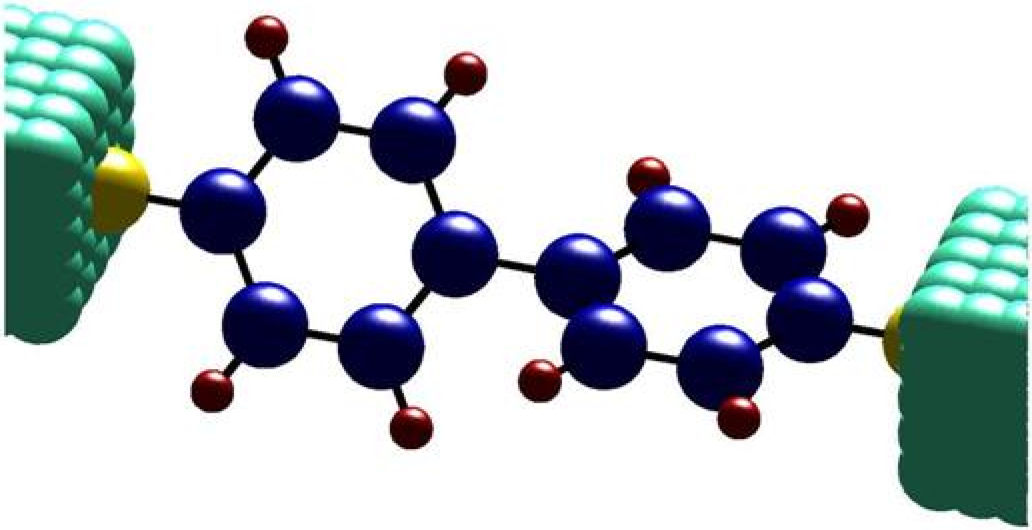
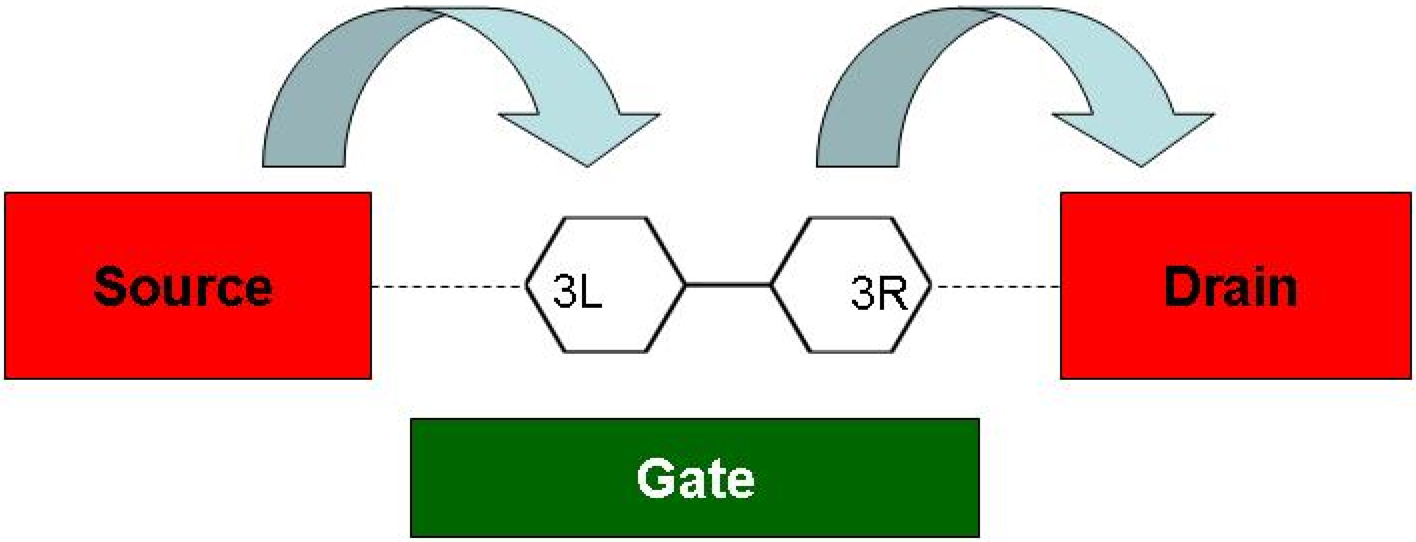


The model



- Weak coupling to the leads + low temperature -> Coulomb blockade
- Gate voltage
- Torsional (the softest mechanical) degree of freedom of the molecule

The Hamiltonian

The hamiltonian of the device can be written as

$$H = H_{Mol} + H_{Leads} + V$$

Where

$$H_{Mol} = T_\theta + H_{PPP}(\hat{\theta})$$

$$H_{Leads} = \sum_{\alpha k \sigma} \epsilon_{k,\alpha} c^\dagger_{\alpha k \sigma} c_{\alpha k \sigma}$$

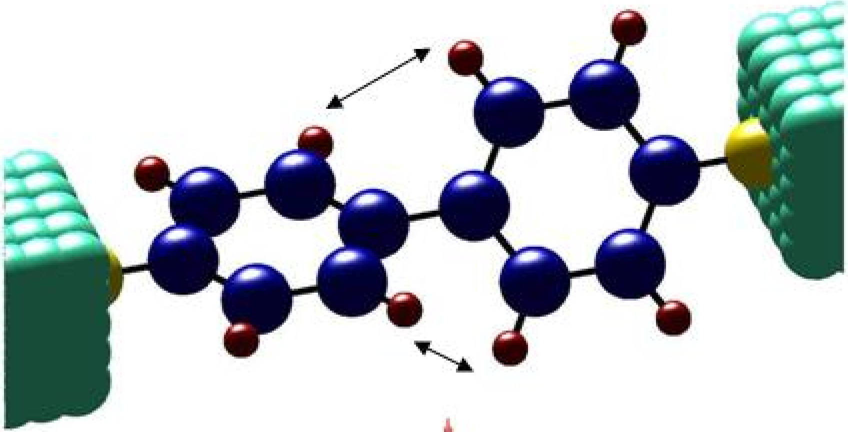
$$V = t \sum_{\alpha k \sigma} (c^\dagger_{\alpha k \sigma} c_{\alpha 3 \sigma} + c^\dagger_{\alpha 3 \sigma} c_{\alpha k \sigma})$$

Isolated biphenyl

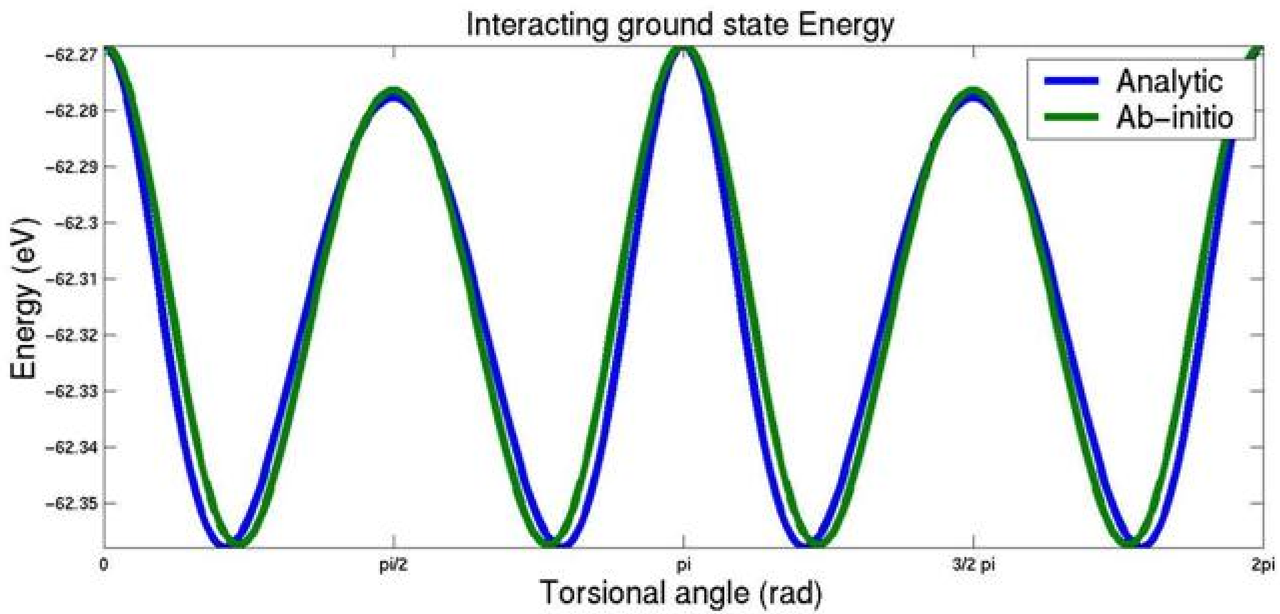
- H_{PPP} is the Pariser-Parr-Pople Hamiltonian

$$H_{PPP}(\theta) = \sum_{i\sigma} b_{i\,i+1}(\theta)(c^\dagger_{i\sigma}c_{i+1\sigma} + c^\dagger_{i+1\sigma}c_{i\sigma}) + \sum_i U_i\left(\hat{n}_{i\uparrow} - \frac{z_i}{2}\right)\left(\hat{n}_{i\downarrow} - \frac{z_i}{2}\right) + \sum_{\langle i<j\rangle} V_{ij}(\theta)(\hat{n}_i - z_i)(\hat{n}_j - z_j)$$

