Quantum Transport and Dissipation Theoretical Physics

Electromechanical properties of a biphenyl transistor

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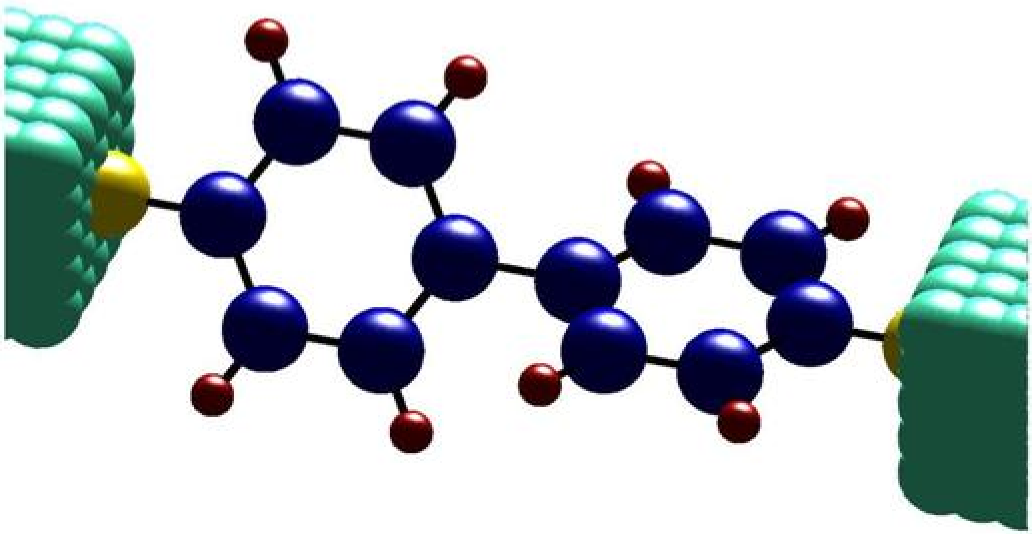
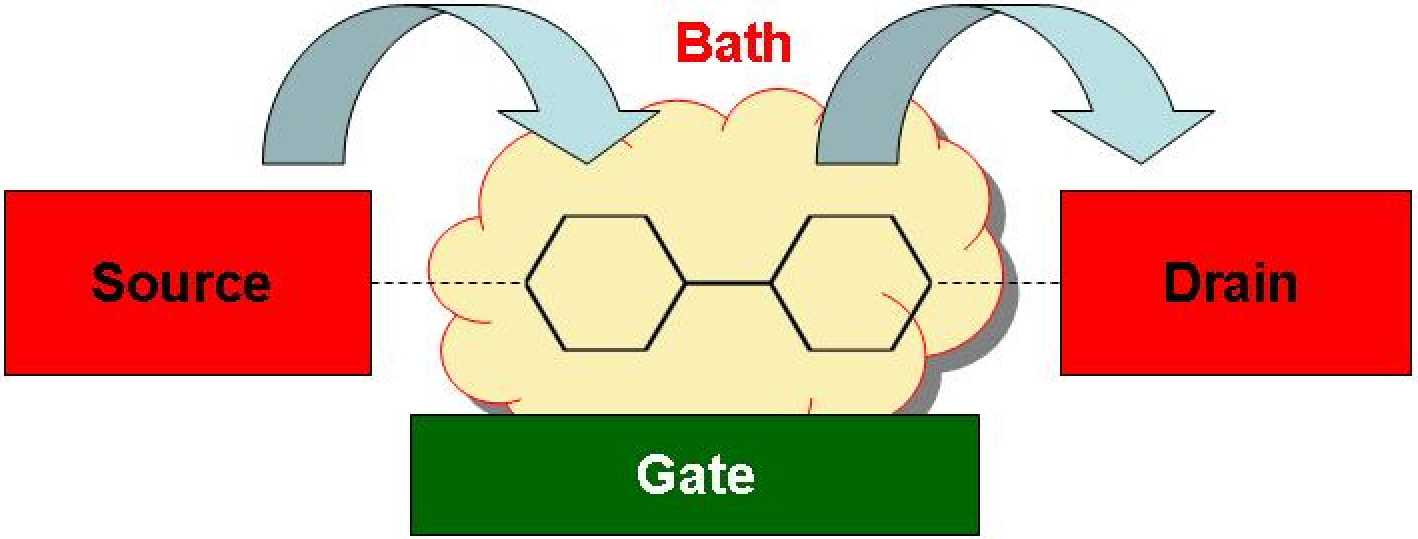


