The gDFTB tool for quantum transport calculations

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Outline

- Introduce NEGF extensions in DFTB
- Overlook on applications
- Electron-phonon interactions and heating in molecules
- Multiscale device simulations in TiberCAD

DFTB = DFT based Tight-Binding method

Kohn-Sham equation:

$$\sum_{\nu}\left[H^{0}_{\mu\nu} + H^{Scc}_{\mu\nu}[\delta n] - E_k S_{\mu\nu}\right]c^{k}_{\nu} = 0$$

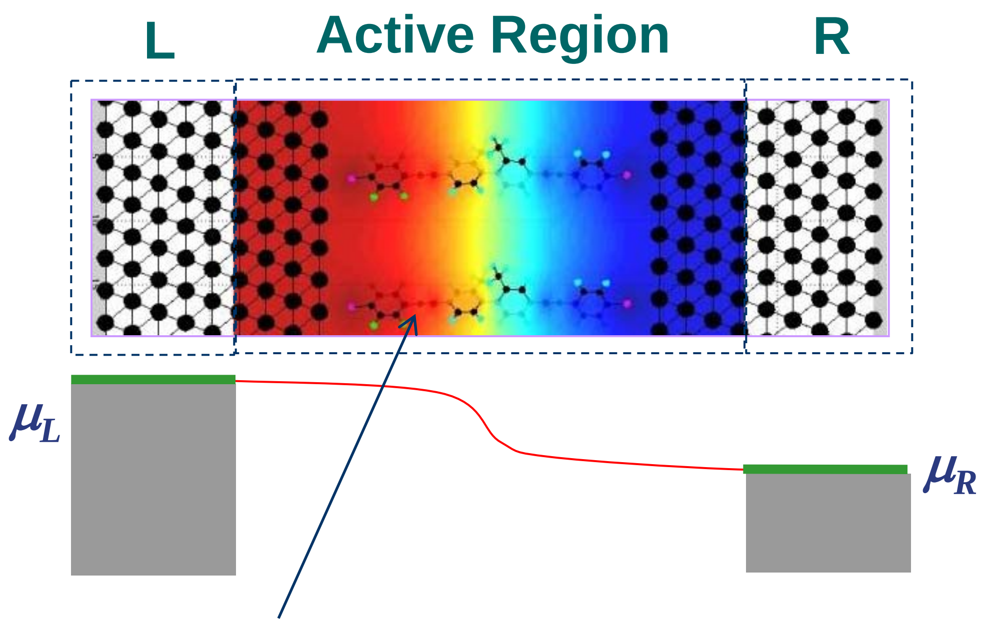
$$H_{\mu\nu} = \begin{cases} \varepsilon_{\mu} & \text{onsite atomic energy levels} \\ \langle \mu | V[n^0_{\mu} + n^0_{\nu}] | \nu \rangle & \text{two-centre density superposition} \end{cases}$$

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

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

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

Self-consistent loop (gDFTB)

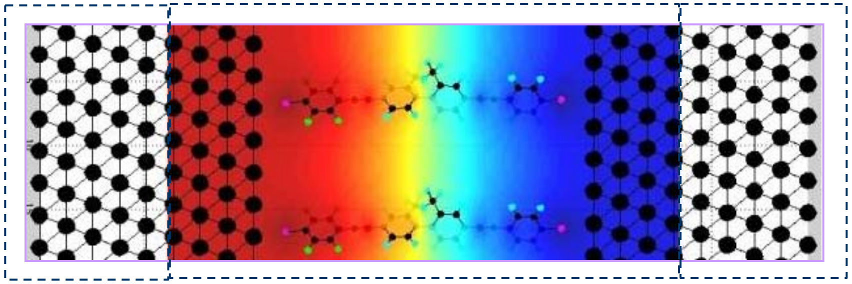


Poisson

Density matrix

$$\rho_{\mu\nu} = \frac{1}{2\pi i} \int dE \;\; G^{<}_{\mu\nu}(E)$$

Green's operators



$$G^r(E) = \left(ES - H - \Sigma_L - \Sigma_R\right)^{-1}$$

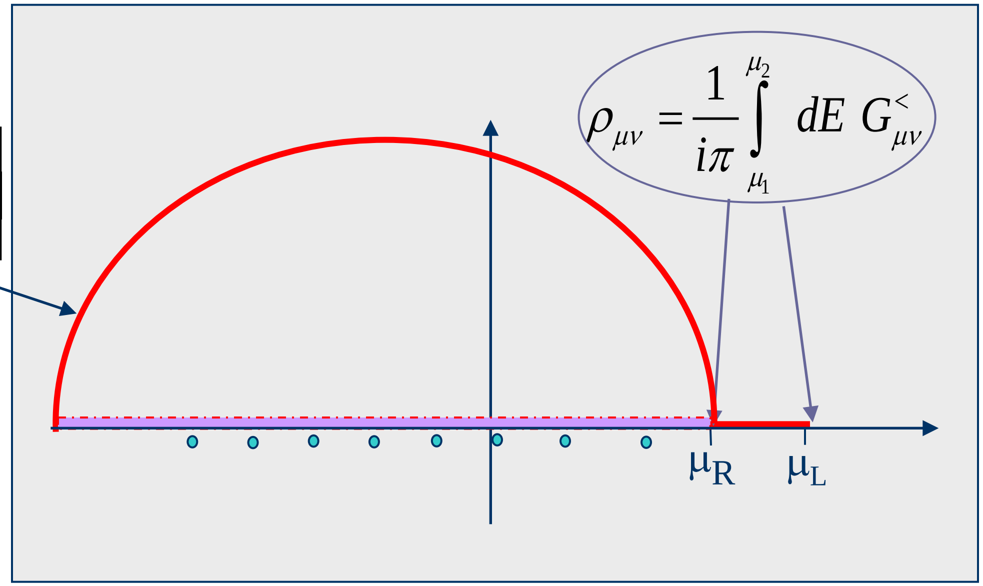
$$G^<(E) = if_L(E)\underbrace{G^r(E)\Gamma_L(E)G^a(E)}_{\text{L -incoming DOS}} + if_R(E)\underbrace{G^r(E)\Gamma_R(E)G^a(E)}_{\text{R -incoming DOS}}$$

Equilibrium limit: $G^<(E) = if(E)[G^r(E) - G^a(E)]$

Contour integration

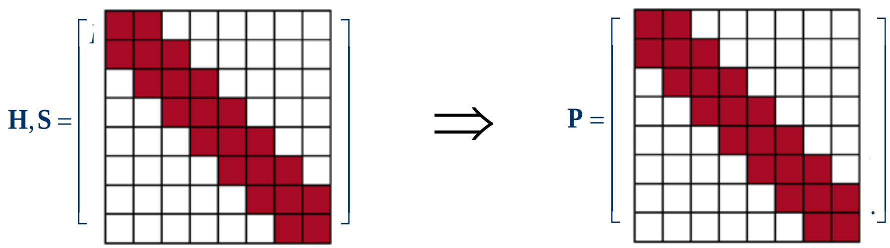
$\int G^{<}$

$$\rho_{\mu\nu} = -\frac{1}{\pi}\,\mathrm{Im}\left[\int_C dE\; G^{r}_{\mu\nu}\right]$$

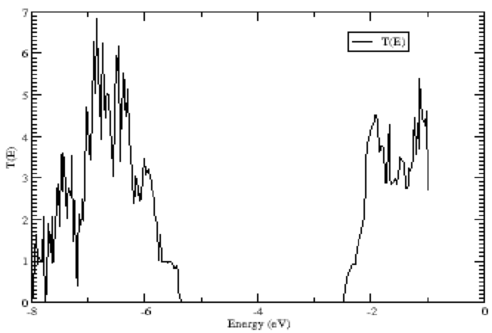


- The largest part of the integration is performed away from the real axis
- The integral is performed numerically via gaussian quadrature
- Parallelized with MPI

Iterative scheme



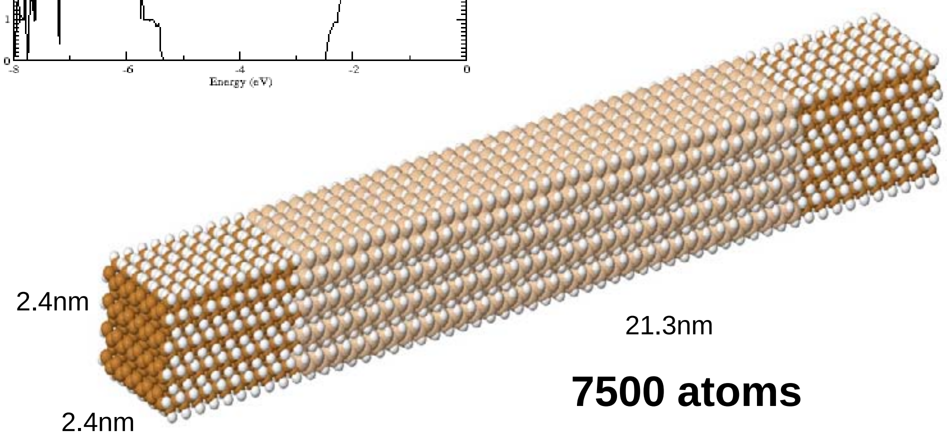
$$q_\mu = \sum_\nu \mathbf{P}_{\mu\nu} \mathbf{S}_{\nu\mu}$$