+ H-H steric repulsion (Lehnard-Jones potential)



Neutral state



- ground state electronic energy for the neutral molecule: four tilted equilibrium configurations

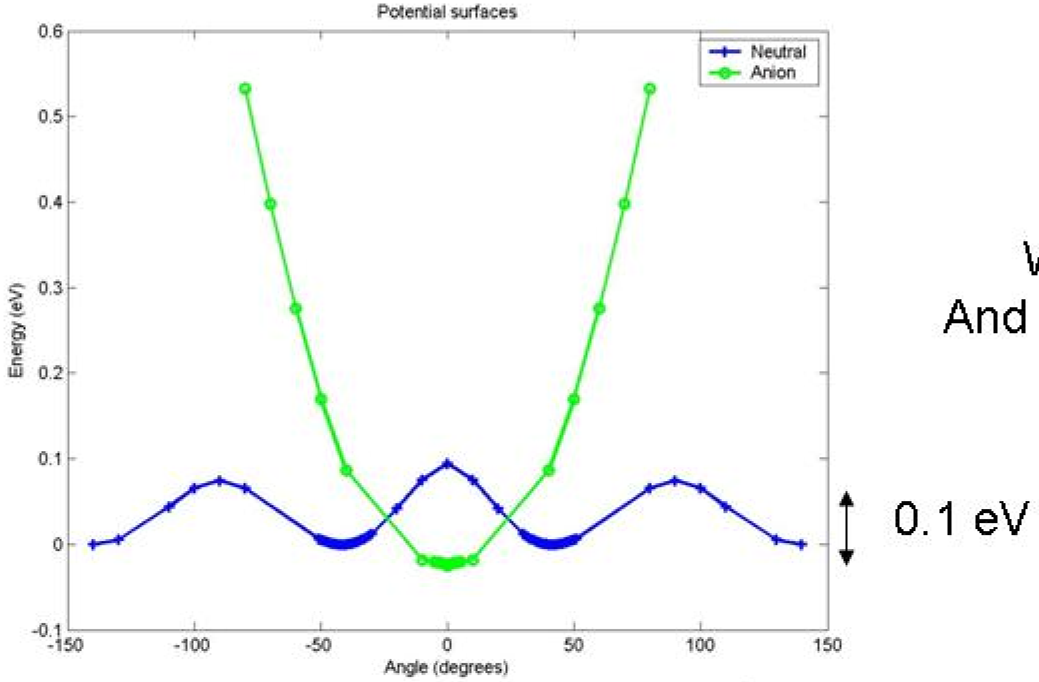
- Technique: Hartree-Fock approximation of the PPP Hamiltonian

Anionic state

- Planar equilibrium configuration for the anionic state
- The Hartree-Fock approximation fails for the anionic state

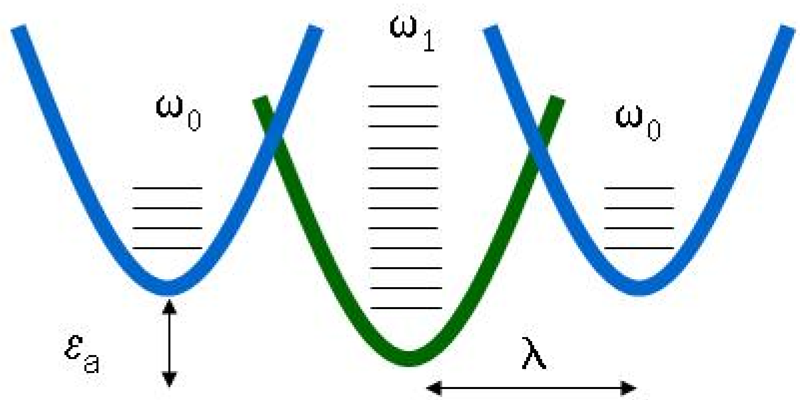
CORRELATIONS!!!

(work in progress to go beyond Hartree-Fock)



DFT calculations
With gaussian basis set
And B3LYP exchange potential

Model ... of the model



- Harmonic approximation for the anionic state
- Low laying states of a double (harmonic) well potential for the neutral state
- The model is identifyed by three parameters:

$$\alpha = \sqrt{\frac{\omega_1}{\omega_0}} \qquad \lambda = \frac{\Delta\theta}{\sqrt{\theta_{z0}\theta_{z1}}} \qquad \epsilon_a = \frac{EA}{\hbar\sqrt{\omega_0\omega_1}}$$

Definition of Unity

- We write the previous approximation in terms of unity operator:

$$\mathbf{1} = |N\rangle\langle N|(\mathcal{P}_+ + \mathcal{P}_-) + |N+1\rangle\langle N+1|\mathcal{P}_0$$

- where $\mathcal{P}_\pm = \sum_{n=0}^{N_\pm} |n_\pm\rangle\langle n_\pm|$, $\mathcal{P}_0 = \sum_{n=0}^{\infty} |n_0\rangle\langle n_0|$

that define the effective Hilbert space.

Effective Hamiltonian

$$H_{eff} = \sum_{\alpha k \sigma} \epsilon_{k,\alpha} c^\dagger_{\alpha k \sigma} c_{\alpha k \sigma} +$$
$$+ \sum_{\alpha k \sigma, n, m} \left[t^{(+)}_{n,m} c^\dagger_{\alpha k \sigma} c_{d\sigma} |n_+\rangle\langle m_0| + t^{(-)}_{n,m} c^\dagger_{\alpha k \sigma} c_{d\sigma} |n_-\rangle\langle m_0| + t^{(+)}_{n,m} c^\dagger_{d\sigma} c_{\alpha k \sigma} |m_0\rangle\langle n_+| + t^{(-)}_{n,m} c^\dagger_{d\sigma} c_{\alpha k \sigma} |m_0\rangle\langle n_-| \right] +$$
$$+ \left(1 - \sum_\sigma c^\dagger_{d\sigma} c_{d\sigma}\right) \left\{ \epsilon_0 + \hbar\omega_0 \left[\mathcal{P}_+ \left(d^\dagger_+ d_+ + \frac{1}{2}\right) \mathcal{P}_+ + \mathcal{P}_- \left(d^\dagger_- d_- + \frac{1}{2}\right) \mathcal{P}_- \right] \right\} +$$
$$+ \sum_\sigma c^\dagger_{d\sigma} c_{d\sigma} \left[\epsilon_1 + \mathcal{P}_0 \left(d^\dagger d + \frac{1}{2}\right) \mathcal{P}_0 \right]$$

where

$$c^\dagger_{d\sigma} |N\rangle \equiv |N+1, \sigma\rangle$$

$$t^{(\pm)}_{n,m} = t\langle N | c_{3\alpha} | N+1\rangle \langle n_\pm | m_0 \rangle$$

Franck-Condon coefficient

We assume it independent of θ

Generalized Master Equation

We study the dynamics of the system with the Generalized Master Equation:

$$\dot{\sigma} = -\frac{i}{\hbar}[H_{Mol}, \sigma] - \frac{1}{\hbar^2}\int_0^\infty \mathrm{d}\tau \mathrm{Tr}_{Leads}\{[V, [\tilde{V}_I(-\tau), \sigma]] \otimes \rho_{Leads}\}$$

- coherences between different charge states vanish

- due to the mechanical degeneracies of the neutral state we MUST keep coherences between displaced mechanical states

- we write the GME in the basis that diagonalize the molecule hamiltonian (Bloch-Redfield form)