The model



- 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

$$H = H_{\mathrm{S}} + H_{\mathrm{EL}} + H_{\mathrm{B}} + V_{\mathrm{T}} + V_{\mathrm{SB}}$$

where

$$H_{\mathrm{S}} = \frac{\hat{p}^2}{2m} + |0\rangle\langle 0|\, V_0(\hat{x}) + |1\rangle\langle 1|\, V_1(\hat{x})$$

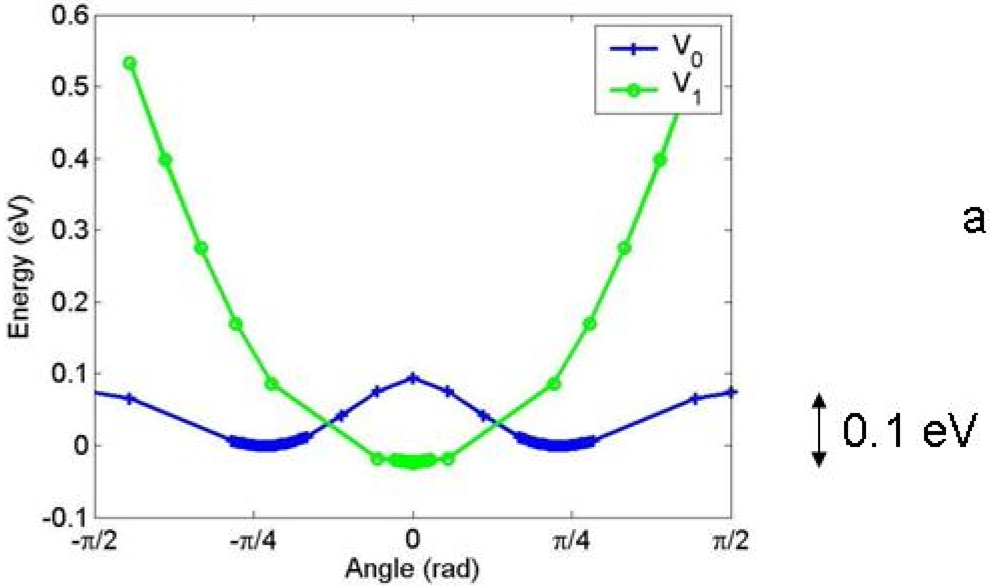
and

$$V_{\mathrm{T}} = t \sum_{k;\alpha=\mathrm{L,R}} \left(|0\rangle\langle 1| c^{\dagger}_{k\alpha} + |1\rangle\langle 0| c_{k\alpha} \right)$$

$$V_{\mathrm{SB}} = \hbar\tilde{g} \sum_{q} \hat{x}(d_q + d_q^{\dagger})$$

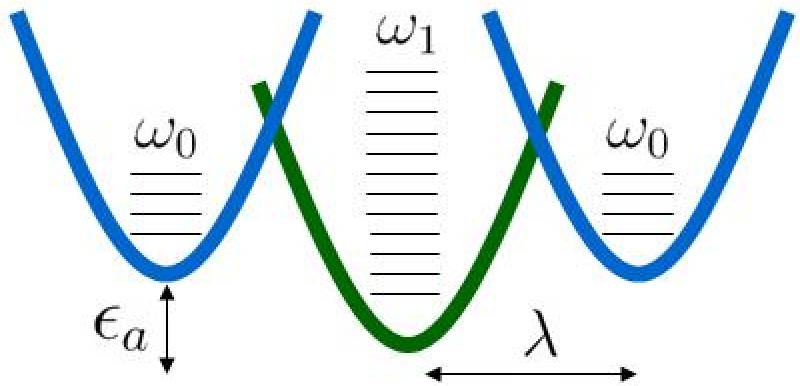
Adiabatic Potentials

- Tilted equilibrium configurations for the neutral molecule
- Planar equilibrium configuration for the anionic molecule



DFT calculations
with gaussian basis set
and B3LYP exchange potential

A generic model



- Harmonic approximation for the anionic state
- Low laying states of a double (harmonic) well potential for the neutral state

