Current-induced forces, Joule heating and heat transport in molecular conductors

A semi-classical Langevin approach

Jing-Tao Lü

Dept. of Micro- and Nanotechnology,
Technical University of Denmark, Denmark

Acknowledgements: Mads Brandbyge, (DTU)

Per Hedegård, (KU)

Jian-sheng Wang (NUS, Singapore)

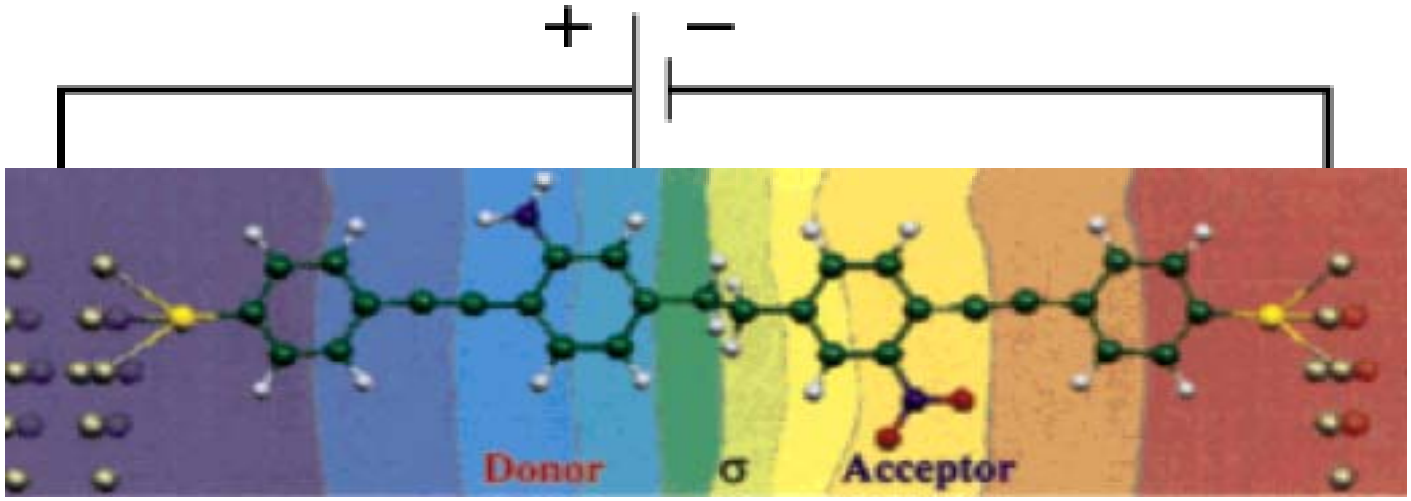
DTU Nanotech
Department of Micro- and Nanotechnology

DTU

Outline

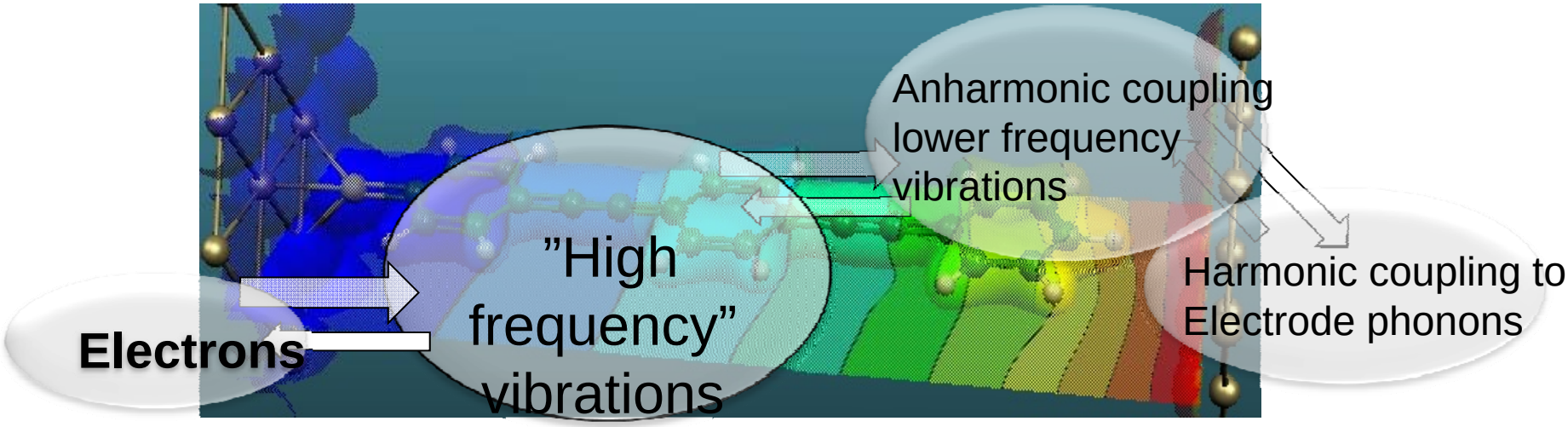
- **Motivation: Current-induced effect**
- **Langevin Approach**
- **Application 1: Current-induced instabilities**
- **Application 2: Heat transport**
- **Summary**

Molecular electronics



Seminal paper in molecular electronics
“Molecular Rectifiers” , A. Aviram and M. A. Ratner, *Chem. Phys. Lett.* **1974**, *29*, 277

