Vibrational effects in the conductance through a molecular bridge

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Aknowledgements:

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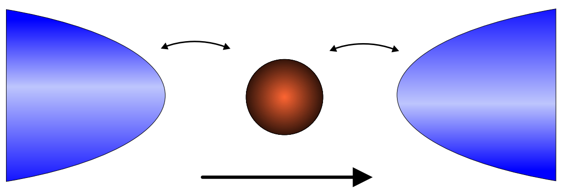
Motivation

- 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')



Our goal

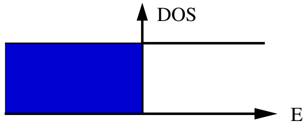
nonequilibrium transport properties of vibrating bridge with finite band leads

$\surd$ method: Keldysh formalism
$\rightarrow$ nonequilibrium inelastic current

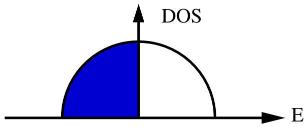
$\surd$ molecule: single level coupled to a bosonic degree of freedom

$\rightarrow$ leads: semi-infinite linear chain:

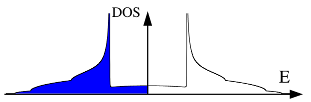
- WBL

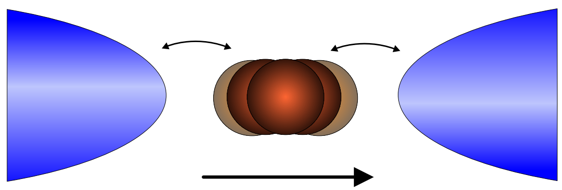


- chain

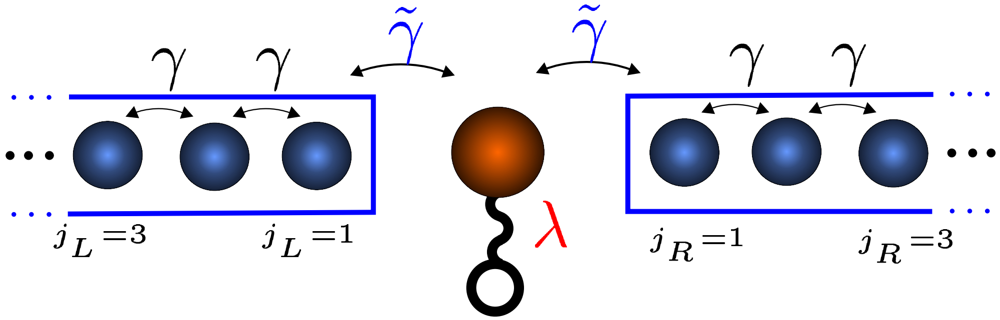


- CNT





Model



$$H = H_L + H_R + H_M + H_T$$

leads ($\alpha = L/R$):

$$H_\alpha = \sum_{j_\alpha} \left\{ eV_\alpha \, c^\dagger_{j_\alpha} c_{j_\alpha} + \gamma \left(c^\dagger_{j_\alpha} c_{j_\alpha+1} + c^\dagger_{j_\alpha+1} c_{j_\alpha} \right) \right\}$$

single site coupled to a phonon:

$$H_M = \epsilon_0 d^\dagger d + \lambda d^\dagger d \left(b^\dagger + b \right) + \hbar\omega \left(b^\dagger b + \tfrac{1}{2} \right)$$

tunneling Hamiltonian:

$$H_T = \tilde{\gamma} \left(c^\dagger_{j_L=1} d + d^\dagger c_{j_L=1} \right) + \tilde{\gamma} \left(c^\dagger_{j_R=1} d + d^\dagger c_{j_R=1} \right)$$

Nonequilibrium current

current: $J = J_L - J_R \qquad J_\alpha = -e \left\langle \left(\frac{\mathrm{d}N_M}{\mathrm{d}t} \right)_\alpha \right\rangle$

$$J = \frac{e}{2h} \int \mathrm{d}\epsilon \{ \Gamma_L(\epsilon) \left(f_L^0(\epsilon) - f(\epsilon) \right) - \Gamma_R(\epsilon) \left(f_R^0(\epsilon) - f(\epsilon) \right) \} A(\epsilon)$$

spectral function:

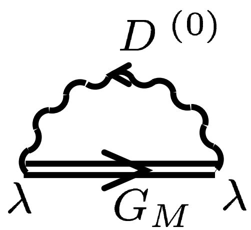
$$\begin{aligned} A(\epsilon) &= -2 \,\mathrm{Im}\, G_M^r(\epsilon) \\ &= -2 \,\mathrm{Im} \left(\epsilon - \epsilon_0 - \Sigma_L^r - \Sigma_R^r - \Sigma_{ph}^r \right)^{-1} \end{aligned}$$