- The effective Hilbert space is defined by the unity operator:

$$\mathbb{1} = |1\rangle\langle 1| + |0\rangle\langle 0|(\mathcal{P}_+ + \mathcal{P}_-)$$

where

$$\mathcal{P}_\pm = \sum_{n=0}^{N} |n,\pm\rangle\langle n,\pm|$$

Effective Hamiltonian

The only components of the Hamiltonian modified by the projection are:

$$H_{\mathrm{S}}^{eff} = |0\rangle\langle 0| \sum_{\tau=+,-} \left[\mathcal{P}_\tau(\tfrac{1}{2} + d_\tau^\dagger d_\tau)\mathcal{P}_\tau\right] \hbar\omega_0 + |1\rangle\langle 1| \left[(\tfrac{1}{2} + d^\dagger d)\hbar\omega_1 + eV_g - \epsilon_a\right]$$

and

$$V_{\mathrm{T}}^{eff} = \sum_{k;\alpha=\mathrm{L,R}} \left[t|0\rangle\langle 1|c_{k\alpha}^\dagger(\mathcal{P}_+ + \mathcal{P}_-) + h.c.\right]$$

$$V_{\mathrm{SB}}^{eff} = \hbar g \sum_q (d_q + d_q^\dagger)\left\{|1\rangle\langle 1|(d + d^\dagger) + |0\rangle\langle 0| \sum_{\tau=-,+} \left[\mathcal{P}_\tau(d_\tau + d_\tau^\dagger + \tau 2\lambda)\mathcal{P}_\tau\right]\right\}$$

Generalized Master Equation

$$\dot{\sigma}(t) = \mathcal{L}\,\sigma(t) = (\mathcal{L}_{\rm coh} + \mathcal{L}_{\rm driv} + \mathcal{L}_{\rm damp})\,\sigma(t)$$

- The Generalized Master Equation is derived in second order perturbation in the interaction Hamiltonians $V_{\rm T}^{eff}$ and $V_{\rm SB}^{eff}$
- Coherences between different charge states vanish
- Due to the mechanical (quasi-)degenerate neutral states we MUST keep coherences between displaced mechanical states

Electrical leads

$$(\mathcal{L}_{\rm driv}\,\sigma)_{11} = \sum_{\alpha,\tau}\Big[2(\Gamma^{\alpha}_{\rm in}\,\sigma_{00}\,\mathcal{P}_\tau + \mathcal{P}_\tau\,\sigma_{00}\,\Gamma^{\alpha\,\dagger}_{\rm in}) - (\mathcal{P}_\tau\,\Gamma^{\alpha}_{\rm out}\,\sigma_{11} + \sigma_{11}\,\Gamma^{\alpha\,\dagger}_{\rm out}\mathcal{P}_\tau)\Big]$$

$$(\mathcal{L}_{\rm driv}\,\sigma)^{\tau\tau'}_{00} = \sum_{\alpha}\mathcal{P}_\tau\Big[\Gamma^{\alpha}_{\rm out}\,\sigma_{11} + \sigma_{11}\,\Gamma^{\alpha\,\dagger}_{\rm out} - 2(\Gamma^{\alpha}_{\rm in}\,\sigma_{00} + \sigma_{00}\,\Gamma^{\alpha\,\dagger}_{\rm in})\Big]\mathcal{P}_{\tau'}$$

where

$$\Gamma^{\alpha}_{\rm in} = \underbrace{\frac{\Gamma_\alpha}{2}}_{\text{bare tunneling rate}}\sum_{m,n\tau}|m\rangle\Big\{\underbrace{f_\alpha[eV_g - \epsilon_a + \hbar\omega(m-n)]}_{\text{Fermi factor}}\underbrace{\langle m|n,\tau\rangle}_{\text{Franck-Condon coefficient}}\Big\}\langle n,\tau|$$

$$\Gamma^{\alpha}_{\rm out} = \frac{\Gamma_\alpha}{2}\sum_{m,n\tau}|n,\tau\rangle\langle m| - \Gamma^{\alpha\,\dagger}_{\rm in}$$

$$\left(\,\mathcal{L}_{\rm coh}\,\sigma = -\frac{i}{\hbar}[H^{eff}_{\rm S},\sigma]\,\right)$$