Electron transport, inelastic effect



Molecular Dynamics in the presence of current

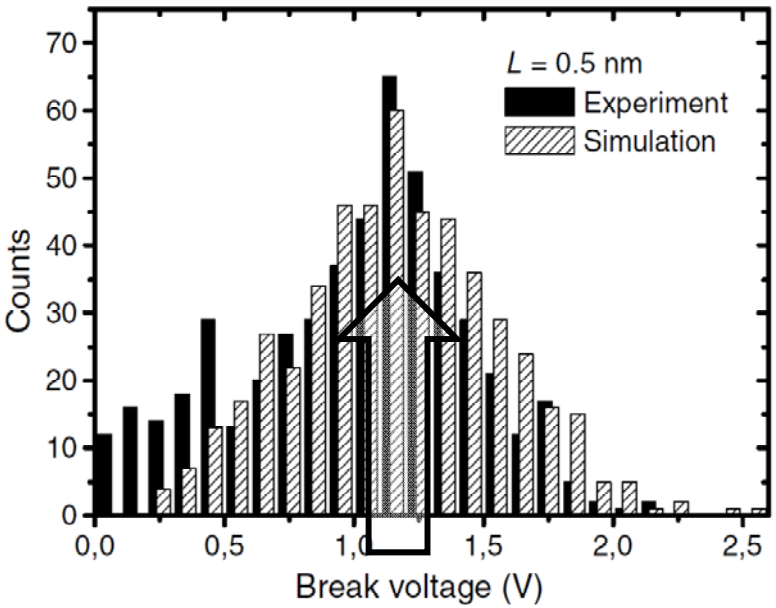
Breaking of Au chain

Gold

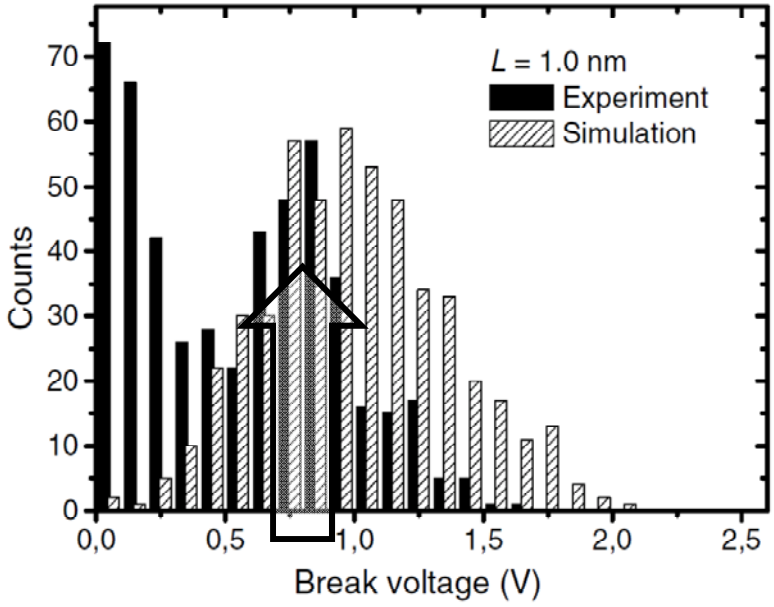
Early exp.: Yasuda, Sakai, PRB **56**, 1069 (1997)

Smit et al, Nanotechnology, **15**, S472 (2004)

Short chains ~ 3-5 atoms



Long chains ~ 5-7 atoms



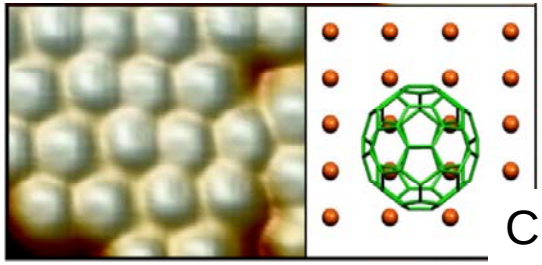
Why/How do the chains break?

Current-induced ”decomposition” of C60

PRL 100, 136801 (2008) PHYSICAL REVIEW LETTERS week ending 4 APRIL 2008

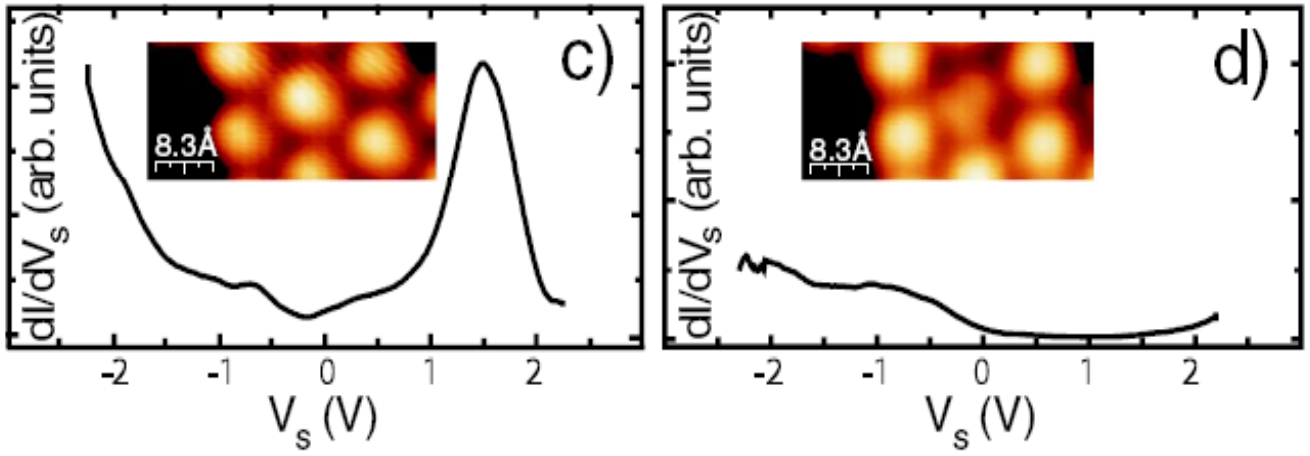
Resonant Electron Heating and Molecular Phonon Cooling in Single C_{60} Junctions

G. Schulze,[1] K. J. Franke,[1] A. Gagliardi,[2] G. Romano,[3] C. S. Lin,[2] A. L. Rosa,[2] T. A. Niehaus,[2] Th. Frauenheim,[2] A. Di Carlo,[3] A. Pecchia,[3] and J. I. Pascual[1]