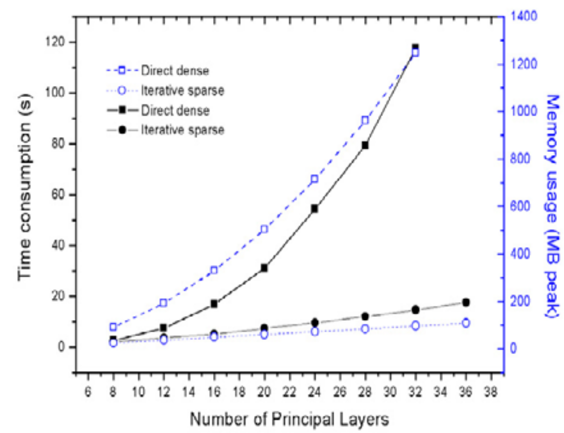
PROFILING

Charge density and potential:	20 h
Density of States (350 points):	6 h
Peak memory:	876 MB

Calculations on single PC Linux core
Intel(R) Core(TM)2 CPU 6600 @ 2.40GHz



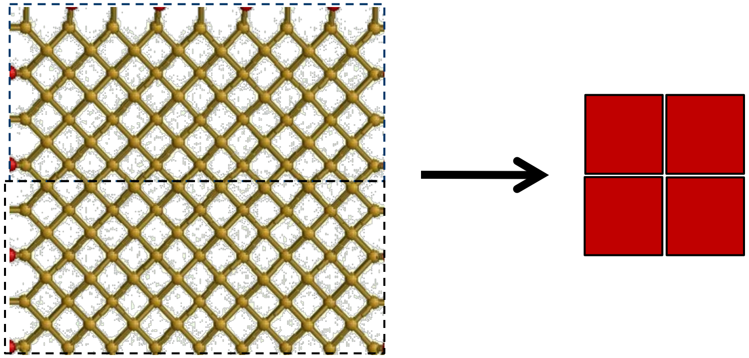
7500 atoms



O(N m³)

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

Sub-partitioning of each layer

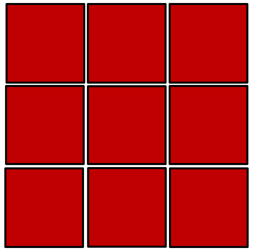


$$G_{11}(E) = \left(ES_{11} - H_{11} - H_{12} g_{22} H_{21}\right)^{-1}$$

$$g_{22}(E) = \left(ES - H_{22}\right)^{-1}$$

$G_{11}(E)$	$G_{12}(E) = -G_{11} H_{12} g_{22}$
$G_{21}(E) = -g_{22} H_{21} G_{11}$	$G_{22}(E) = g_{22} + g_{22} H_{21} G_{11} H_{12} g_{22}$

2 $O(m^3)/8$ + MM mult $\approx O(m^3)/3$



$O(m^3)/9$ + MM mult $\approx O(m^3)/6$

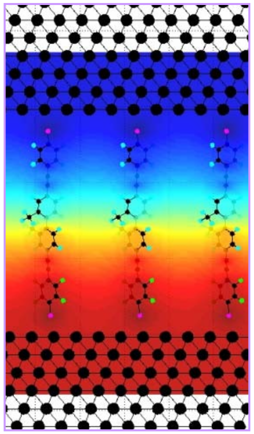
LibNEGF

- General Sparse Matrices (CSR)
- Automatic partitioning (METIS)
- Parallel computations (MPI/OpenMP)
- GPU acceleration (?)

… work in progress…

Poisson solver

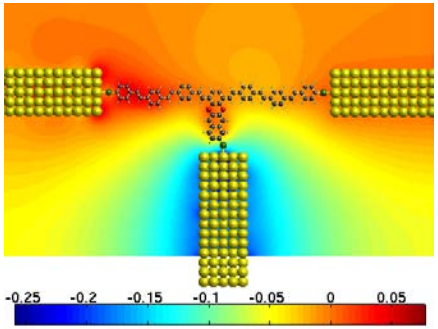
2-terminals



Discretize in real space

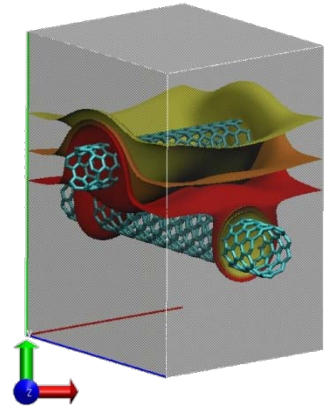
$$\Delta\rho(r) = \sum_{\mu} \Delta q_{\mu} n_{\mu}(r)$$

gated (3-term.)

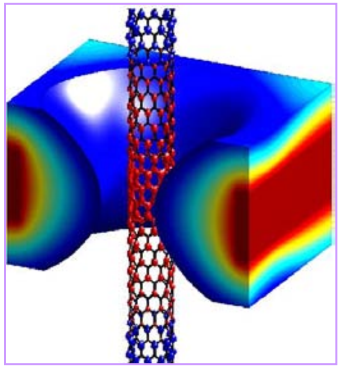


The Poisson equation is solved with a multi-grid algorithm (MUDPACK).

4-terminals



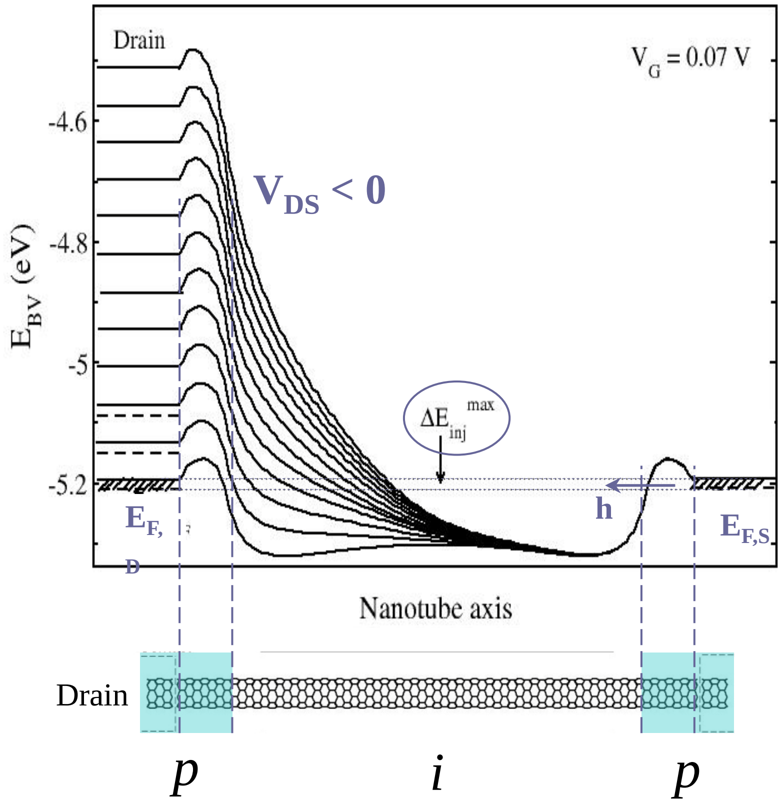
coaxially-gated

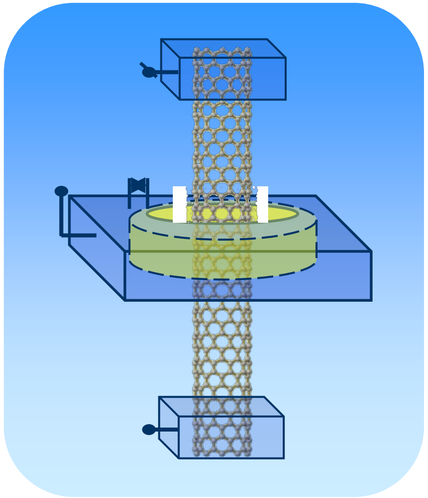


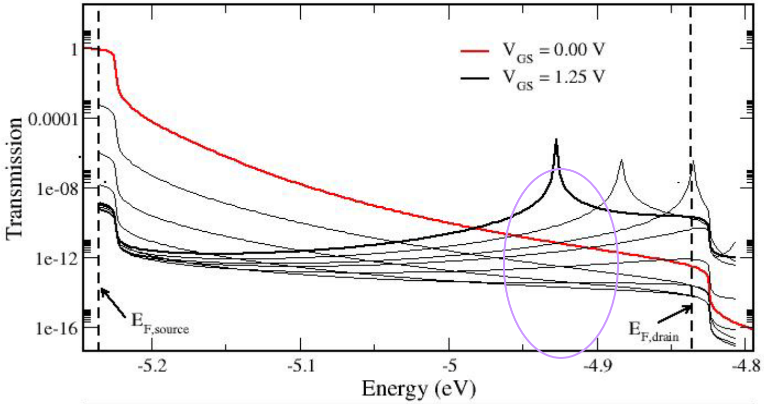
This allows to solve complex boundary conditions (bias, gate)

