Vibrational effects in the conductance through a molecular bridge

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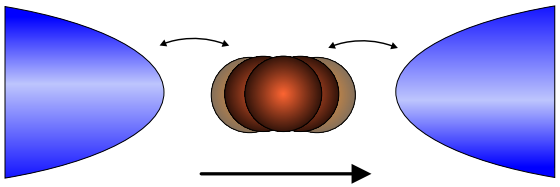
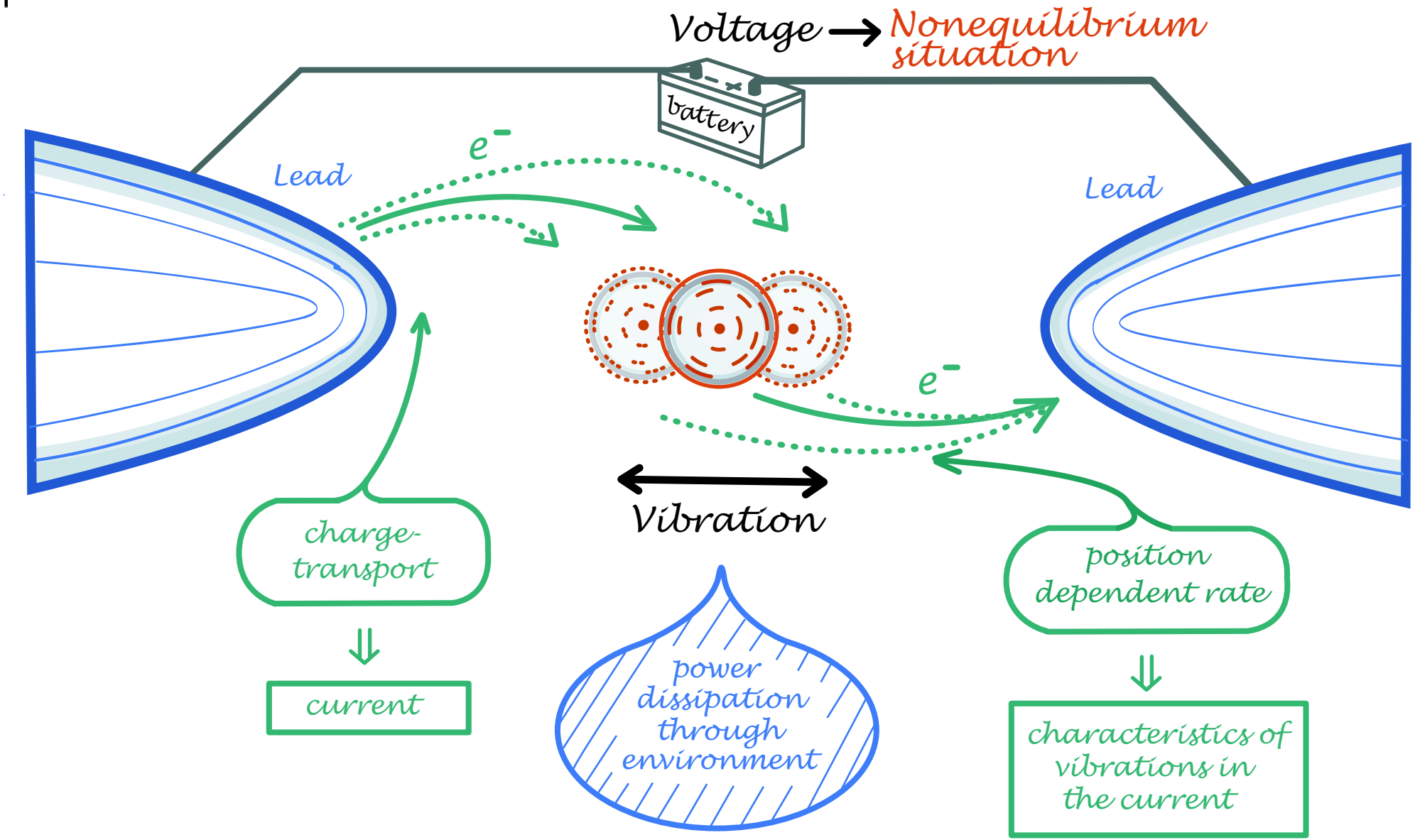
Motivation