$\Rightarrow$ Y. Meir, N. S. Wingreen PRL **68**, 2512 (1992)

H. Haug, A.-P. Jauho 'Quantum Kinetics in Transport ...', Springer (1996)

selfconsistent distribution function

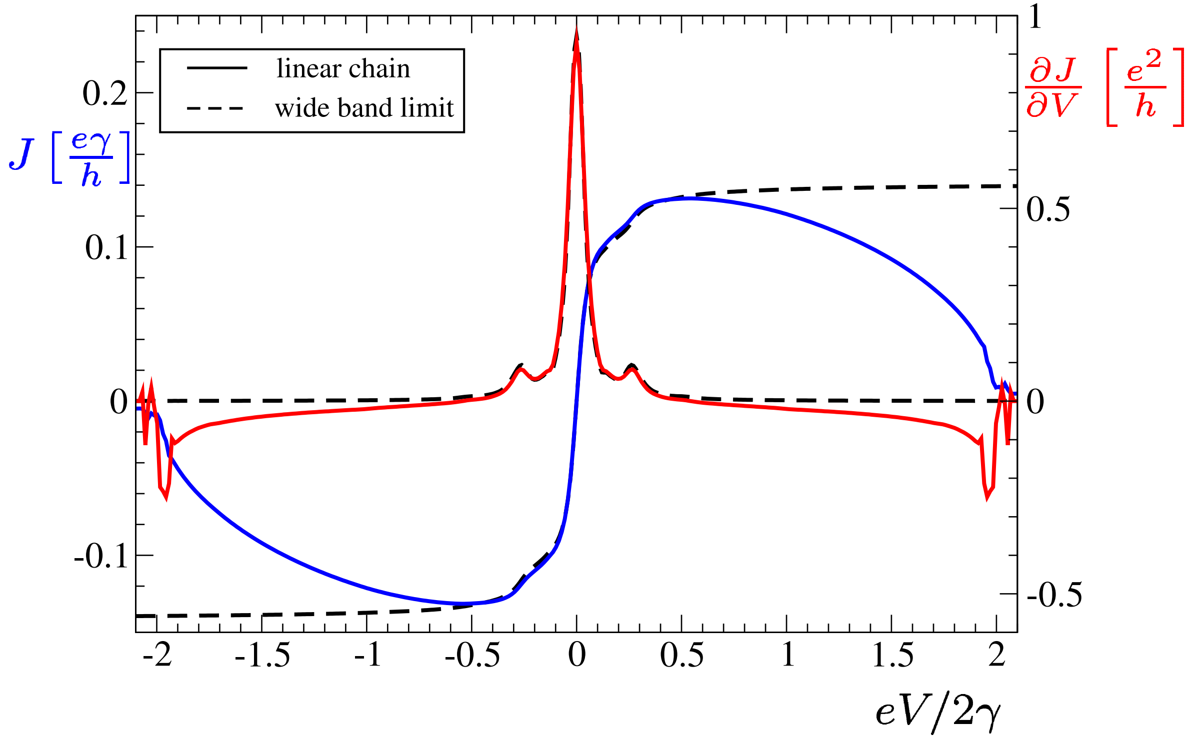
$\Rightarrow$ Dmitri Ryndyk, TT 11.9

phonon self energy: $\Sigma =$ 

S.C. Born approx.

Results

the current through the interacting region:



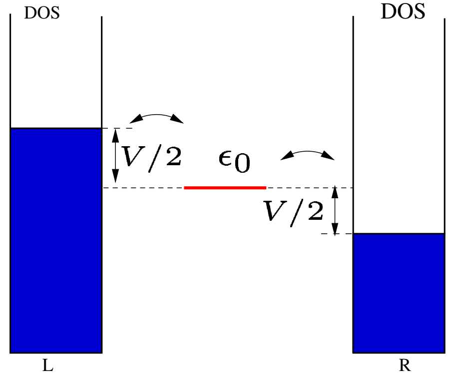
vibrational frequency: $\omega = 0.25\gamma$

vibrational coupling strength: $\lambda = 0.1\gamma$

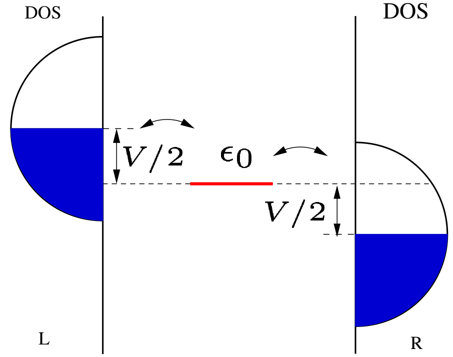
level coupling: $\tilde{\gamma} = 0.15\gamma$

level offset: $\epsilon_0 = 0$

wide band limit:



linear chain:



Results

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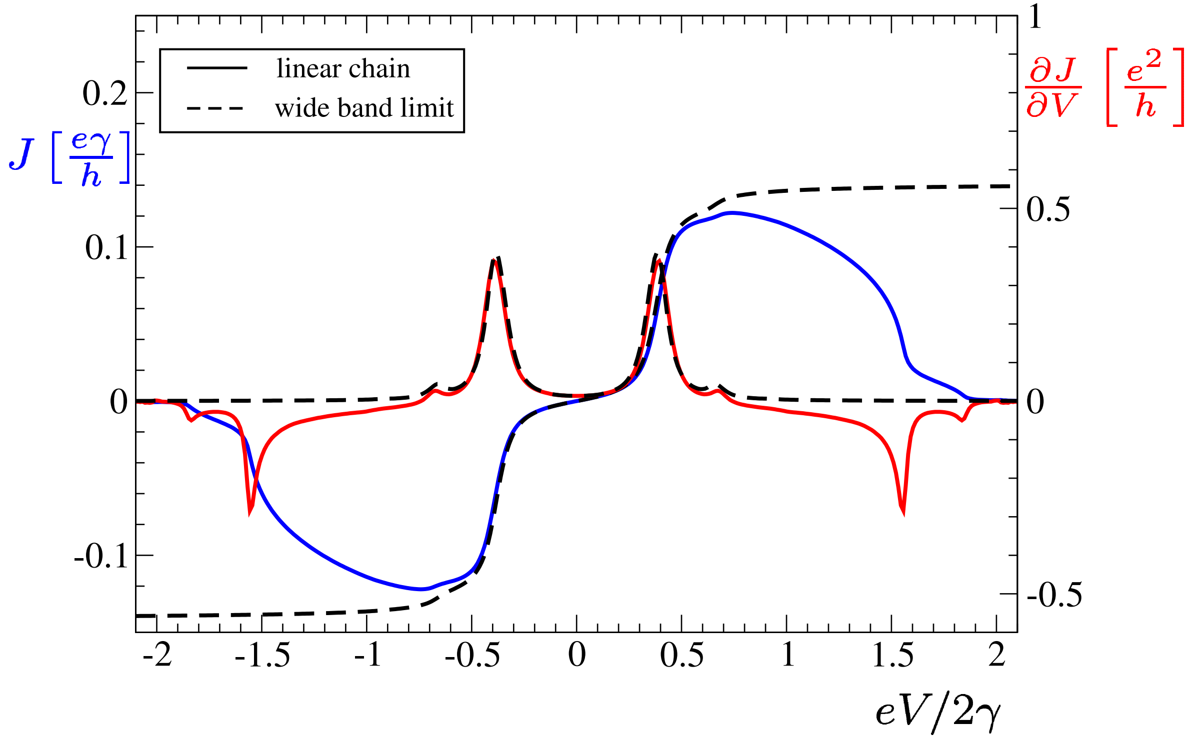