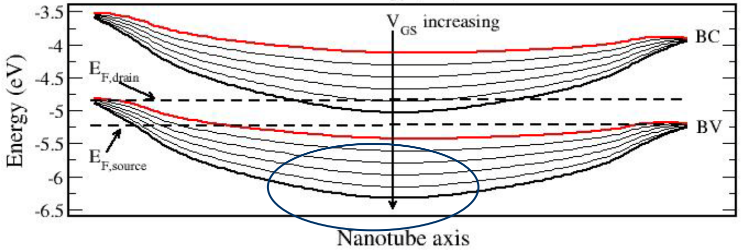
$$\nabla^2 V = -4\pi\Delta\rho$$

CNT devices







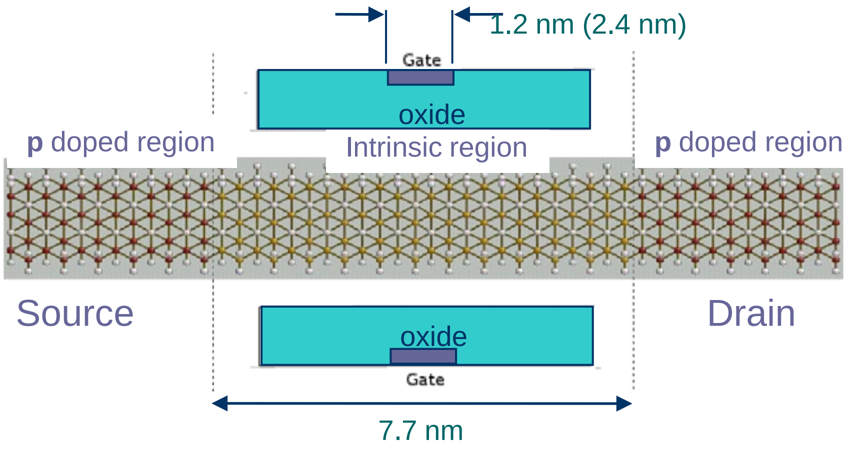


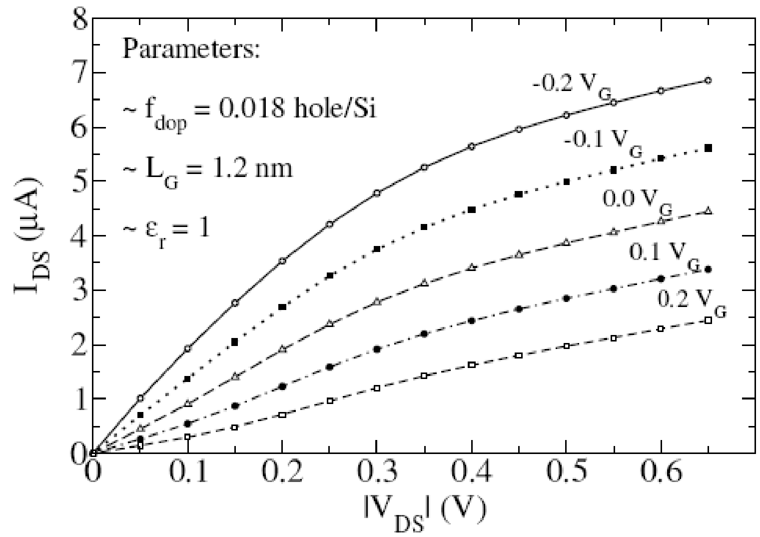
Self- consistent barriers

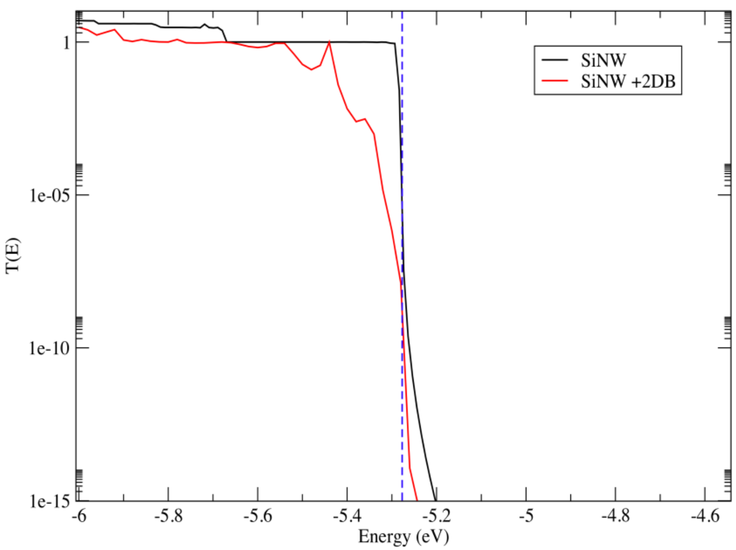
Band to band tunneling

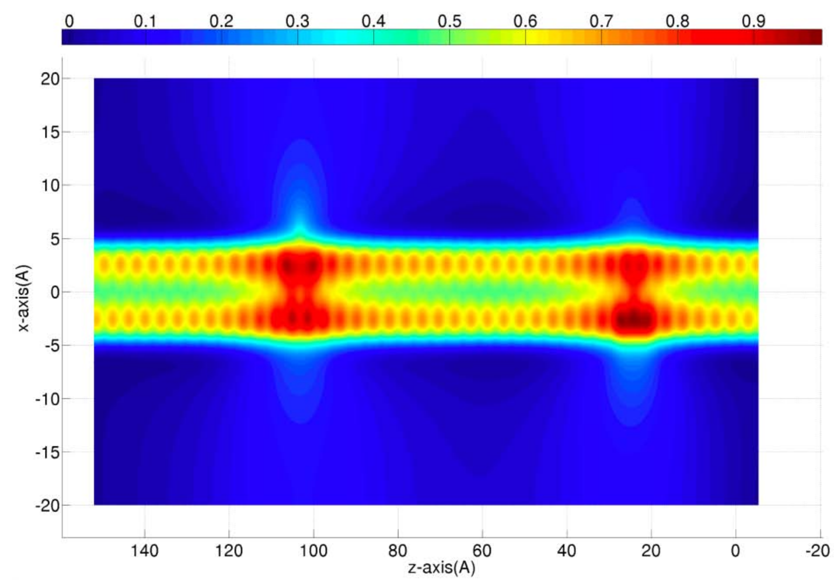
Negative Quantum Capacity

L. Latessa et al., PRB 72, 035455 (2005)

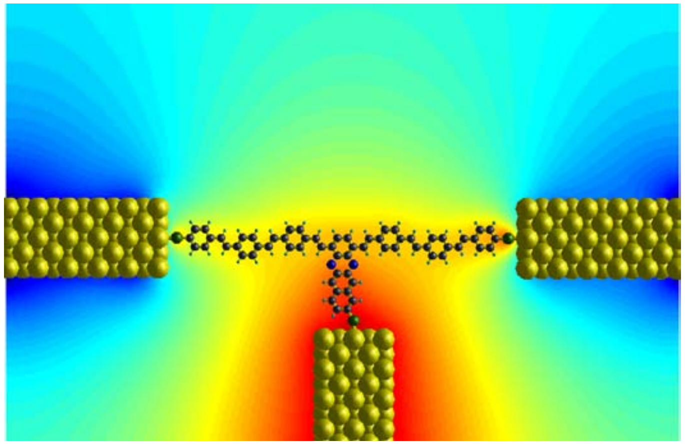

1.2 nm (2.4 nm)
Gate
oxide
p doped region
Intrinsic region
p doped region
Source
Drain
oxide
Gate
7.7 nm

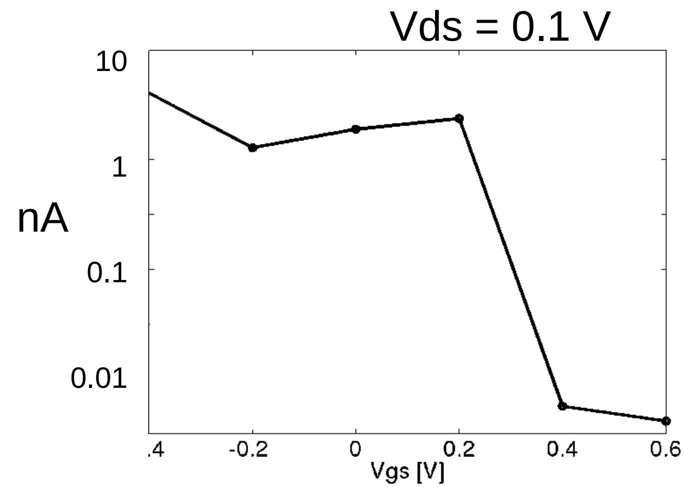

Parameters:
~ f_{dop} = 0.018 hole/Si
~ L_G = 1.2 nm
~ ε_r = 1
-0.2 V_G
-0.1 V_G
0.0 V_G
0.1 V_G
0.2 V_G
I_{DS} (µA)
$|V_{DS}|$ (V)