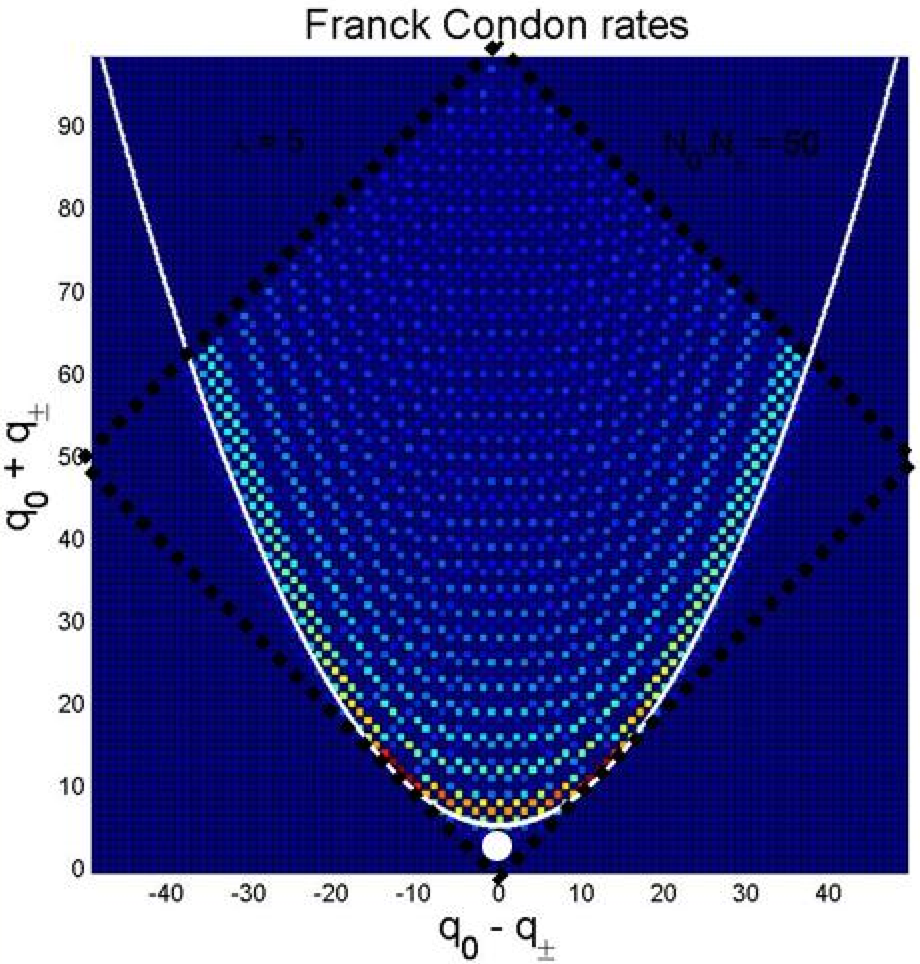
$$\dot{\sigma}_{ij} = \sum_{mn} R_{ijmn}\sigma_{mn}$$

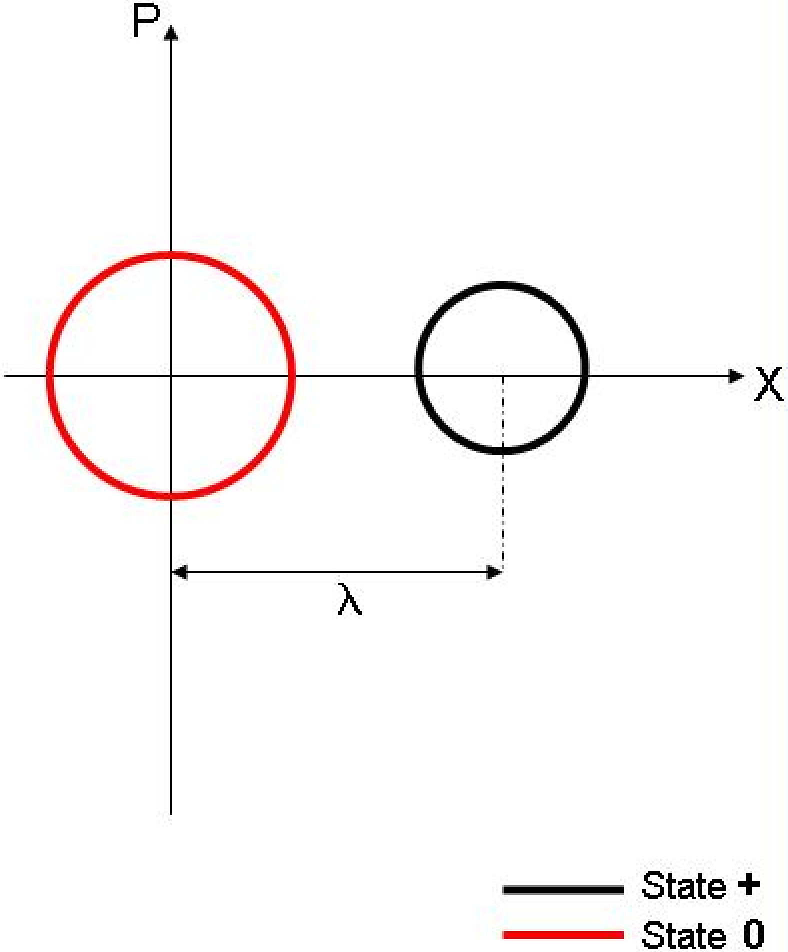
GME (II)

The Redfield tensor R_{ijmn} depends on

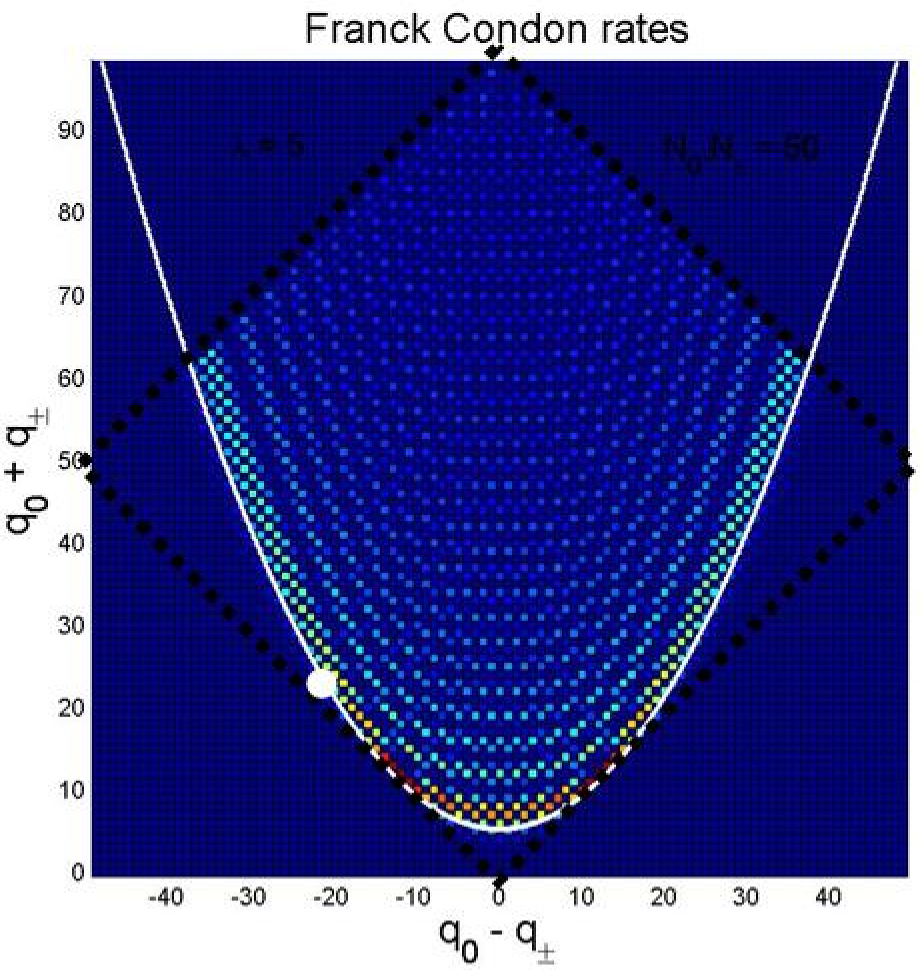
- Fermi factors - the Pauli esclusion principle prevents some transitions to occur
- Bare tunneling rates - given by the density of states in the leads times the electrical coupling leads-molecule
- Franck-Condon coefficients – each state transition in the system is electromechanical

