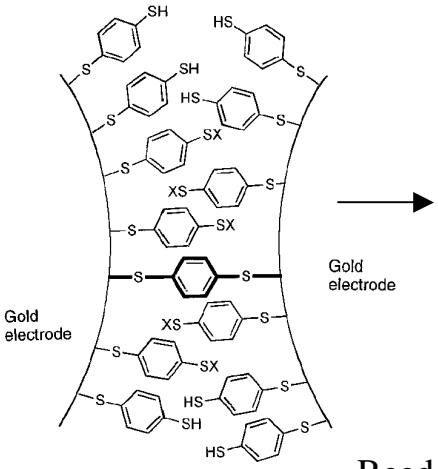
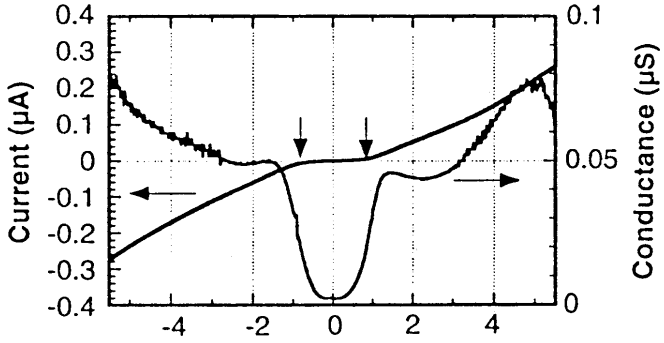
Transport in molecular structures: an overview of present understanding

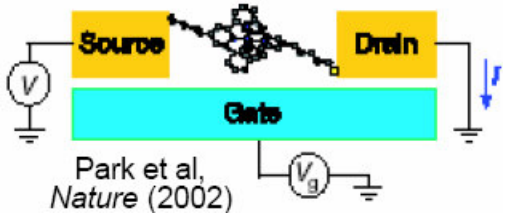
Massimiliano Di Ventra
Department of Physics, University of California, San Diego

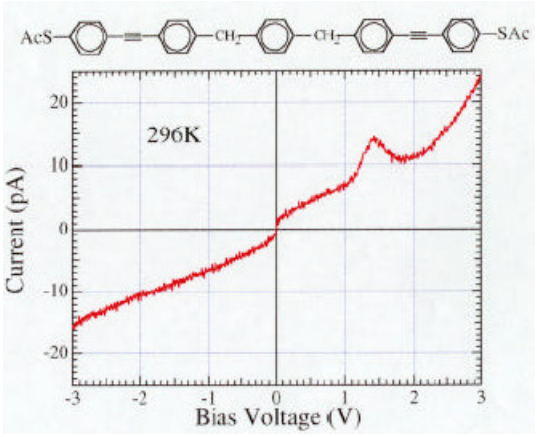
Experimental realization of molecular devices



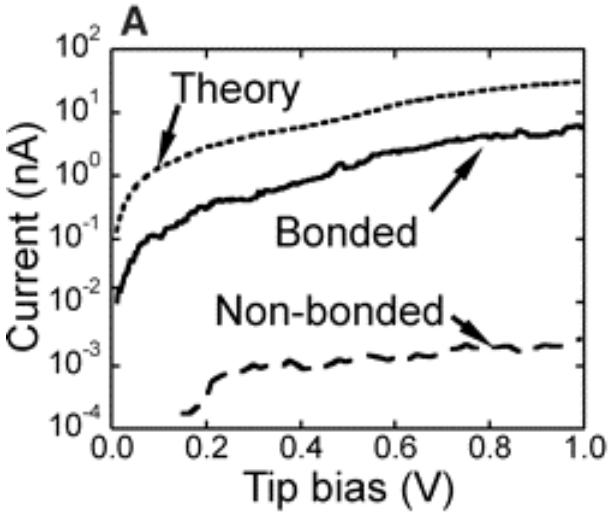


Reed *et al.*, *Science*, 1997





Zhou *et al.*, *APL*, 1997

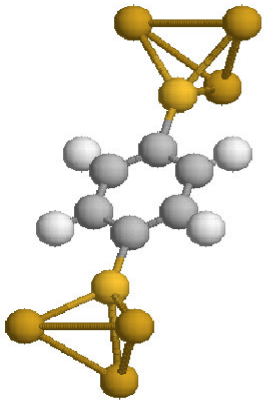


Cui *et al.*, *Science*, 2001

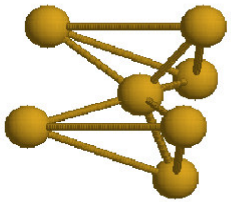
Current status of comparison between Th. (static DFT) and Exp.

UCSD

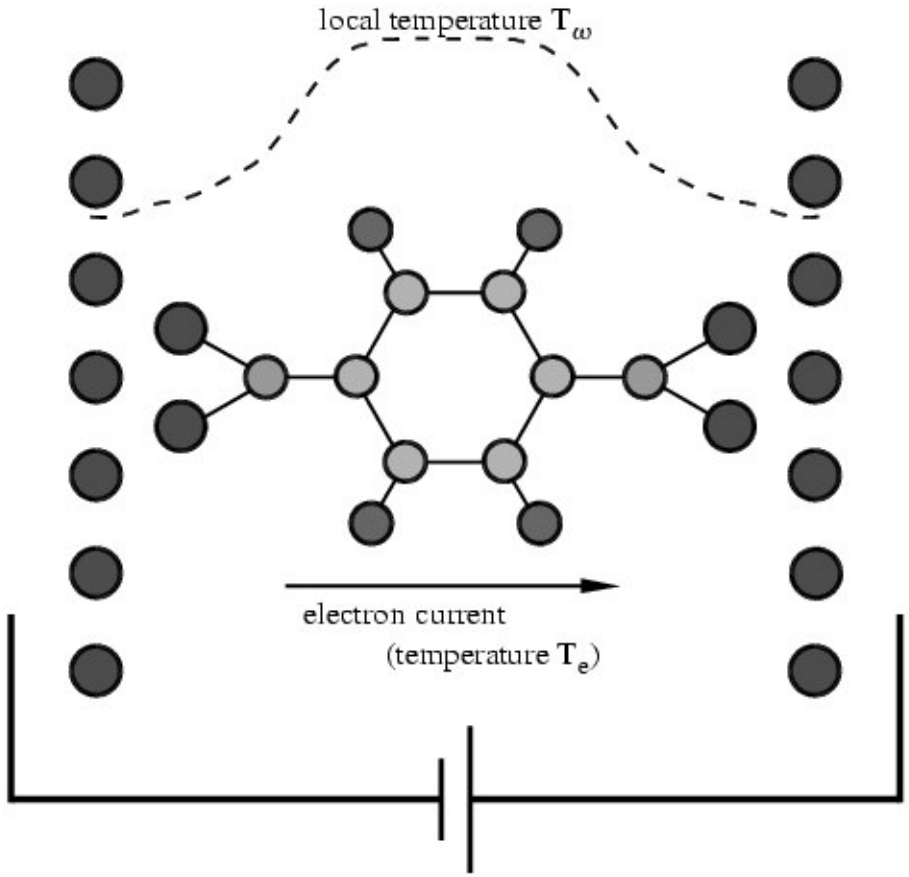
• Molecular structures: Apart from few claims of "perfect" agreement between th. and exp., theoretical estimates of the conductance are way off (larger values than experimental ones). Results very sensitive to geometry and local environment.



• Quantum point contacts: reasonable agreement (Au, Al, Pt, ….)