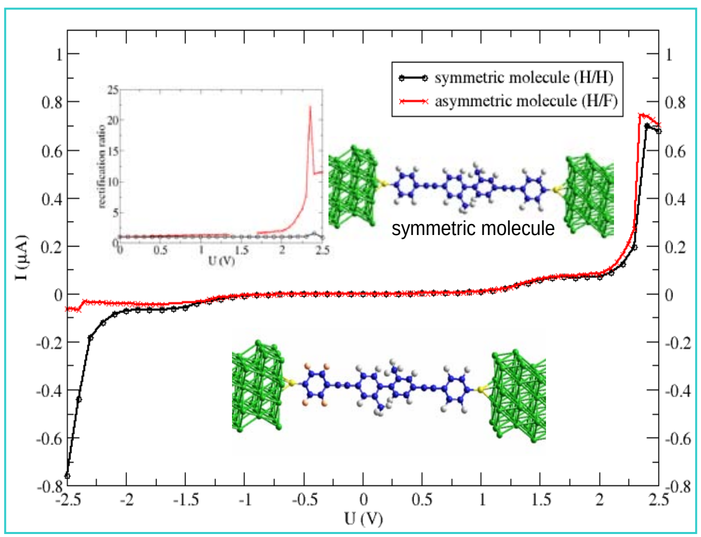

SiNW
SiNW +2DB
T(E)
Energy (eV)

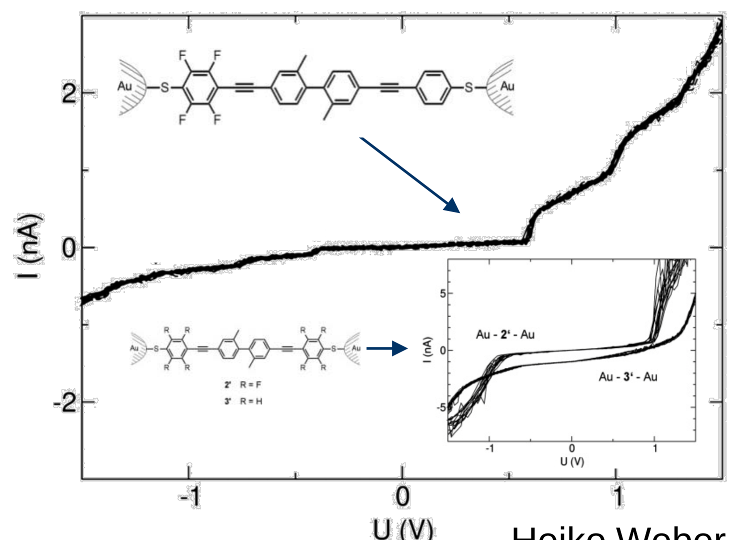

x-axis(A)
z-axis(A)

OPV – based transistor







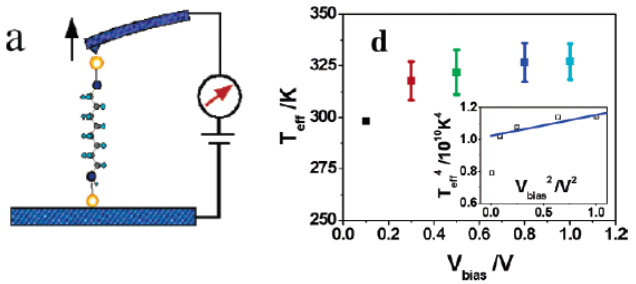


F. Pump, G. Cuniberti

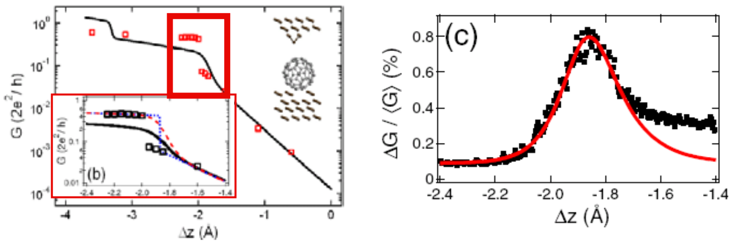
Heiko Weber

Power Dissipation

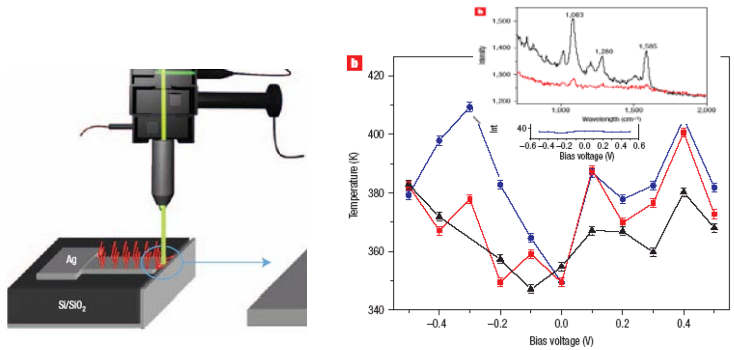
- Thermal effects at the molecular scale represent an increasingly ‘hot’ topic
- Theoretical and experimental challenges to measure nanoscale temperatures



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

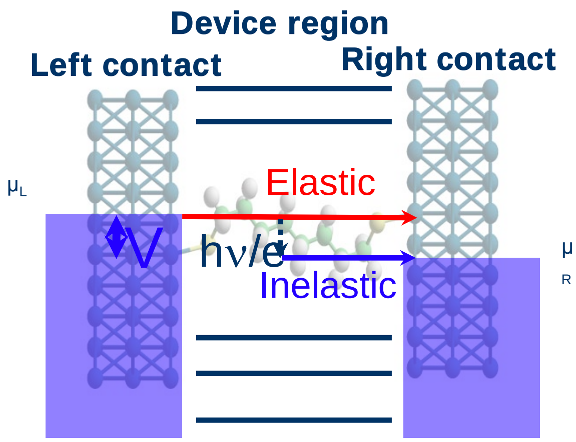


N. Néel et al. PRL 98, 065502 (2007)



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

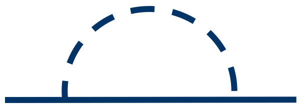
electron-phonon scattering



$$G^r(E) = [ES - H^{DFT} - \Sigma_L^r - \Sigma_R^r - \Sigma_{scatt}^r]^{-1}$$

$$G^<(E) = G^r[\Sigma_L^< + \Sigma_R^< + \Sigma_{scatt}^<]G^a$$

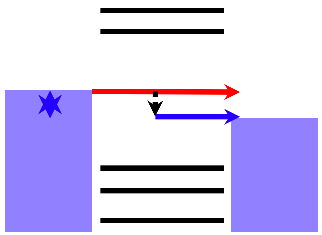
$$\Sigma_{L,R}^< = if_{L,R}\Gamma_{L,R}$$

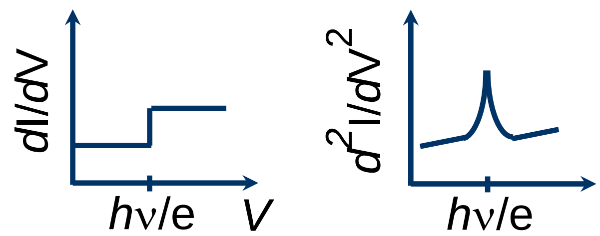


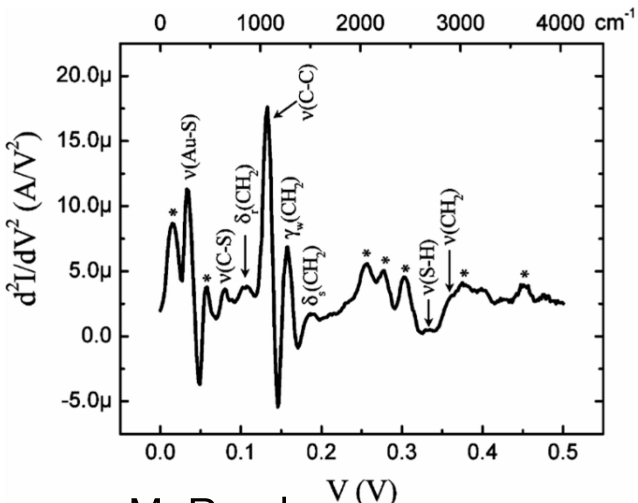
$$\Sigma_{ph}^<(E) = \frac{i}{2\pi}\sum_q \int dE' \alpha^q G^<(E-E')\alpha^q D_q^<(E')$$

$$\Sigma_q^<(E) = N_q\gamma_q G^<(E-\omega_q)\gamma_q + (N_q+1)\gamma_q G^<(E+\omega_q)\gamma_q$$

