Dynamical symmetry breaking in transport through molecules

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Even or Odd?

Current

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The system

- Weak coupling to the leads + low temperature $\rightarrow$ Coulomb blockade
- Gate voltage: a three terminal device
- Molecule can move: consider the softest mode
The Hamiltonian

The Hamiltonian of the device can be written as

\[ H = H_S + H_{EL} + H_B + V_T + V_{SB} \]

where

\[ H_S = \frac{\hat{p}^2}{2m} + \langle 0 \vert 0 \rangle V_0(\hat{x}) + \langle 1 \vert 1 \rangle V_1(\hat{x}) \]

and

\[ V_T = t \sum_{k; \alpha = L, R} \left( \langle 0 \vert 1 \rangle c_{k\alpha}^\dagger + \langle 1 \vert 0 \rangle c_{k\alpha} \right) \]

\[ V_{SB} = \hbar \tilde{g} \sum_q \hat{x}(d_q + d_q^\dagger) \]
Symmetry of the adiabatic potentials

\[ V_0(x) \quad \text{and} \quad V_1(x) \]

\[ \lambda \]

\[ \omega_0 \quad \omega_1 \]

\[ \epsilon_a \]

energy

mechanical coordinate

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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:

\[ 1 = |1\rangle\langle1| + |0\rangle\langle0|(P_+ + P_-) \]

where

\[ P_\pm = \sum_{n=0}^{N} |n, \pm\rangle\langle n, \pm| \]
Effective Hamiltonian

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

\[ H_{S}^{\text{eff}} = |0\rangle\langle 0| \sum_{\tau=+, -} \left[ \mathcal{P}_{\tau} \left( \frac{1}{2} + d_{\tau}^\dagger d_{\tau} \right) \mathcal{P}_{\tau} \right] \hbar \omega_{0} + |1\rangle\langle 1| \left[ \left( \frac{1}{2} + d_{\tau}^\dagger d_{\tau} \right) \hbar \omega_{1} + eV_{g} - \epsilon_{a} \right] \]

and

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

\[ V_{SB}^{\text{eff}} = \hbar g \sum_{q} (d_{q} + d_{q}^\dagger) \left\{ |1\rangle\langle 1|(d + d_{q}^\dagger) + |0\rangle\langle 0| \sum_{\tau=-, +} \left[ \mathcal{P}_{\tau}(d_{\tau} + d_{\tau}^\dagger + \tau 2\lambda) \mathcal{P}_{\tau} \right] \right\} \]
The Generalized Master Equation is derived in second order perturbation in the interaction Hamiltonians $V_{TT}$ and $V_{SB}^{eff}$.

- Coherences between different charge states vanish.
- Due to the mechanical (quasi-)degenerate neutral states we MUST keep coherences between displaced mechanical states.
Electrical leads

\[
\begin{align*}
(L_{\text{driv}} \sigma)_{11} &= \sum_{\alpha, \tau} \left[ 2(\Gamma_{\text{in}}^\alpha \sigma_{00} P_{\tau} + P_{\tau} \sigma_{00} \Gamma_{\text{in}}^{\alpha \dagger}) - (P_{\tau} \Gamma_{\text{out}}^\alpha \sigma_{11} + \sigma_{11} \Gamma_{\text{out}}^{\alpha \dagger} P_{\tau}) \right] \\
(L_{\text{driv}} \sigma)_{00}^{\tau \tau'} &= \sum_{\alpha} P_{\tau} \left[ \Gamma_{\text{out}}^\alpha \sigma_{11} + \sigma_{11} \Gamma_{\text{out}}^{\alpha \dagger} - 2(\Gamma_{\text{in}}^\alpha \sigma_{00} + \sigma_{00} \Gamma_{\text{in}}^{\alpha \dagger}) \right] P_{\tau'}
\end{align*}
\]

where

bare tunneling rate

\[
\Gamma_{\text{in}}^\alpha = \frac{\Gamma_{\alpha}}{2} \sum_{m, n, \tau} |m\rangle \left\{ f_{\alpha} [eV_g - \epsilon_{\alpha} + \hbar \omega (m - n)] \langle m | n, \tau \rangle \right\} \langle n, \tau |
\]

\[
\Gamma_{\text{out}}^\alpha = \frac{\Gamma_{\alpha}}{2} \sum_{m, n, \tau} |n, \tau \rangle \langle m| - \Gamma_{\text{in}}^{\alpha \dagger}
\]

Fermi factor

Franck-Condon coefficient

\[
(L_{\text{coh}} \sigma = -\frac{i}{\hbar} [H_{\text{eff}}, \sigma])
\]
Mechanical bath

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

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

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

\[
- 8\gamma \frac{k_B T}{\hbar \omega} \lambda^2 (\mathcal{P}_+ + \sigma_{00}^{\tau\tau'} \mathcal{P}_- + \mathcal{P}_- \sigma_{00}^{\tau\tau'} \mathcal{P}_+)
\]

Interwell dephasing term
Observables

- **Current:**

  \[
  I^{\text{stat}} = \text{Tr}_S[\sigma^{\text{stat}} \hat{I}_L]
  \]

  \[
  \hat{I}_L = \sum_{\tau} \left[ 2|0\rangle\langle0| (P_{\tau} \Gamma_{\text{in}}^L + \Gamma_{\text{in}}^L \dagger P_{\tau}) - |1\rangle\langle1| (P_{\tau} \Gamma_{\text{out}}^L + \Gamma_{\text{out}}^L \dagger P_{\tau}) \right]
  \]

- **Parity:**

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

  \[
  P_1 = \sum_{n=0}^{\infty} \langle 1, 2n|\sigma^{\text{stat}}|1, 2n \rangle - \langle 1, 2n + 1|\sigma^{\text{stat}}|1, 2n + 1 \rangle
  \]
Electromechanical states
Symmetry breaking
Gate voltage asymmetry
Un-blocking rates

(a) $2 < \Delta V < 4, V_g = 0$

(b) $2 < \Delta V < 4, V_g = 1$

Neutral state Parity

$\Delta V$

$V_g$

|0, even$

|1$

|0, odd$

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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

Scaling

Maxima

Abs max
Rel max
$\exp(-\lambda^2/2)$
$1/(1+\lambda)^{1.2}$
$1/(1+\lambda)^{1.5}$

$\lambda$