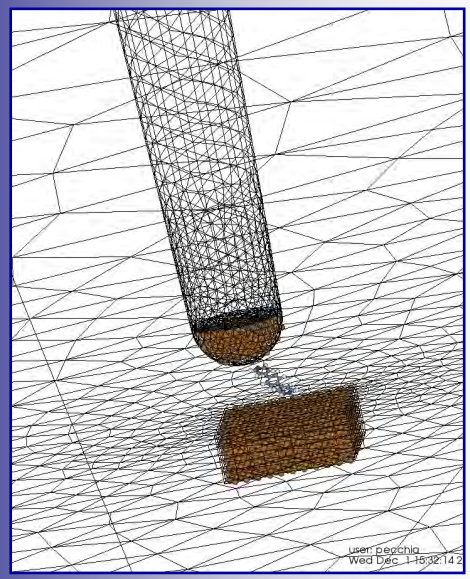
Heat dissipation in molecular junctions: linking molecules to macroscopic contacts



Alessandro Pecchia

CNR - ISMN ***Institute for Nanostractured Materials***

University of Roma "Tor Vergata"

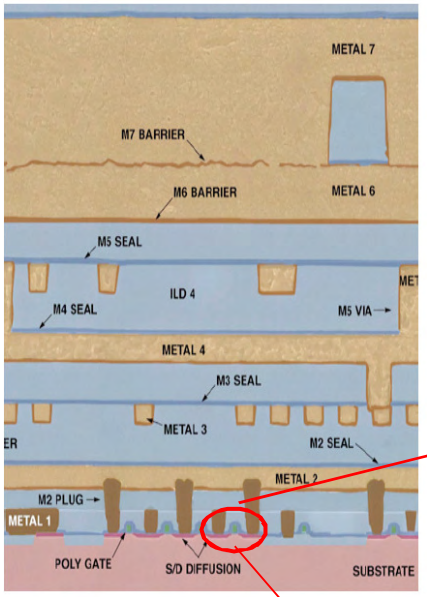
A Gagliardi, G. Romano, G. Penazzi,
M. Auf der Maur, Aldo Di Carlo

Outline

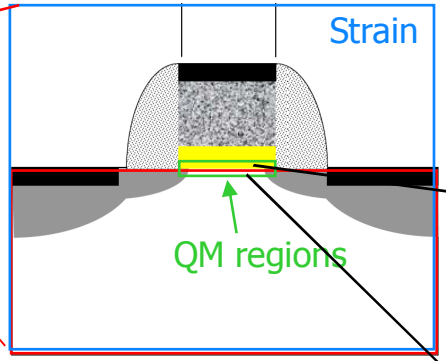
- Intro: Coupling Micro/macro scales
- The TiberCAD project
- Power dissipation at the nanoscale
- Example: heat dissipation in a STM junction