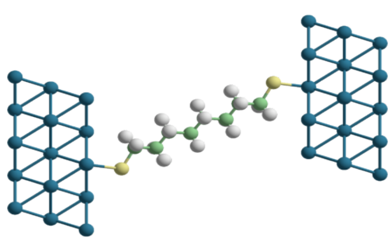
IETS simulations

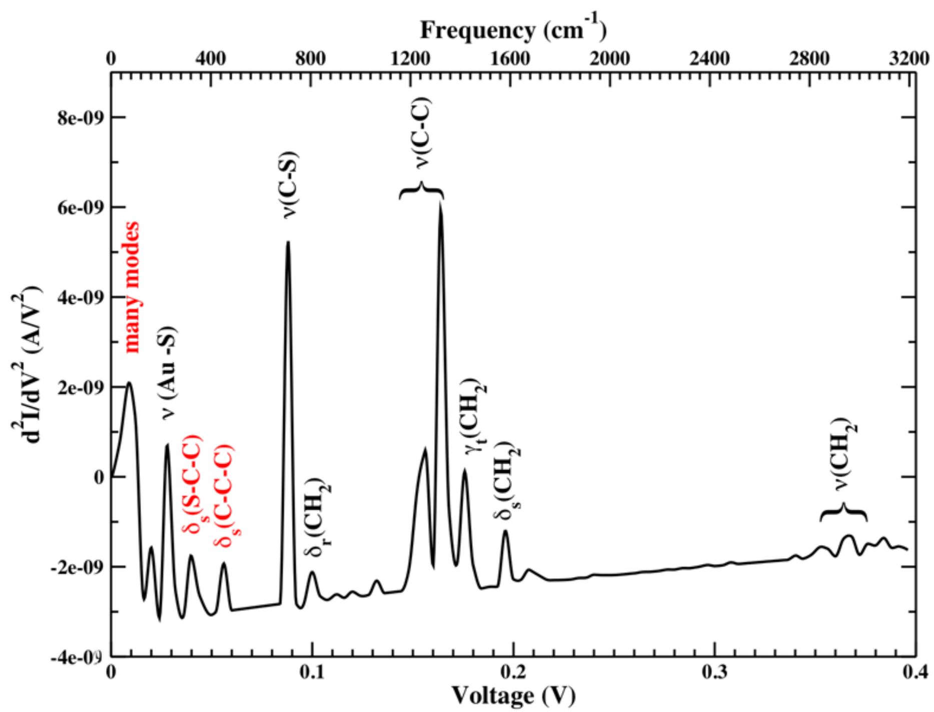






M. Reed





Solomon *et al.*, J. Chem Phys 124, 094704 (2006)

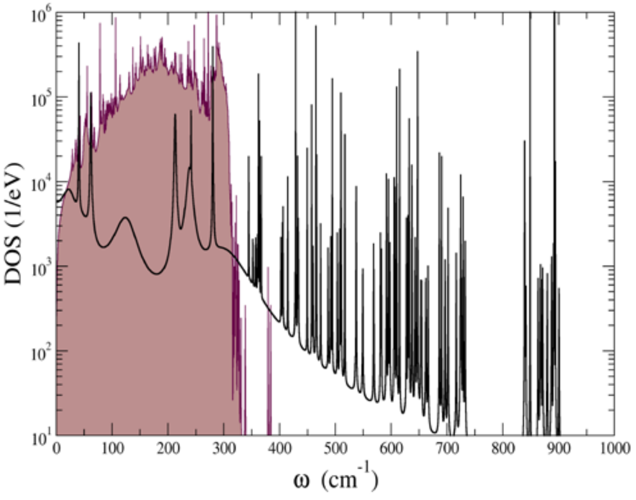
Phonon population

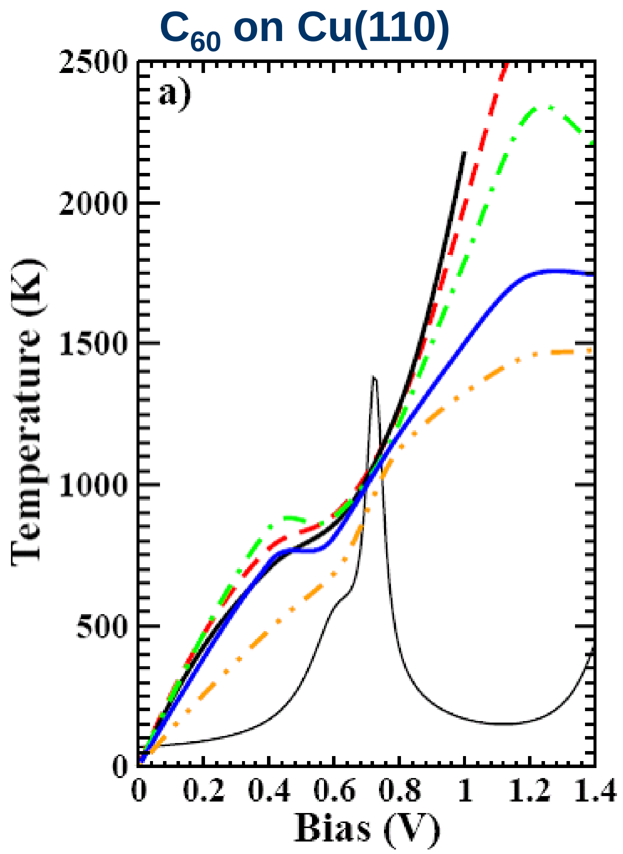
Set up a steady-state solution for the vibronic populations

Rate equation:

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

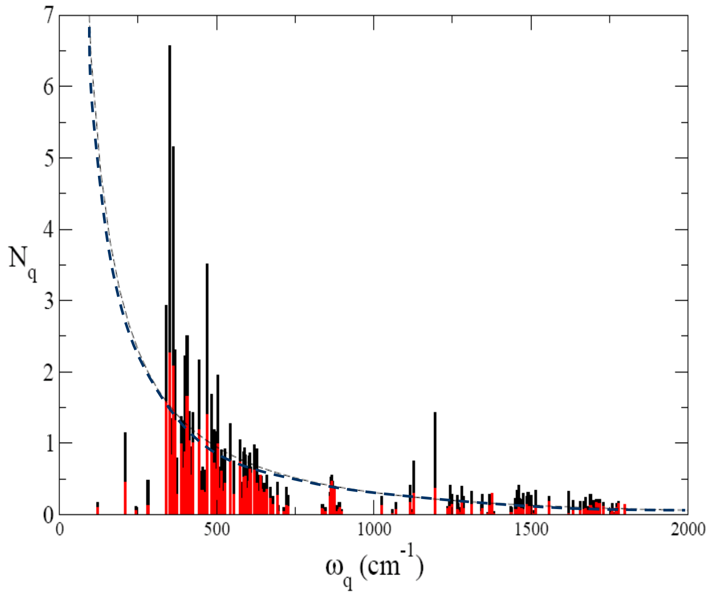
$$R_q = (N_q + 1)E_q - N_q A_q$$

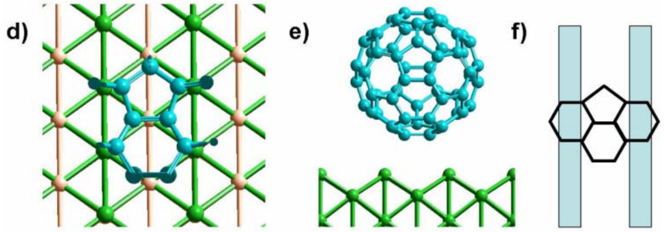




- **Definition of molecular temperature:**

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

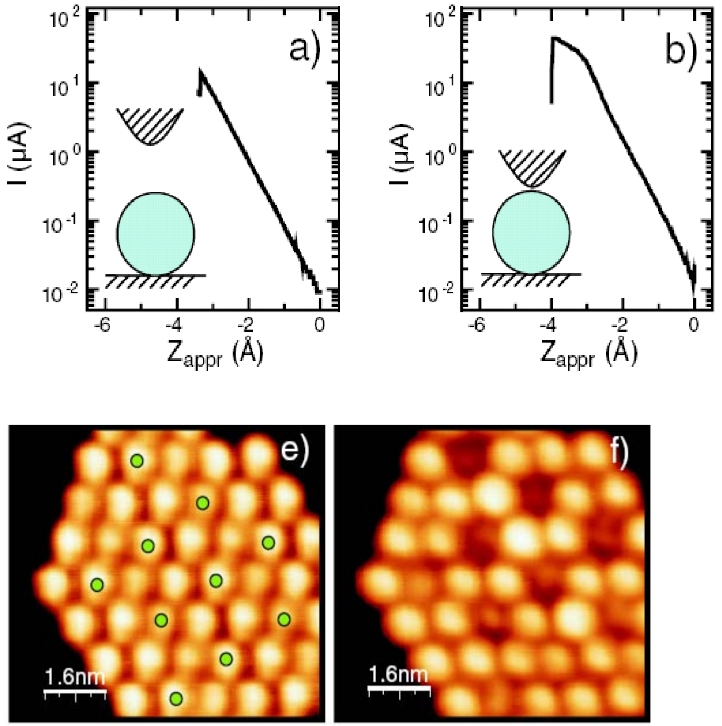


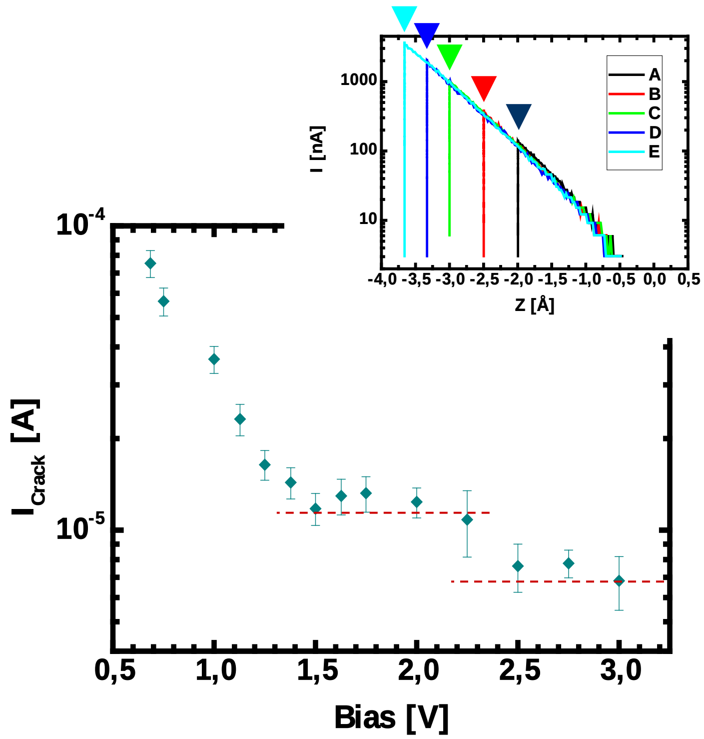


Bath coupling:

$$\sum_q \hbar\omega_q W_q \left[N_q - n_q (T) \right] = 0$$

Tip approaches at fixed V until C_{60} cracks.
Molecules can be selectively burned