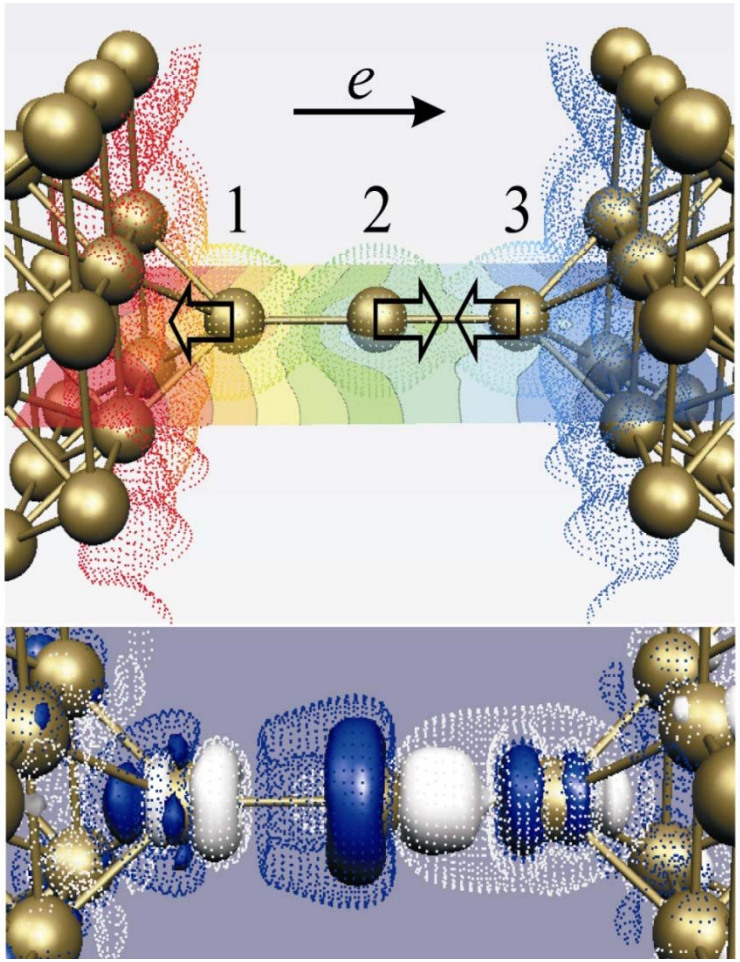


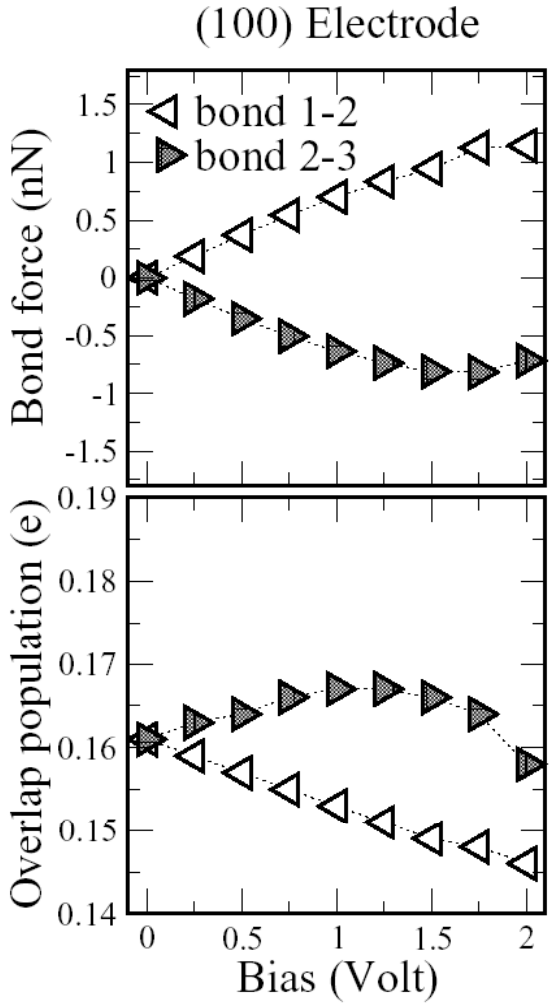
Cu(110)

Before high current (micro amps) After high current (micro amps)