Different physical models are needed to describe electronic devices

Packadge Level

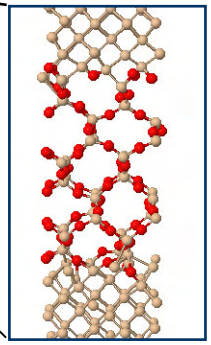


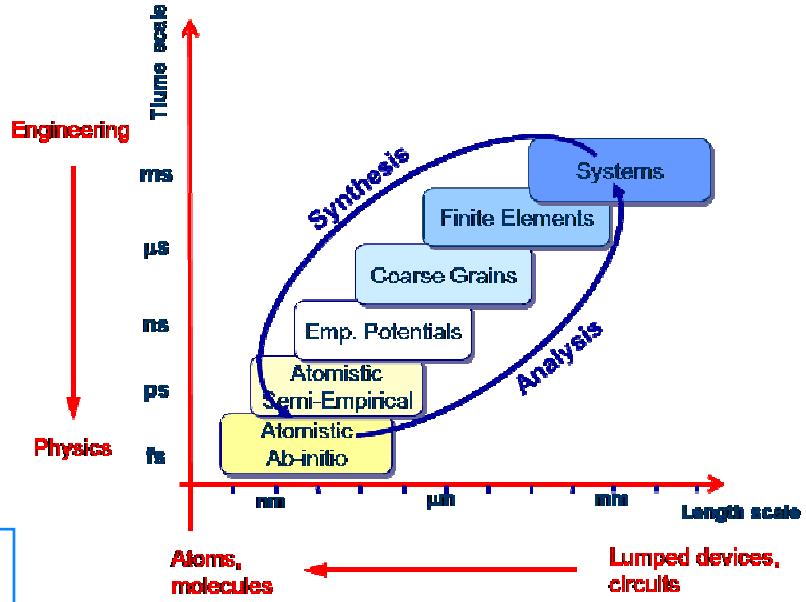
Device Level



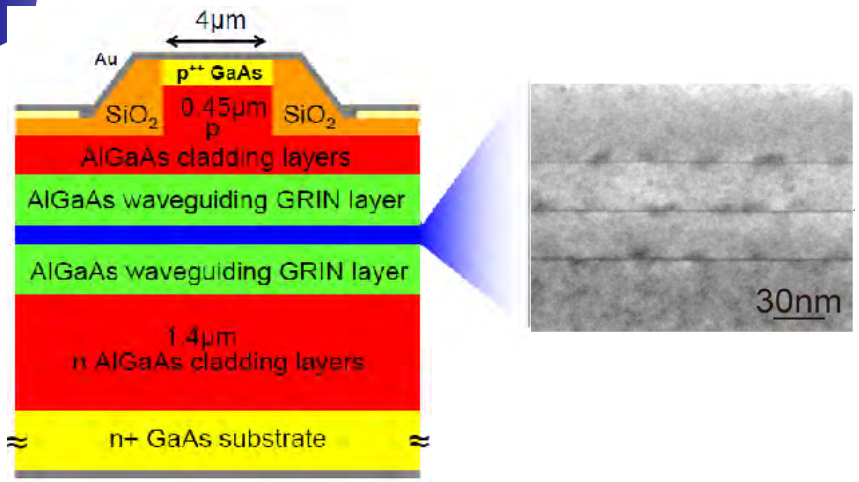
Poisson/Drift Diffusion

Atomistic Level

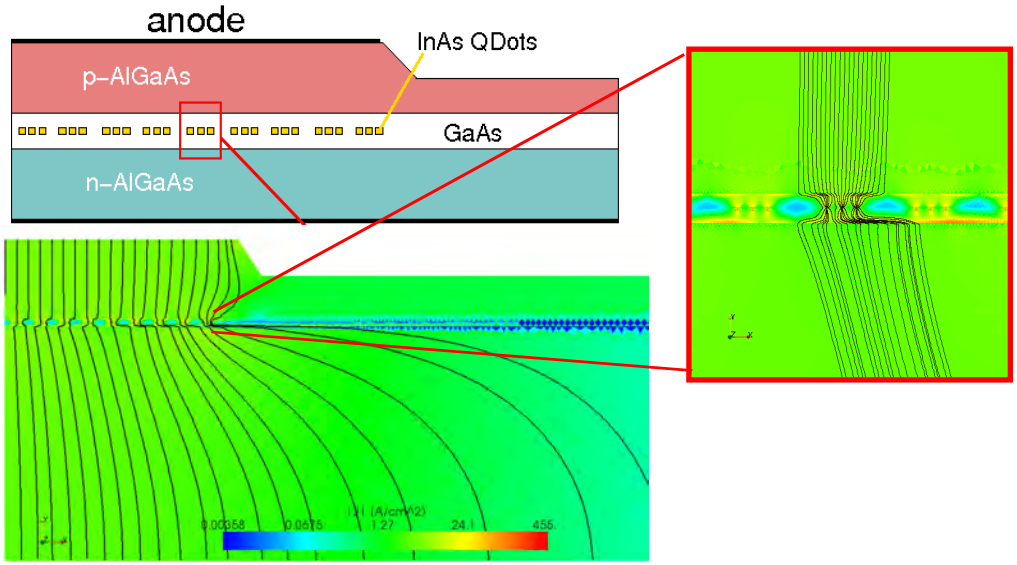


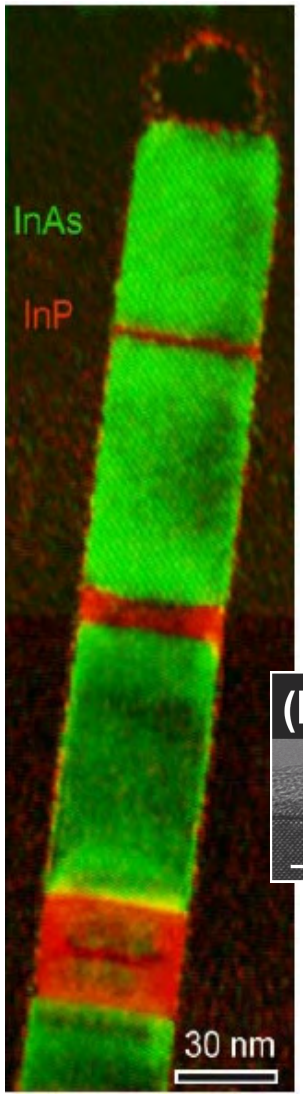


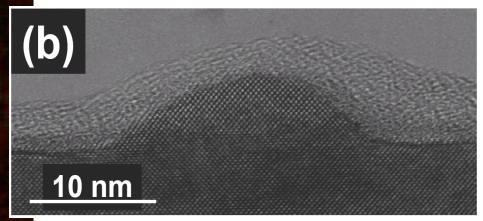
Optoelectronic devices



M. Buda et. al., IEEE Journal of Quantum Electronics, 2003

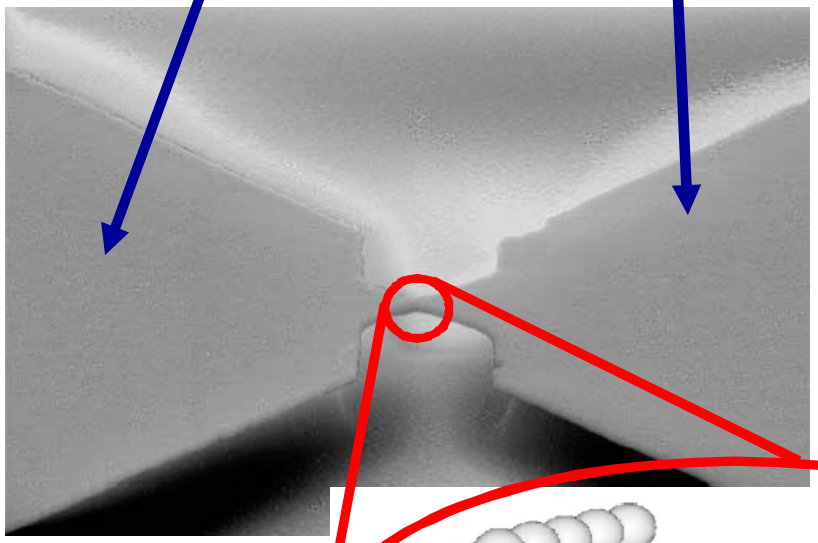


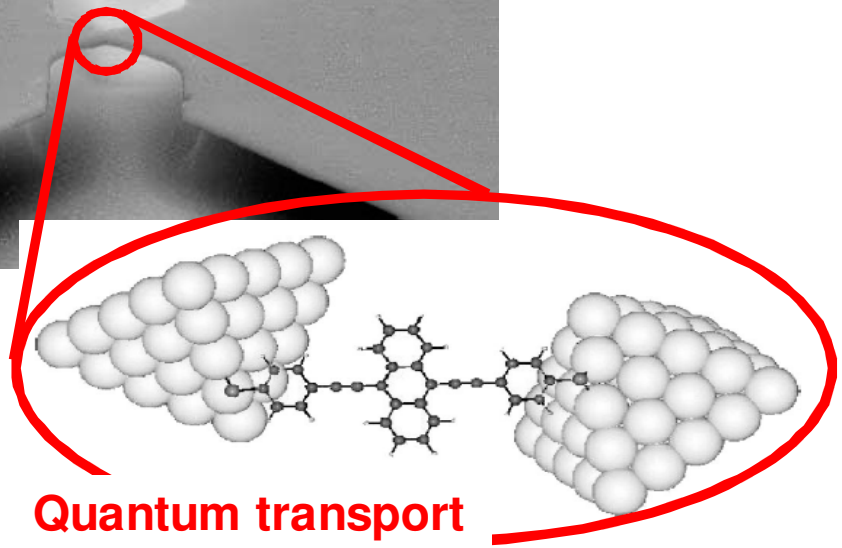




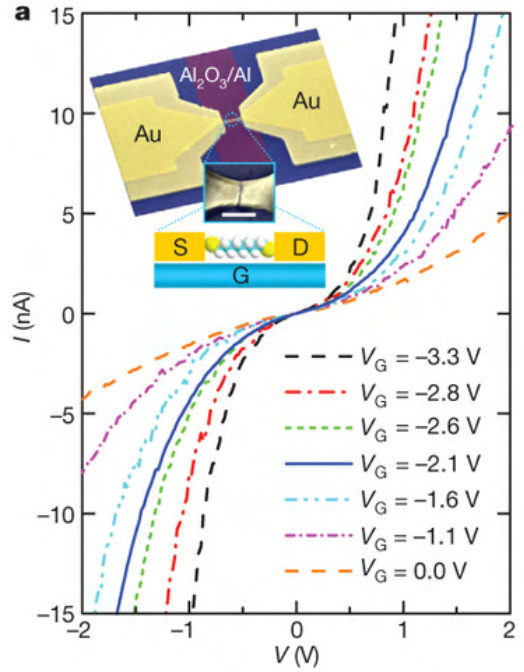
Molecular electronics

Electrodes: classical transport





Quantum transport

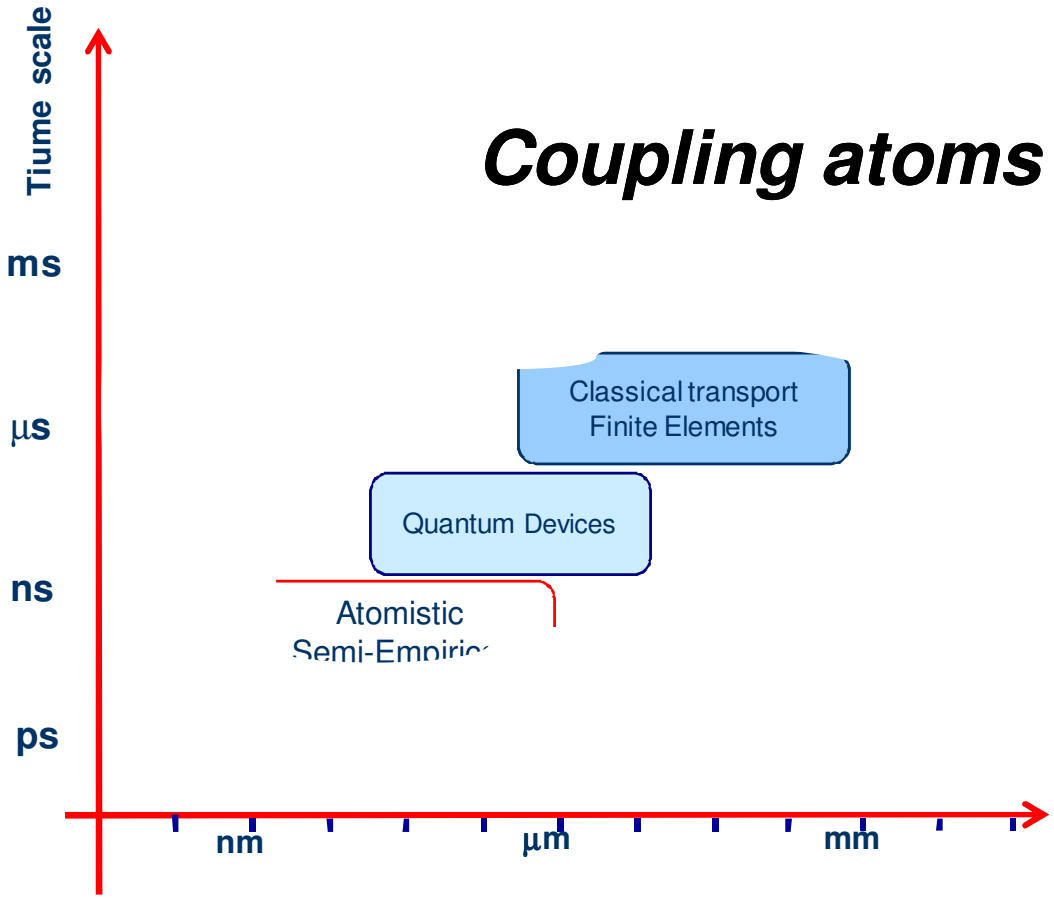


H.Song et al. *Nature* **462**, 1039-1043 (2009)

Atomically controlled junctions can now be gated showing I-V modulations

Precise modelling of the electrostatics of the whole device are important for accurate predictions of the electronic transport properties

Coupling atoms with finite elements



TiberCAD people

Prof. Aldo Di Carlo

Dr. A. Pecchia

Dr. M. Auf der Maur

tiberCAD

Multiscale Device Simulator

http://www.tibercad.org

Dr. A. Gagliardi

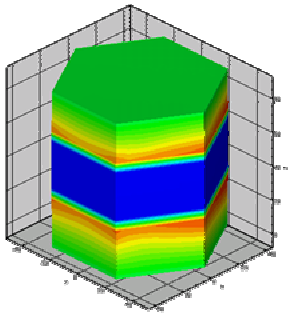
Dr. F. Sacconi

Dr. G. Romano

Dr. G. Penazzi

FEM/atomistic approaches

FEM



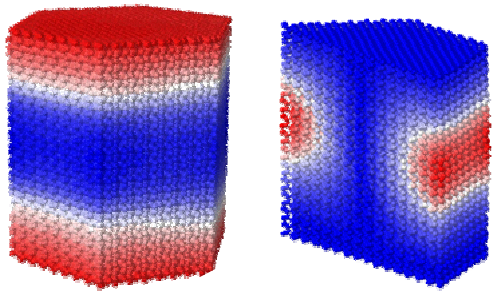
Potentials
(electrostatic, piezo, …)

Strain and deformations

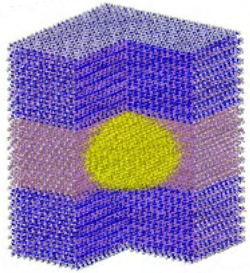
Projection on atoms

Projection on FEM

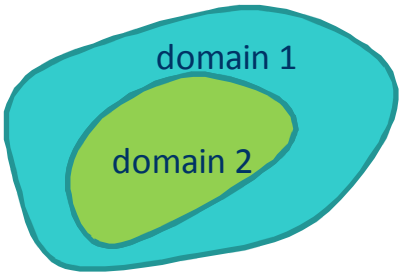
ATOMISTIC



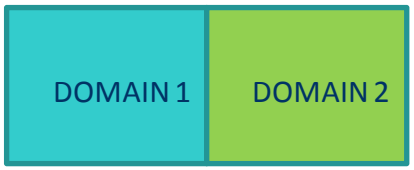
Electrostatic map
Deformation map
(Wavefunctions)
Charge density
Current