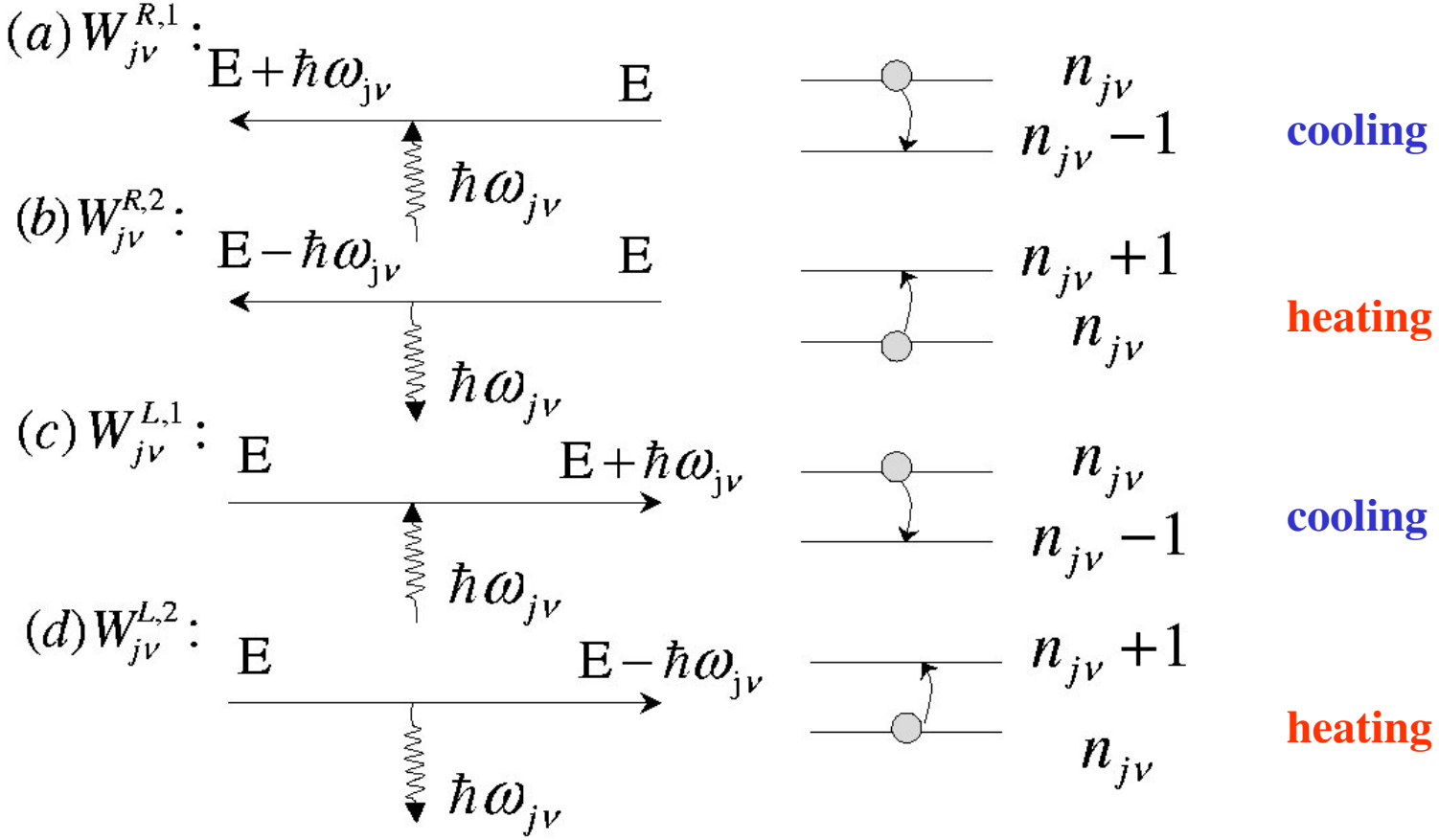


Current-induced mechanical effects



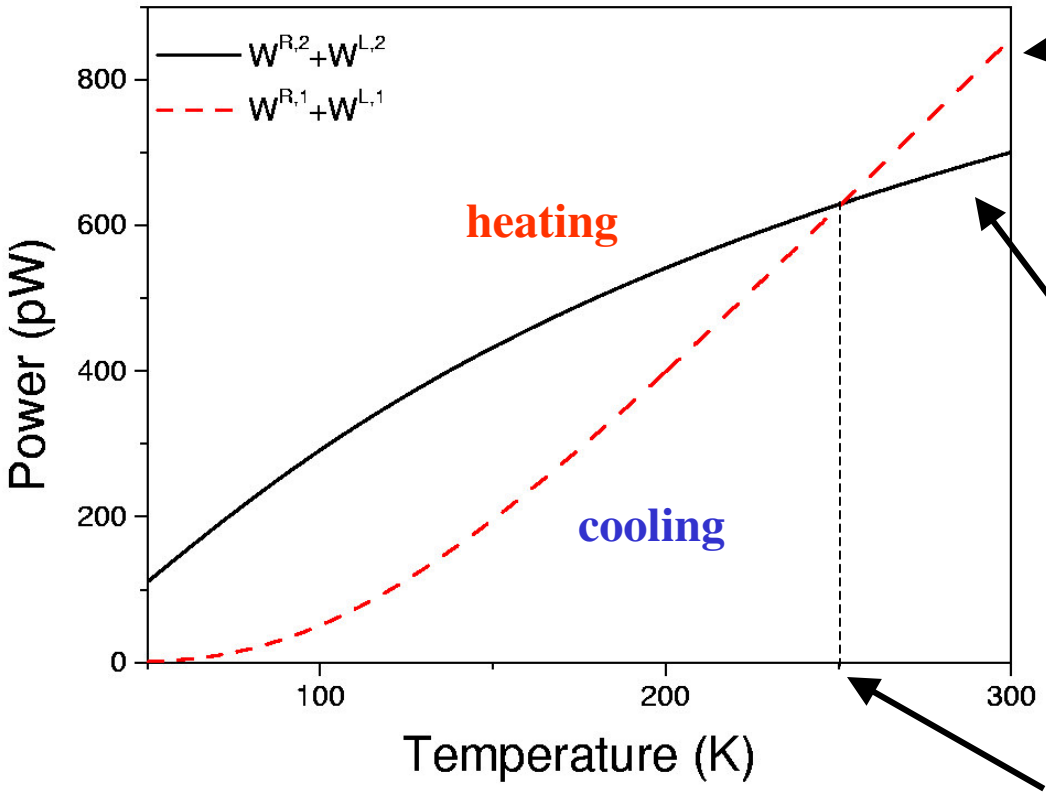
M. Di Ventra, T.N. Todorov, *Electrical Conduction in Nanoscale Systems* (Oxford University Press, to be published)

Local Heating



Local Heating

$$P = \sum_{j\nu \in vib} (W_{j\nu}^{R,2} + W_{j\nu}^{L,2} - W_{j\nu}^{R,1} - W_{j\nu}^{L,1})$$