Current-induced force: static

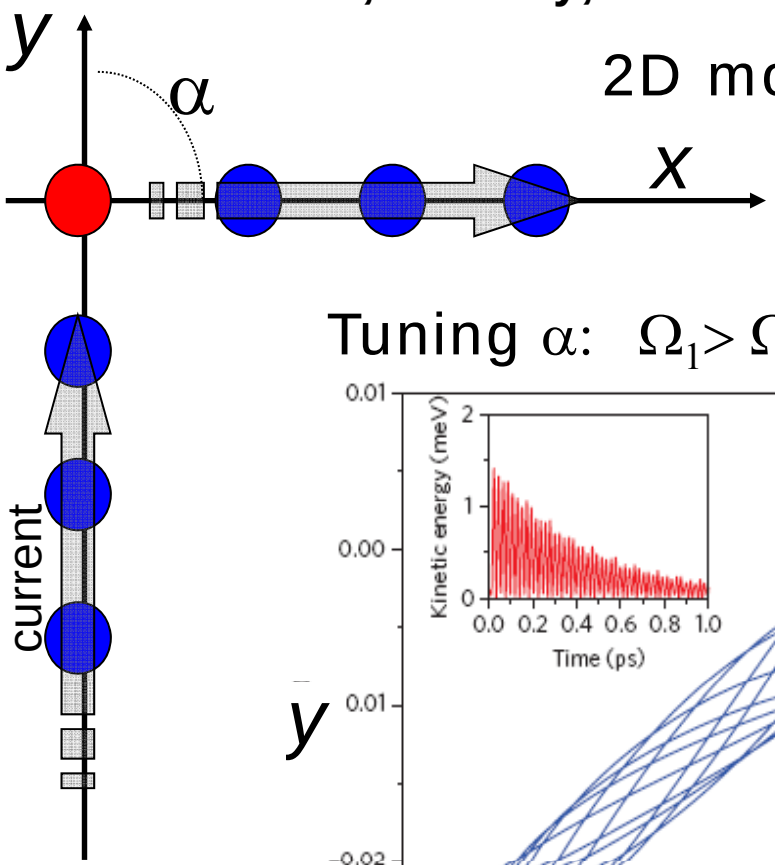




M. Brandbyge *et al*., PRB **67**, 193104 (2003)

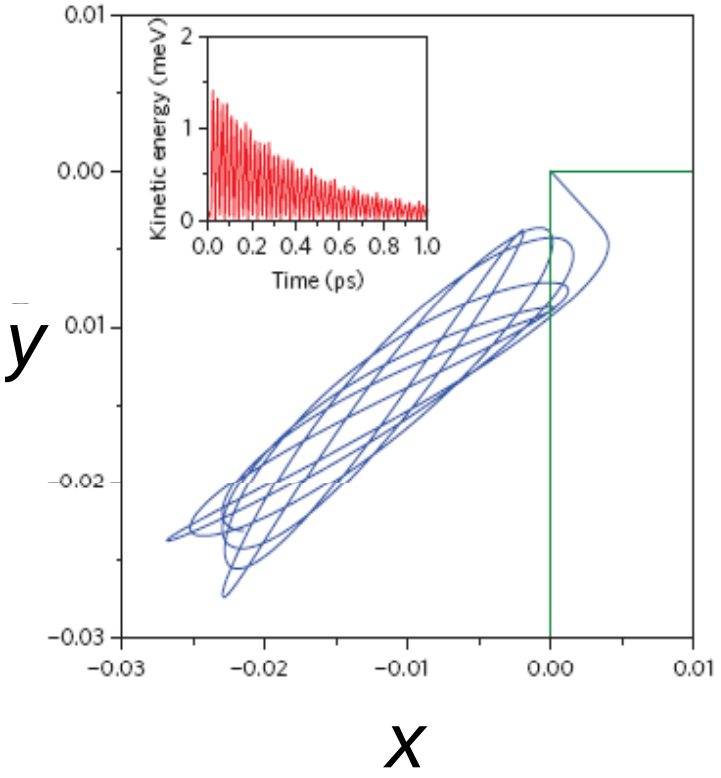
Current-driven waterwheel

Dundas, McEniry, Todorov, NATURE NANOTECH 4, 99, 2009

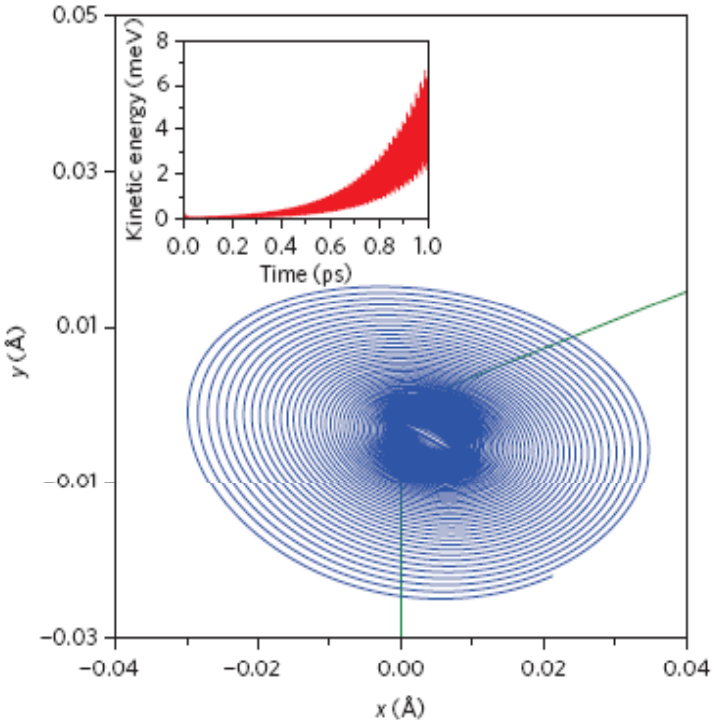


2D model

Tuning α: $\Omega_1 > \Omega_2$



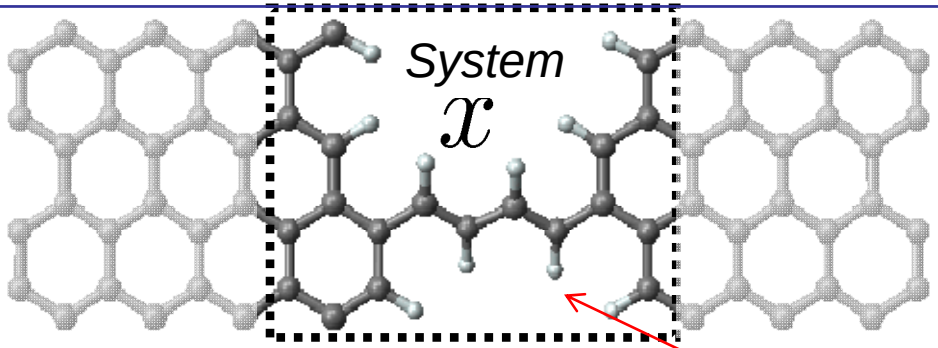
$\Omega_1 = \Omega_2$



Outline

- **Motivation: Current-induced effect**
- **Langevin Approach**
- **Application 1: Current-induced instabilities**
- **Application 2: Heat transport**
- **Summary**

System-Bath Model



$$H = H_{sys} + H_{bath} + V_{sb} + H_e(x=0) + \nabla_x H_e \cdot x$$

Harmonic phonon bath!!!

Nonequilibrium electron bath!!!

Langevin equation: $\ddot{Q}(t) + V'(Q(t)) = -\int^t \tilde{\Pi}(t-t')Q(t')dt' + f(t)$

Feynman, Vernon, Ann. Phys. 24, 118 (1963);Caldeira, A.; Leggett, A. Physica A, 121, 587 (1983)
Schmid, A. J. Low Temp. Phys., 49, 609 (1982);M. Brandbyge, P. Hedegård, Phys. Rev. Lett. **72**, 2919 (1994)

Langevin eaquation

$$\ddot{Q}(t) + V'(Q(t)) = -\int^{t} \tilde{\Pi}(t-t')Q(t')dt' + f(t)$$

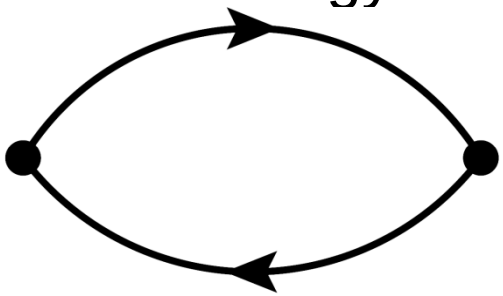
Damping kernel: $\tilde{\Pi} = \tilde{\Pi}_{ph} + \tilde{\Pi}_{e}$

Noise correlation: $\langle f(t)f(t')\rangle = \check{\Pi}(t-t') = \check{\Pi}_{ph} + \check{\Pi}_{e}$

Relation with self-energies in NEGF:

$$\tilde{\Pi} = \Pi^{>} - \Pi^{<}, \quad \check{\Pi} = \frac{i}{2}\left(\Pi^{>} + \Pi^{<}\right)$$

Electrons self-energy:

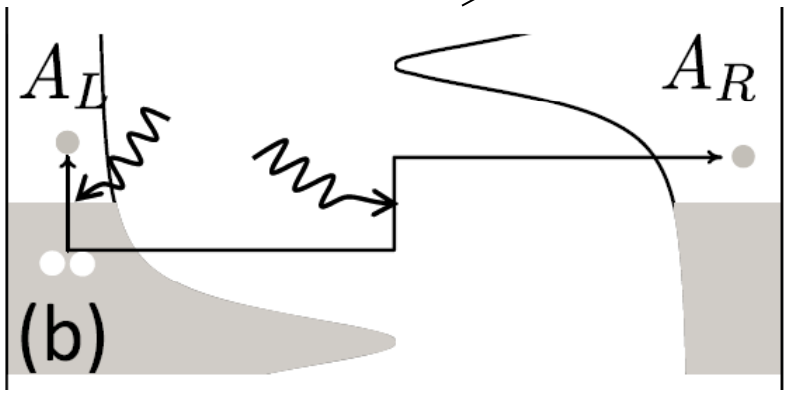


One mode: Equilibrium electrons

”Reservoir”: Equilibrium Electrons

$$\ddot{x} + \Omega^2 x = -\gamma_0 \dot{x} + f$$

$$\check{\Pi}(\omega) = \gamma_0 \, \omega \coth\left(\frac{\hbar\omega}{2kT}\right)$$