Mechanical bath

$$(\mathcal{L}_{\text{damp}}\,\sigma)_{11} = -\frac{i\gamma}{2\hbar}[\hat{x},\{\hat{p},\sigma_{11}\}] - \frac{\gamma m\omega}{\hbar}(\bar{N}+\tfrac{1}{2})[\hat{x},[\hat{x},\sigma_{11}]]$$

$$(\mathcal{L}_{\text{damp}}\,\sigma)_{00}^{\tau\tau'} = -\frac{i\gamma}{2\hbar}\mathcal{P}_\tau[\hat{x},\{\hat{p},\sigma_{00}^{\tau\tau'}\}]\mathcal{P}_{\tau'}$$

$$-\frac{\gamma m\omega}{\hbar}(\bar{N}+\tfrac{1}{2})\mathcal{P}_\tau[\hat{x},[\hat{x},\sigma_{00}^{\tau\tau'}]]\mathcal{P}_{\tau'}$$

$$-8\gamma\frac{k_{\text{B}}T}{\hbar\omega}\lambda^2(\mathcal{P}_+\,\sigma_{00}^{\tau\tau'}\,\mathcal{P}_- + \mathcal{P}_-\,\sigma_{00}^{\tau\tau'}\,\mathcal{P}_+)$$

Interwell dephasing term

Observables

- Current:

$$I^{stat} = \mathrm{Tr_S}[\sigma^{stat}\hat{I}_{\mathrm{L}}]$$

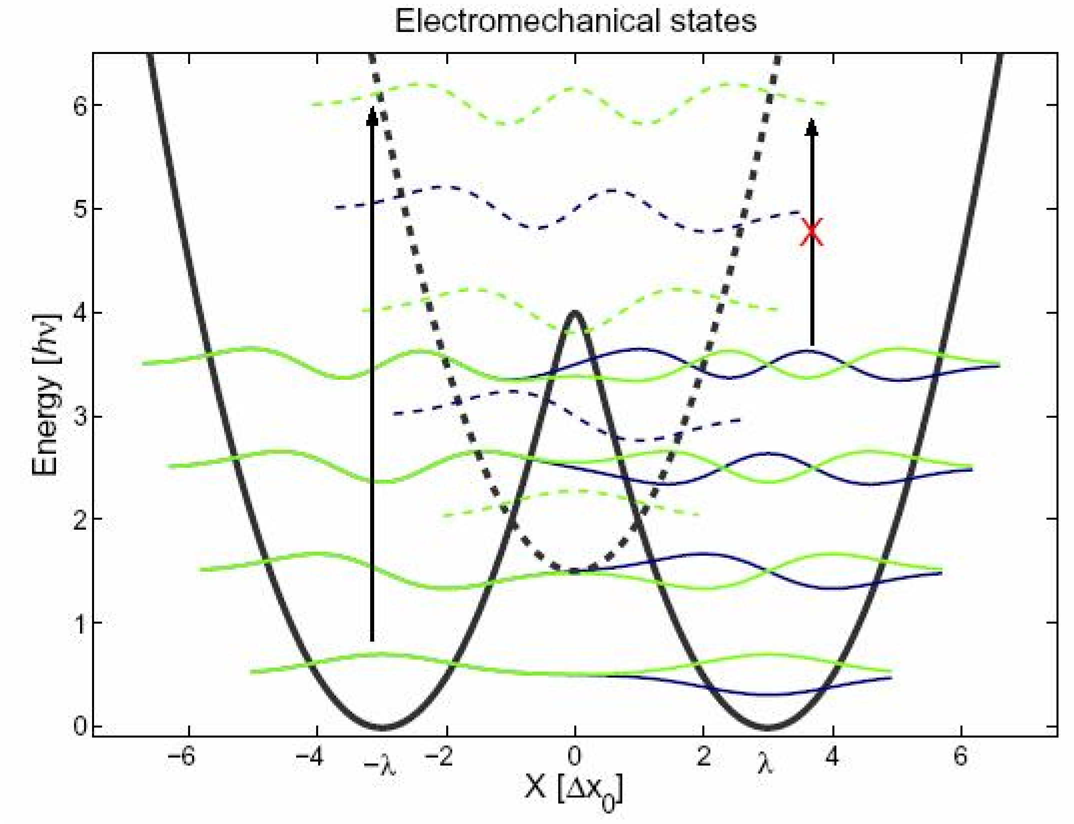
$$\hat{I}_{\mathrm{L}} = \sum_{\tau}\left[2|0\rangle\langle 0|(\mathcal{P}_\tau\,\Gamma^{\mathrm{L}}_{\mathrm{in}} + {\Gamma^{\mathrm{L}}_{\mathrm{in}}}^{\dagger}\mathcal{P}_\tau) - |1\rangle\langle 1|(\mathcal{P}_\tau\,\Gamma^{\mathrm{L}}_{\mathrm{out}} + {\Gamma^{\mathrm{L}}_{\mathrm{out}}}^{\dagger}\mathcal{P}_\tau)\right]$$