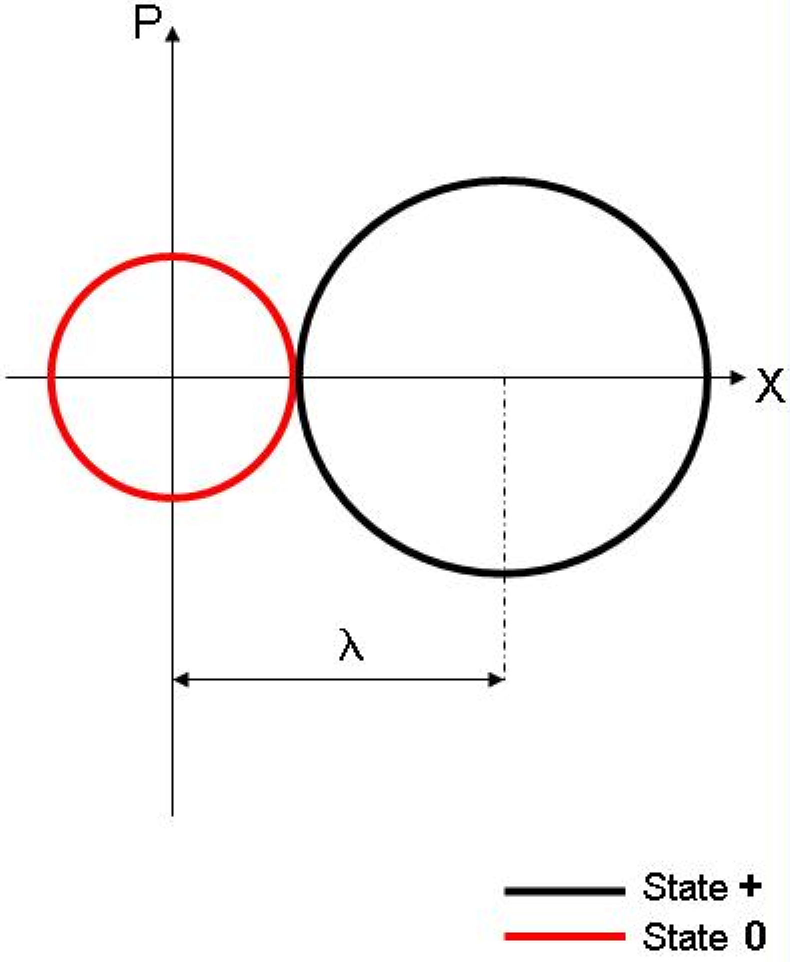
The Franck-Condon parabola





The Franck-Condon parabola





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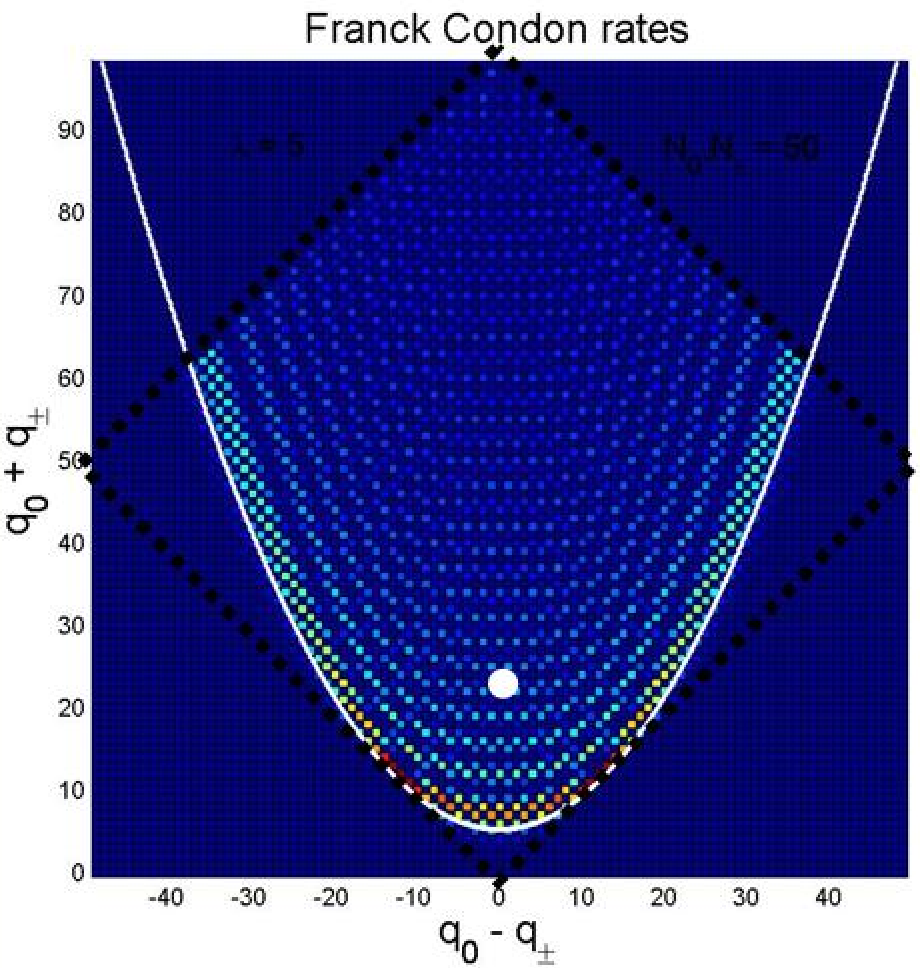
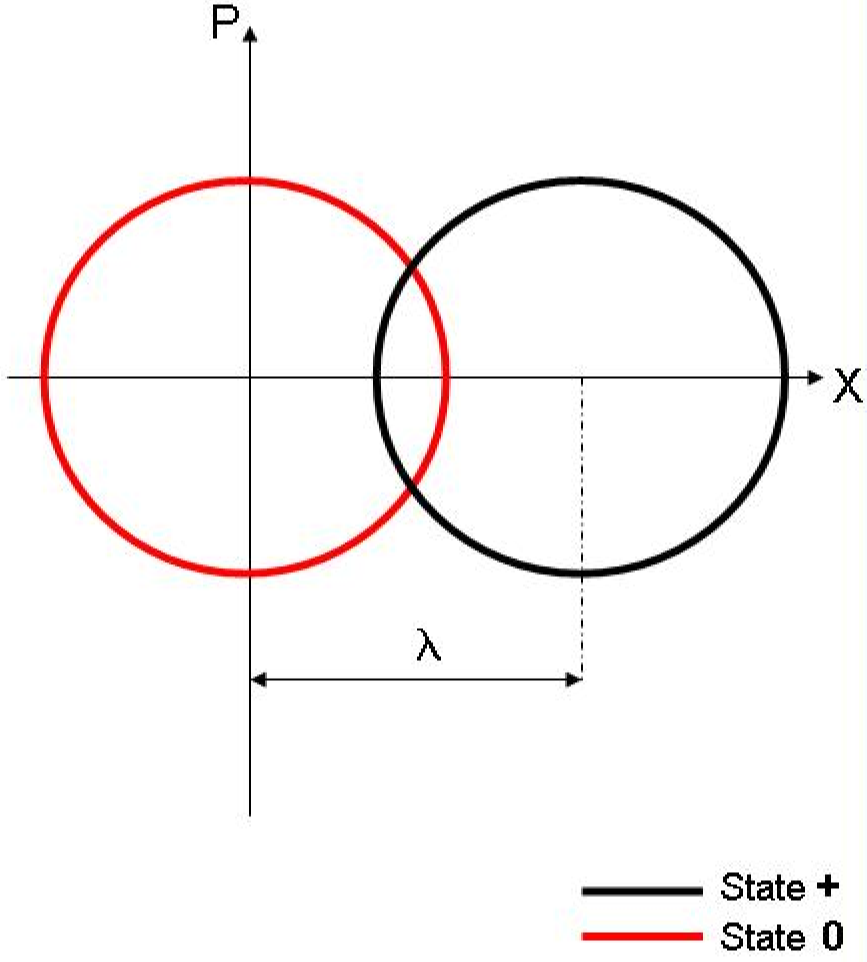
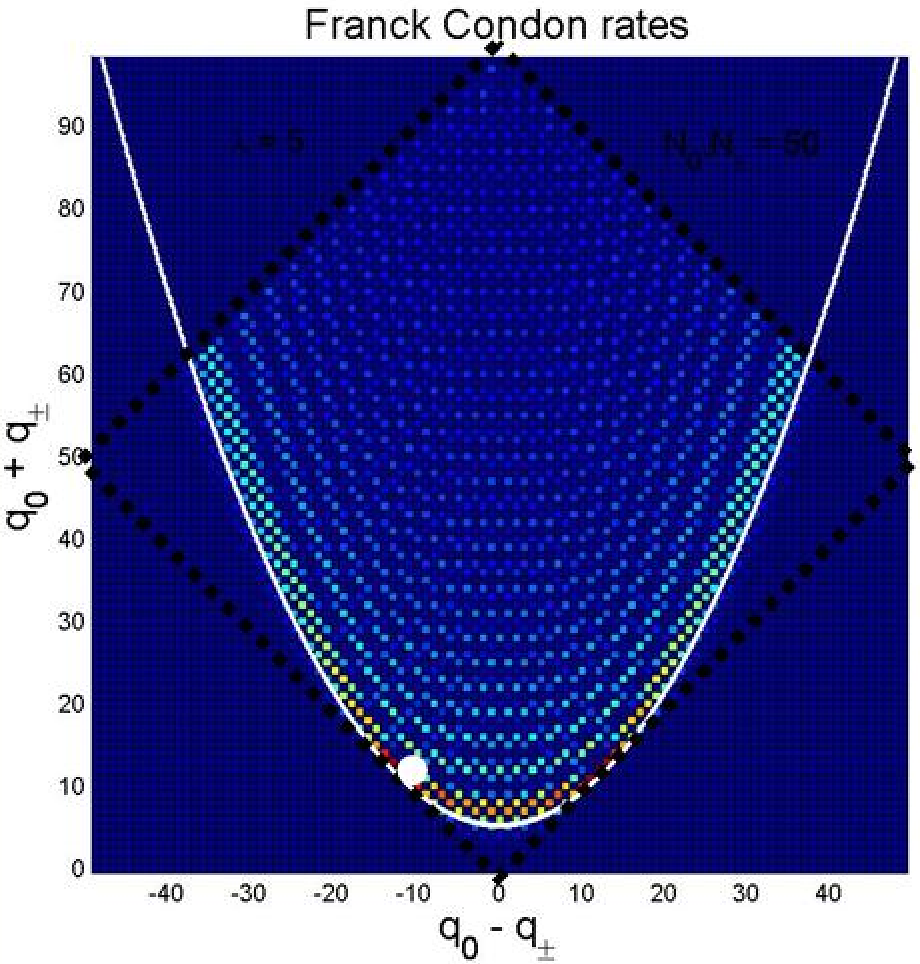
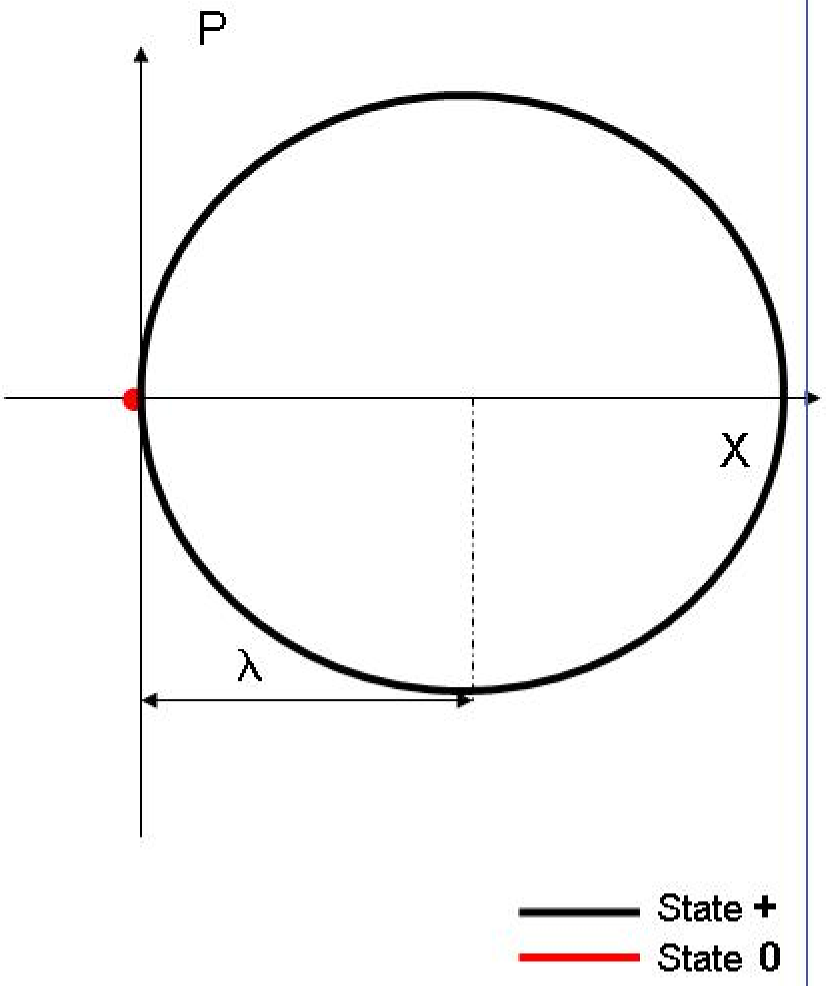