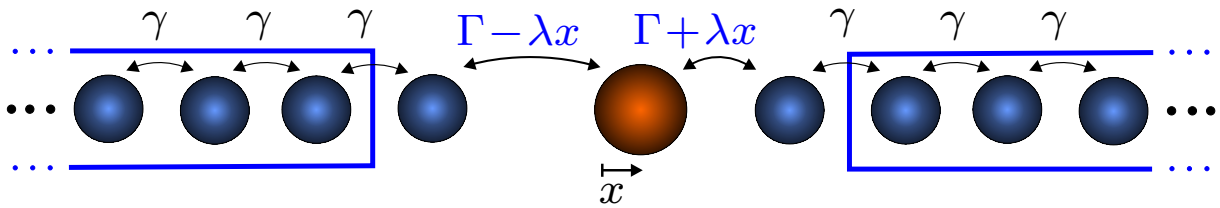
- standard calculations of quantum transport have reached a sophisticated level
- seldom the modelling of molecular vibrations are taken into accout
- importance of vibrations demonstrated
  e.g. by J. van Ruitenbeek, Nature **419**, 906 (2002)
  ('Conductance through a single H_2 molecule')



Model


Voltage → Nonequilibrium situation
battery
Lead
e⁻
Lead
e⁻
Vibration
charge-transport
current
power dissipation through environment
position dependent rate
characteristics of vibrations in the current

Model



$$H = H_{\rm MV} + \hbar\omega_0(b^\dagger b + \tfrac{1}{2}) + H_{\rm leads} + H_{\rm bath}$$

central region coupled to a phonon mode:

$$H_{\rm MV} = \begin{pmatrix} \epsilon_{\rm L} & \Gamma & 0 \\ \Gamma & \epsilon_0 & \Gamma \\ 0 & \Gamma & \epsilon_{\rm R} \end{pmatrix} + \begin{pmatrix} 0 & -\lambda x & 0 \\ -\lambda x & \lambda_0 x & \lambda x \\ 0 & \lambda x & 0 \end{pmatrix}$$

bond, onsite } coupling

$H_{\rm leads}$: semi-infinite linear chains
in a tight-binding description
and coupling to central region

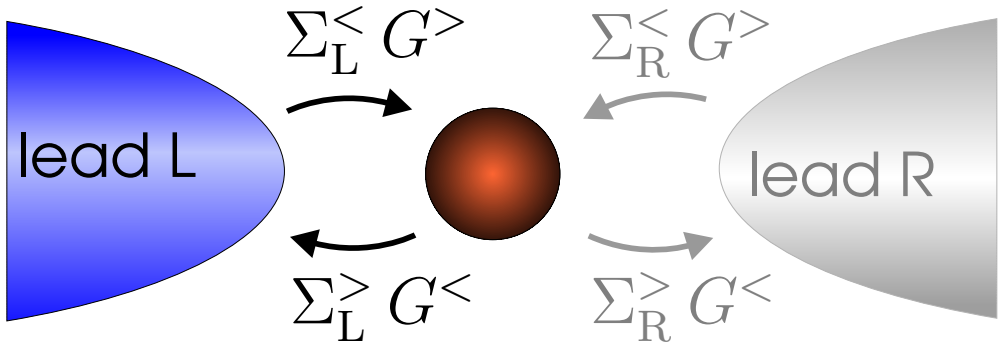
$H_{\rm bath}$: dissipative bath; set of harmonic oscillators
coupled to a phonon mode