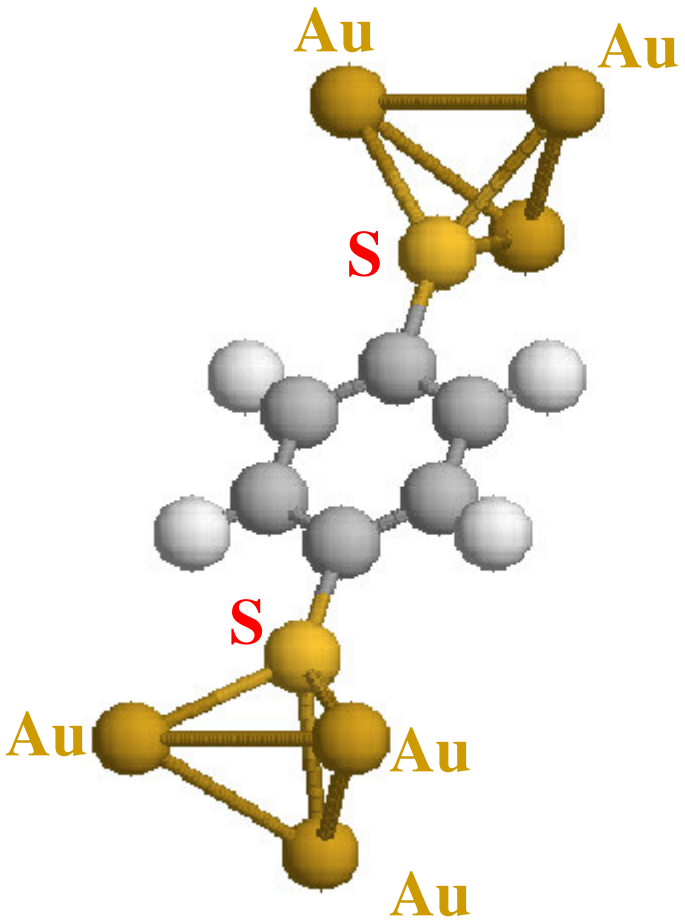
$\propto g(T_w) \times V_{bias}$

$\propto (1 + g(T_w)) \times V_{bias}$

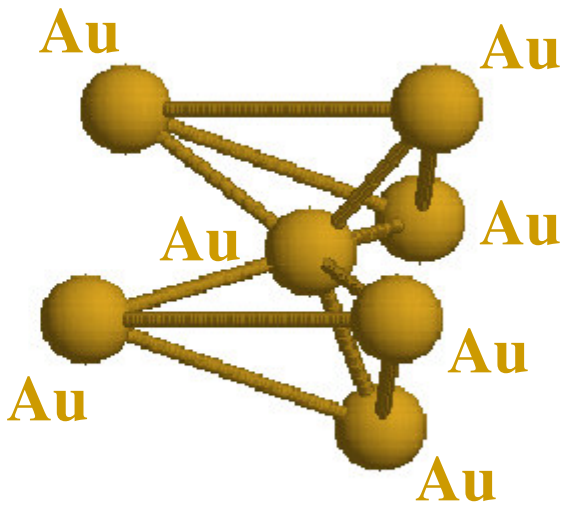
Steady-state T

Gold contact, T_e = 50K, Bias = 7 mV

Local Heating: examples



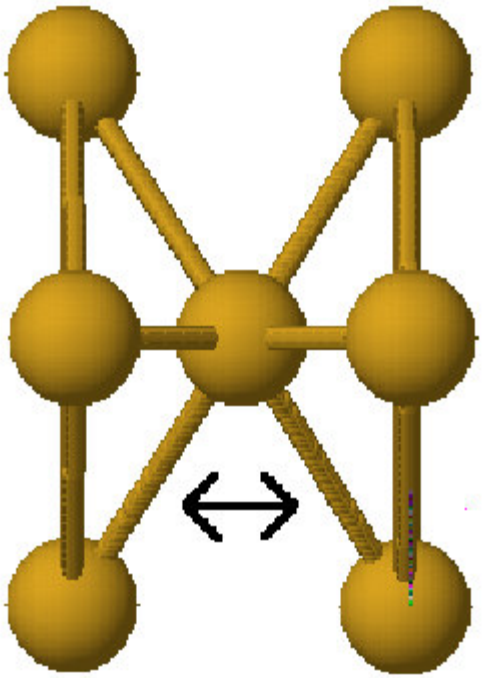
Phenyldithiolate device



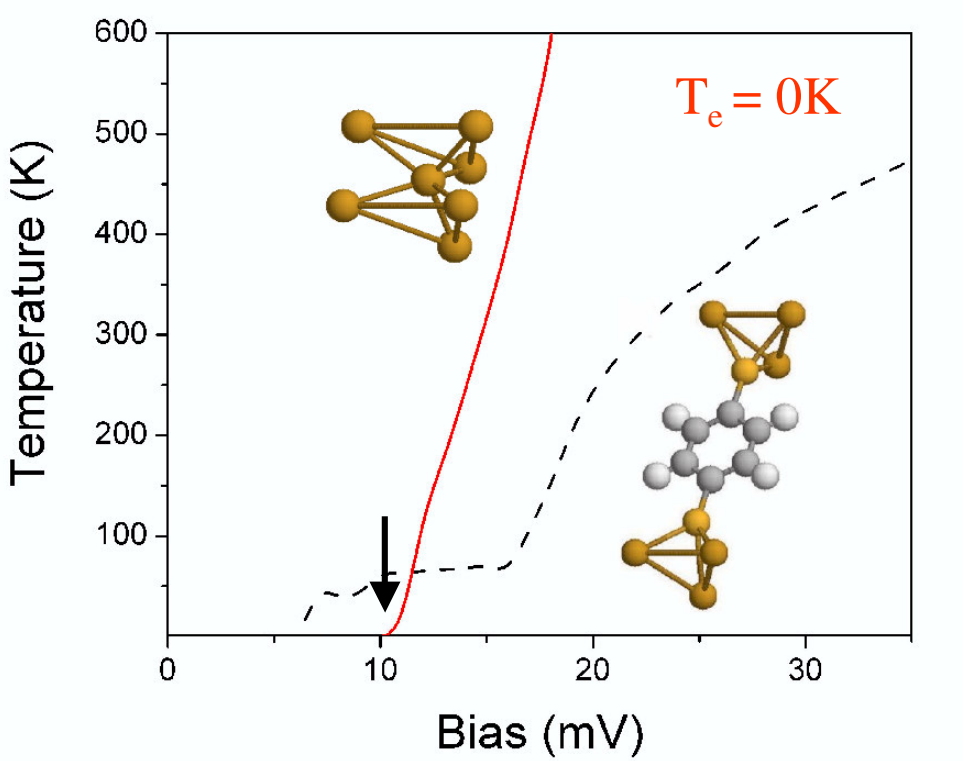
Gold point contact

Local Heating: point contact

No Goldstone mode => onset bias



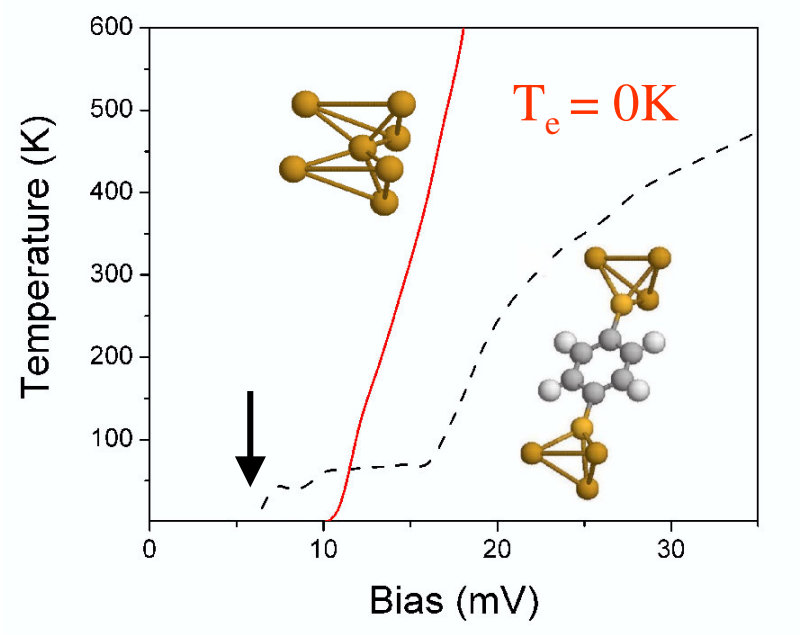
E = 11 meV



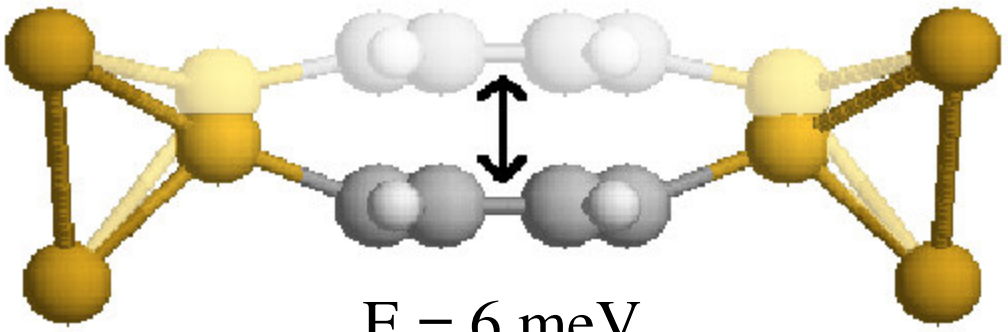
Onset bias ~ 11 mV

Exp: 11-16 mV
Agrait et al. PRL 2002

Local Heating: molecule

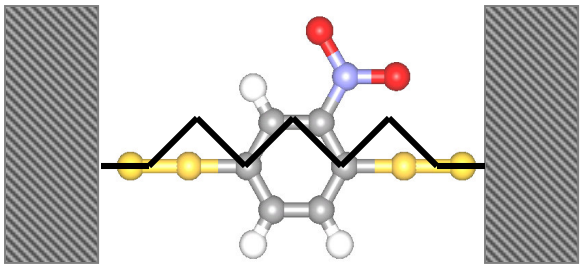


Onset bias ~ 6 mV



E = 6 meV

Local Heating and Heat Dissipation



Thermal current:

$$I_{th} = \frac{4\pi K^2}{\hbar} \int d\varepsilon \varepsilon N_L(\varepsilon) N_R(\varepsilon) [n_L(\varepsilon) - n_R(\varepsilon)]$$

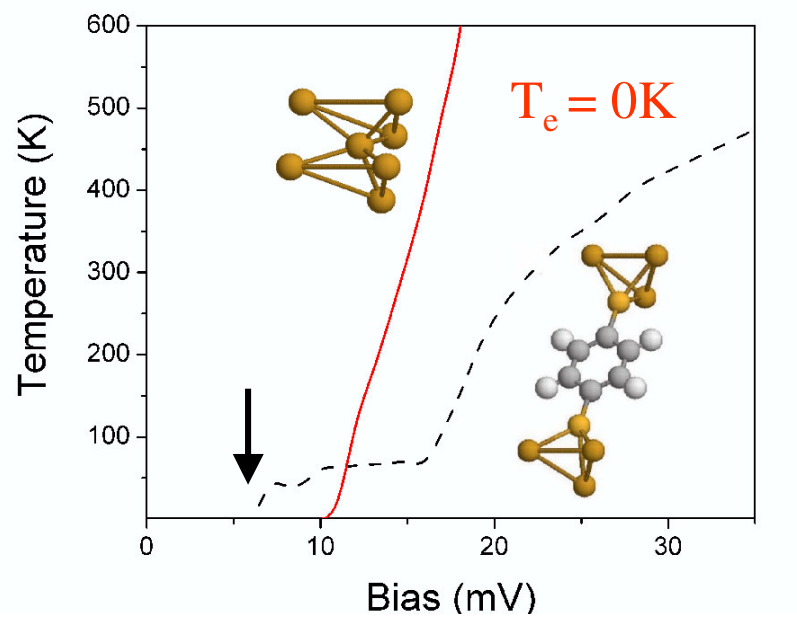
Phonon local spectral DOS

K = stiffness

Patton, Geller
PRB, 64 155320 (2001)