The Franck-Condon parabola





The current calculation

1. Computation of the stationary reduced density matrix σ^{stat}

2. Identification of the current operators:

$$
\begin{aligned}
I &= \frac{dQ}{dt} = \sum_m \dot{\sigma}_{11}^{m\,m} \\
&= -\sum_{k\,l\,\alpha} \left(W_{k\,l}^{\alpha} \sigma_{11}^{l\,k} + \sigma_{11}^{k\,l} W_{l\,k}^{\alpha} \right) + \sum_{k\,n'\tau'\,n\tau\,\alpha} \left(R_{k\,k\,n'\tau'\,n\tau}^{\alpha} + R_{k\,k\,n\tau\,n'\tau'}^{\alpha} \right) \sigma_{00}^{n'\tau'\,n\tau} \\
&= -2\mathrm{Tr}_{Mech}\{W_1^L \sigma_{11}\} + 2\mathrm{Tr}_{Mech}\{W_0^L \sigma_{00}\} - 2\mathrm{Tr}_{Mech}\{W_1^R \sigma_{11}\} + 2\mathrm{Tr}_{Mech}\{W_0^R \sigma_{00}\} \\
&= \langle \hat{I}_L \rangle - \langle \hat{I}_R \rangle
\end{aligned}
$$

3. Calculation of the stationary current:

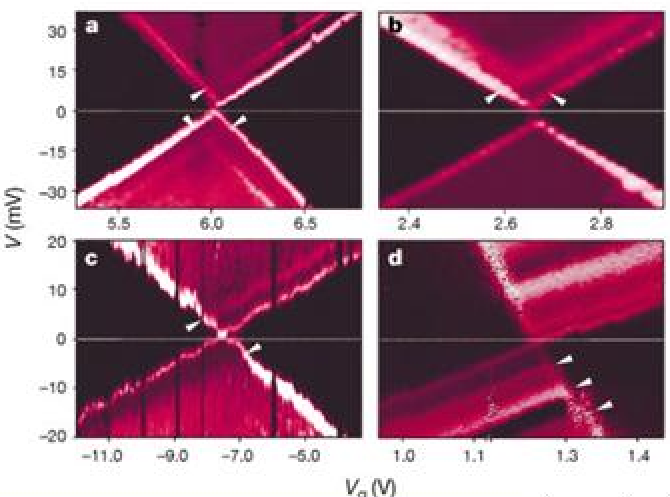
$$
I_L^{stat} = \mathrm{Tr}_{Mech}\{\sigma^{stat} \hat{I}_L\}
$$
$$
I_R^{stat} = \mathrm{Tr}_{Mech}\{\sigma^{stat} \hat{I}_R\}
$$

Stability diagrams

- Plot of differential conductance as a function of bias and gate voltage:

$$G = \frac{dI}{dV_b}(V_b, V_g)$$

- Helpful representation of tunneling spectroscopy:



Nanomechanical oscillations in a single-C_{60} transistor

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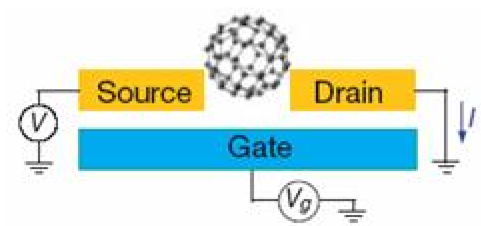