Nonequilibrium current

total current: $I = \frac{1}{2}(I_{\mathrm{L}} - I_{\mathrm{R}})$

$$I_\alpha = -2e \left\langle \frac{d}{\mathrm{d}t} N_\alpha \right\rangle$$

$$= \frac{2e}{h} \int \mathrm{d}\epsilon \Big(\Sigma_\alpha^{<}(\epsilon)\, G^{>}(\epsilon) - \Sigma_\alpha^{>}(\epsilon)\, G^{<}(\epsilon) \Big)$$



Meaning of greater and lesser Green functions:

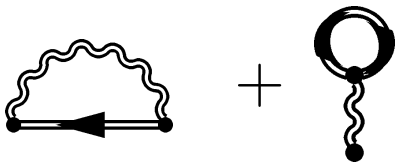
$G^{<}(\epsilon) = \mathrm{i} f(\epsilon) A(\epsilon)$ density of electrons

$G^{>}(\epsilon) = -\mathrm{i}(1 - f(\epsilon)) A(\epsilon)$ density of holes

Dyson equation

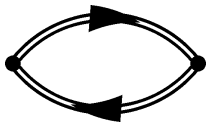
calculation of the electronic Green function:

$$\bar{G} = \bar{G}_0 + \bar{G}_0\big(\bar{\Sigma}_{\rm L} + \bar{\Sigma}_{\rm R} + \bar{\Sigma}_{\rm ph}\big)\bar{G}$$

with $\Sigma_{\rm ph} =$ 

accounting for finite oscillation amplitudes:

$$\bar{D} = \bar{D}_0 + \bar{D}_0\big(\bar{\Pi}_{\rm el} + \bar{\Pi}_{\rm bath}\big)\bar{D}$$

with $\Pi_{\rm el} =$ 

$$\bar{G} = \begin{pmatrix} G^{\rm r} & G^{<} + G^{>} \\ 0 & G^{\rm a} \end{pmatrix}$$

Link to equilibrium theory

approximation for small voltages:

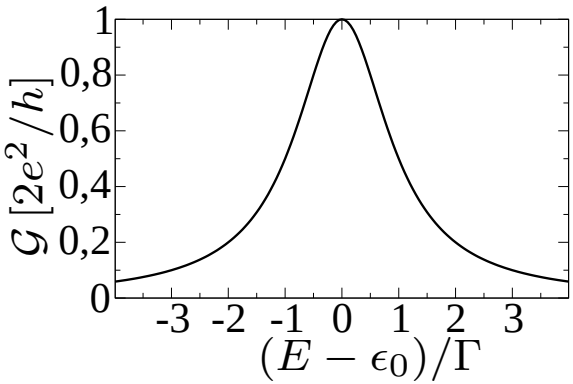
$$I = I_{\mathrm{L}} - I_{\mathrm{R}} = \frac{2e}{h} \int \mathrm{d}\epsilon \left(f_{\mathrm{L}}(\epsilon) - f_{\mathrm{R}}(\epsilon) \right) \underbrace{\mathrm{Tr}\left(G^{\mathrm{a}} \Gamma_{\mathrm{R}} G^{\mathrm{r}} \Gamma_{\mathrm{L}} \right)}_{\text{transmission}}$$

the zero-temperature conductance:

$$\mathcal{G} = \frac{\partial I}{\partial V} = \frac{2e^2}{h} T(\mu) = \frac{2e^2}{h} \mathrm{Tr}\left(G^{\mathrm{a}} \Gamma_{\mathrm{R}} G^{\mathrm{r}} \Gamma_{\mathrm{L}} \right)$$