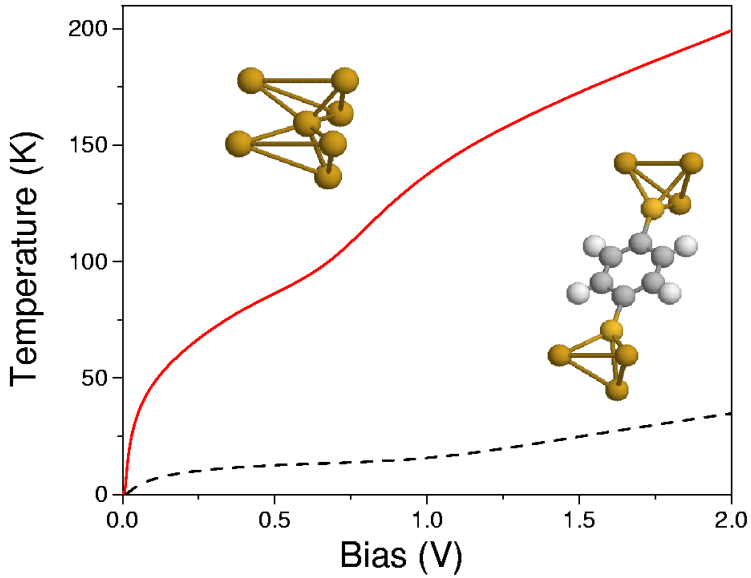
Local heating and dissipation

w/o dissipation



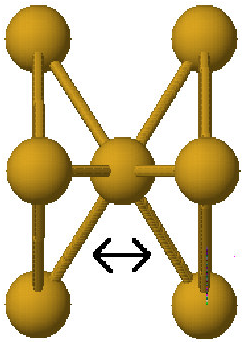
➡ Can lead to low-V instabilities

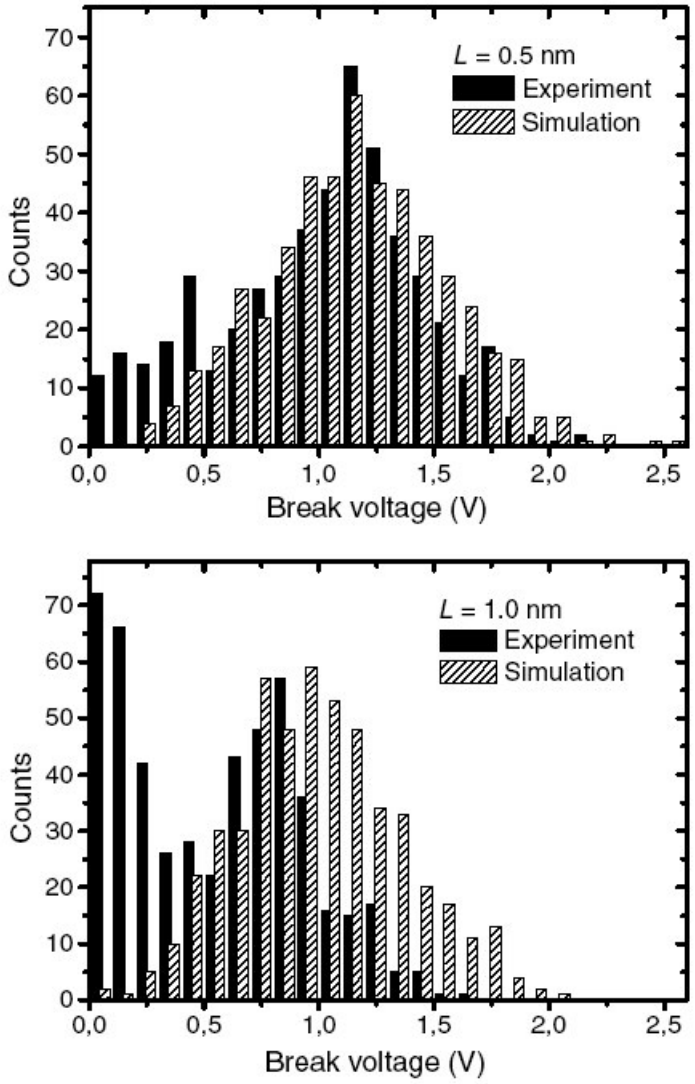
with dissipation



Molecule heats up less:
lower current &
larger dissipation into the electrodes

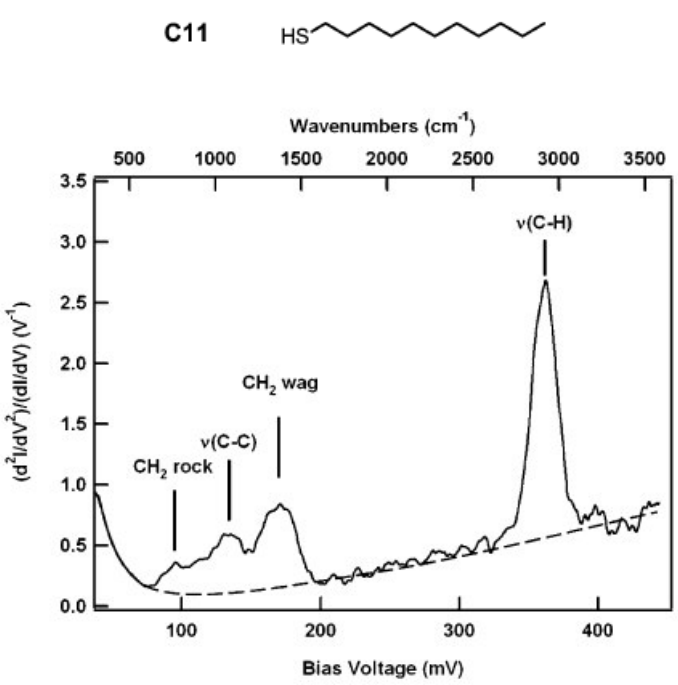
Stability: experiments on point contacts



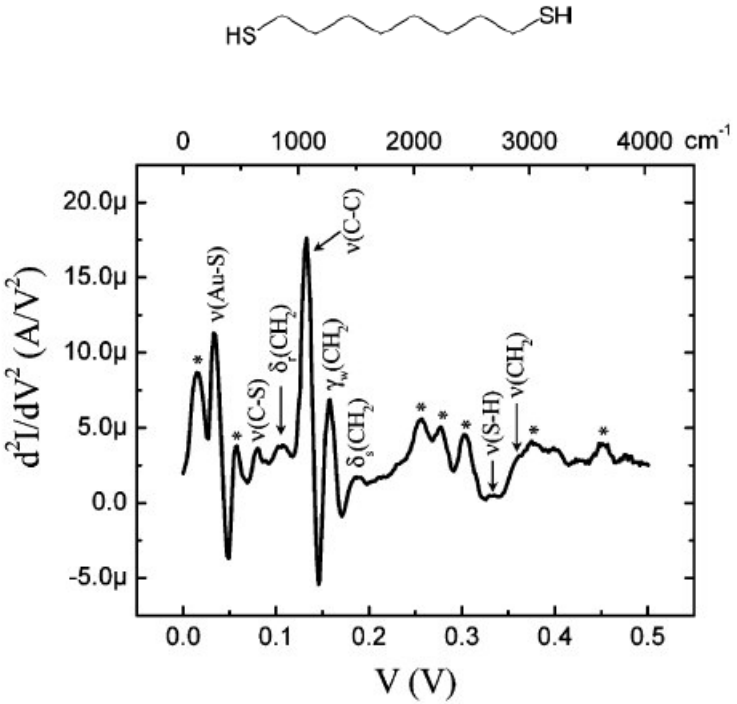


Smit *et al.*, Nanotechnology (2004)