- Parity:

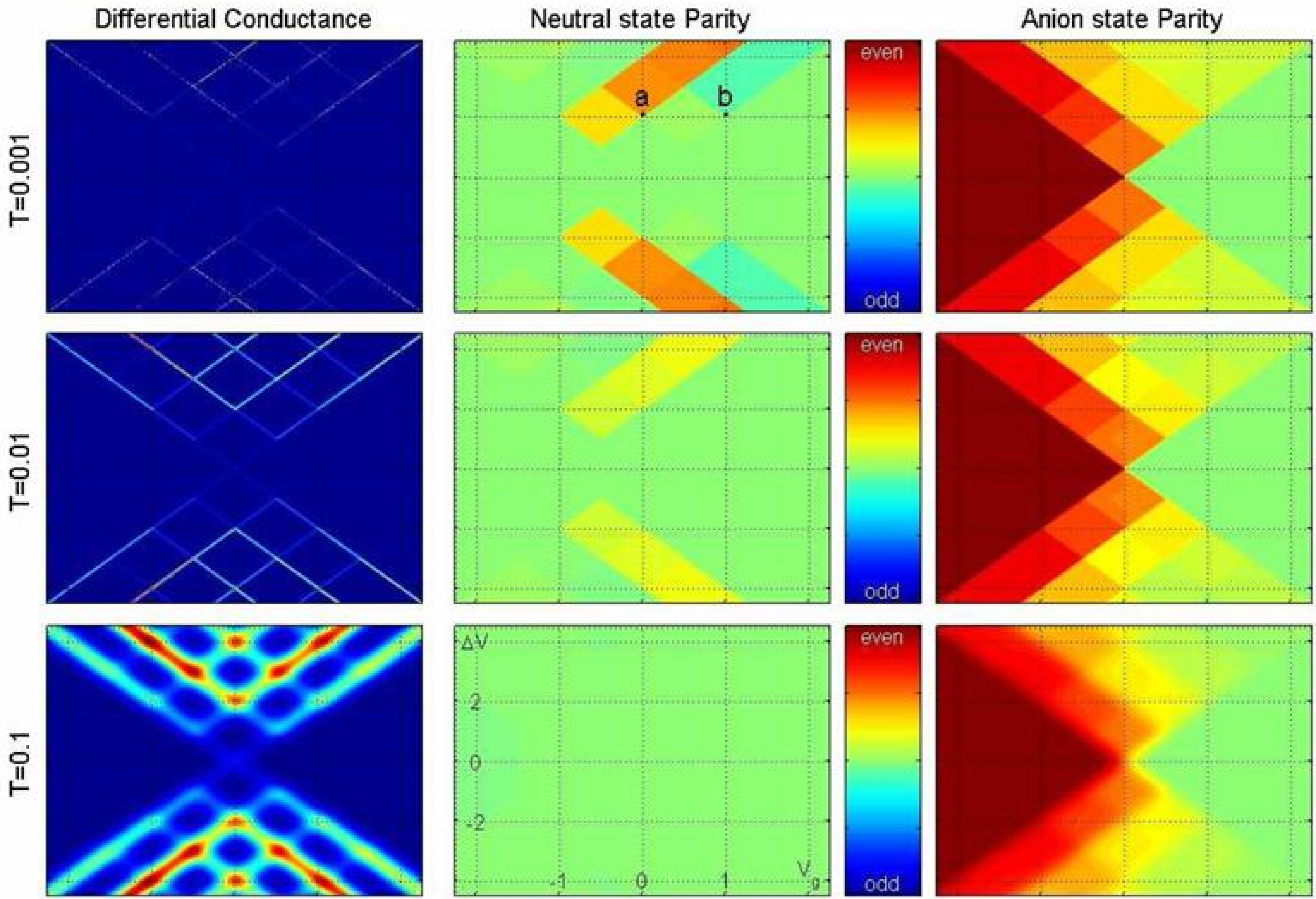
$$P_0 = \sum_{n=0}^{N}\langle 0,e,n|\sigma^{stat}|0,e,n\rangle - \langle 0,o,n|\sigma^{stat}|0,o,n\rangle$$

$$P_1 = \sum_{n=0}^{\infty}\langle 1,2n|\sigma^{stat}|1,2n\rangle - \langle 1,2n+1|\sigma^{stat}|1,2n+1\rangle$$