Different simulation domains exchange data

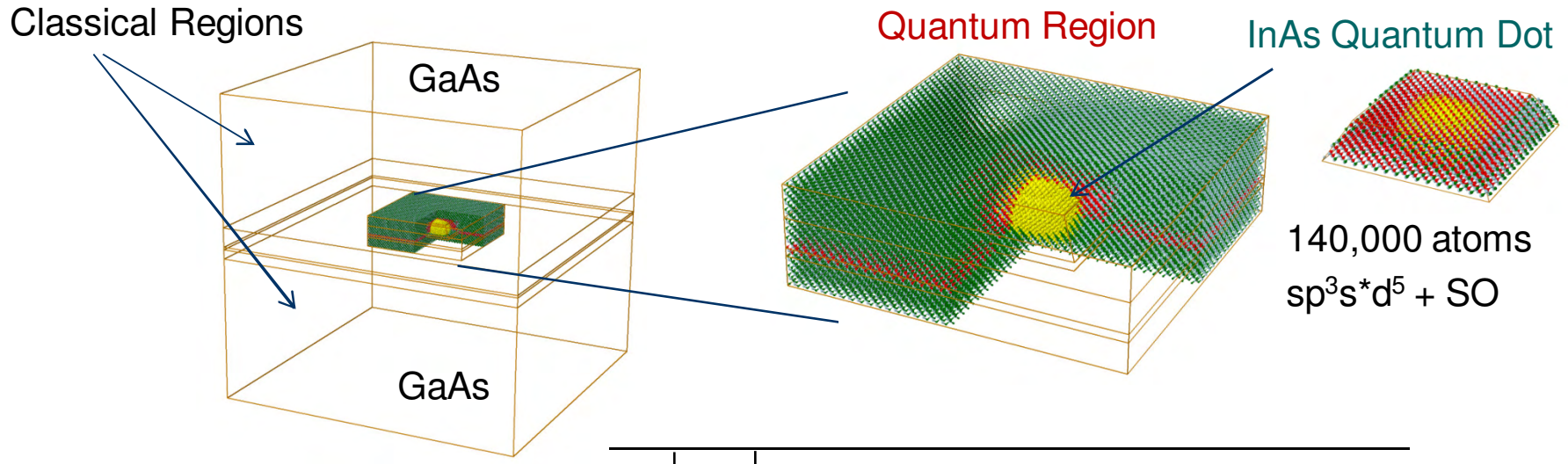


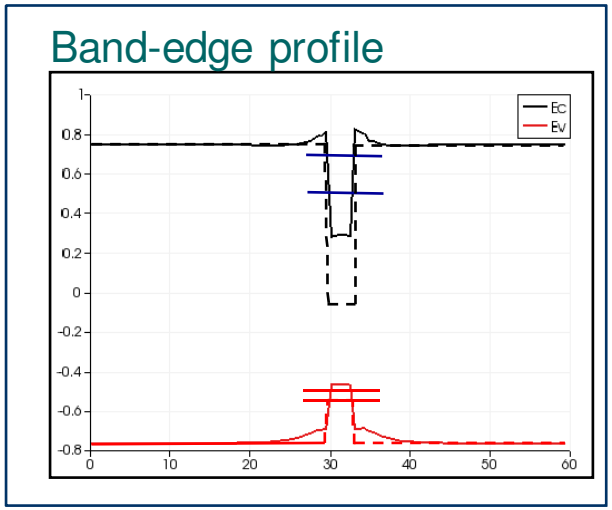
Overlap

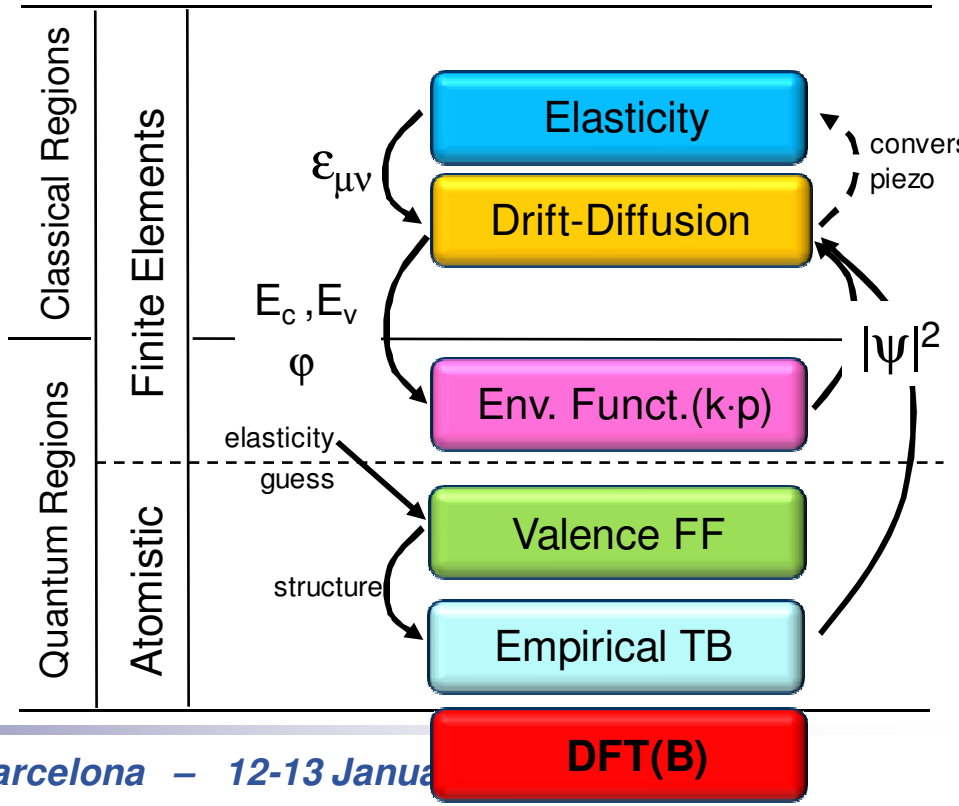


Bridge

Coupling models and scales

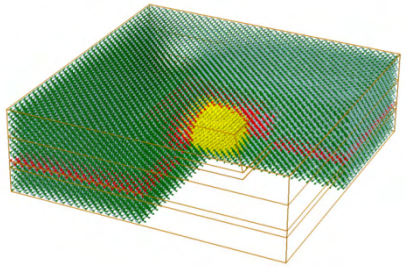


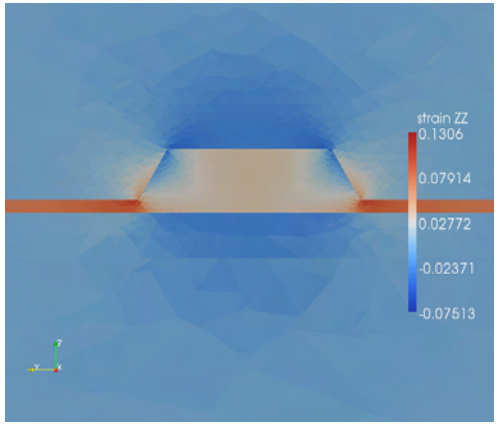




M. Auf der Maur IEEE Trans Elect. Dev. (2011)

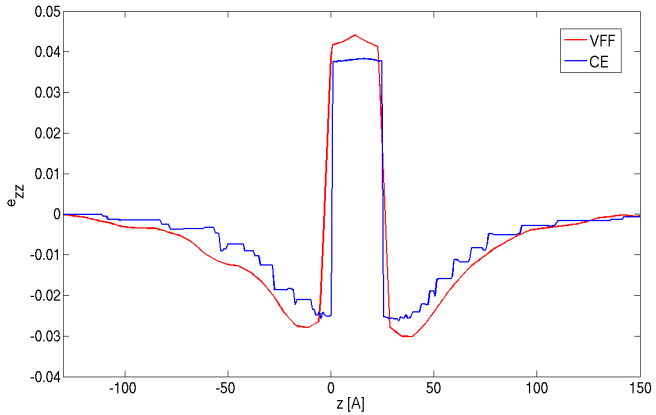
InAs Qdot in GaAs





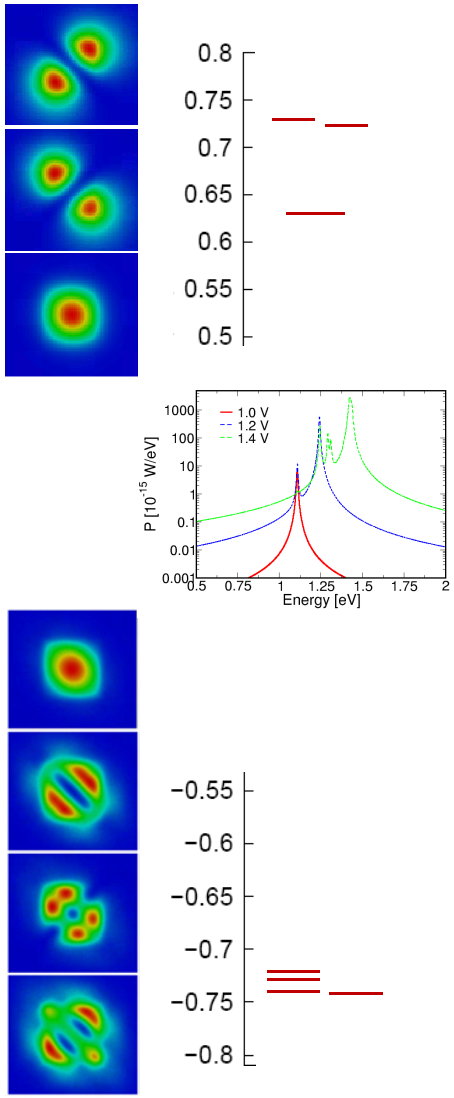
Strain map due to lattice mismatches

Projection of strain on the atoms and VFF relaxations

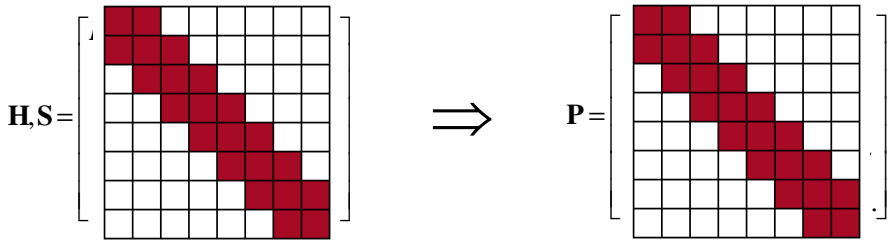


up to **15%** differences due to VFF corrections

TB eigenstates



Coherent Transport

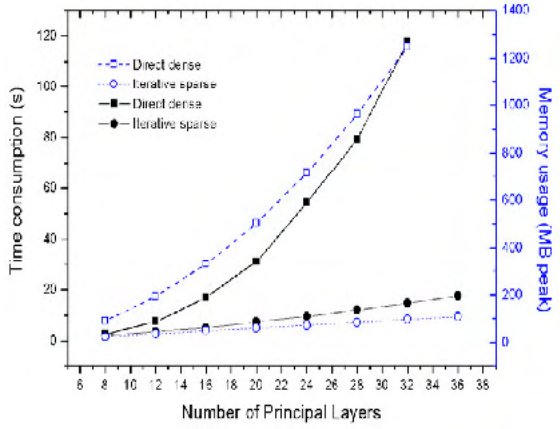


$$q_\mu = \sum_\nu \mathbf{P}_{\mu\nu}\mathbf{S}_{\nu\mu}$$

O(N m^3)

- **General API to accept any Hamiltonian (overlap)**
- Thread safe
- OpenMP parallel (via MKL)
- *MPI on energy integration*

Linear scaling with wire length



PROFILING: SiNW of 5000 atoms

Charge density and potential:	**46 h**
(**37** SCC iterations of	**1h15**)
Density of States (150 points):	**6 h**
Peak memory:	**1.2 Gb**

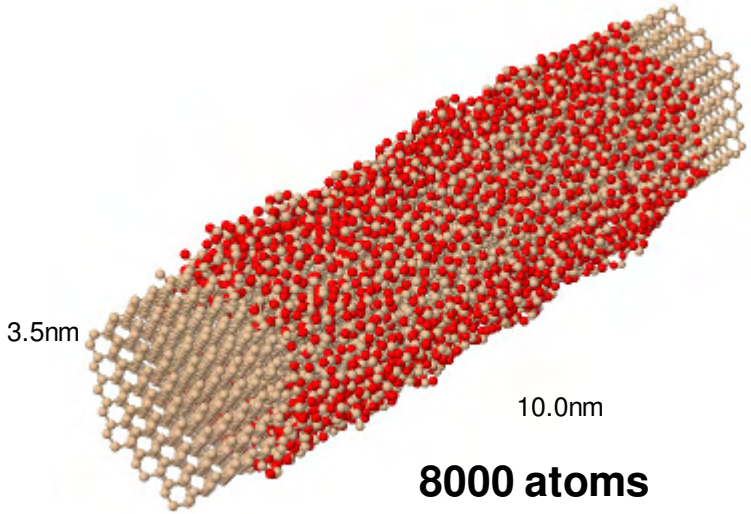
Calculations on single PC Linux Quad-Core Intel(R) Xeon Qcore W3530 @ 2.80 GHz

[Penazzi, et al. New J. Phys. 10 (2008)]

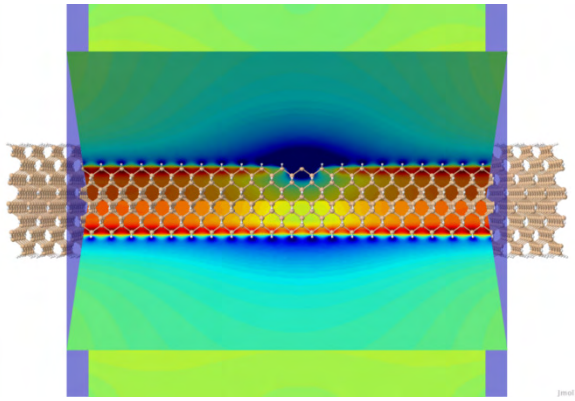
Transport in dftb+

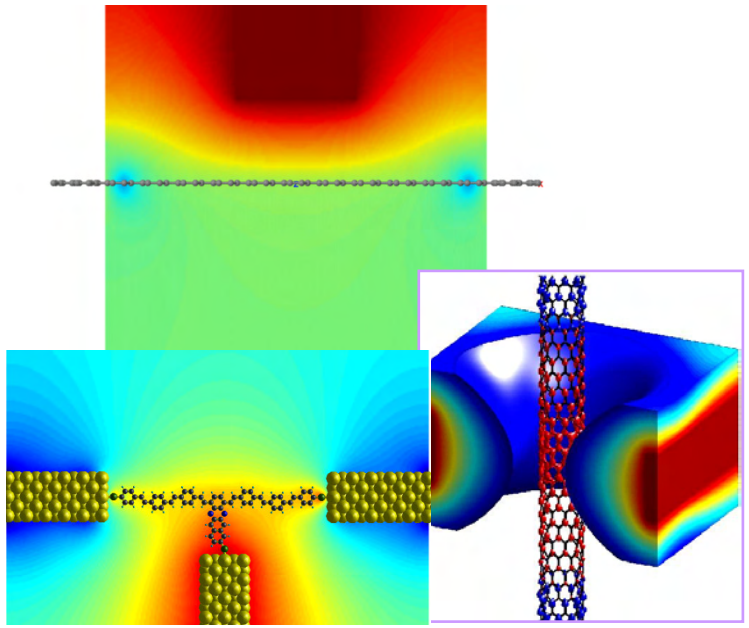
NEGF Transport has been embedded in the dftb+ code

http://www.dftb.org



8000 atoms





- dftb+ user friendly input
- k-point integrations
- real-space poisson with different BC like gates
- Efficient OpenMP parallel (via MKL)
- *MPI on energy integration -> to do*