Inelastic current: experiments on alkanethiols

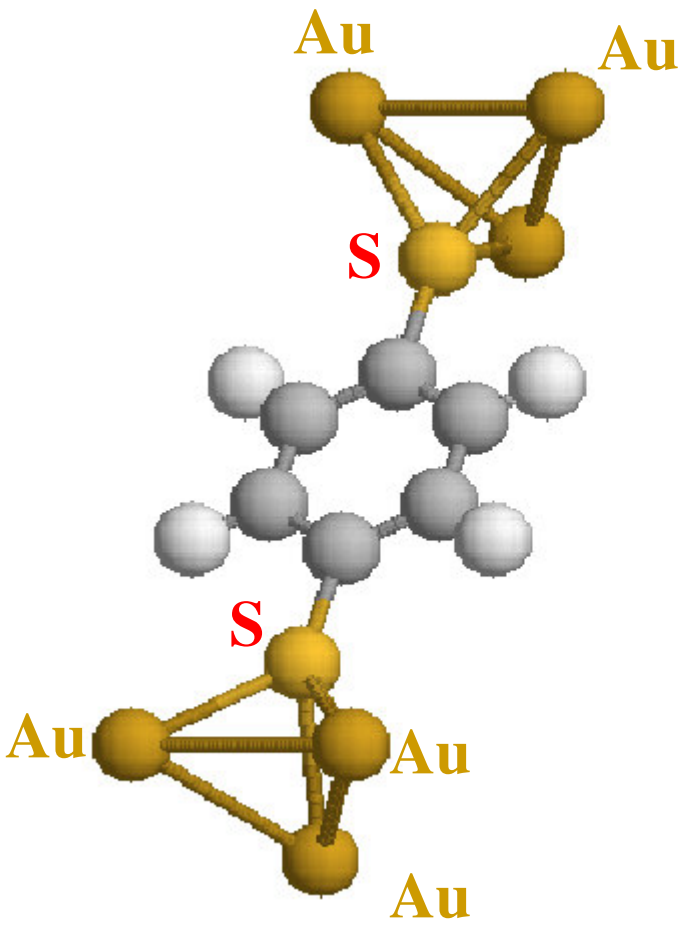


Kushmerick *et al*., Nanoletters (2004)



Wang *et al*., Nanoletters (2004)

Inelastic current: examples



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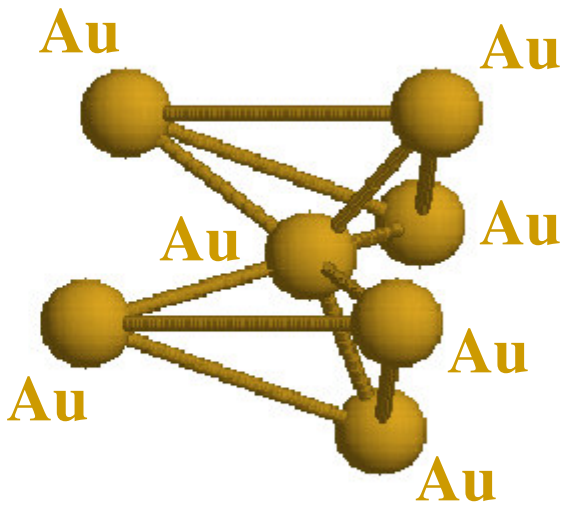
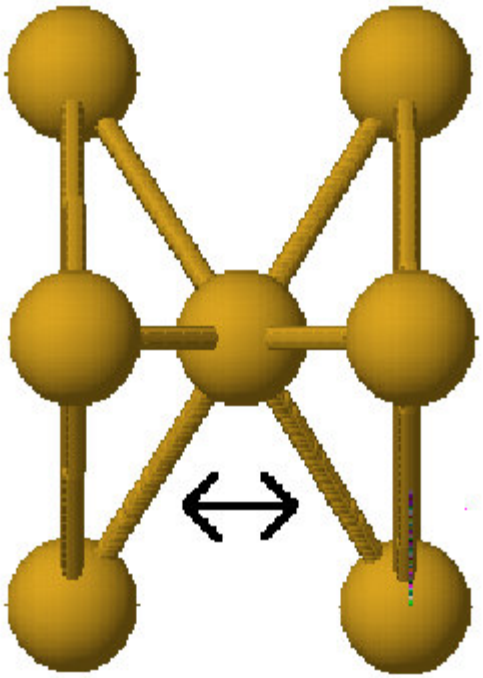
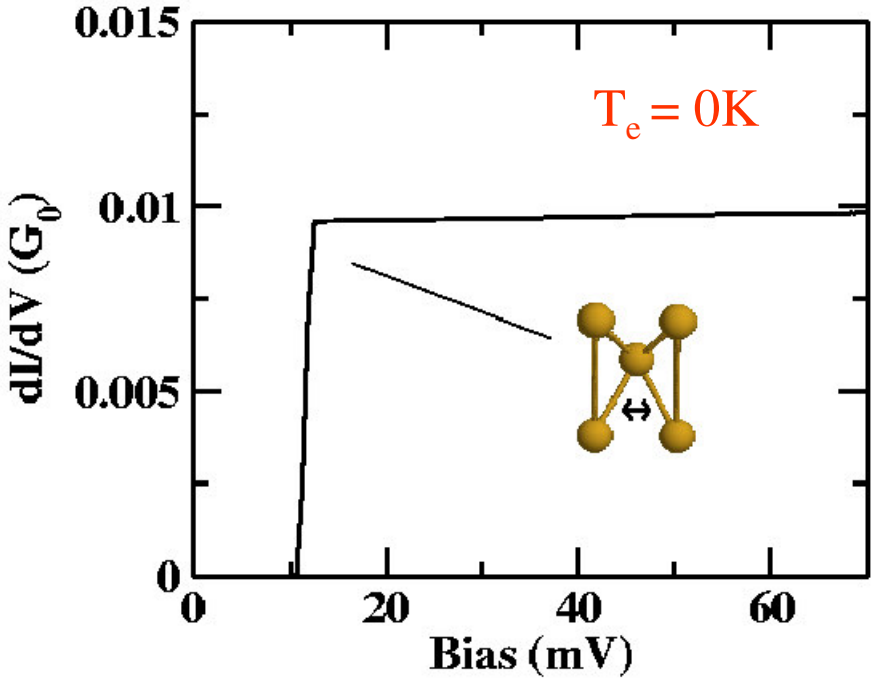