- High T: $\sim 2\gamma_0 kT$
- Low T: $\sim \frac{1}{2}\gamma_0\omega$

Equilibrium: ”MD with electronic frictions at equilibrium”,
M. Head-Gordon, J. C. Tully, J. Chem. Phys. **103** (1995)

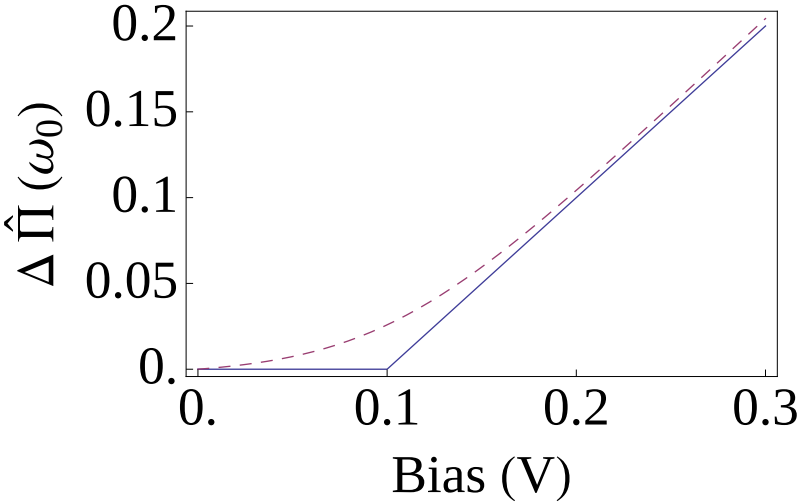
One-mode: nonequilibrium electrons

”Reservoir”: Electrons with steady-state current (Landauer picture)

$$\ddot{x} + \Omega^2 x = -(\gamma_0 + \delta\gamma)\,\dot{x} + f$$

$$\check{\Pi}(\omega) = (\gamma_0 + \delta\gamma)\,\omega \coth\left(\frac{\hbar\omega}{2kT}\right) + \Delta\check{\Pi}(\omega, V)$$

$$\Delta\check{\Pi}(\omega, V) \sim (\hbar\omega \pm eV)\left(\coth\left(\frac{\hbar\omega \pm eV}{2kT}\right) - \coth\left(\frac{\hbar\omega}{2kT}\right)\right)$$

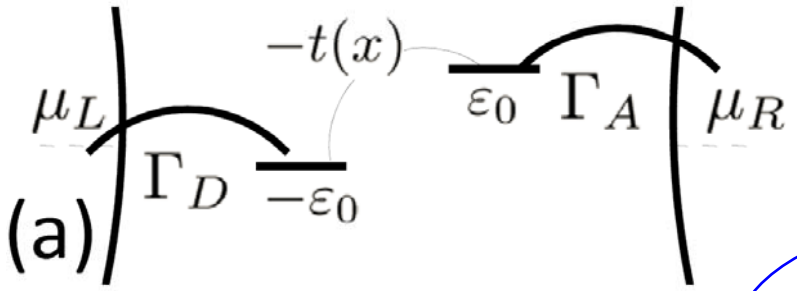


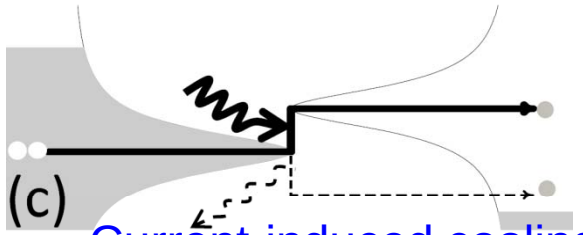
Outline

- **Motivation**
- **Langevin equation**
- **Application 1: Current-induced instabilities**
- **Application 2: Heat transport**
- **Summary**

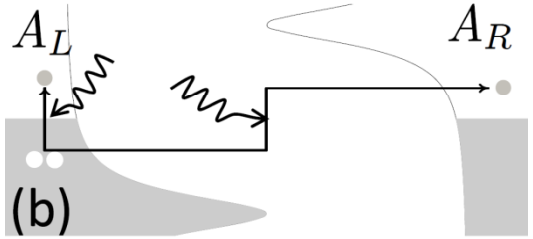
NEQ correction to friction

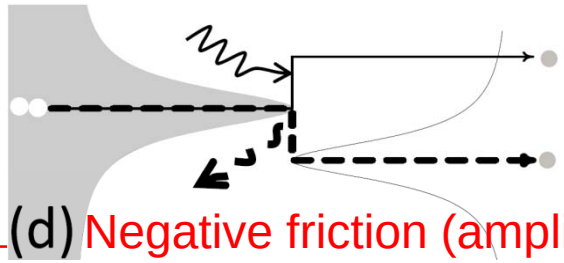
$$\ddot{x} + \Omega^2 x = -(\gamma_0 + \delta\gamma)\,\dot{x} + f$$



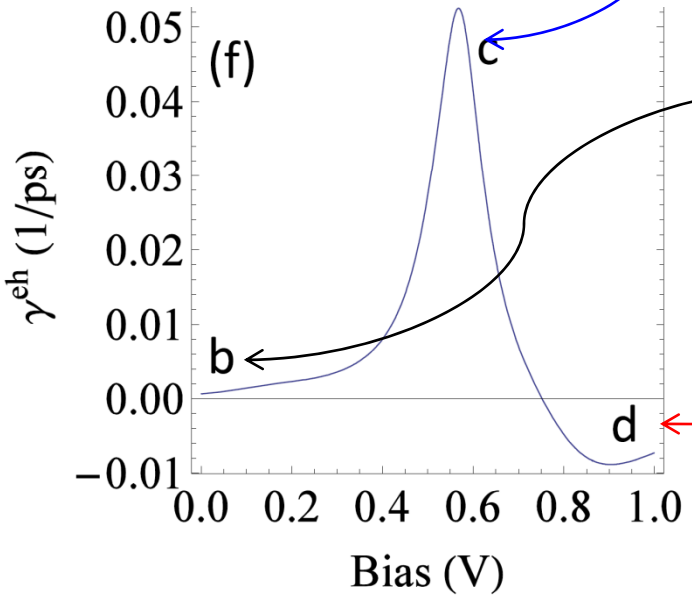


Current-induced cooling





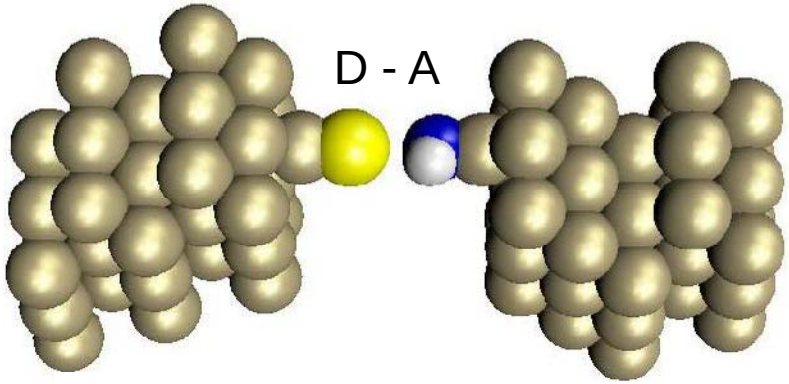
Negative friction (amplification)