DFTB = DFT based Tight-Binding method

Kohn-Sham equation: $\Longrightarrow$ $$\sum_{\nu}\left[H^{0}_{\mu\nu}+H^{Scc}_{\mu\nu}[\delta n]-E_{k}S_{\mu\nu}\right]c^{k}_{\nu}=0$$

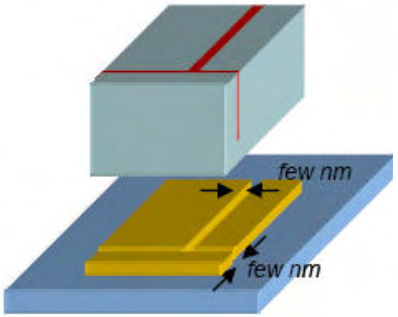
$$H_{\mu\nu}=\begin{cases}\varepsilon_{\mu} & \text{onsite atomic energy levels}\\ \langle\mu|V[n^{0}_{\mu}+n^{0}_{\nu}]|\nu\rangle & \text{two-centre density superposition}\end{cases}$$

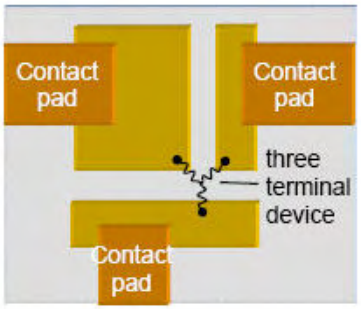
$$S_{\mu\nu}=\langle\mu||\nu\rangle$$

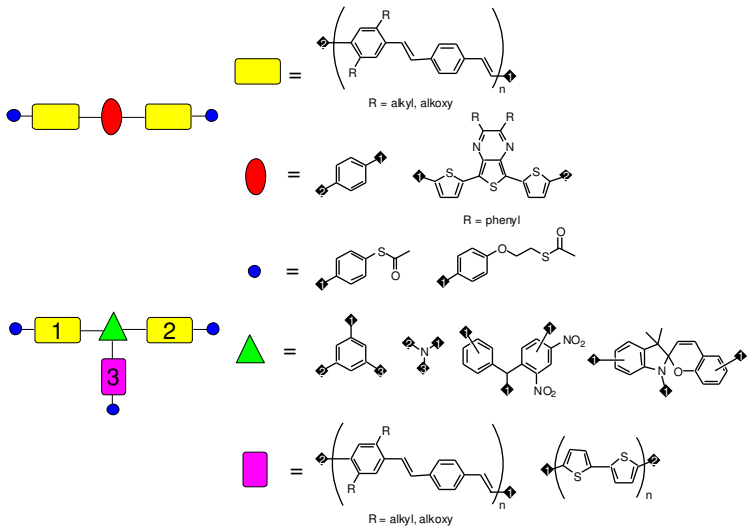
$$H^{Scc}_{\mu\nu}=\frac{1}{2}S_{\mu\nu}\sum\left(\gamma_{\bar{\mu}\bar{\sigma}}+\gamma_{\bar{\nu}\bar{\sigma}}\right)\Delta q_{\bar{\sigma}}$$

[Elstner, et al. Phys. Rev. B 58 (1998) 7260]

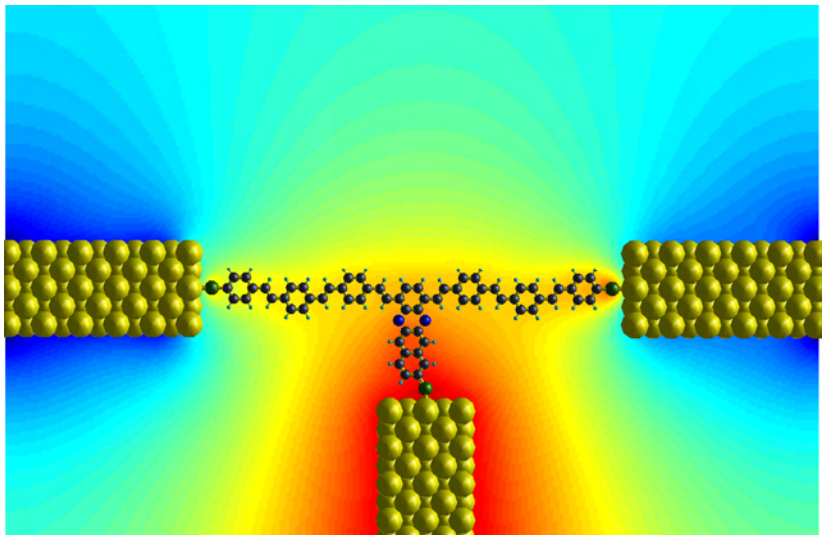
Three terminal devices

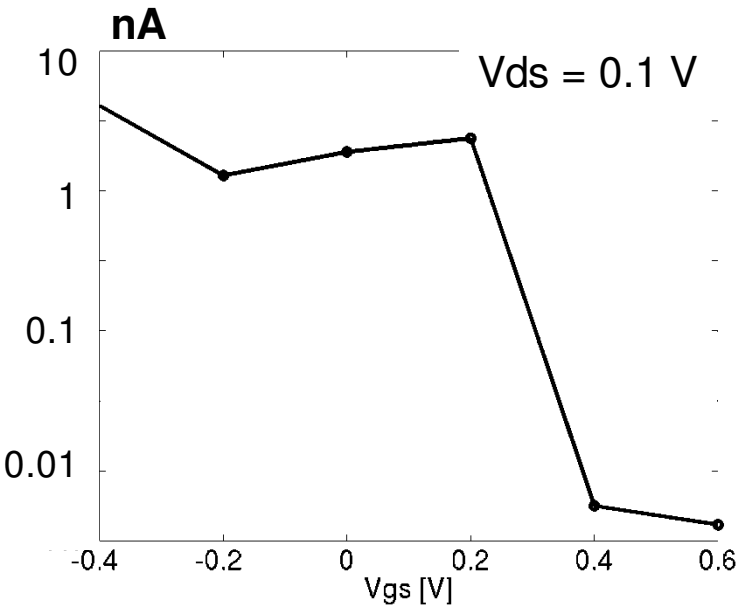




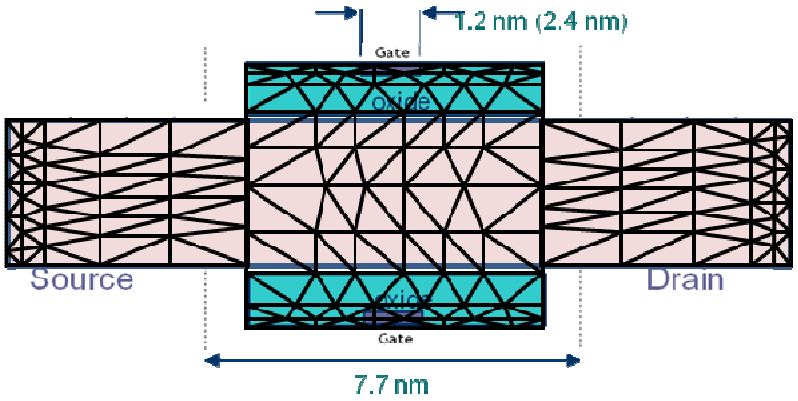


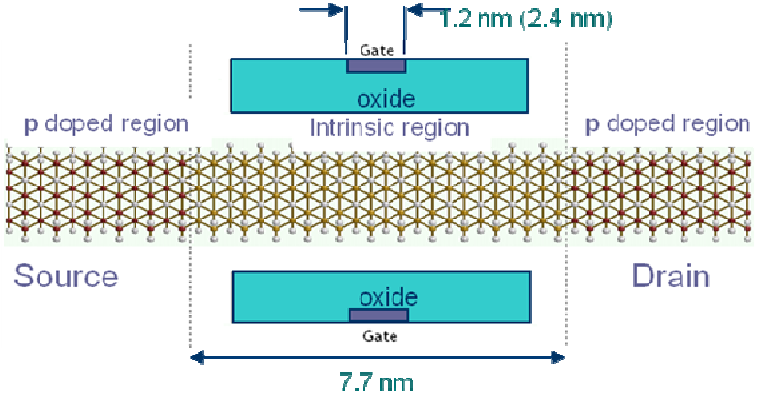
OPV – based transistor





FEM/ATOMISTIC

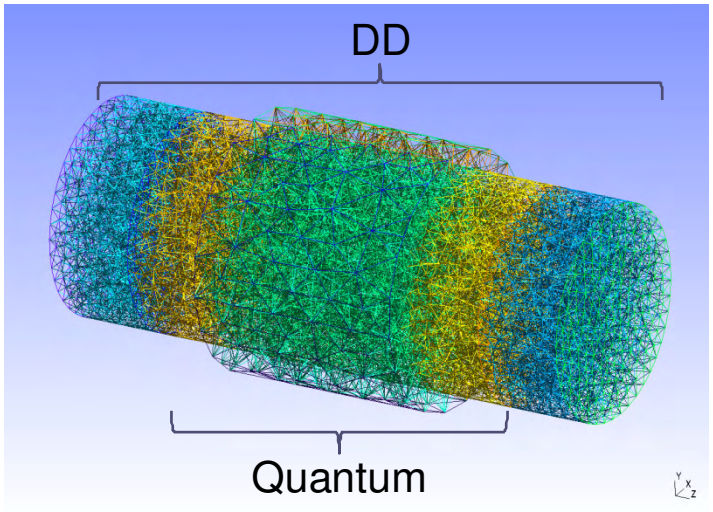


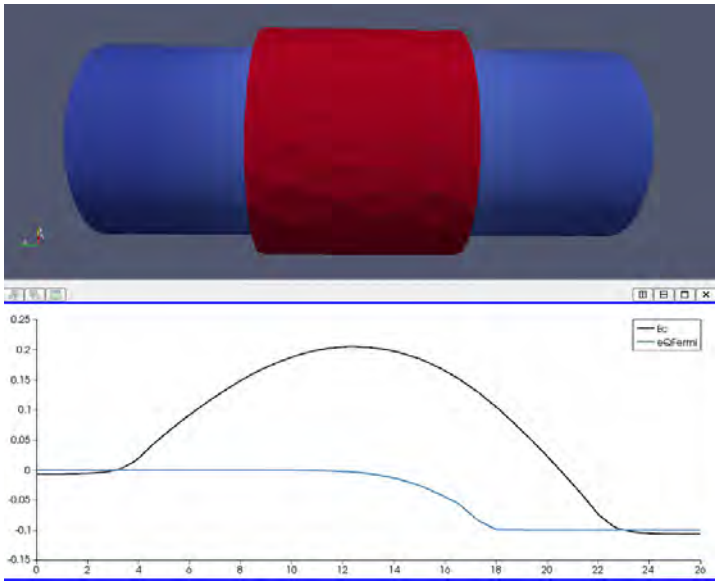


Integration of *lib*NEGF in TiberCAD almost completed

- Automatic mesh extensions for contact definitions ✓
- Automatic mesh partitioning taking into account contacts ✓
- C++ interface to the library ✓
- Automatic treatment of atomistic contacts
- Port all developed electron-phonon scattering