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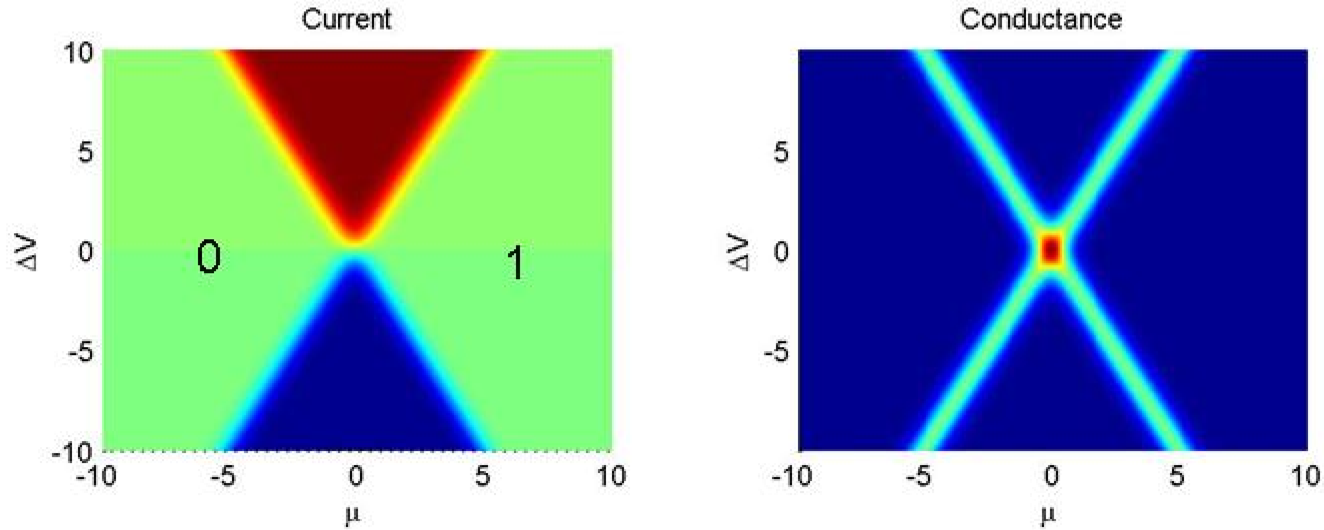
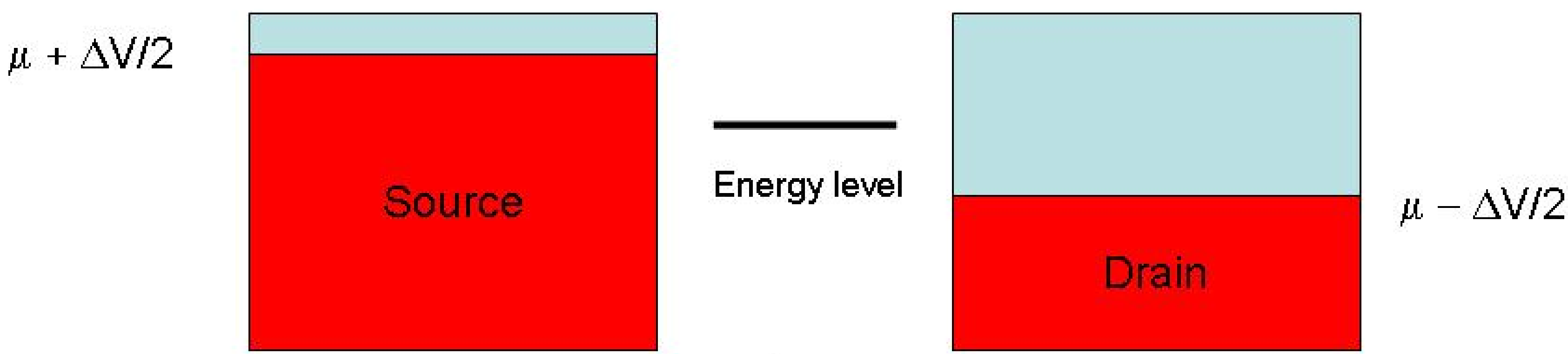
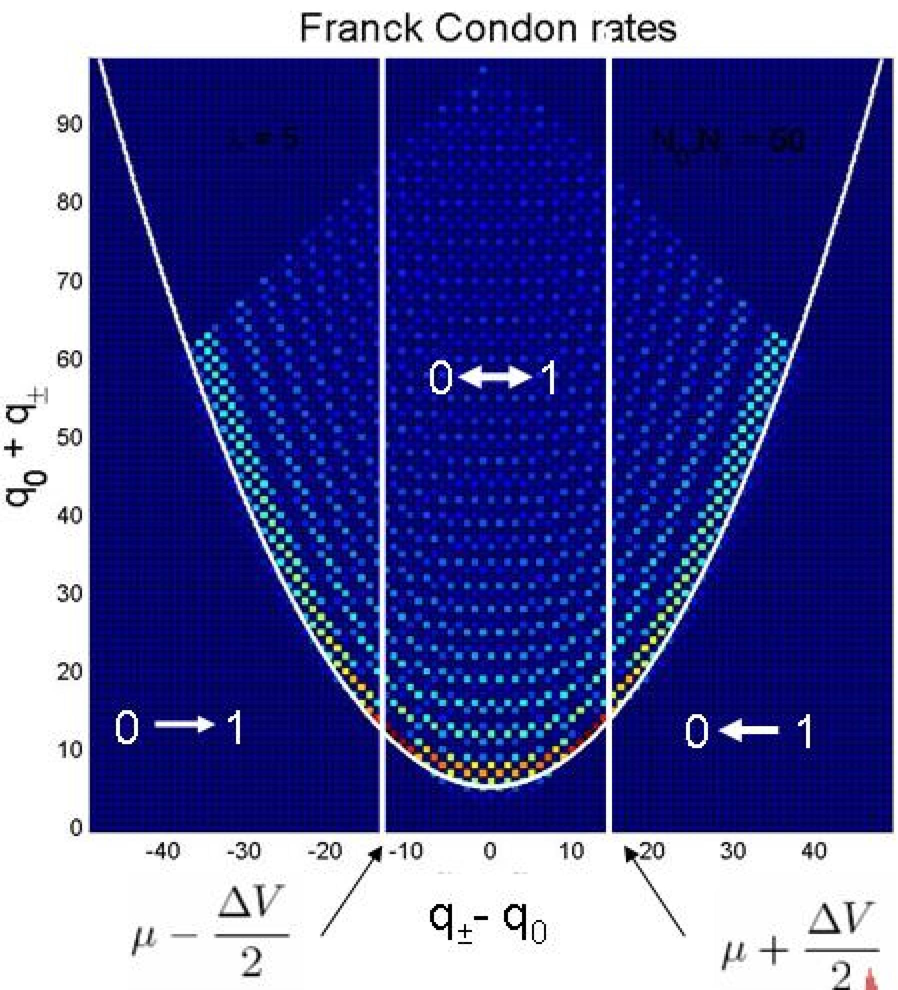
Single level





Franck-Condon parabola (II)



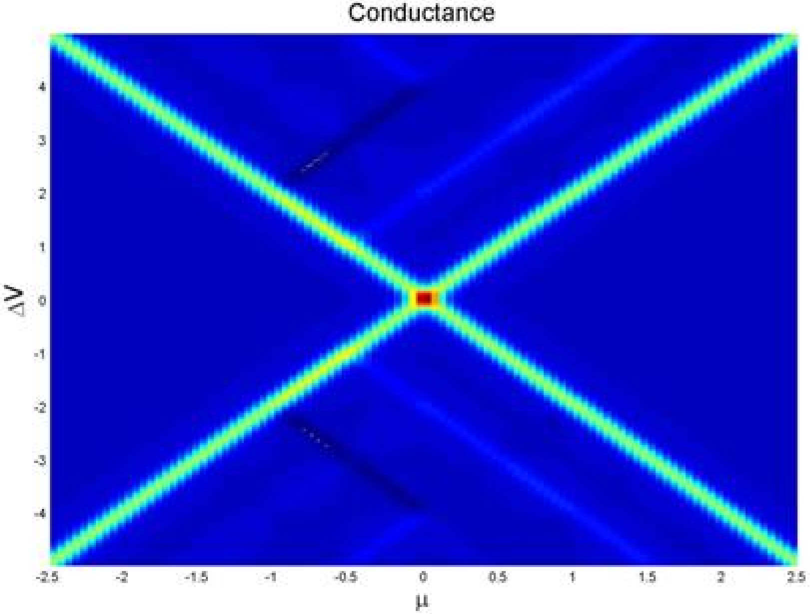
Incoming electrons

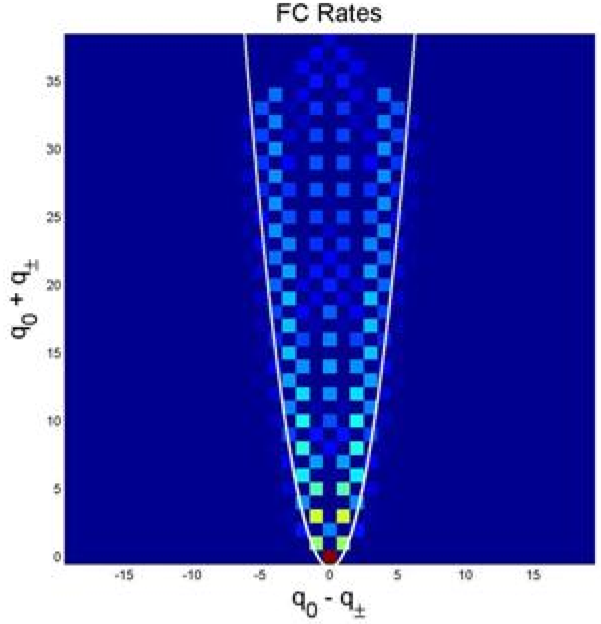
$$\mu_L = \mu + \frac{\Delta V}{2} \geq \epsilon_1 - \epsilon_0 + (q_\pm - q_0)\hbar\omega$$

$$\mu_R = \mu - \frac{\Delta V}{2} \leq \epsilon_1 - \epsilon_0 + (q_\pm - q_0)\hbar\omega$$