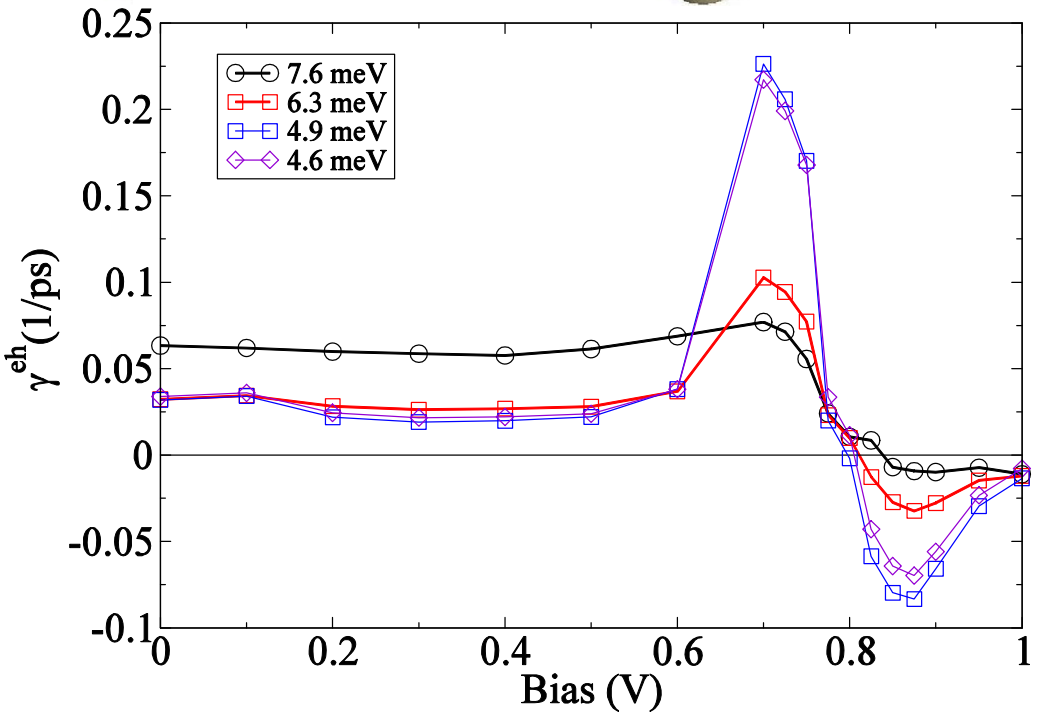
J.-T. Lü, P. Hedegård, M. Brandbyge, PRL, 107, 046801 (2011)

DFT calculation

Experimental setup:
Functionalized STM-tip
and adsorbed molecule



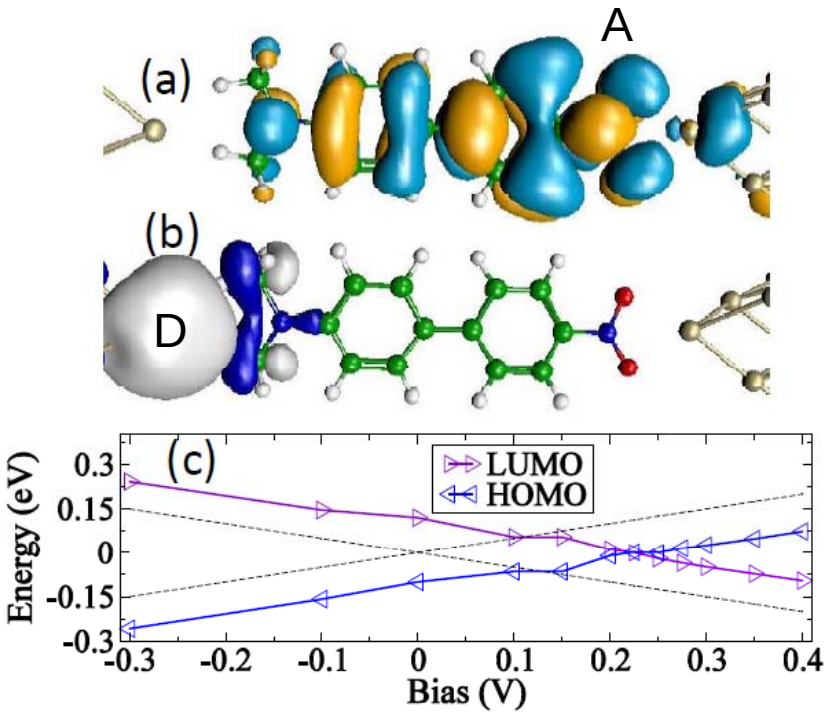
DFT-NEGF calculation
using tranSIESTA

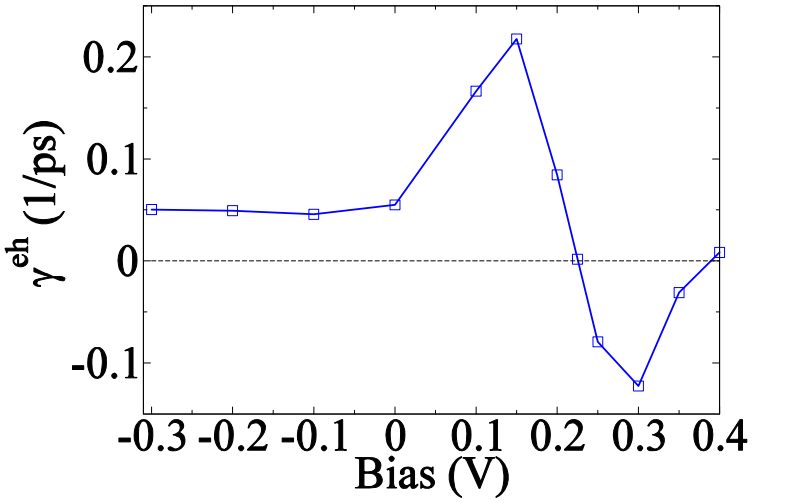


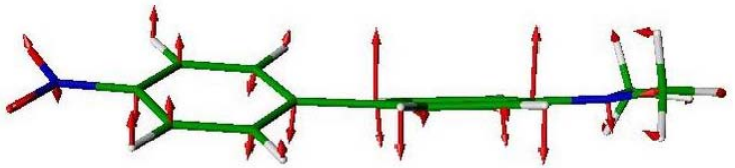
J-T. Lü, P. Hedegård, M. Brandbyge, Phys. Rev. Lett., 107, 046801 (2011)