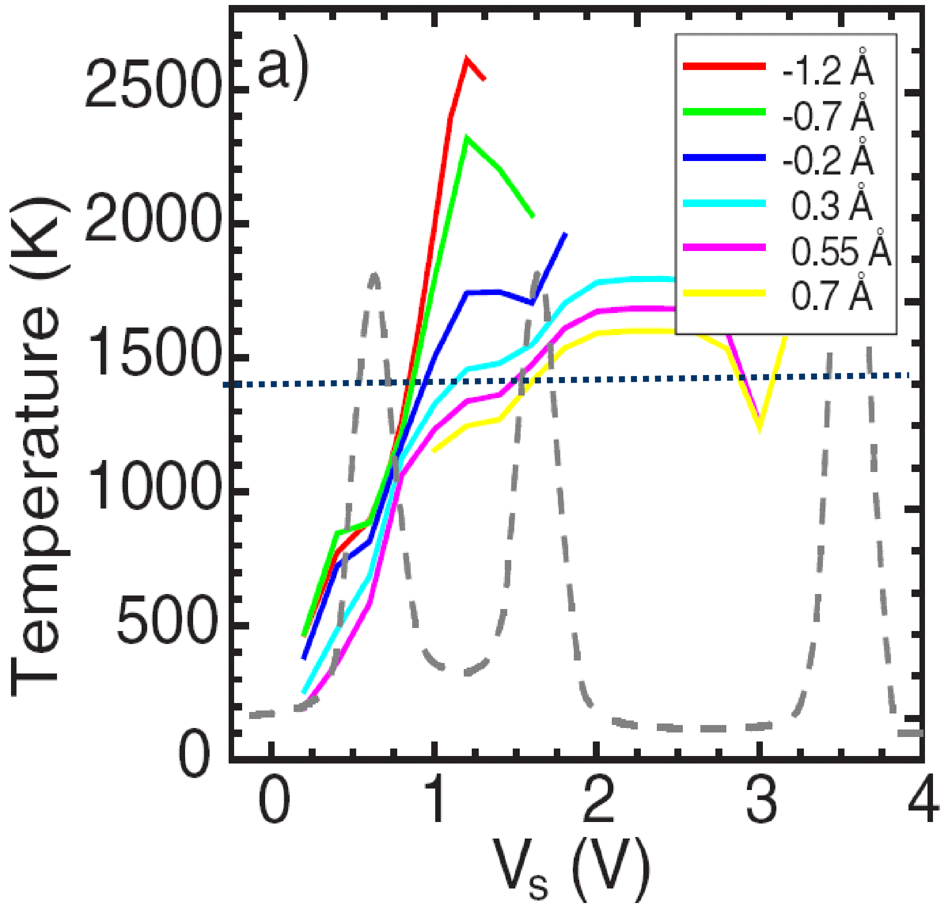


$P = I_{crack} \cdot V_{crack}$ is not a constant but shows features and plateaux

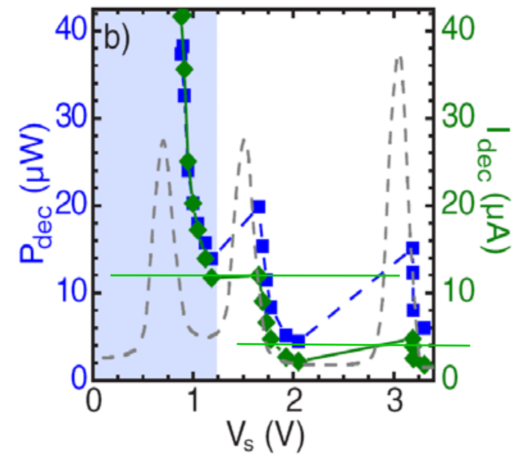
G. Shulze et al., Phys. Rev. Lett. **100,** 136801 (2008)