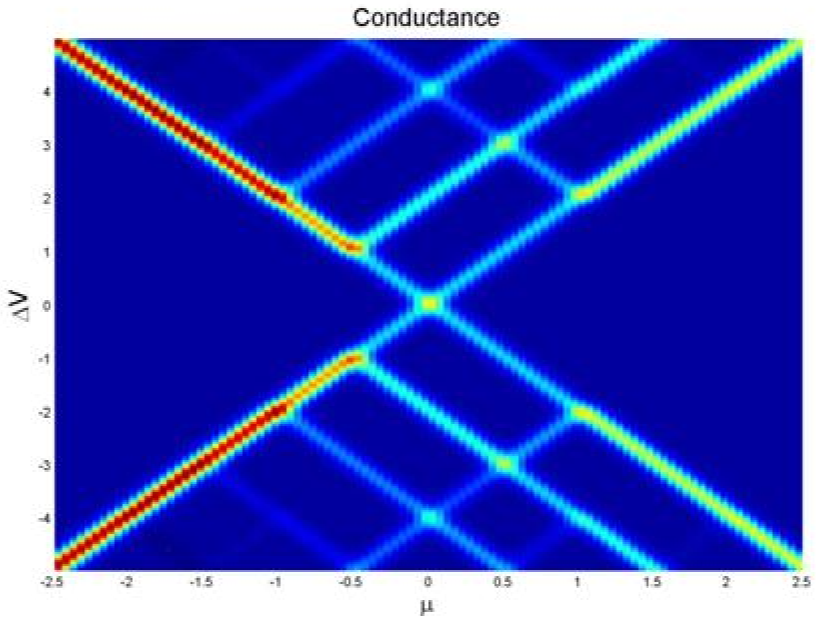
Outgoing electrons

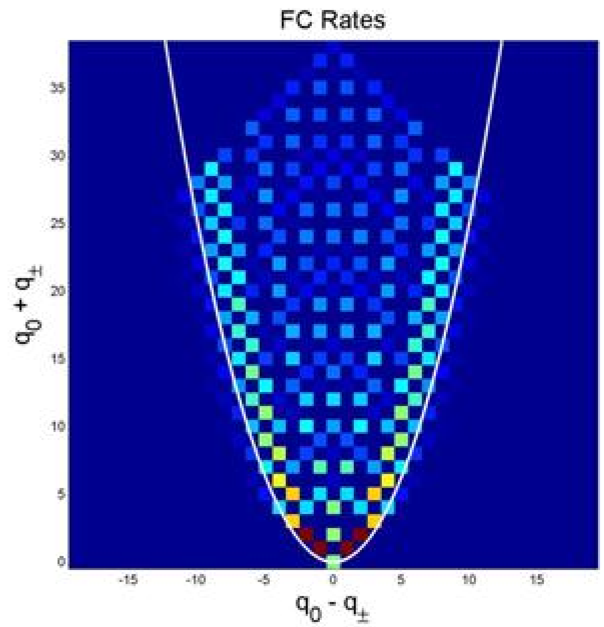
Weak coupling $\lambda = 1$



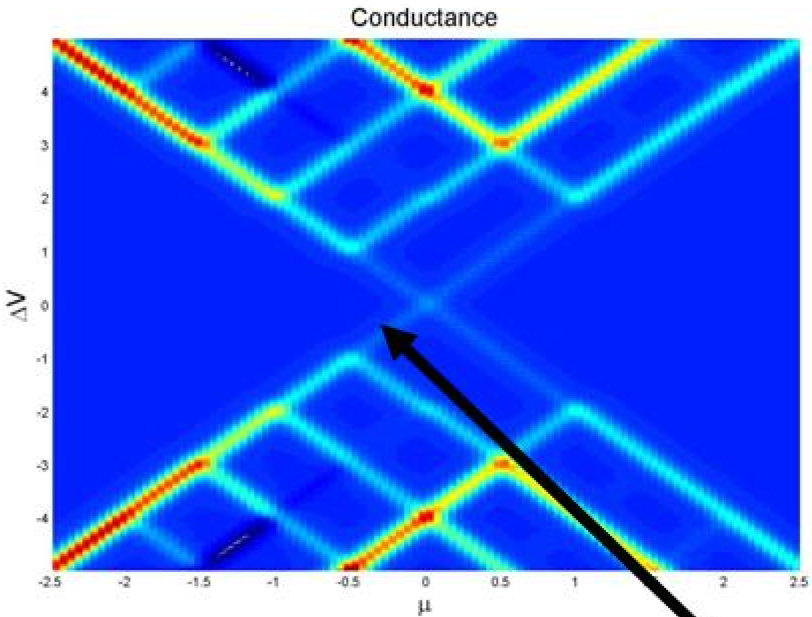


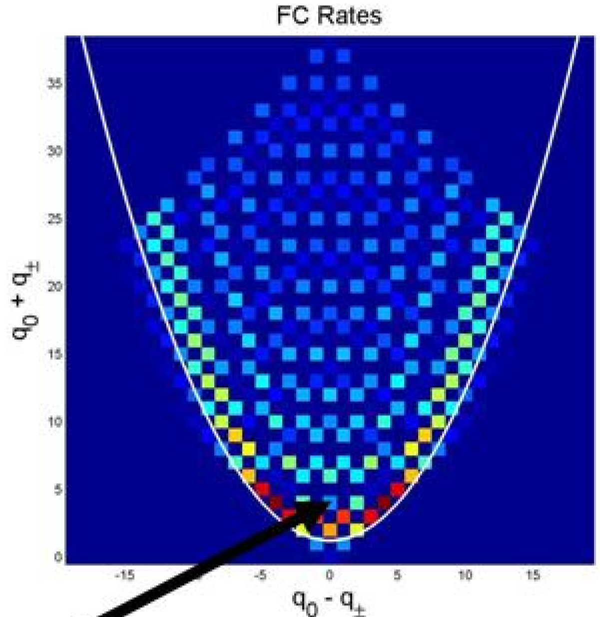
$\lambda = 2$





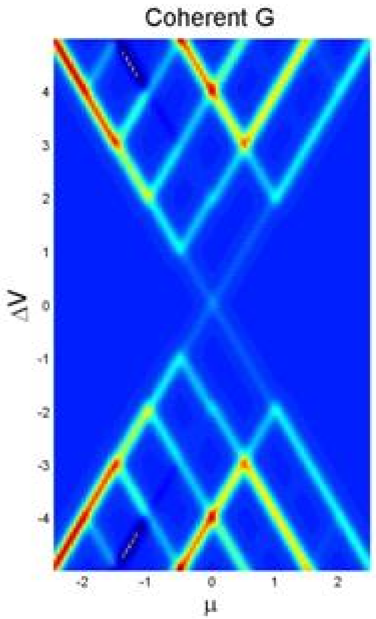
$\lambda = 3$

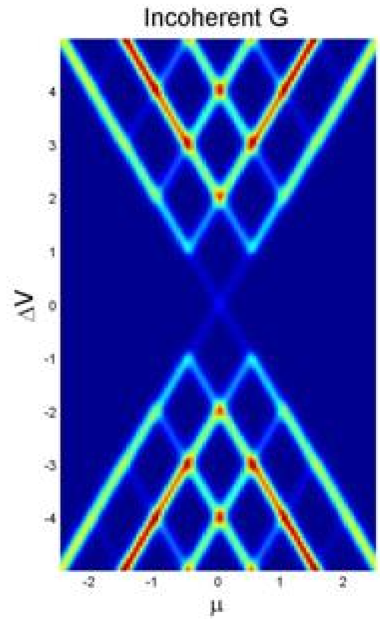




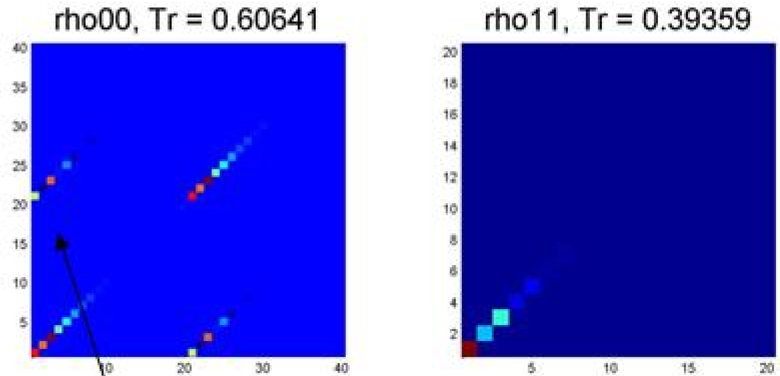
Franck Condon Blockade

Coherences...examples





$\lambda = 1 \quad \Delta V = 5, \ \mu = -1$

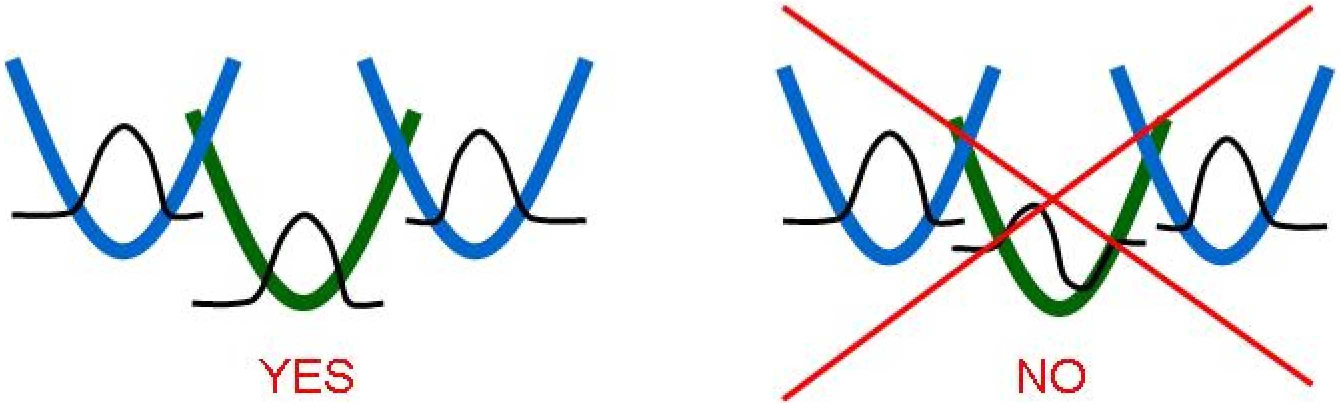


Conductance evaluated neglecting coherences

coherences

Coherences...proof

- Tunneling preserves parity of the mechanical state
- In the even and odd sector degeneracies are lifted and a the stationary reduced density matrix is diagonal