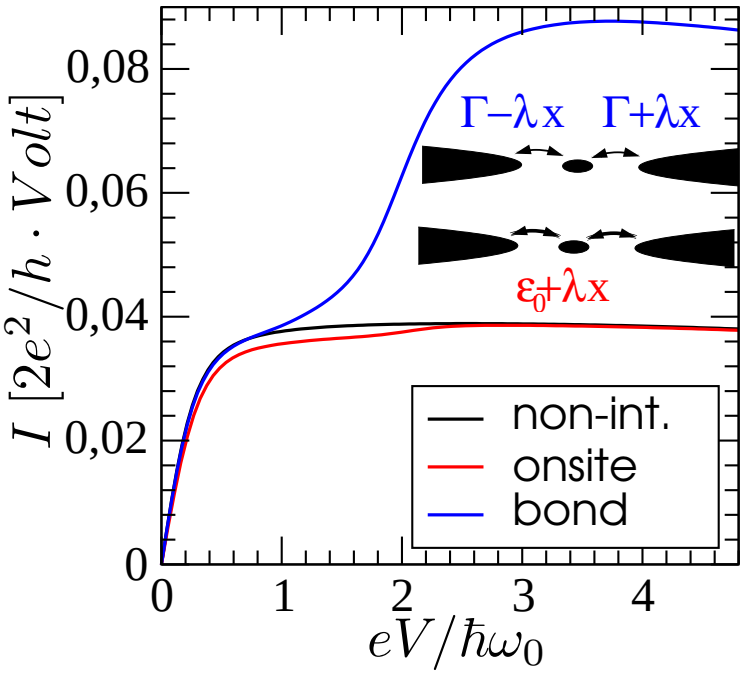
single energy level between two leads:

$$\mathcal{G} = \frac{2e^2}{h} \frac{\Gamma_{\mathrm{L}} \Gamma_{\mathrm{R}}}{(E - \epsilon_0)^2 + (\Gamma_{\mathrm{L}} + \Gamma_{\mathrm{R}})^2/4}$$