Gold point contact

Inelastic current: point contact

No Goldstone mode => onset bias



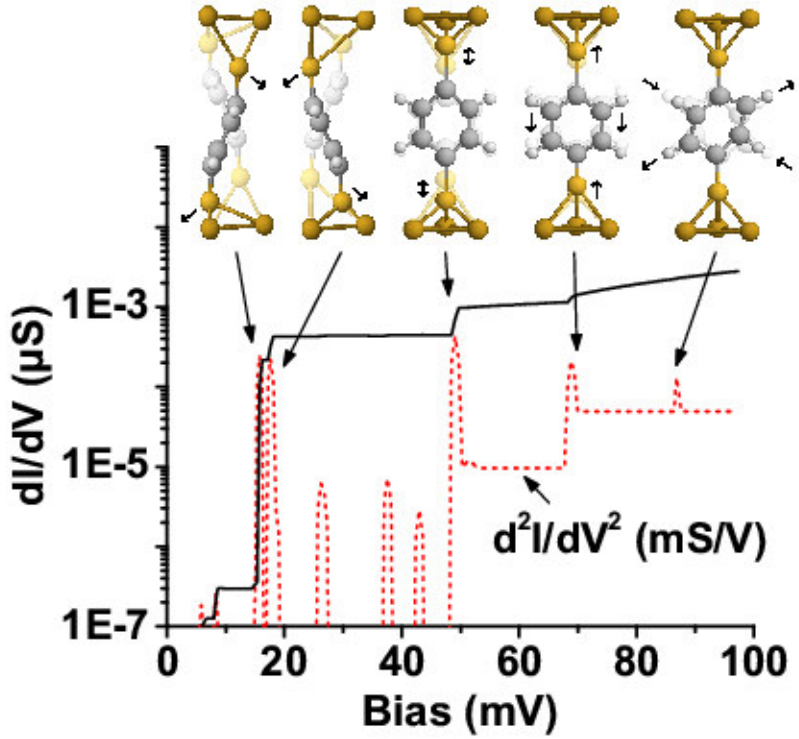


E = 11 meV

Onset bias ~ 11 mV

Exp: 1%
Agrait et al. PRL 2002

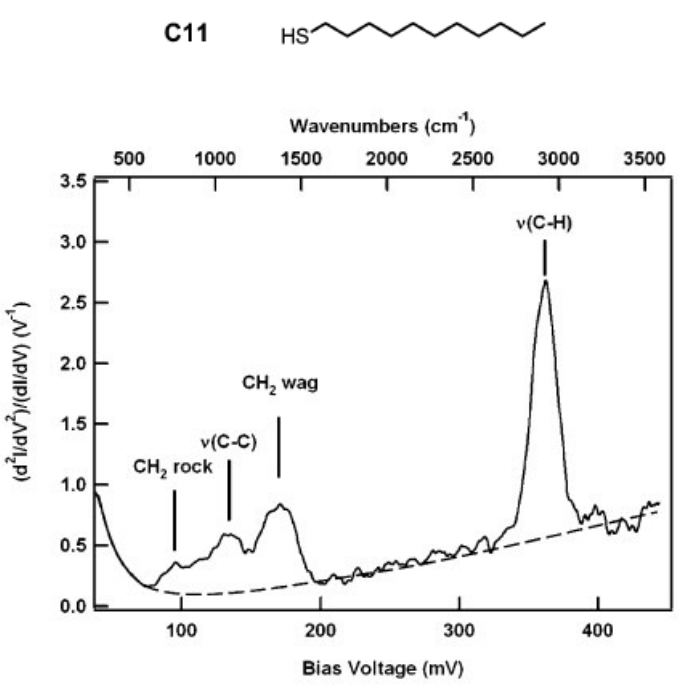
Inelastic current: molecule



Larger contribution from S and C displacements

$$H_{el-ion} = \sum_{\alpha\beta=L,R} \sum_{E_1E_2} \sum_{i\mu,j\nu} \sqrt{\frac{\hbar}{2\omega_{j\nu}}} A_{i\mu,j\nu} J^{i\mu,\alpha\beta}_{E_1E_2} a^{\alpha+}_{E_1} a^{\beta}_{E_2} (b_{j\nu} + b^{+}_{j\nu})$$

Inelastic current: experiments on alkanethiols



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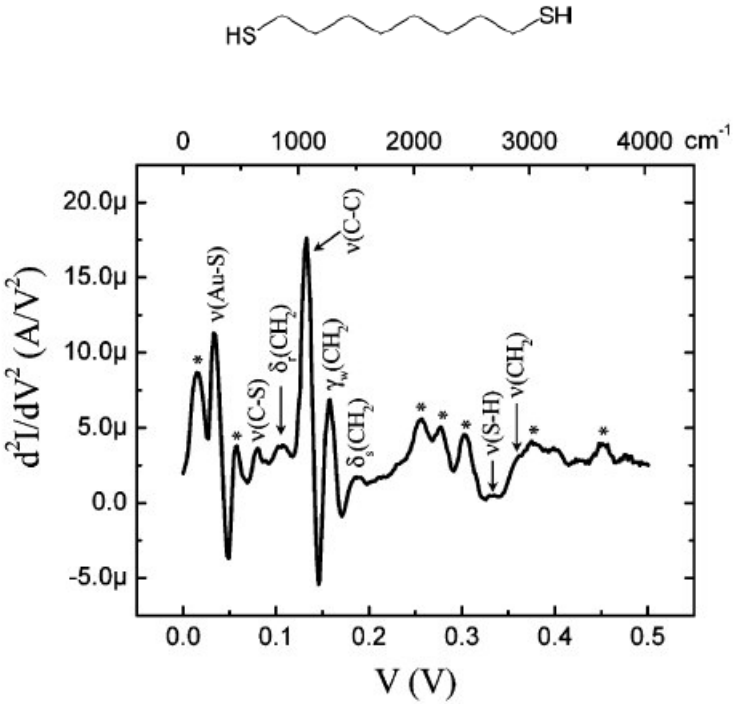

Wang *et al*., Nanoletters (2004)

Inelastic current: effect of contacts

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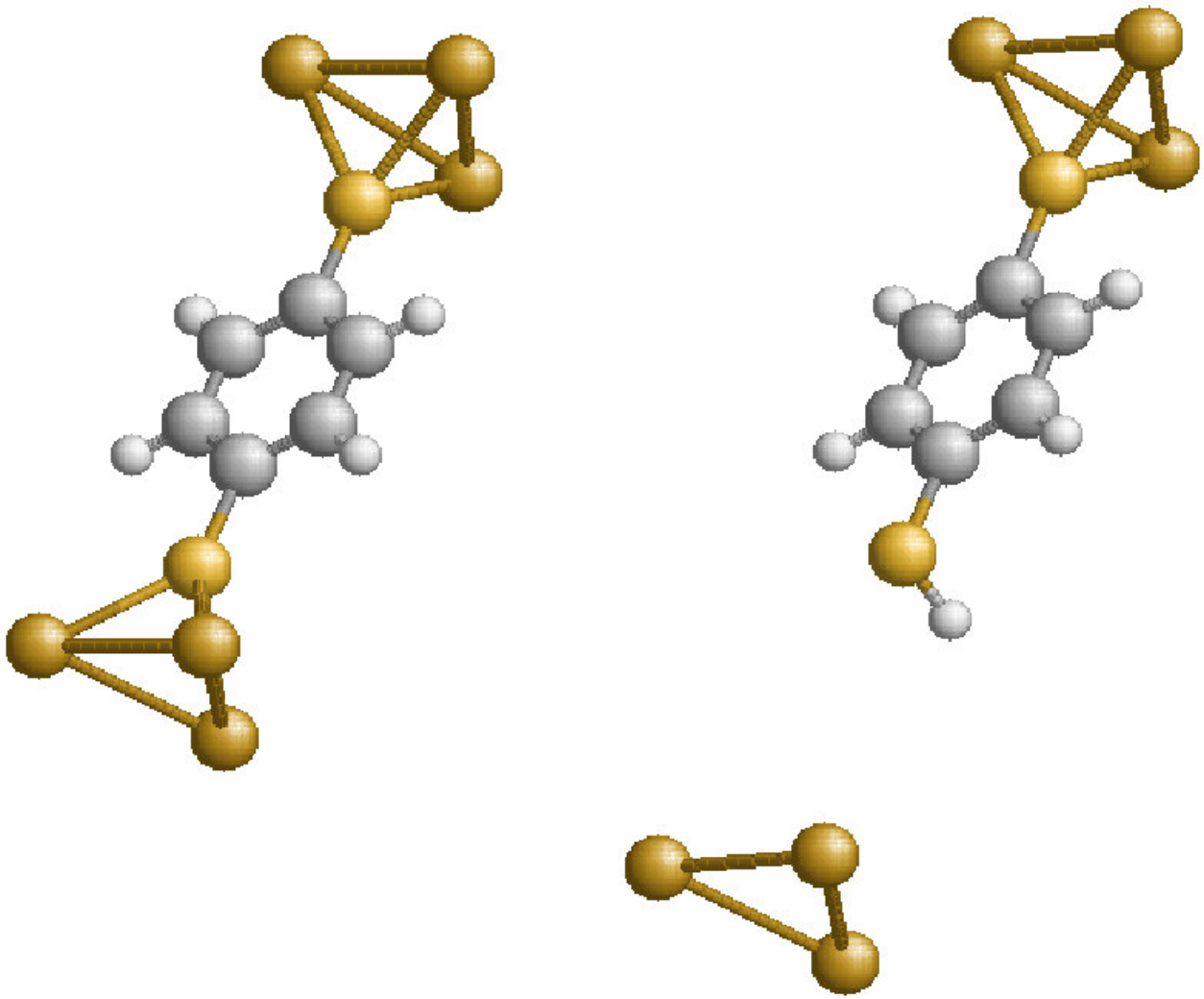
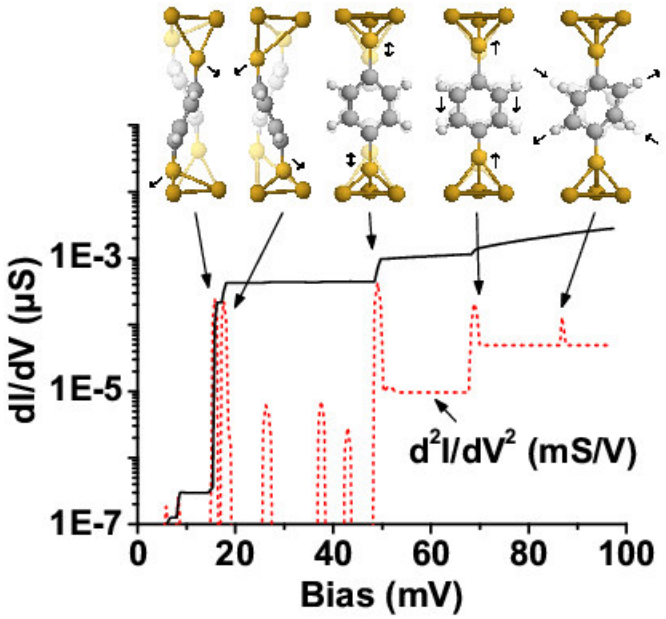
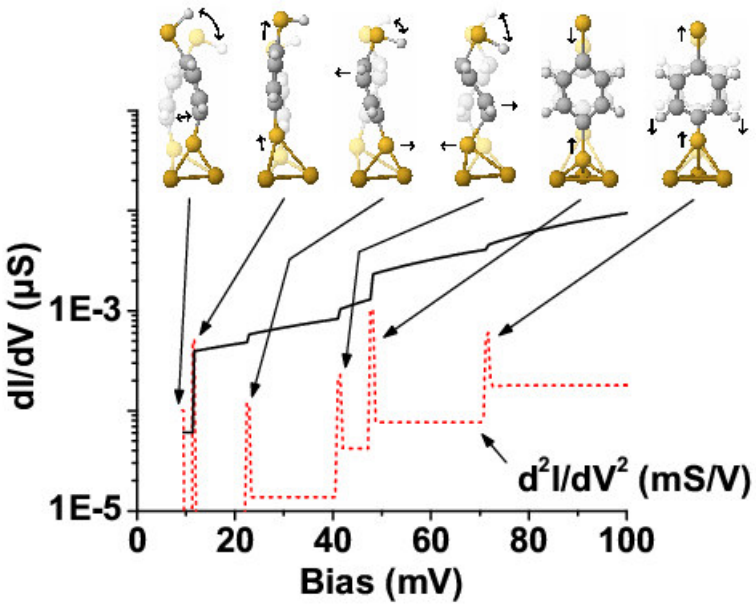
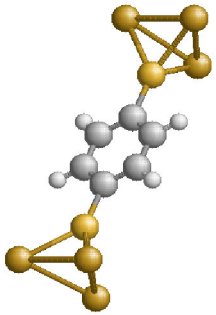
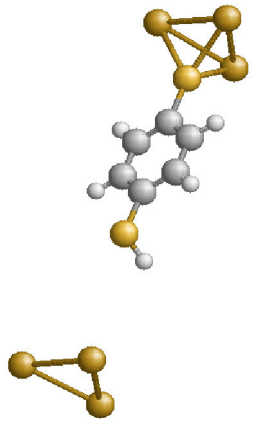