- Franck-Condon coefficients are different in the even and odd sectors and give different stationary solutions
- Thus in the + / - basis we must have coherences

Summary

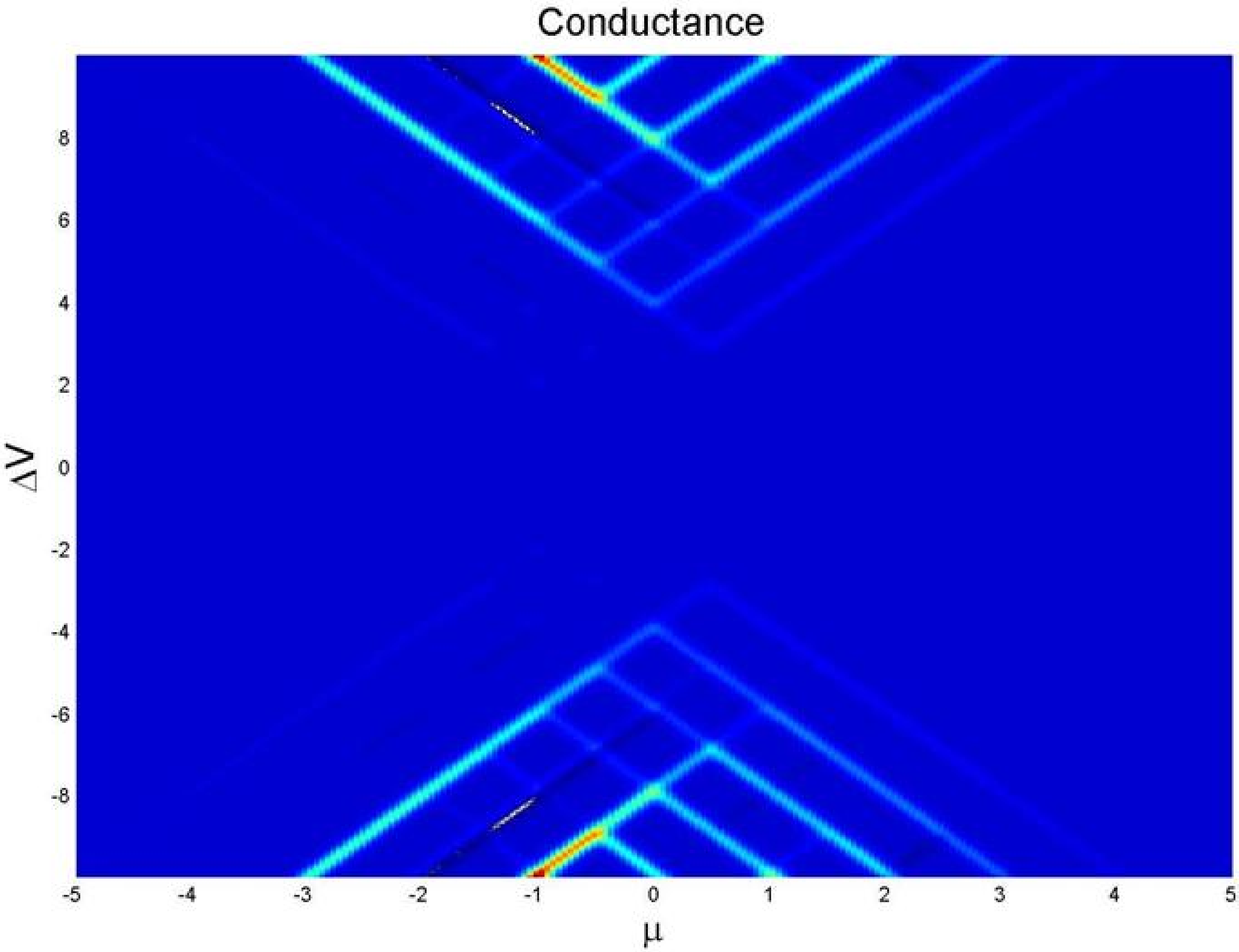
- We derived an effective Hamiltonian for the biphenyl transistor
- We obtained the description of the electromechanical dynamics in terms of a GME
- We observed the Franck-Condon blockade and gave an interpretation in terms of the Franck-Condon rates
- We observed the relevance of the mechanical coherences in the transport mechanism

Still (a lot) to do!

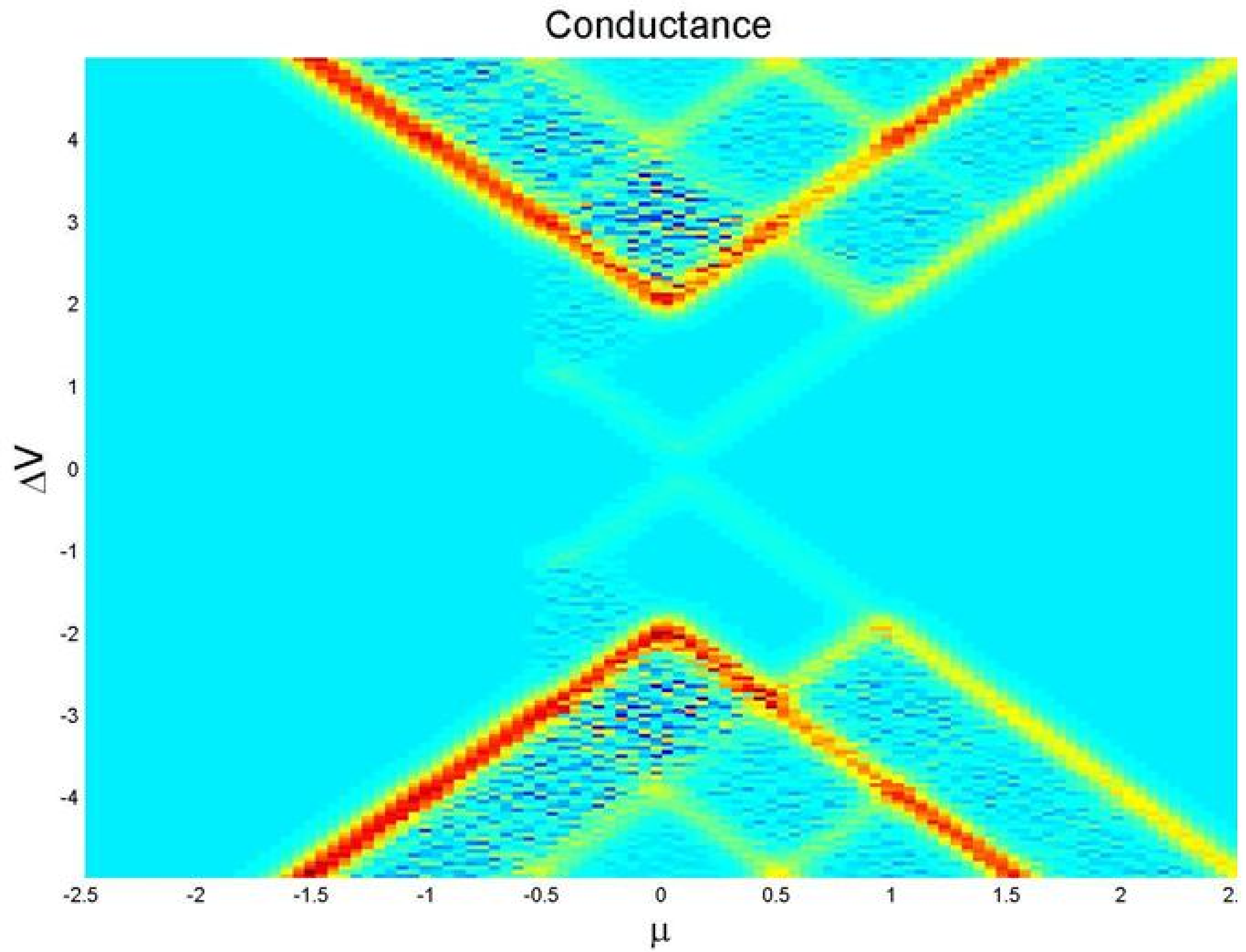
- Understand the anionic potential surface in terms of the PPP Hamiltonian
- Understand the role of mechanical coherences in transport through biphenyl
- Use realistic values for the electron-vibron coupling in biphenyl ($\lambda \sim 50$)
- Investigate the role of the mechanical bath

Thanks for your attention!

$\lambda = 5$



Bath ($\lambda = 3$)



Lambda scaling