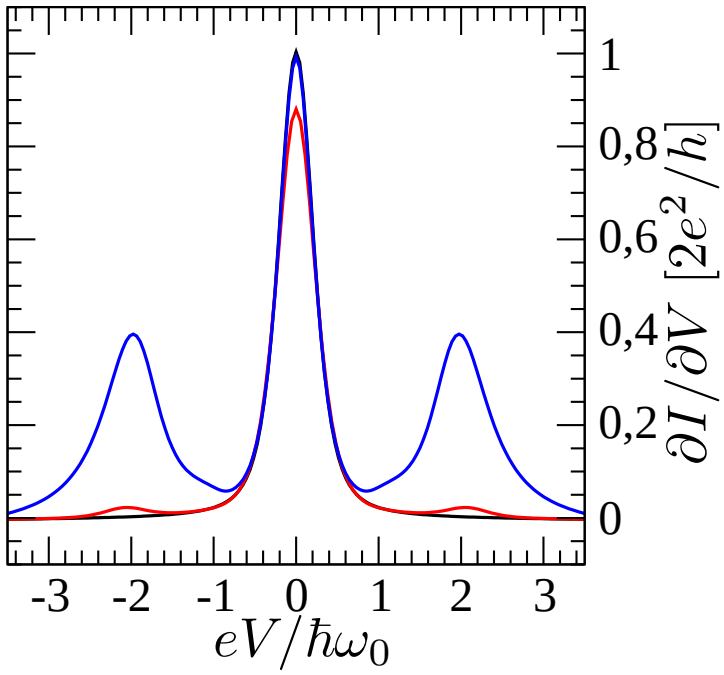


Results I: coupling mechanism

current

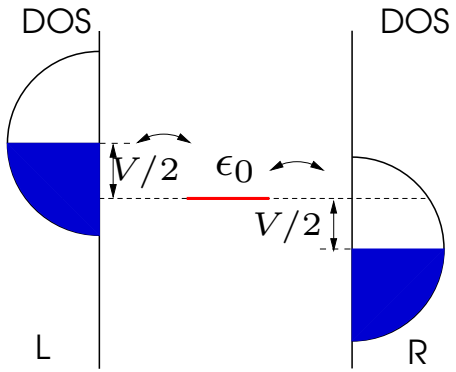


conductance

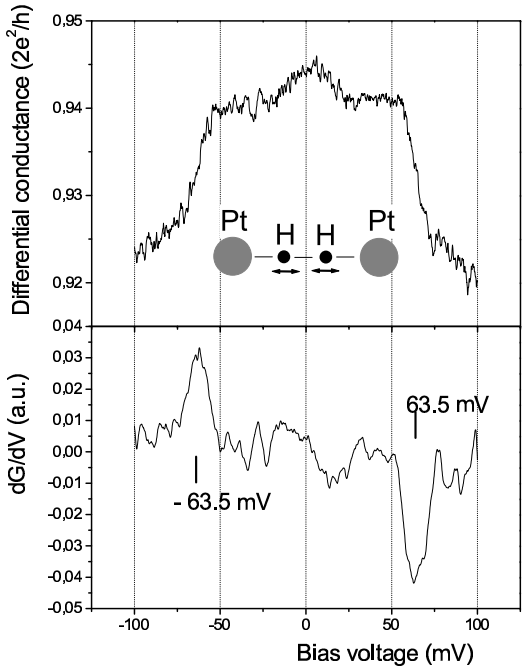


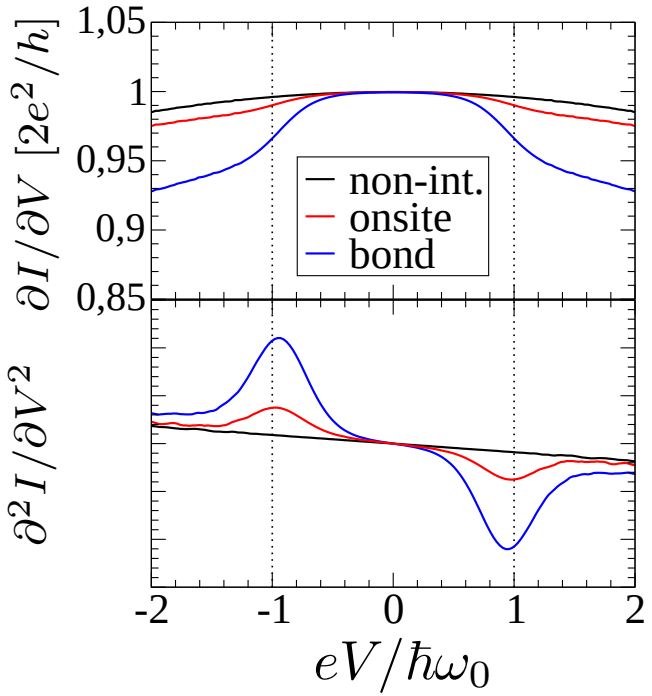
vibrational coupling:

$\lambda = 0.24\ \hbar\omega_0$



Results II: strong coupling regime





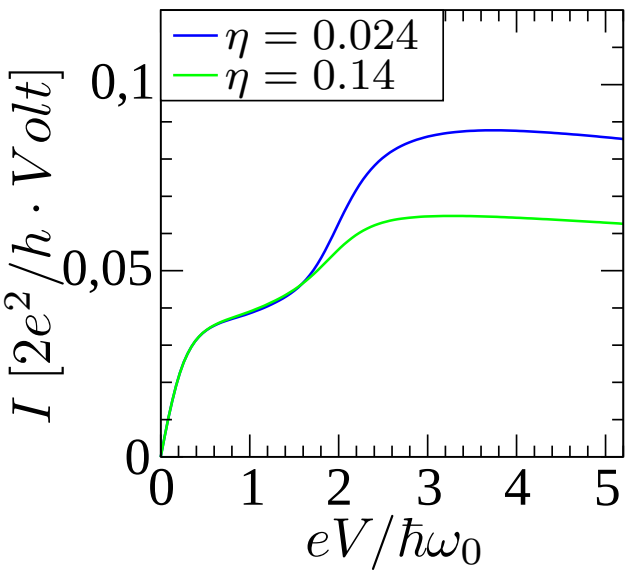
Conductance through a single H_2-molecule.