Model vs Experiments

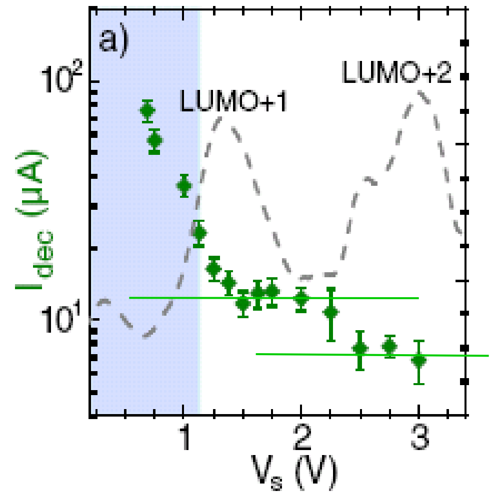
Fix critical T for degradation



Theory

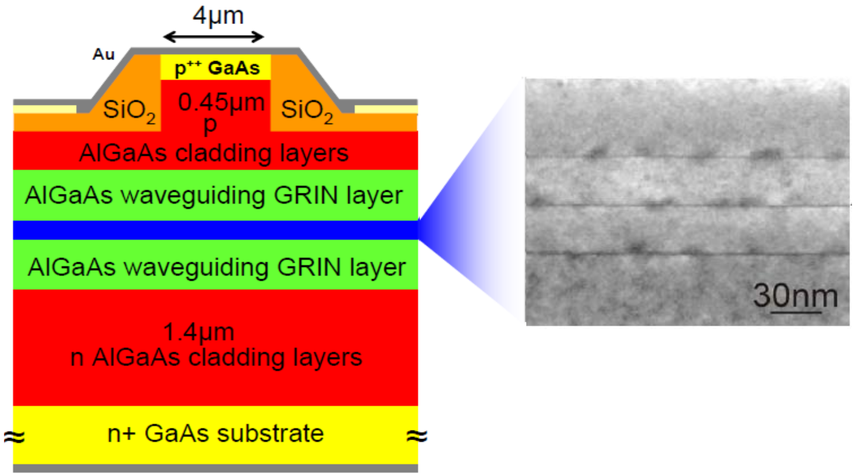


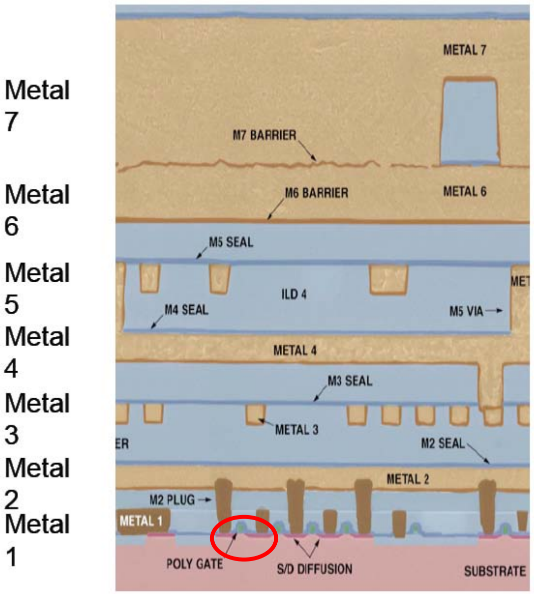
Experiment



Multiscale/multiphysics outlook

Micro/macro scale

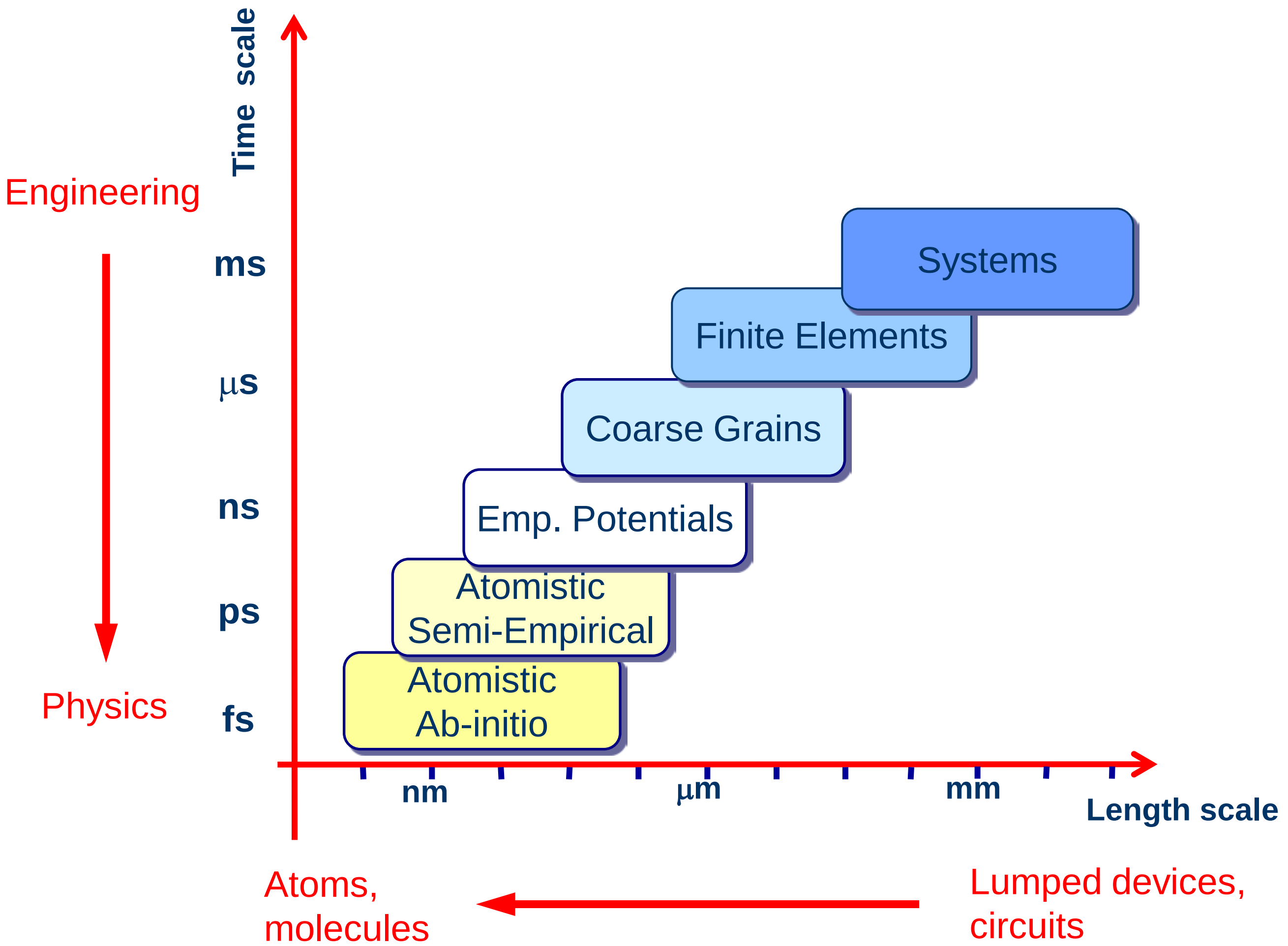




- ***Device should be accessible from a macro scale***
- ***Number of atoms cannot grow to much in simulations***
- ***Micro/macro scale details are as important as nanoscale features***

Introduction: Multiscale/multiphysics

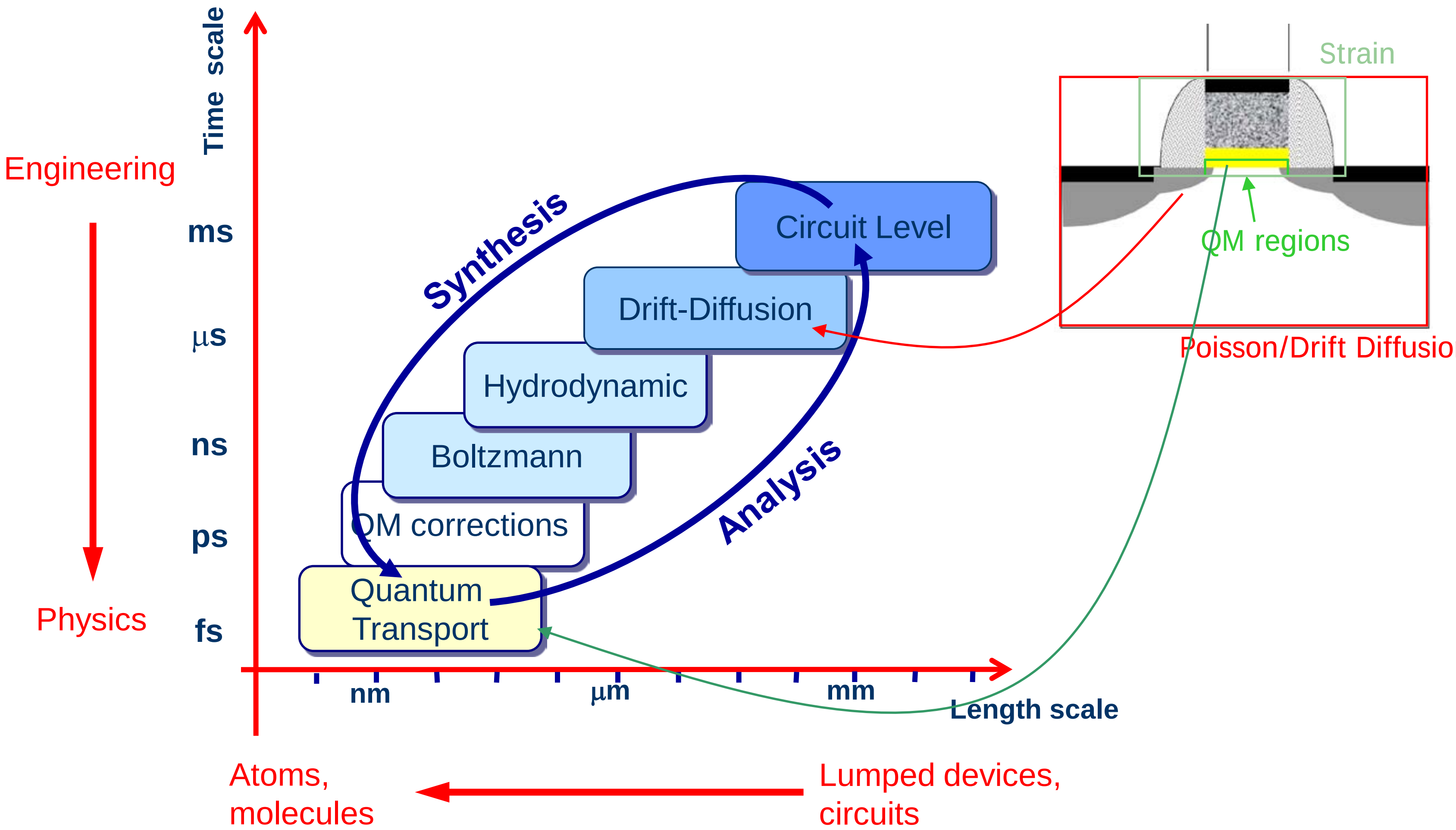
Length and time scale hierarchy



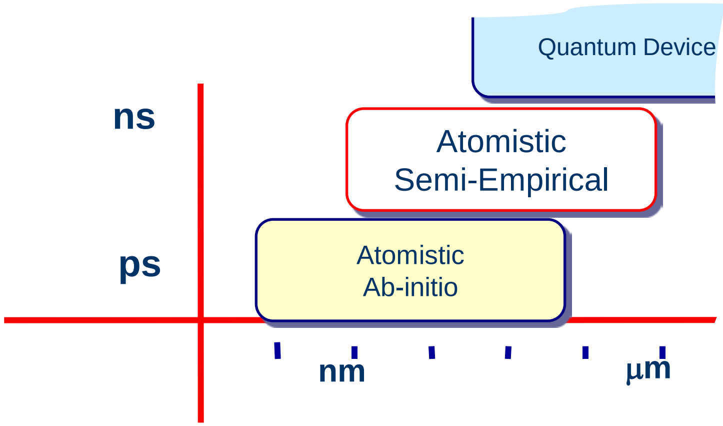
Hierarchy of transport models

TIBER CAD

http://www.tibercad.org

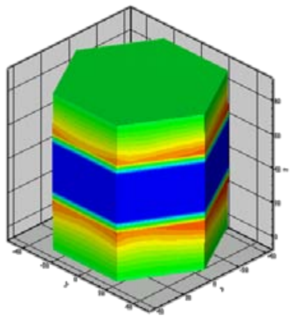


DFTB as intermediate method



FEM/atomistic approaches

FEM



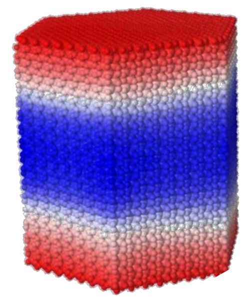
Potentials (electrostatic, piezo, …)