Molecular diode

4-dimethylamino-4'-nitrobiphenyl







63 meV

J-T. Lü, P. Hedegård, M. Brandbyge, Phys. Rev. Lett., 107, 046801 (2011)

More Modes

Langevin equation for the vibrational modes (Q):

$$\ddot{Q}_I + \Omega_I^2 Q_I = -\sum_J \gamma_{IJ}\,\dot{Q}_J - \mathcal{N}_{IJ}\,Q_J - \mathcal{B}_{IJ}\,\dot{Q}_J + f_I$$

Non-conservative "water-wheel" forces

Conservative Lorentz-like forces ("Berry phase" of electrons)

$$\mathcal{N}_{IJ}, \mathcal{B}_{IJ} \propto eV \sum_{ij} \mathrm{Im}\langle L,i|\bigtriangledown_I H_e|R,j\rangle\langle R,j|\bigtriangledown_J H_e|L,i\rangle$$

Anti-symmetric !!!

J.-T. Lü, M. Brandbyge, P. Hedegård, Nano Lett. **10**, 1657 (2010)

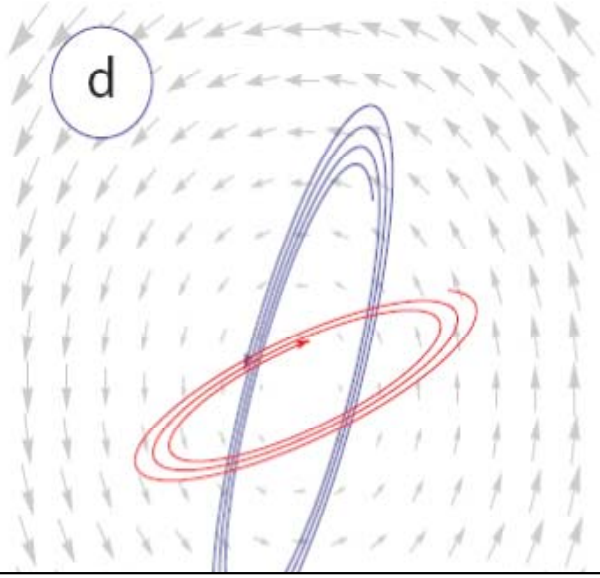
Simple 2D model: modes

$$\begin{pmatrix} \ddot{x} \\ \ddot{y} \end{pmatrix} = -\begin{pmatrix} \Omega_1^2 & a \\ -a & \Omega_2^2 \end{pmatrix}\begin{pmatrix} x \\ y \end{pmatrix} - \begin{pmatrix} \eta & b \\ -b & \eta \end{pmatrix}\begin{pmatrix} \dot{x} \\ \dot{y} \end{pmatrix}.$$

Non-conservative, wind

Berry, Lorentz

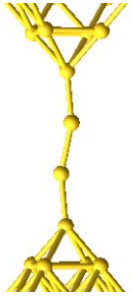
$$\begin{pmatrix} x \\ y \end{pmatrix} = e^{i\omega t}\begin{pmatrix} x_0 \\ y_0 \end{pmatrix}.$$