Example:T(E) in a SiNW FET

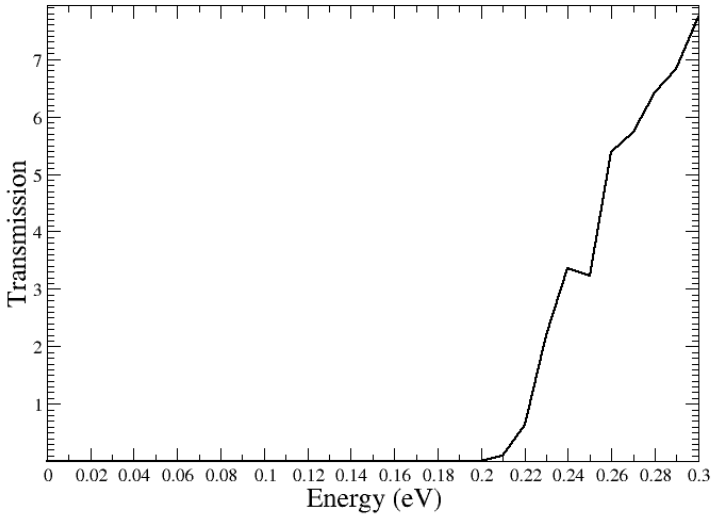




Drift-Diffusion is solved on the whole device

Assemble an effective mass Hamiltonian

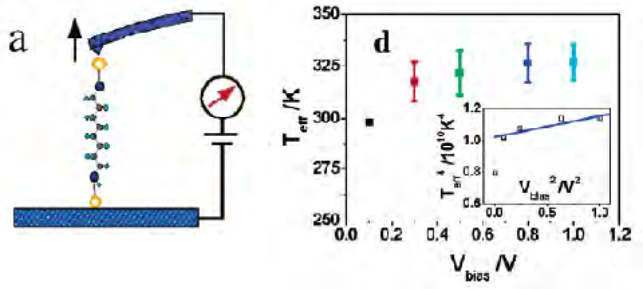
Compute tunneling and current



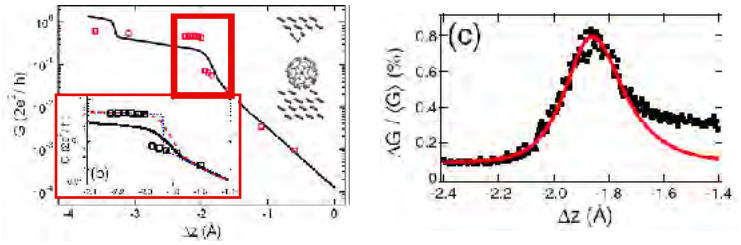
Power Dissipation at molecular scale

Molecular heating&cooling

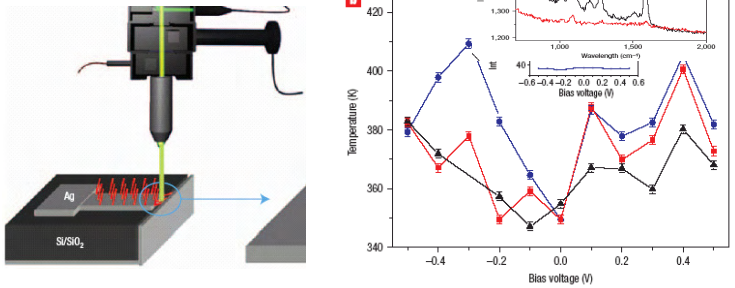
- Thermal effects at the molecular scale represent an increasingly 'hot' topic
- Theoretical and experimental challenges to measure nanoscale temperatures



Z. Huang et al. Nano Lett. 6, 1240 (2006)

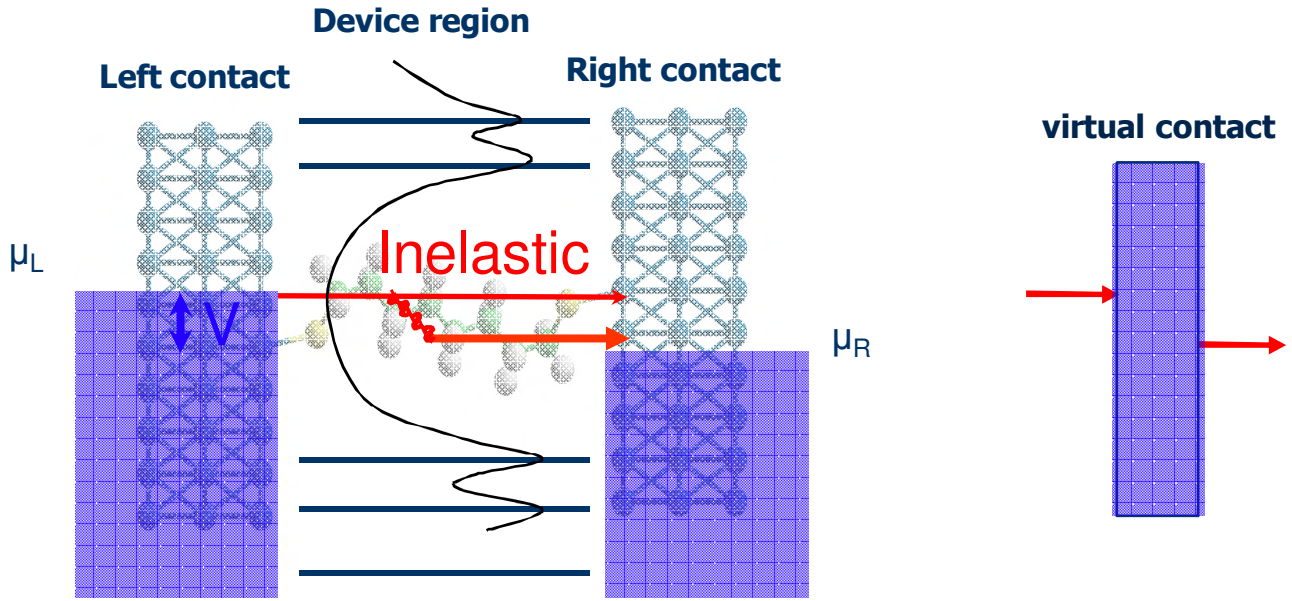


N. Néel et al. PRL 98, 065502 (2007)



Z. Ioffe et al. Nature Nanotech., on-line doi:10.1038/nnano.2008.304

Heat dissipation



The rate of emitted phonons depends on the power emitted:

$$W_q = \frac{2}{h}\int E \cdot Tr[\Sigma_q^<(E)G^>(E) - \Sigma_q^>(E)G^<(E)]dE$$

$$R_q = \frac{W_q}{\hbar\omega_q} = (N_q + 1)E_q - N_q A_q$$

electron-phonon couplings

$$\hat{H} + \delta\hat{H} = \sum_{\mu} \varepsilon_{\mu} c_{\mu}^{\dagger} c_{\mu} + \sum_{\mu,\nu} \sum_{q} \alpha_{\mu\nu}^{q} c_{\mu}^{\dagger} c_{\nu} (a_q^{+} + a_q)$$

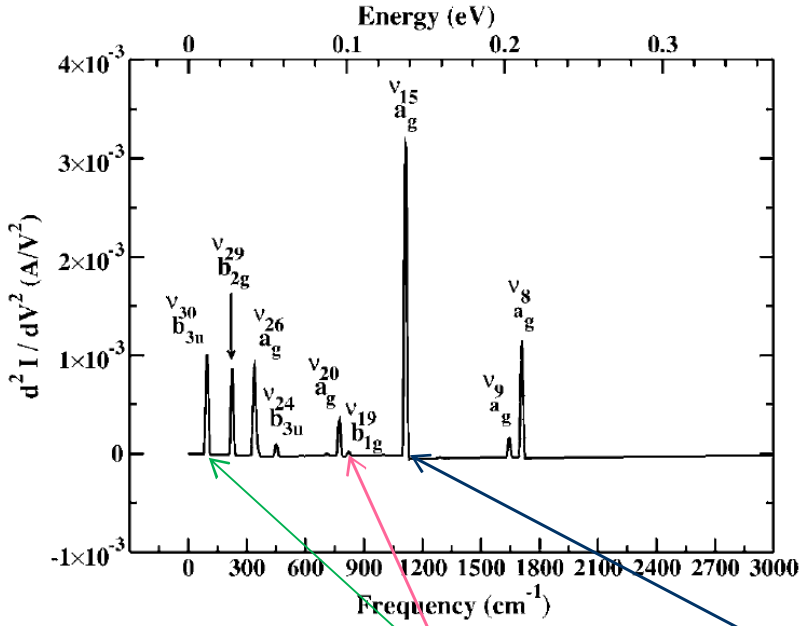
The electron-phonon coupling Hamiltonian is derived by expanding to first order the DFTB-Hamiltonian with respect to the atomic positions.

$$\alpha_{\mu\nu}^{q} = \sum_{I,\beta} \sqrt{\frac{\hbar}{2\omega_q m_I}} \left\langle \mu \left| \frac{\partial \hat{H}}{\partial Q_{I,\beta}} \right| \nu \right\rangle_{Q=0} e_{I,\beta}^{q}$$

Electron phonon self-energy, perturbation theory:

$$\Sigma_{ph}^{<,>}(E) = \frac{i}{2\pi} \sum_{q} \int dE' \alpha^{q} G^{<,>}(E - E') \alpha^{q} D_{q}^{<,>}(E')$$

Propensity rules



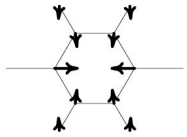
$$I_q = \sum_{ij} A_{ii}^L \mid \alpha_{ij}^q \mid^2 A_{jj}^R$$

Few dominant channels with rep:

A_1 (a_{1g} , b_{1u})

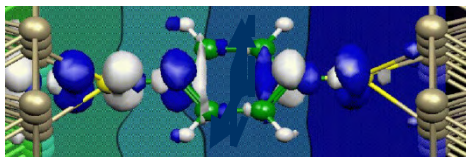
B_1 (b_{3u} ,b_{2g})

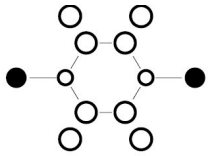
Weak signals: **A_2 (a_{1u}, b_{1g})**



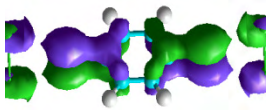
$$A_1 = B_1 \otimes A_1 \otimes B_1$$

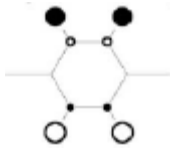
C_{2v}



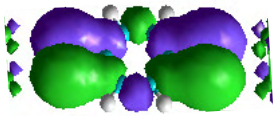


$$A_1 = A_1 \otimes B_1 \otimes B_1$$





$$A_1 = B_1 \otimes A_2 \otimes B_2$$

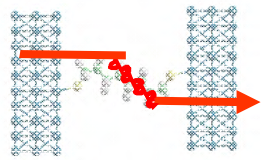


Solomon et al.*J.C.P.* **125** (2006) 184702

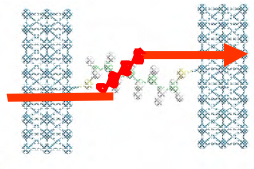
A. Gagliardi et al., *PRB 75* (2007)

Electron-vibron interactions

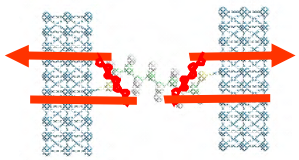
Phonon emission (E_q)



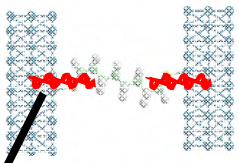
Phonon absorption 1 (A_q)