Measurement by J. van Ruitenbeek, Nature **419**, 906 (2002)

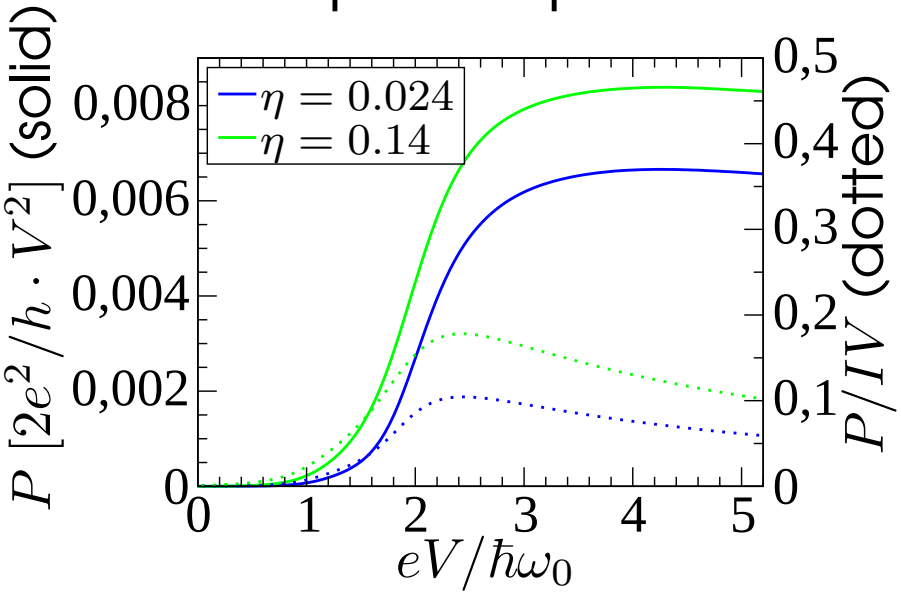
Calculation of the center of mass vibration

Results III: dissipated power

current



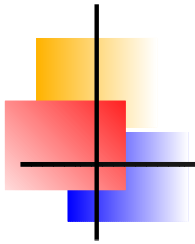
dissipated power



η: coupling strength to dissipative environment

- lead atoms
- other internal molecular vibrations
- surrounding

Modelling of bath: set of harmonic oscillators (ohmic)



Conclusions and outlook

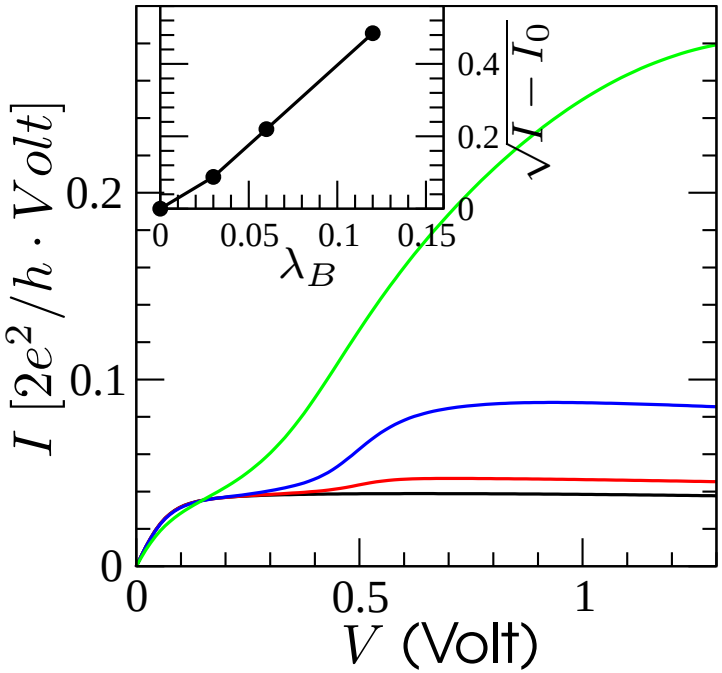
- relevance of bond stretching over onsite electron-phonon coupling
- bond-stretching assisted current
- quantum power $\neq$ classical power