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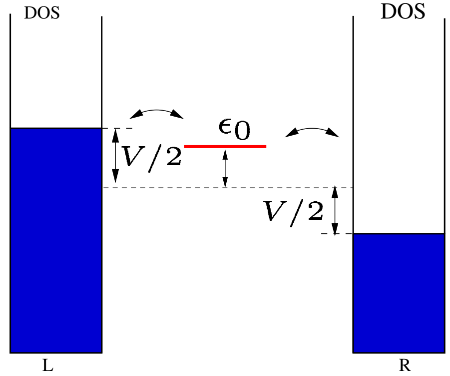
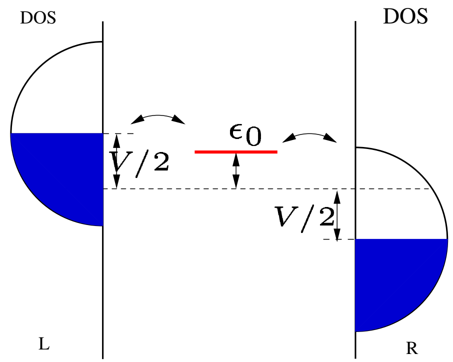
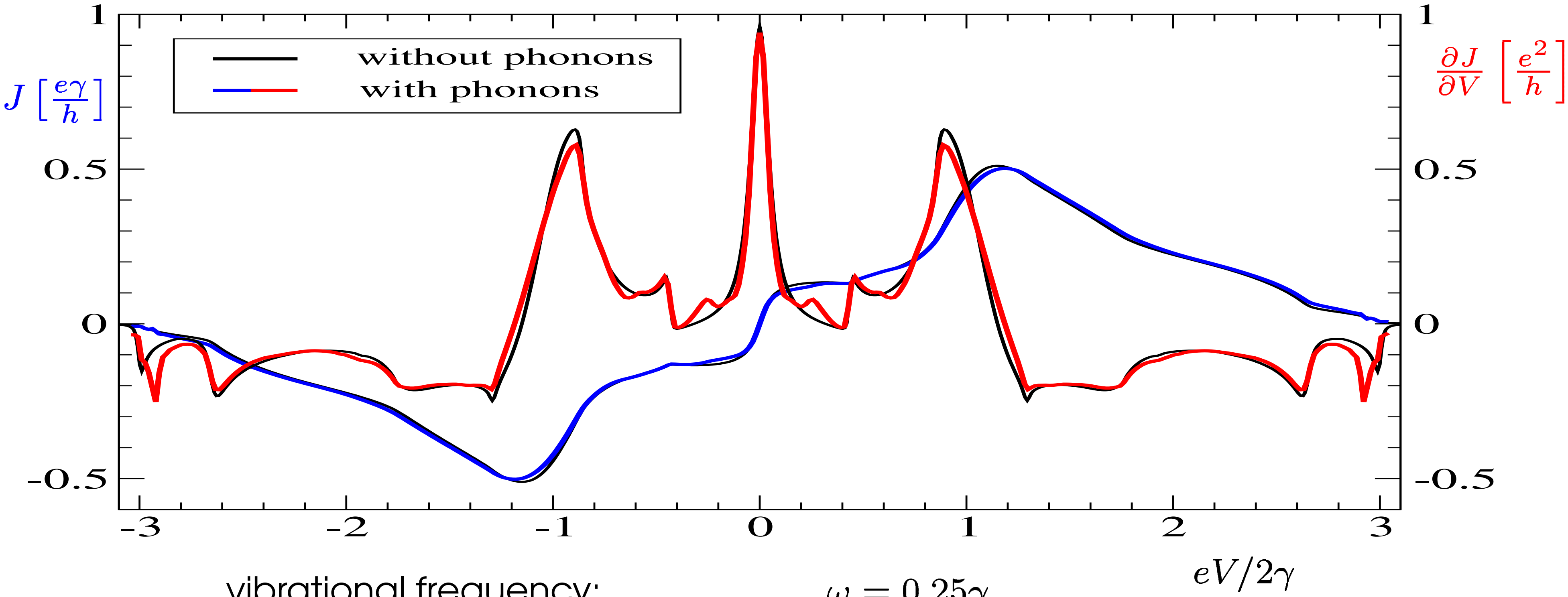


linear chain:



vibrational frequency:	$\omega = 0.25\gamma$
vibrational coupling strength:	$\lambda = 0.1\gamma$
level coupling:	$\tilde{\gamma} = 0.15\gamma$
level offset:	$\epsilon_0 = 0.4\gamma$

Carbon nanotube



vibrational frequency: $\omega = 0.25\gamma$

vibrational coupling strength: $\lambda = 0.1\gamma$

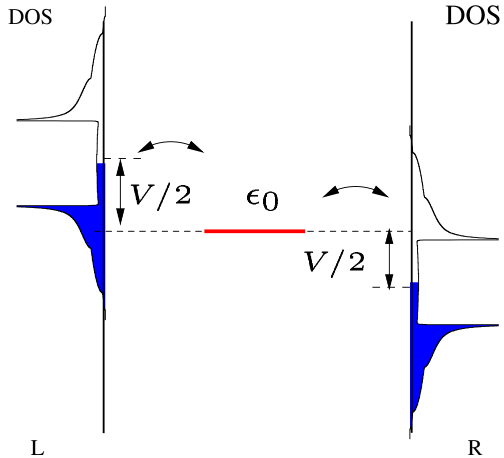
level coupling: $\tilde{\gamma} = 0.3\gamma$

level offset: $\epsilon_0 = 0$

armchair CNT(3,3)

G. Cuniberti, G. Fagas, K.Richter

Chem. Phys. **281**, 465-476 (2002)



Conclusions and outlook

$\surd$ minimal model beyond the static, equilibrium Landauer scheme; including the effects of mesoscopic leads (beyond WBLA)

$\rightarrow$ calculate the phonon distribution self consistently

$\rightarrow$ more realistic lead configurations: (pyramide on a) fcc - surface

$\rightarrow$ transferability to DFT based calculations