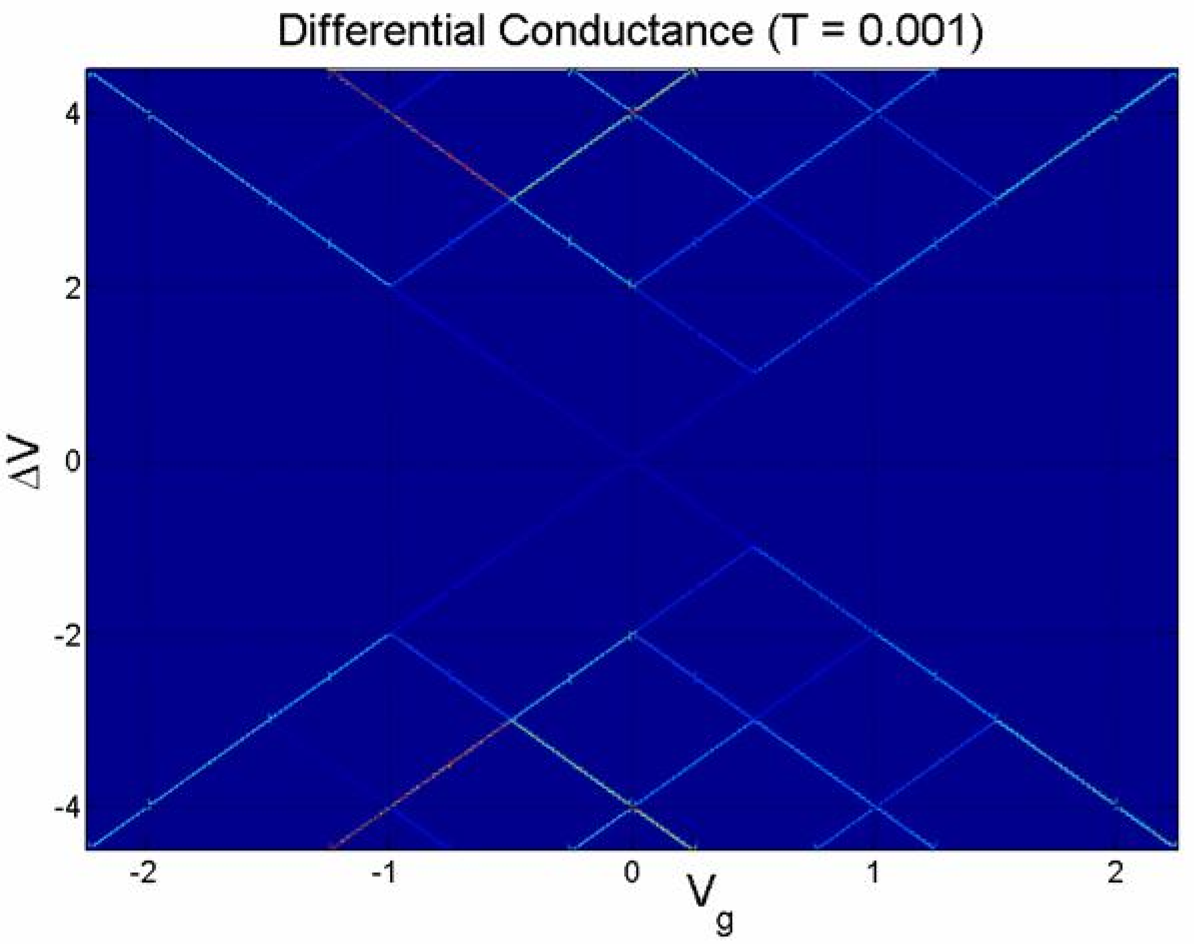
Electromechanical states



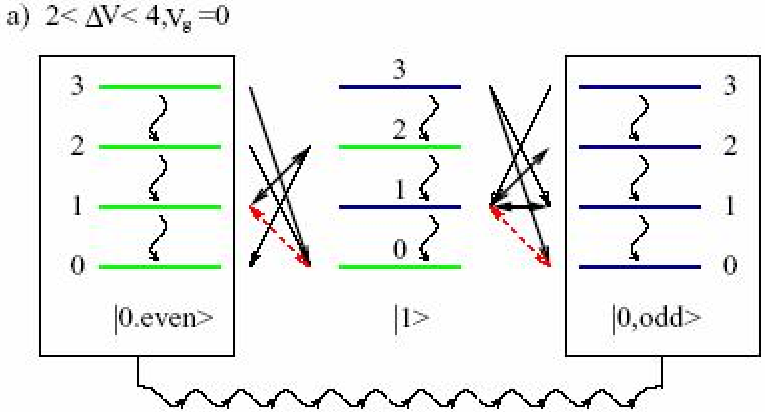
Symmetry breaking

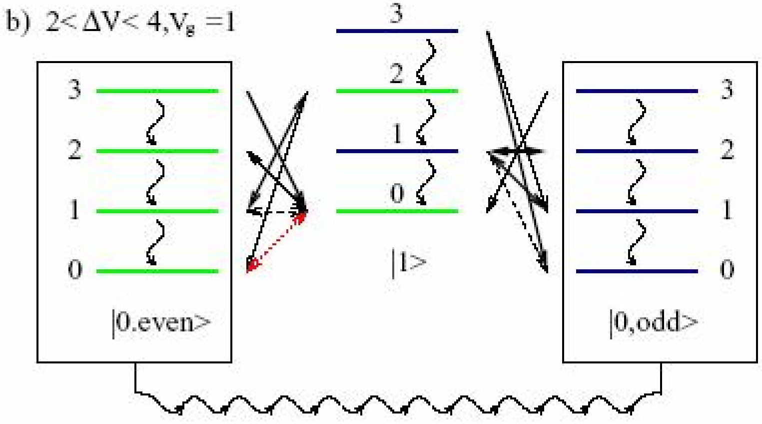


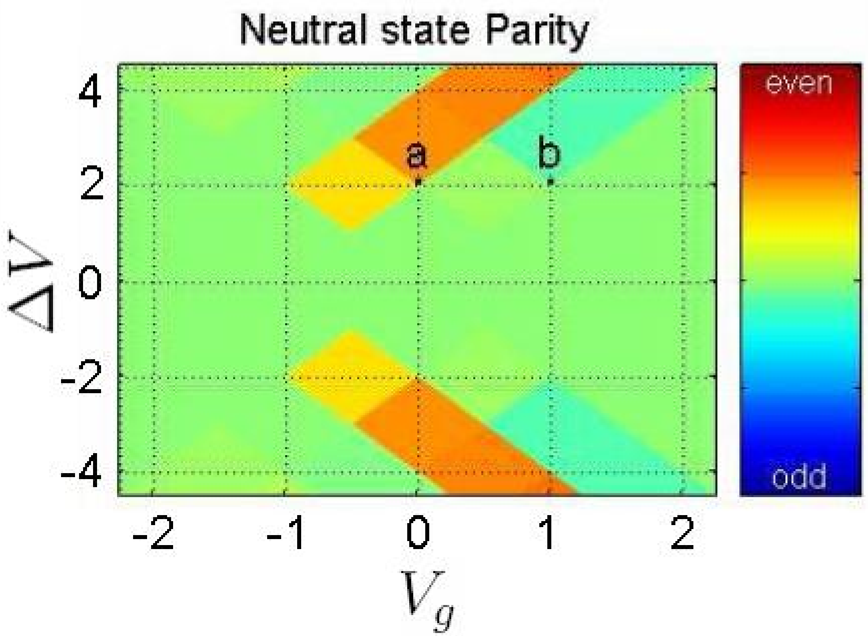
Gate voltage asymmetry



Un-blocking rates







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 parity dynamical symmetry-breaking in different bias and gating conditions
- We investigated the relevance of the mechanical coherences in the transport mechanism: i.e. gate voltage asymmetry of the differential conductance

Thanks for your attention...

Lambda scaling