Inelastic current: effect of contacts

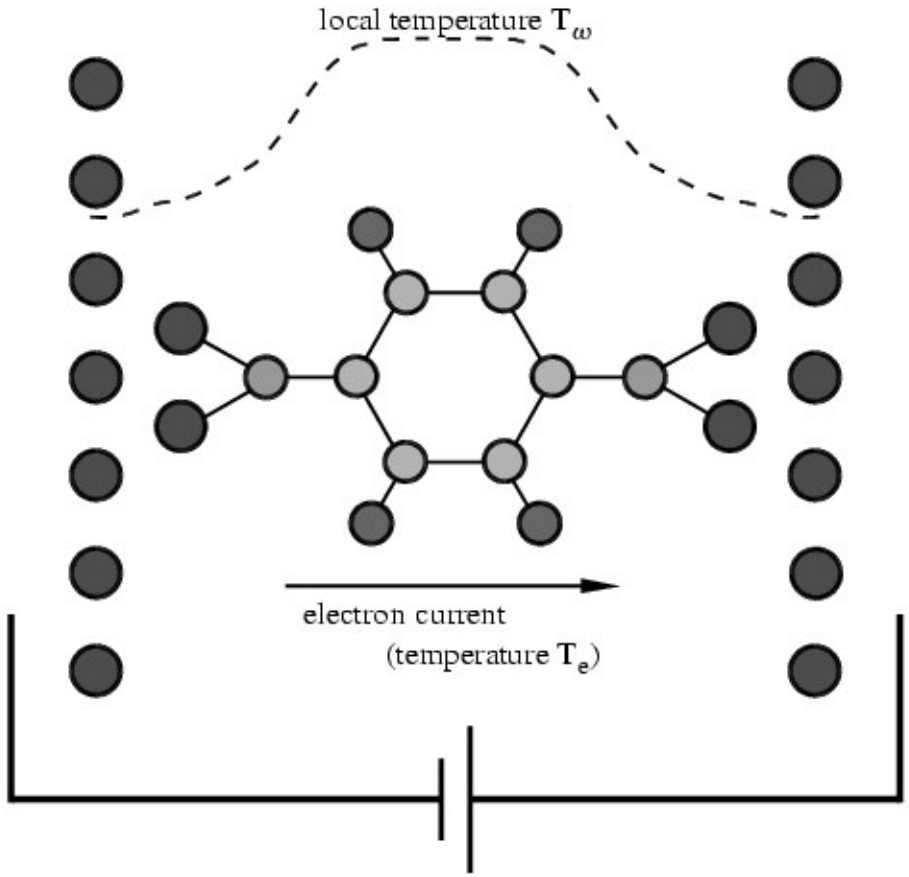








Current-induced mechanical effects



M. Di Ventra, T.N. Todorov, *Electrical Conduction in Nanoscale Systems* (Oxford University Press, to be published)

Current-induced forces

UCSD

Difficult to define forces in a non-equilibrium case

Eherenfest theorem: $$i\hbar \frac{d}{dt}\langle p \rangle = \langle [p, H] \rangle$$

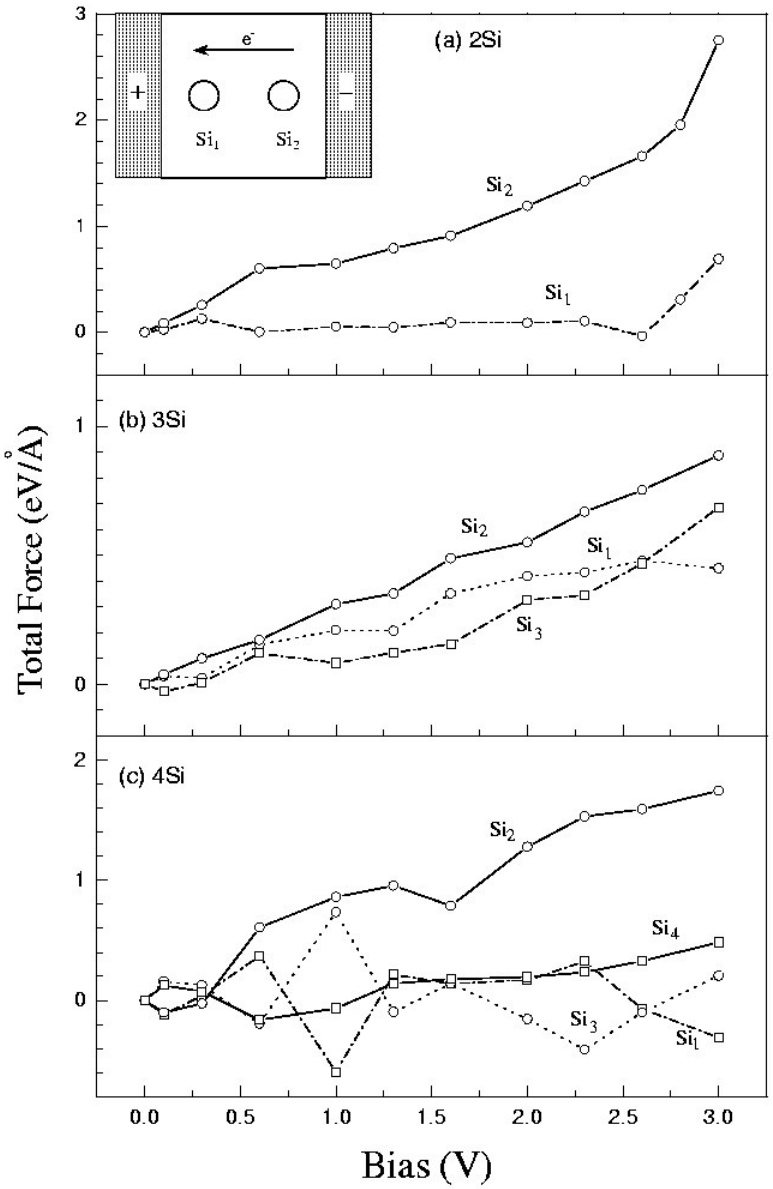
$$\Downarrow$$

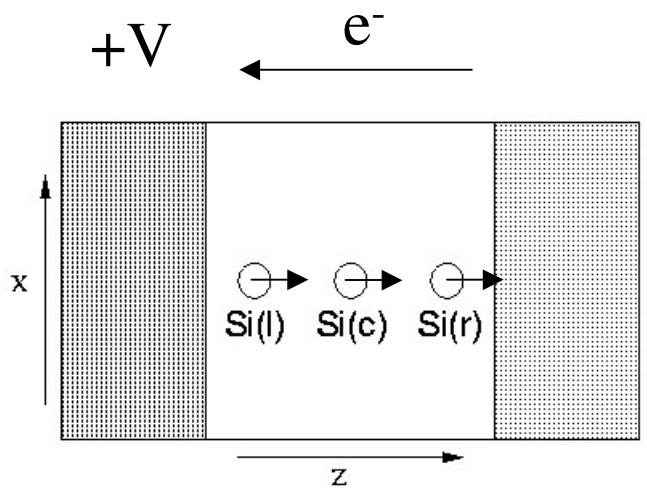
Forces : $$F = -\sum_i \underbrace{\left\langle \Psi_i \left| \frac{\partial H}{\partial R} \right| \Psi_i \right\rangle}_{\text{DISCRETE}} - \underbrace{\lim_{\Delta \to 0} \int_\sigma dE \left\langle \tilde{\Psi} \left| \frac{\partial H}{\partial R} \right| \tilde{\Psi} \right\rangle}_{\text{CONTINUUM+DIRECT}}$$