May obtain growing solutions: ”runaway”
Q-factor<0

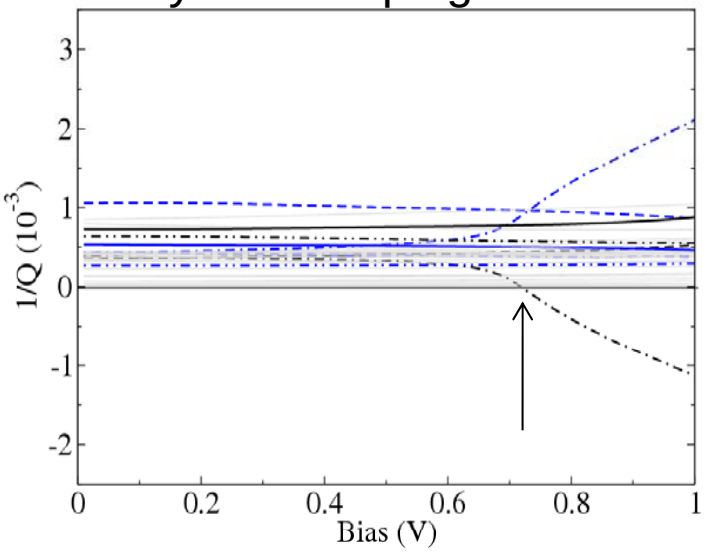
Excitation by the fluctuating force

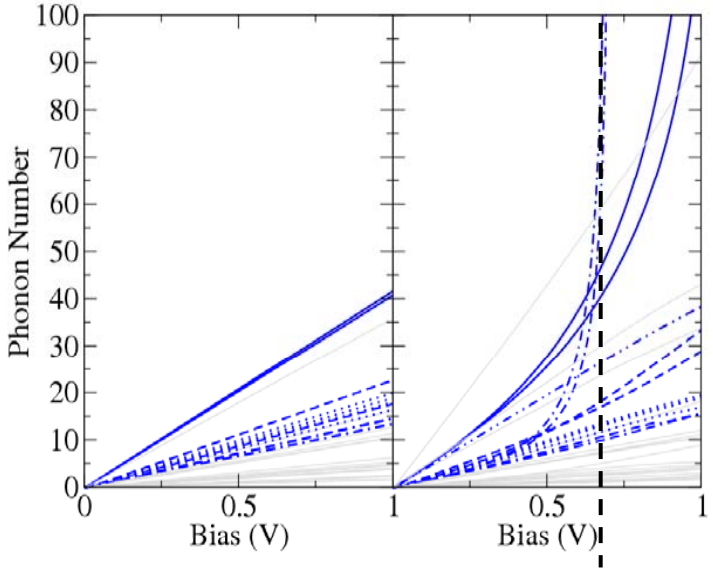
Au

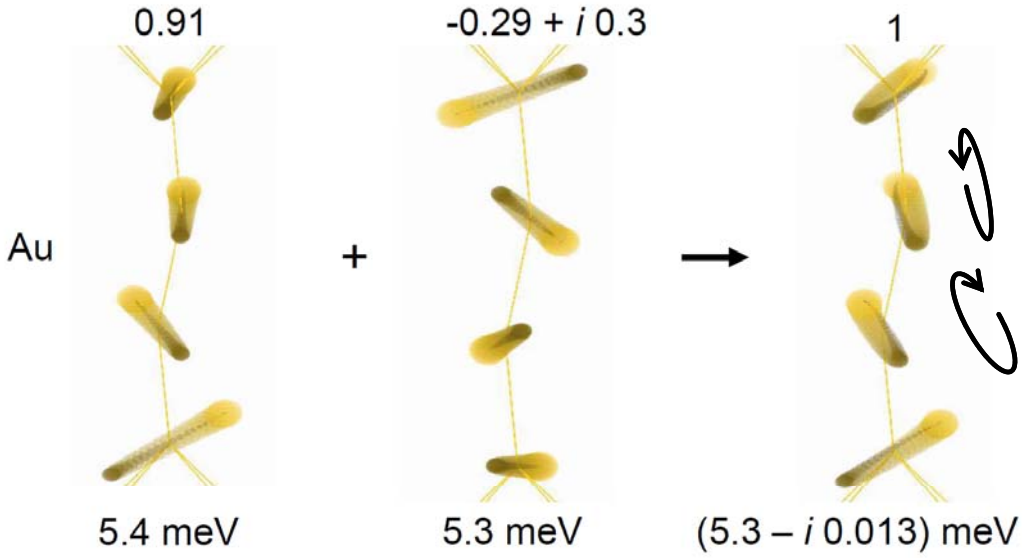


(100)

Only e-h damping:







Normal modes

Runaway mode

Outline

- **Motivation: Current-induced effect**
- **Langevin Approach**
- **Application 1: Current-induced instabilities**
- **Application 2: Heat transport**
- **Summary**

Heat current

Langevin equation for multimodes:

$$\ddot{Q}(t) + V'(Q(t)) = -\int^{t} \tilde{\Pi}(t-t')Q(t')dt' + f(t)$$

Contribution from all baths:

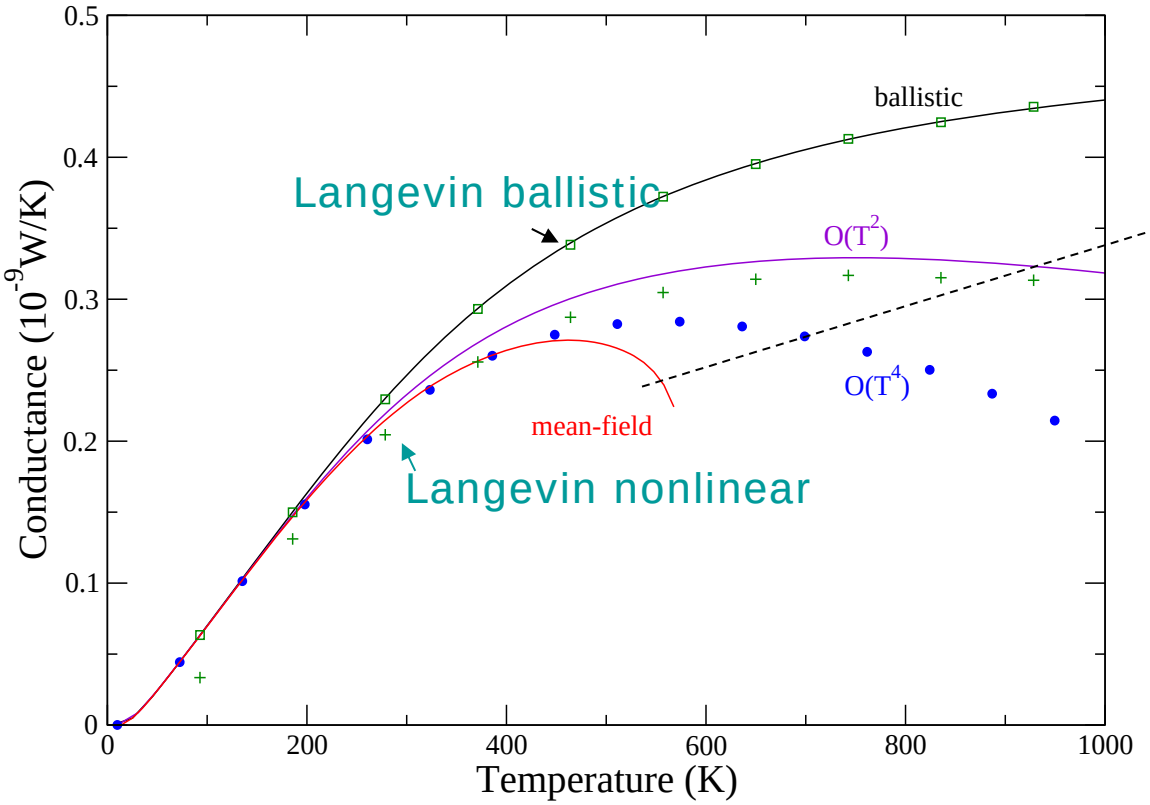
$$\tilde{\Pi} = \sum_{\alpha} \tilde{\Pi}_{\alpha}, \quad f = \sum_{\alpha} f_{\alpha}, \quad \alpha = L, R, e$$

At steady state: $\dot{H}_{ph} = -\dot{Q}^{T}\left(\int^{t} \tilde{\Pi}(t-t')Q(t')dt' - f(t)\right) = 0$

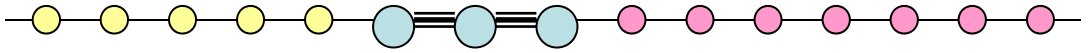
$$\Downarrow$$

Heat current from bath: $$I_{\alpha} = -\dot{Q}^{T}\left(\int^{t} \tilde{\Pi}_{\alpha}(t-t')Q(t')dt' - f_{\alpha}(t)\right)$$

Phonon heat transport: No electrons



NEGF mean field fails for strong anharmonic interaction, or high temperature!!!



k_L=1.56 k_C=1.38, t=1.8 k_R=1.44

Three-atom junction with cubic nonlinearity (FPU-α).

J. S. Wang, PRL, 99, 160601 (2007);

J. S. Wang, J. S. Wang, and J.-T. Lü, Eur. Phys. J. B, 62, 381 (2008).

Summary

A semi-classical Langevin equation

$$\ddot{Q}(t) + V'(Q(t)) = -\int^{t} \tilde{\Pi}(t-t')Q(t')dt' + f(t)$$

has been used to study:

- **Current induced forces**
- **Joule heating**
- **Heat transport**

Future plan:

- **Thermo-electric transport**