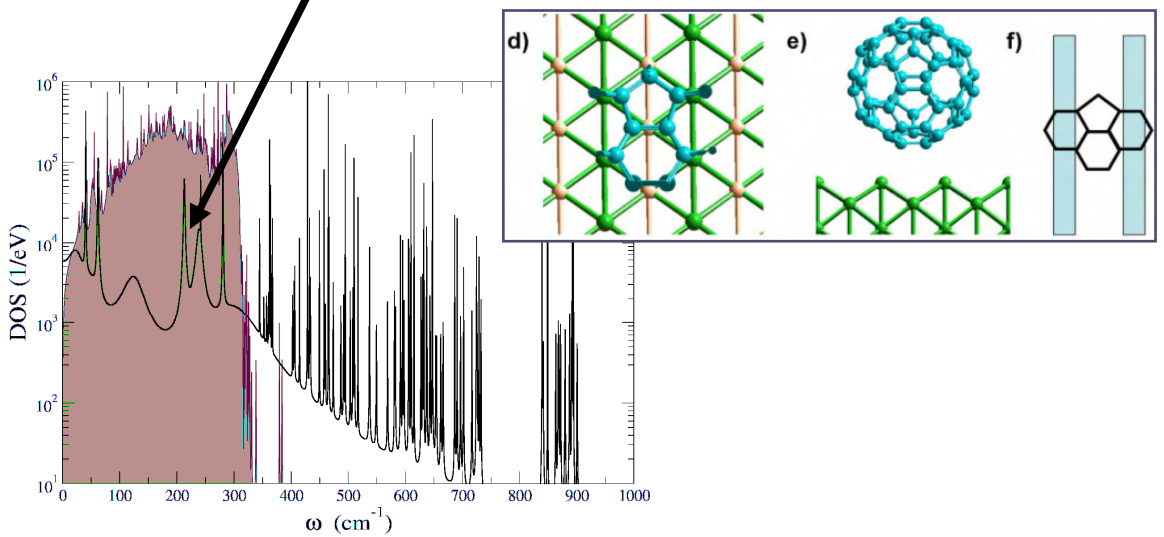


Phonon absorption el-h (A_q)



Phonon dissipation (J_q)





Pecchia, Romano, PRB 75, 035405 (2007)

Non-eq populations

Set up a steady-state solution for the vibronic populations

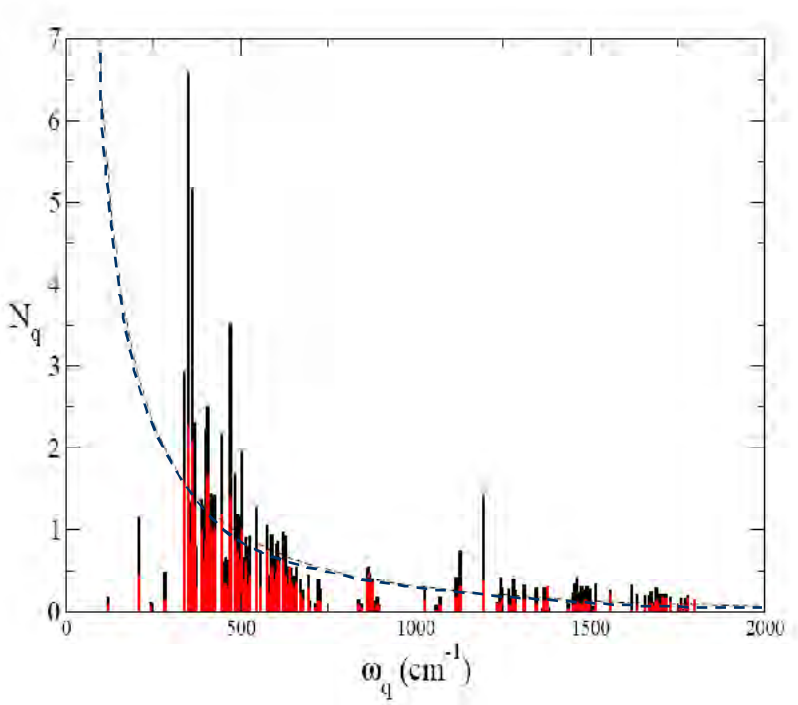
$$\frac{dN_q}{dt} = R_q - J_q[N_q - n_q(T_{eff})] = 0$$

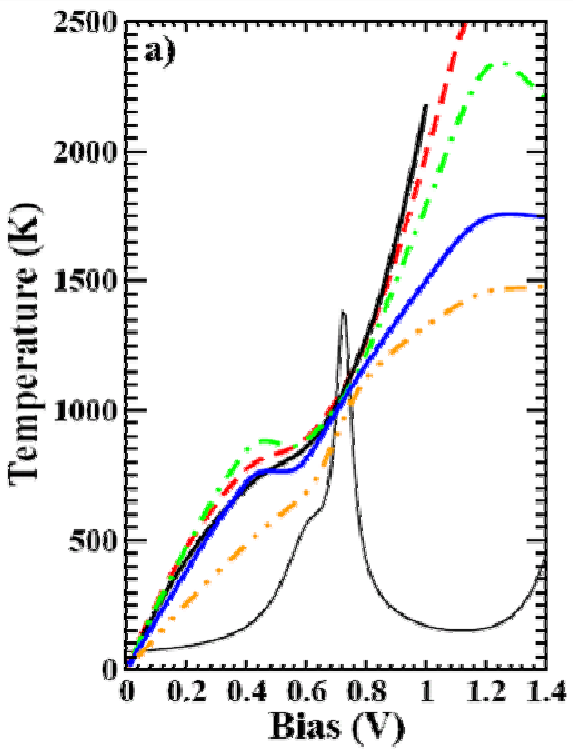
$$R_q = (N_q + 1)E_q - N_q A_q$$

$$R_q = J_q(N_q - n_q(T_0))$$

$$N_q = \frac{n_q(T_0)J_q + E_q}{J_q + A_q - E_q}$$

Effective Temperature





$$U = \sum_q \hbar\omega_q N_q = \sum_q \hbar\omega_q n_q(T_{mol})$$

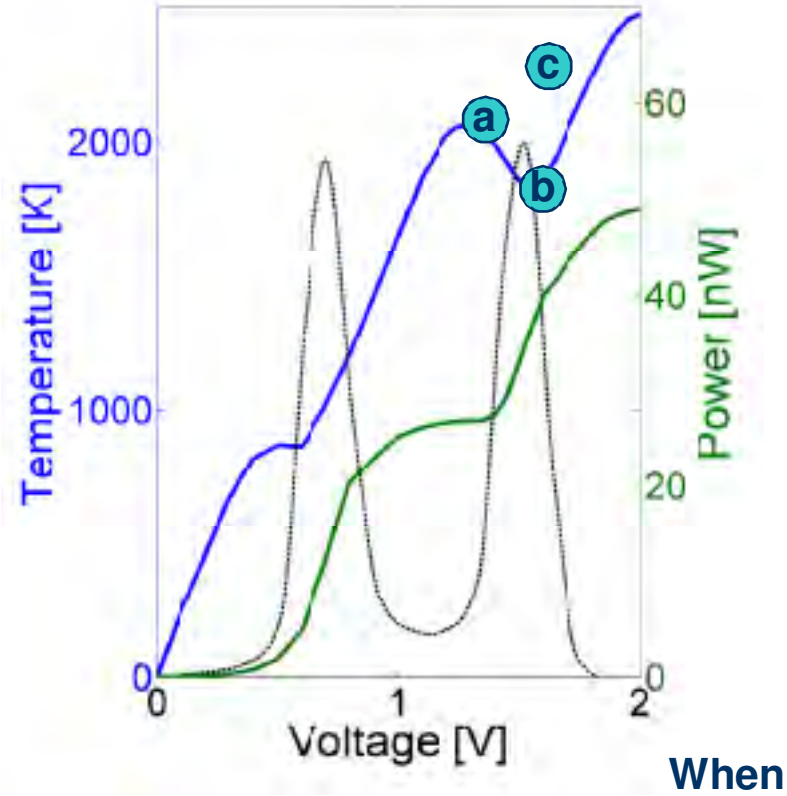
T_{mol} can be seen as a parameter related to the total energy stored in the vibrations

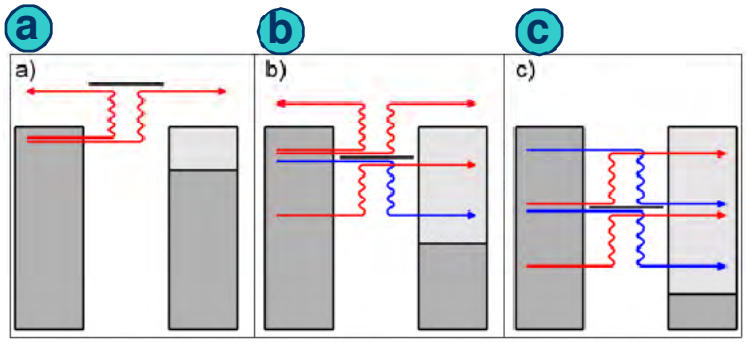
T_{mol} gives the best parameter for an estimator given by a BE distribution

Are there better definitions of T_{mol} thermodinamically meaningful ?

Romano et al., PRB 81, 115438 (2010)

Cooling mechanisms





When the resonance approaches the bias window several regimes take place subsequently:

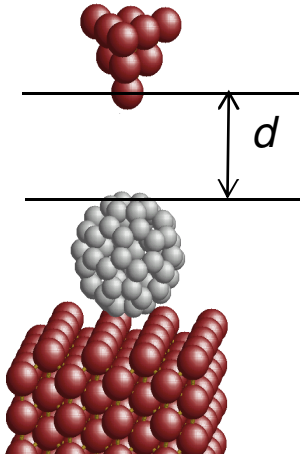
a) Resonant e-h absorption that cools the molecule

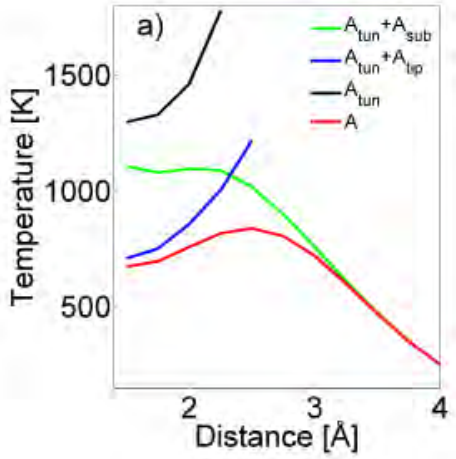
b) Intermediate regime with competing effects

c) Emission take over absorption

Phys Rev B **81, 115438 2010**

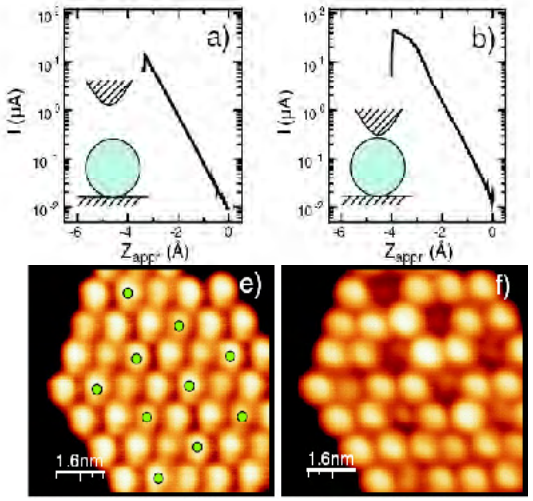
What keeps a molecule stable?