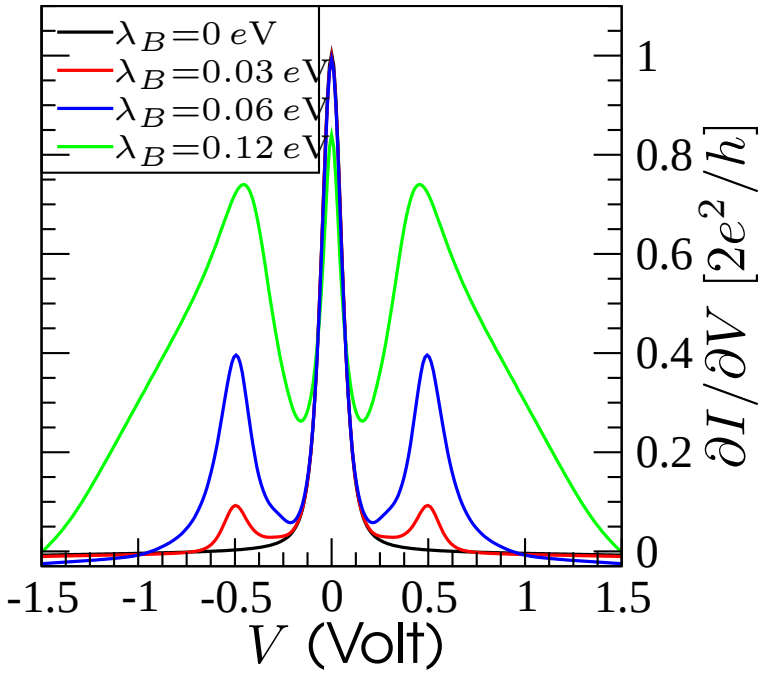
? Transferability to DFT based calculations