$$\left| \tilde{\Psi} \right\rangle = A \int_\Delta \Psi dE \in \ell^2(\Re^3)$$

M. Di Ventra *et al.*
Phys. Rev. B, 2000

Current-induced forces: atomic wires

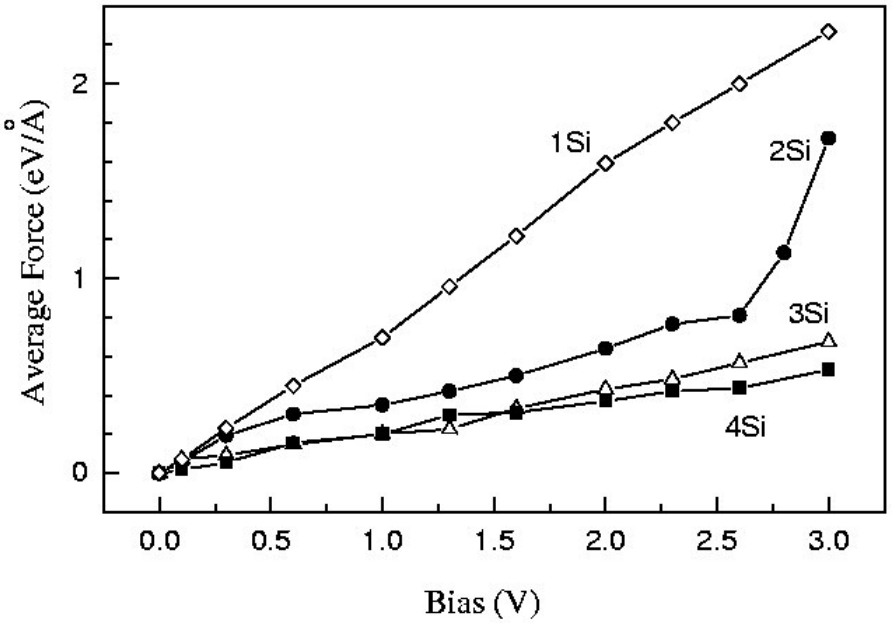




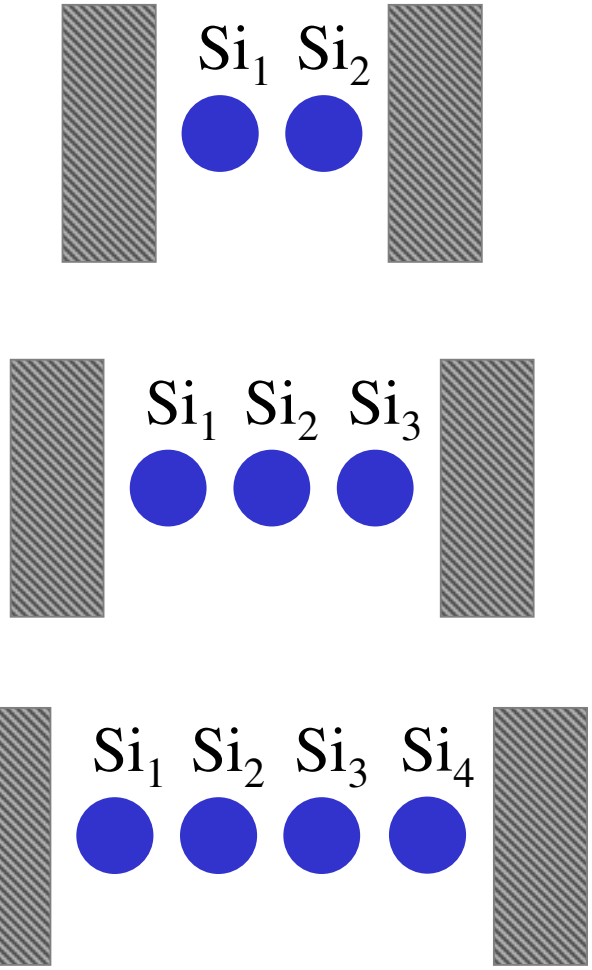
Strong non-linearities in current-induced forces

Z.Yang, M. Di Ventra
PRB, Rapid Comm. (2003)

Current-induced forces: atomic wires



$$\overline{F} = \frac{\sum_n F_n}{N}$$

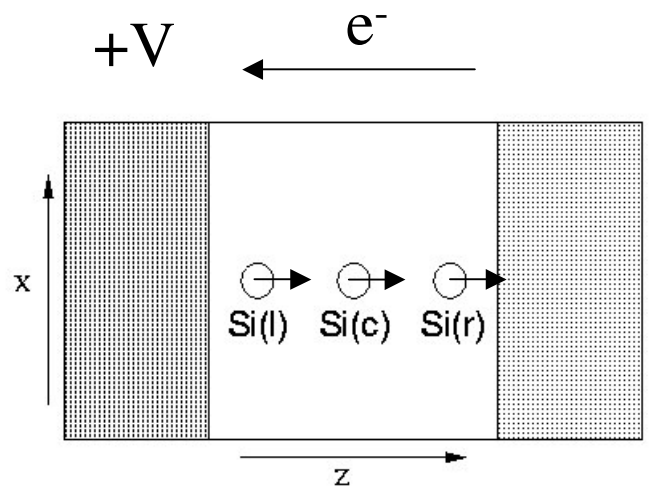


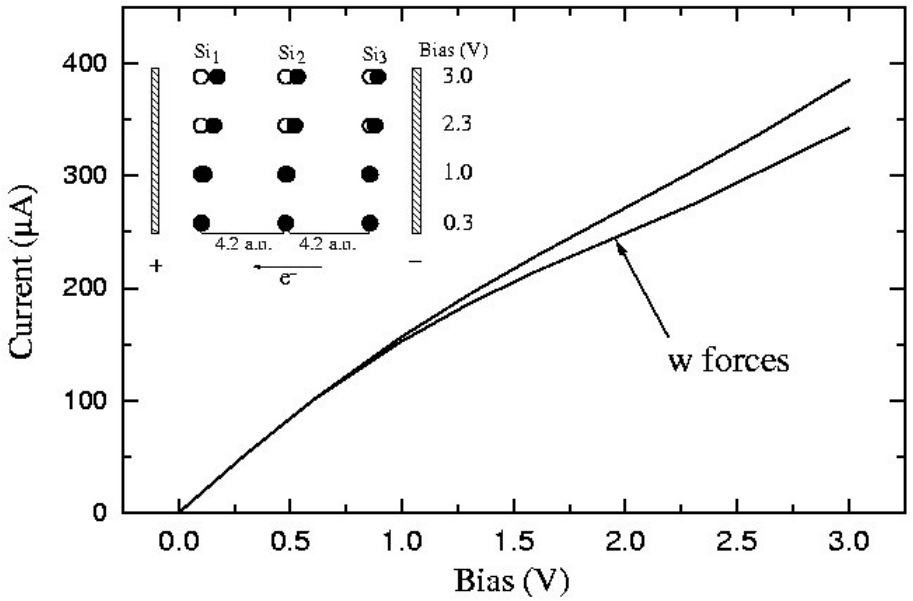
Shorter wires are easier to break

Exp: A.I. Yanson et al. *Nature* (1998)

Z.Yang, M. Di Ventra
PRB, Rapid Comm. (2003)

Current-induced forces: atomic wires





Z.Yang, M. Di Ventra
PRB, Rapid Comm. (2003)

Conclusions

UCSD

- Current-induced effects in nanostructures are very important
- We still need basic understanding of important issues (e.g. forces, heat)

Acknowledgements

Group:

Zhongqin Yang (Fudan Univ.)
Yu-Chang Chen (UCSD)
Mairbek Chshiev (VT)
Na Sai (UCSD)
Johan Lagerqvist (UCSD)
Neil Bushong (UCSD)
Mike Zwolak (CalTech)