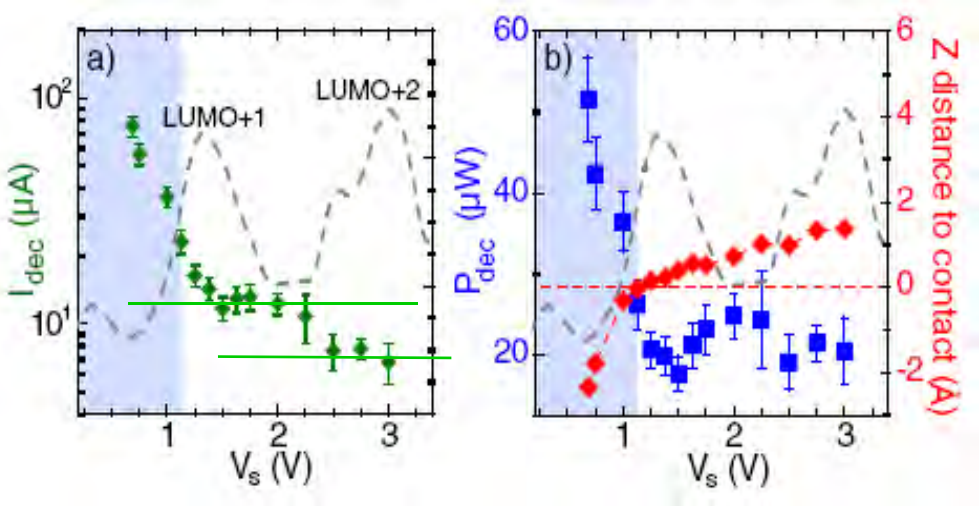




Tip-induced cooling

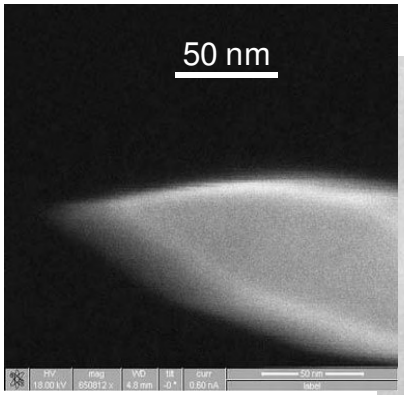
Experiment:



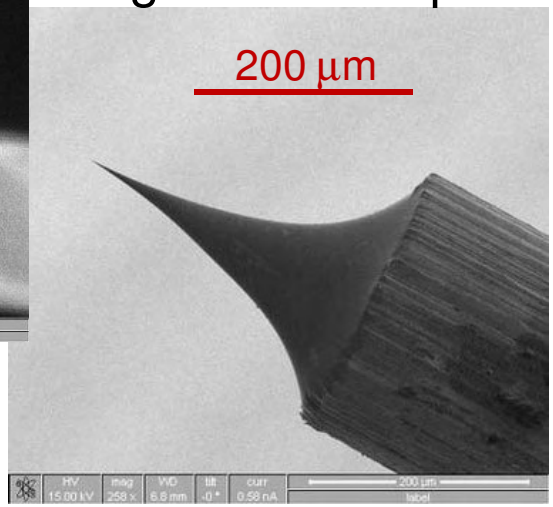


G. Shulze et al., Phys. Rev. Lett. **100,** 136801 (2008)

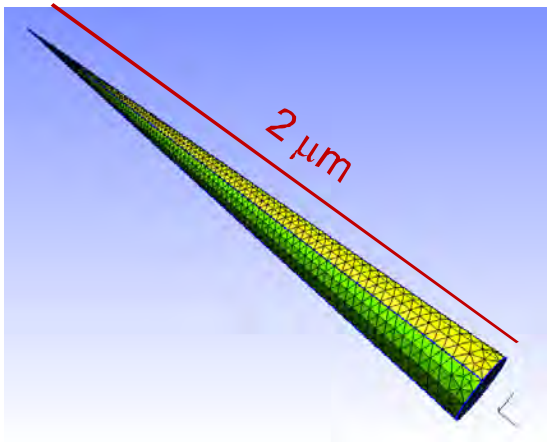
Modeling of STM junction



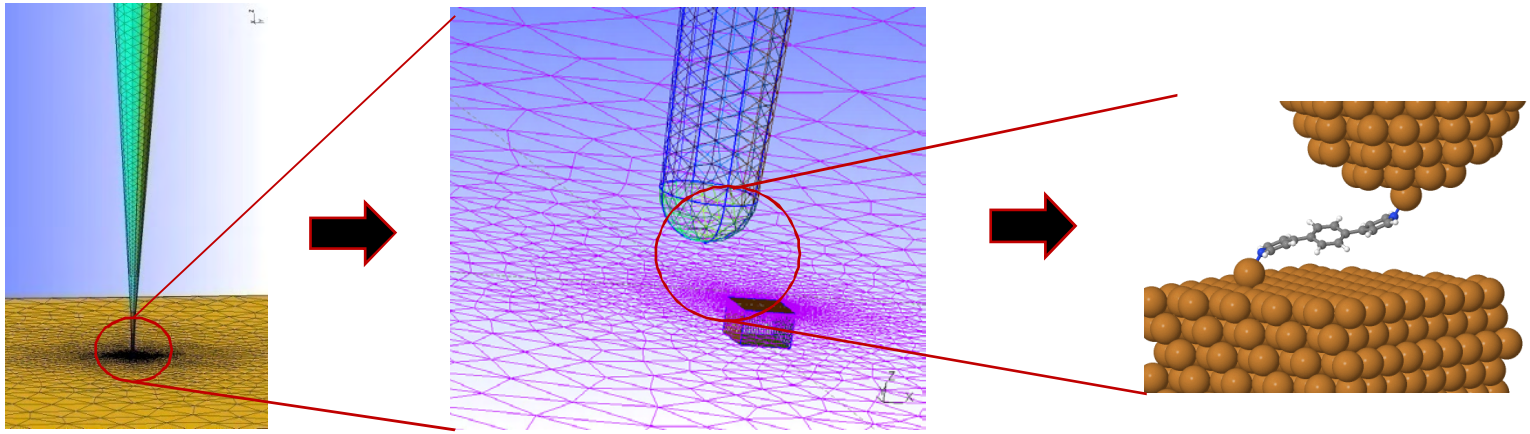
Tungsten STM tip



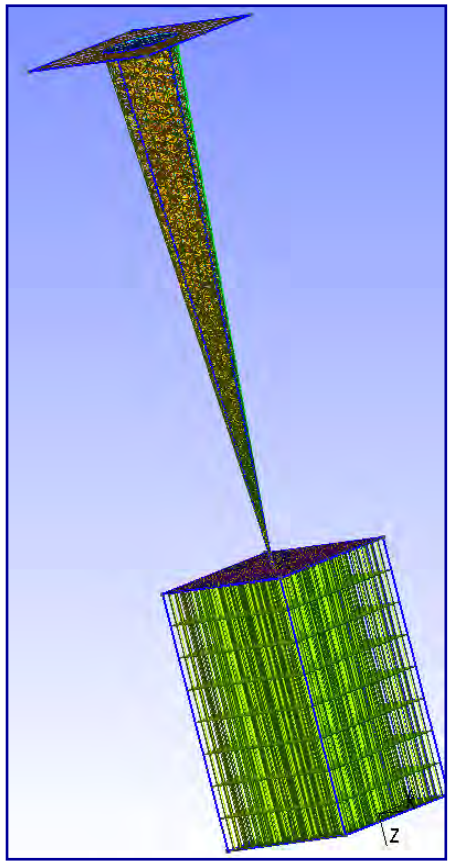
Idealized model of the tip

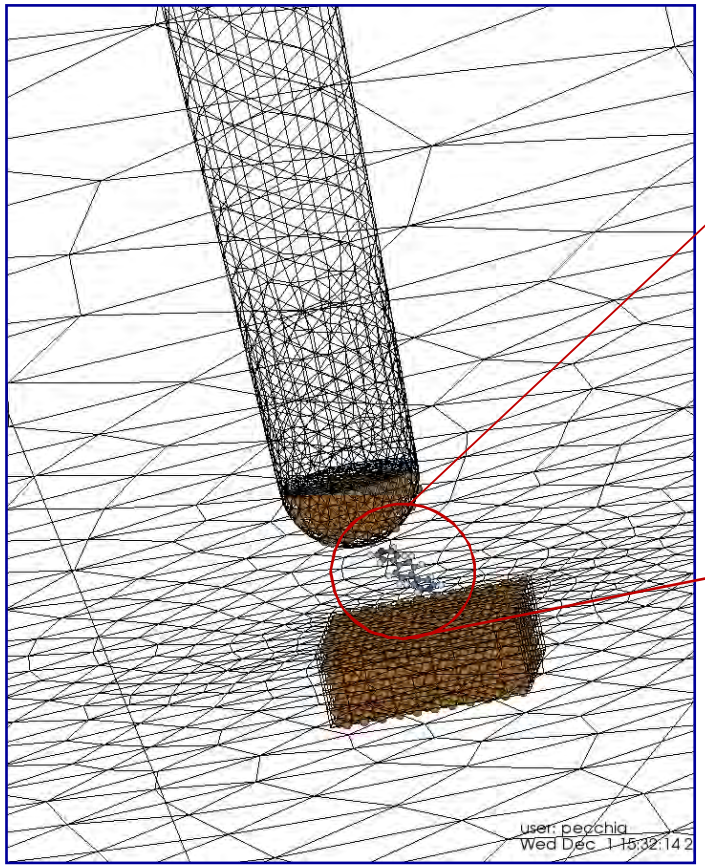


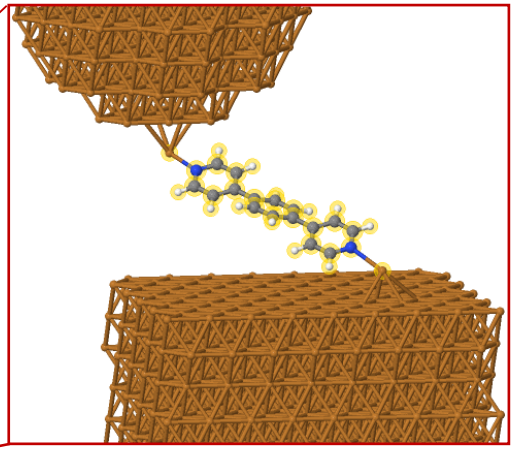
Construction of FEM – Atomistic Model of the STM junction