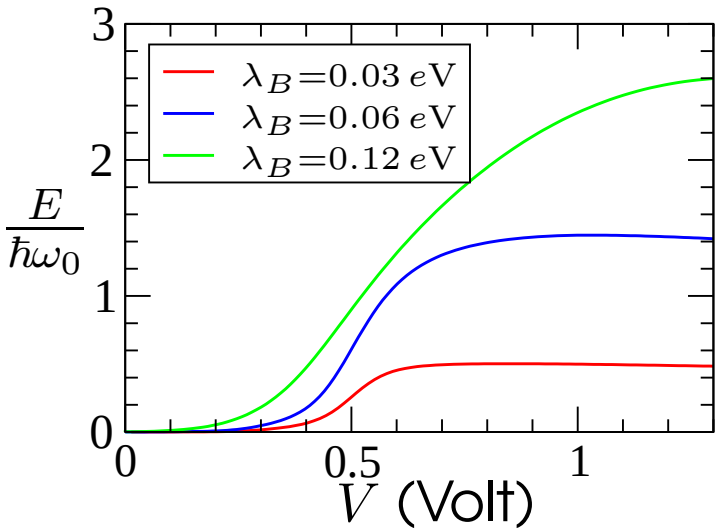
? Strong electron-phonon coupling

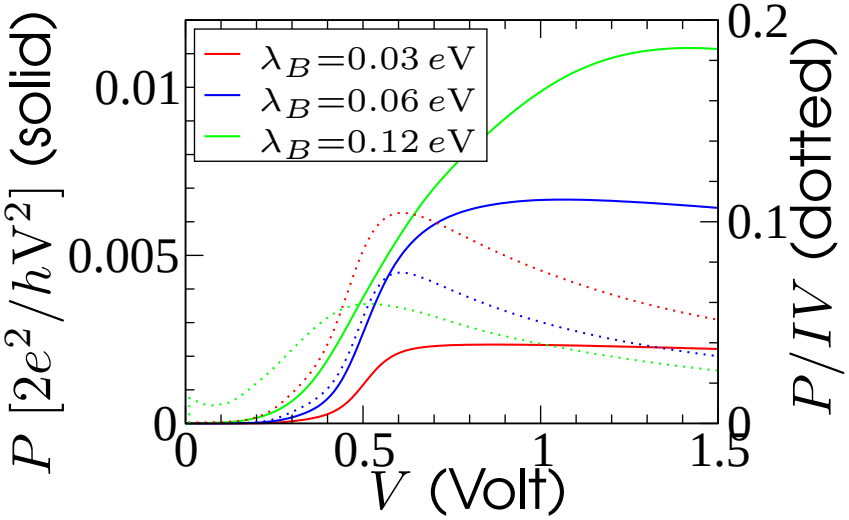
? Coulomb blockade and Kondo regimes

Results IV: coupling strength

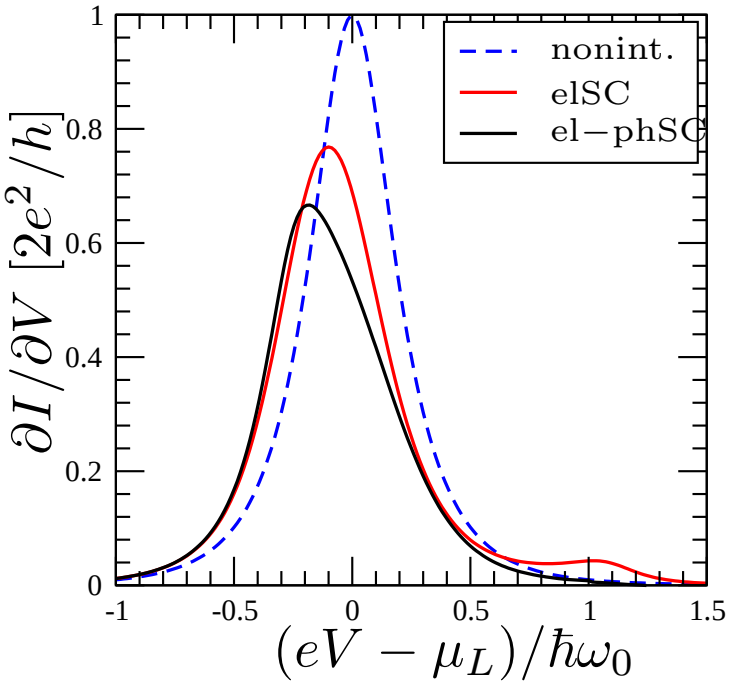


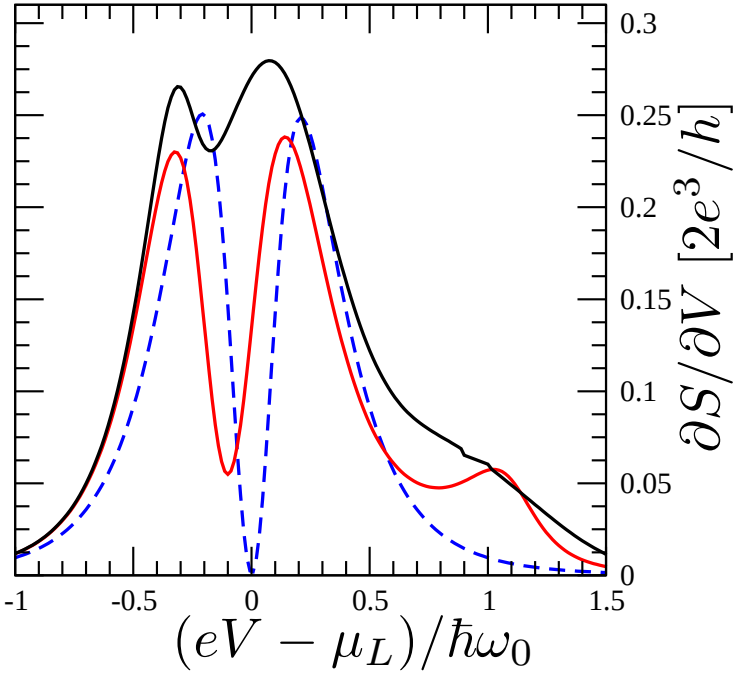






Results V: noise





Results VI: asymmetric