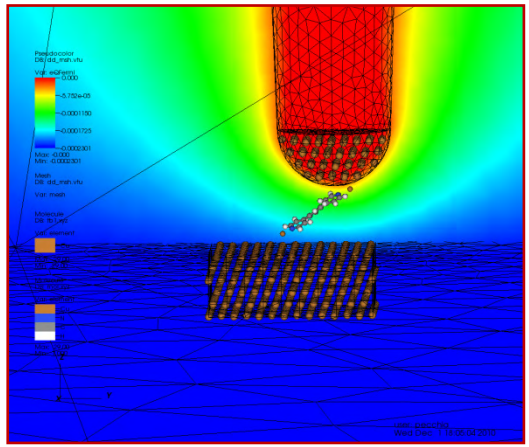


Atomistic-FEM coupling



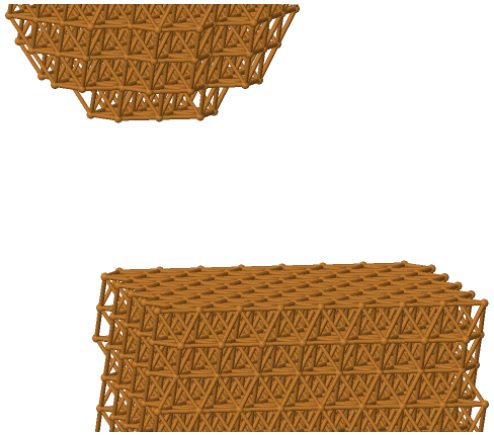




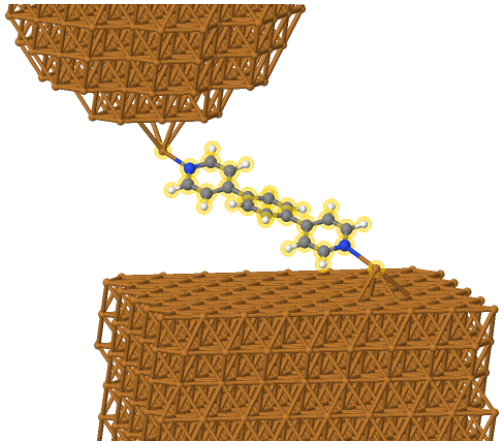


Geometry setup

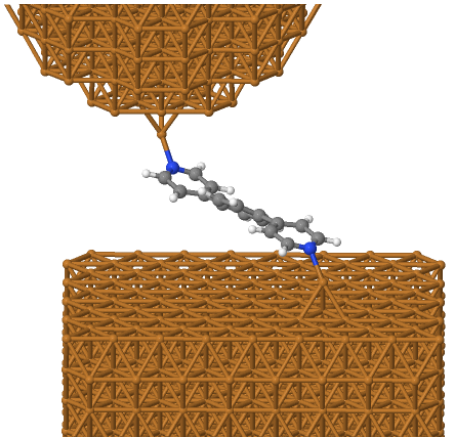
1. Atomistic Generator

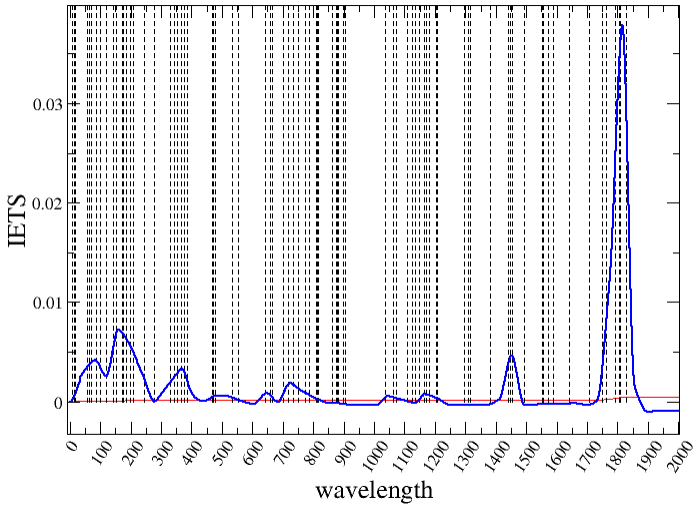


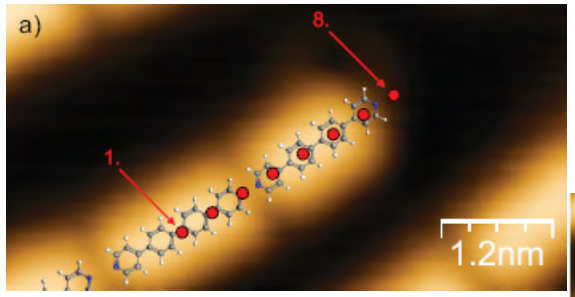
2. Starting guess



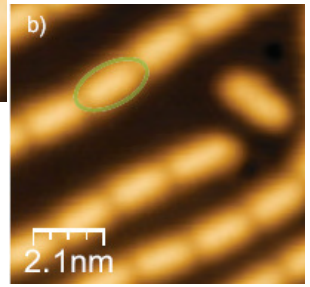
3. Geometry relaxations





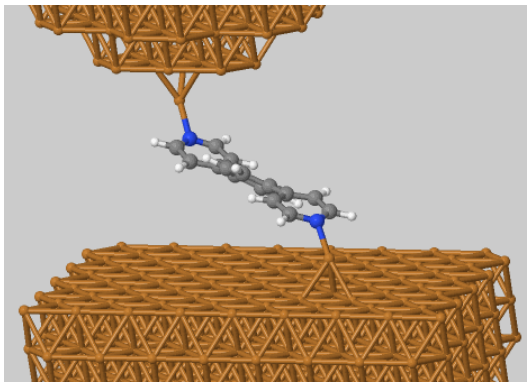


(G. Schulze PhD Thesis
N.Pascual, Frei U. Berlin)

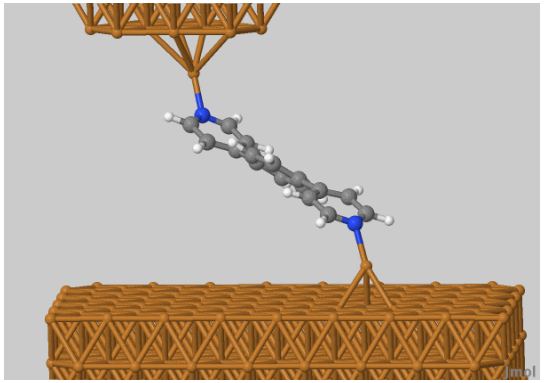


Molecular rising

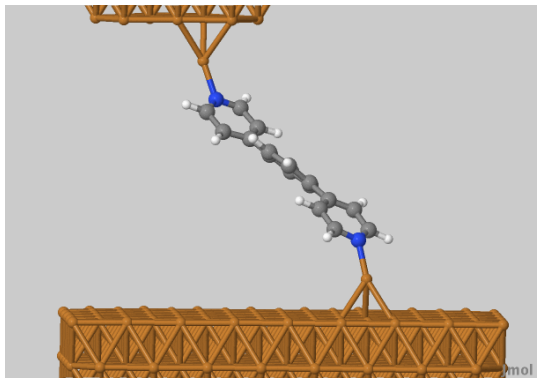
1 nm distance



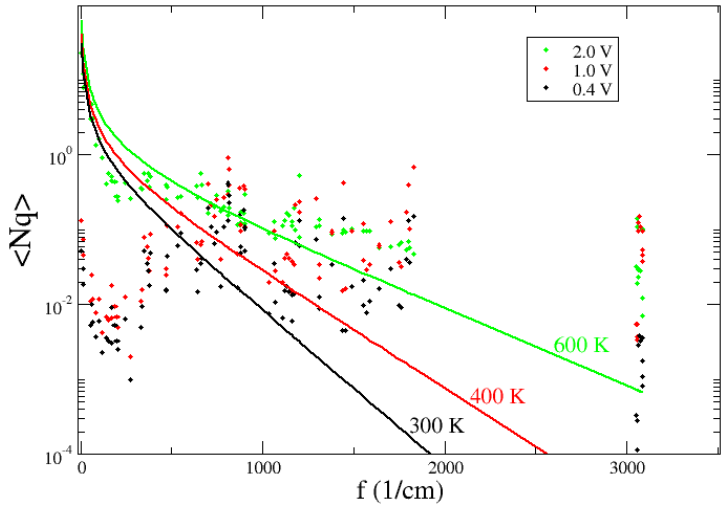
1.2 nm distance



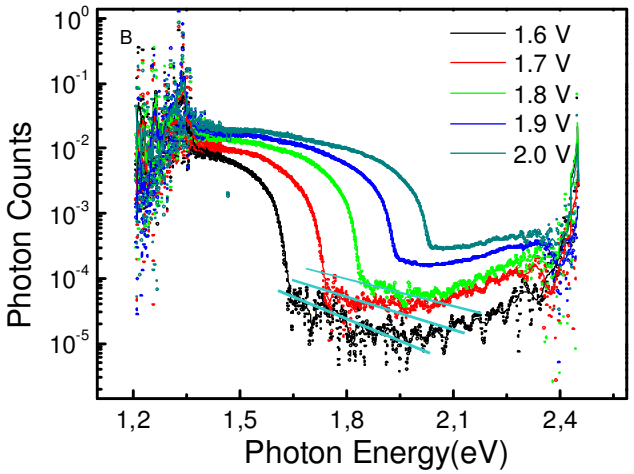
1.4 nm distance

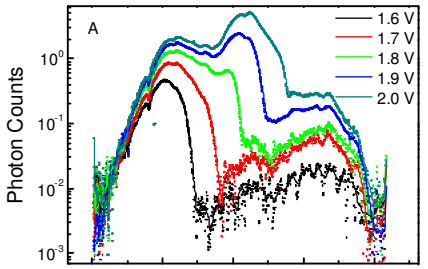


T vs V at 1.0 nm

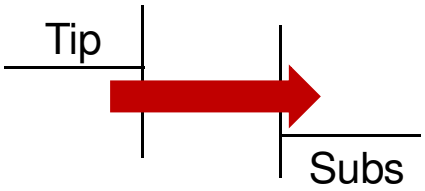


Optical Spectrum vs V



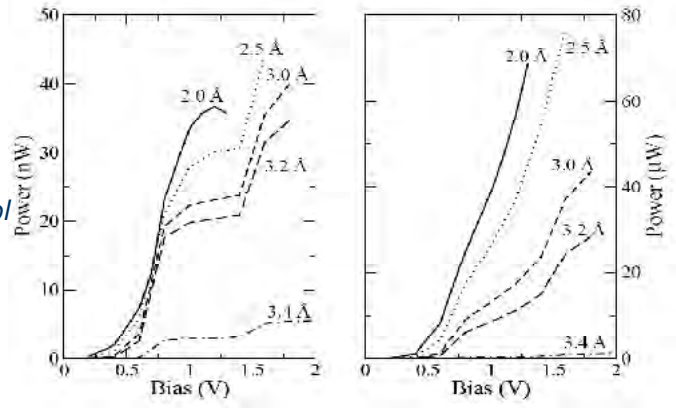


Tip Heating



V = 1.0 Volt

P_{mol}

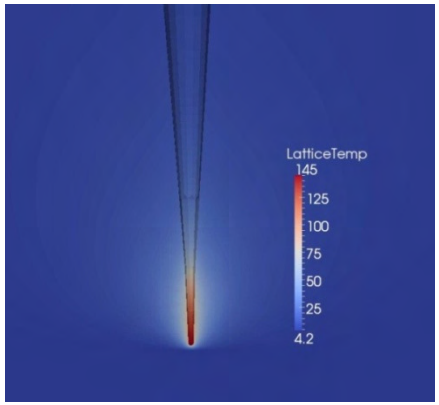


$P=I \cdot V$

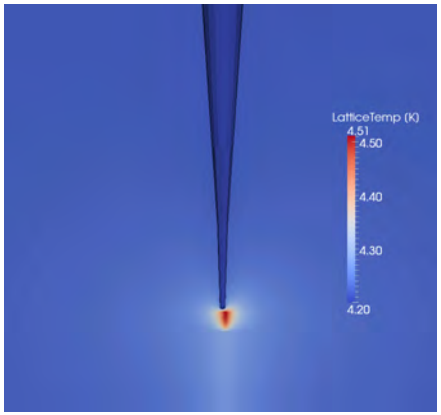
$P_{mol} << I \cdot V$

Only 0.1% of the power is dissipated in the molecule !

Most of the power is dissipated in the contacts



Fourier dissipation



Large heating of the tip

Low heating of the substrate

Reduced thermal conductance of the tip due to size effects

Conclusions

- Multiscale simulations are important for future device modelling
- Finite element have been coupled to TB NEGF transport
- Molecular heating and cooling are key factors in future devices
- Heating of the contacting leads near the molecule can be important

Acknowledgements

Prof. Aldo Di Carlo

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