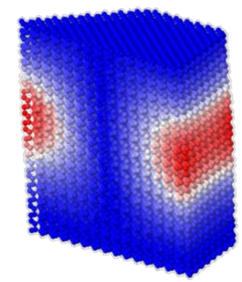
Strain and deformations

Projection on atoms

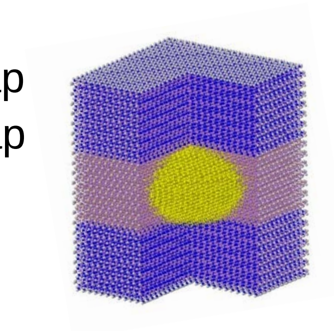
Projection on FEM

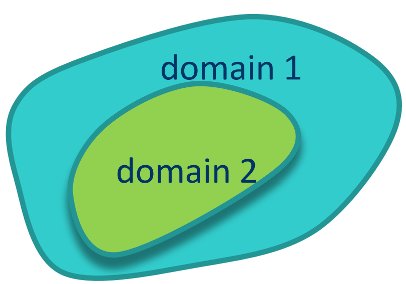
ATOMISTIC

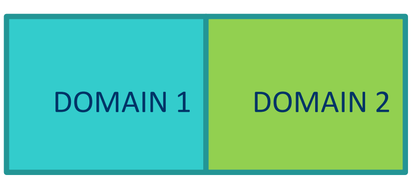




Electrostatic map
Deformation map
(Wavefunctions)
Charge density
Current

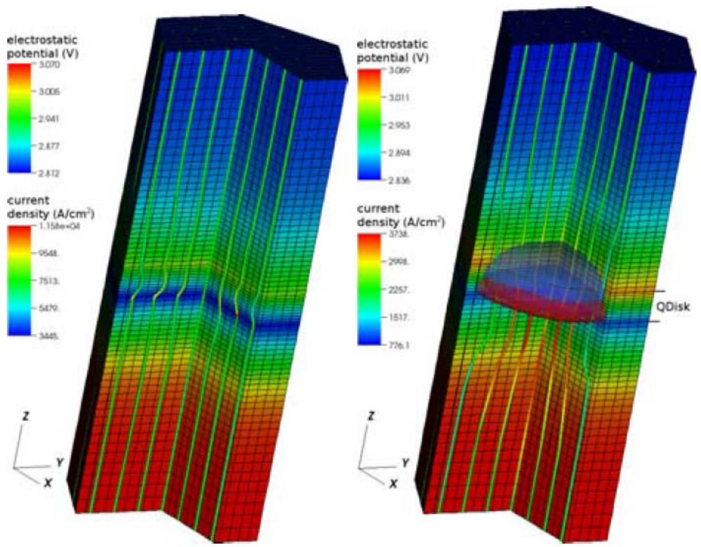


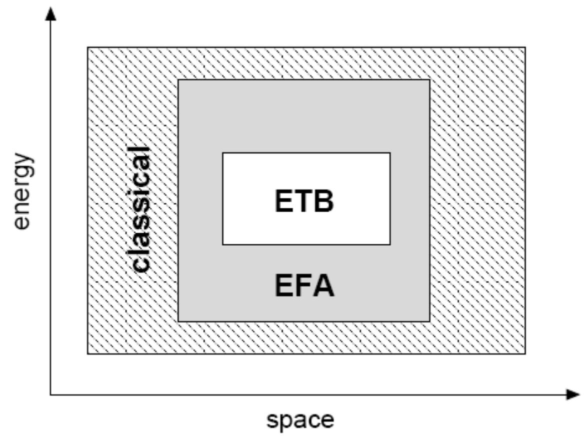


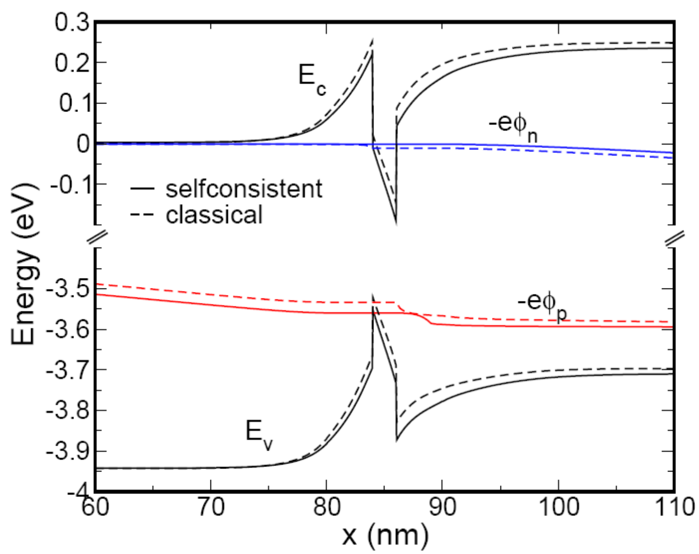


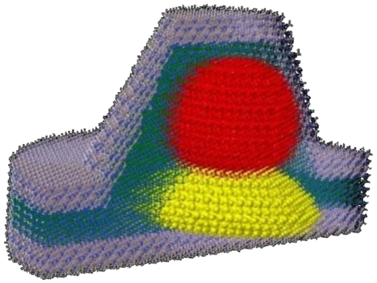
Different simulation domains exchange data

Light emitting GaN/AlGaN QD





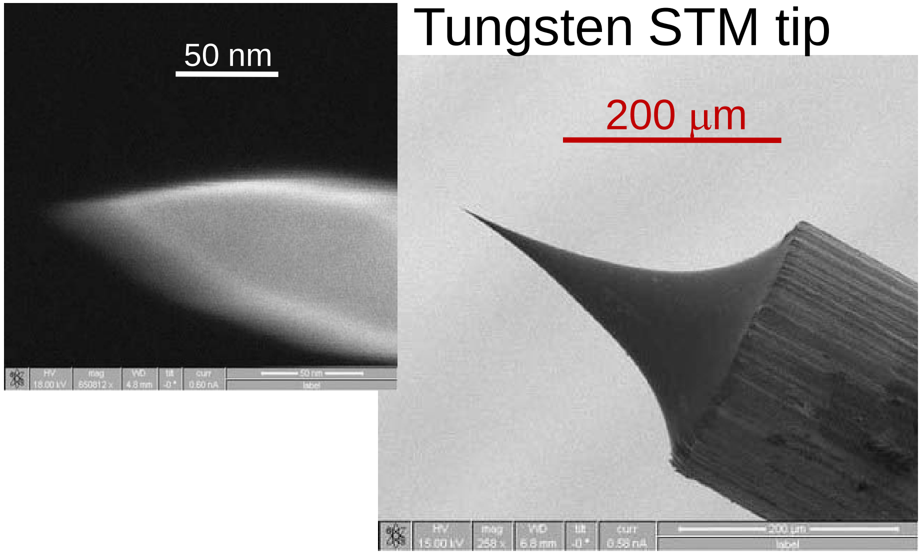


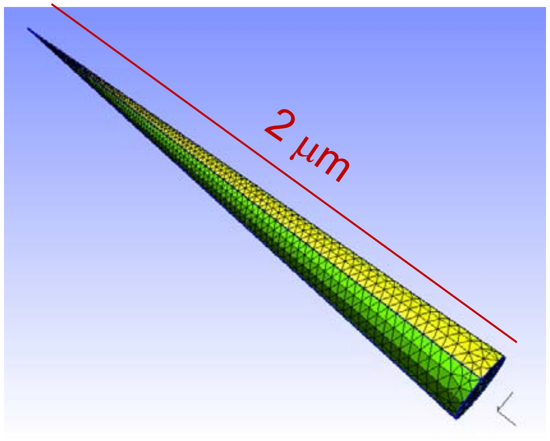


$sp^3d^5s^*$ Empirical TB

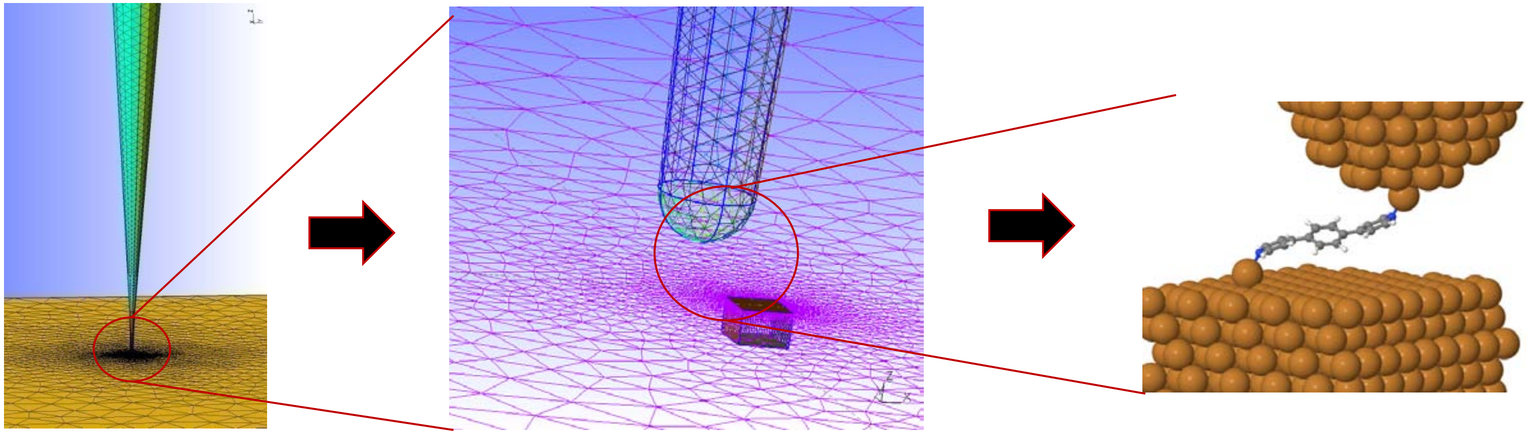
~150.000 atoms

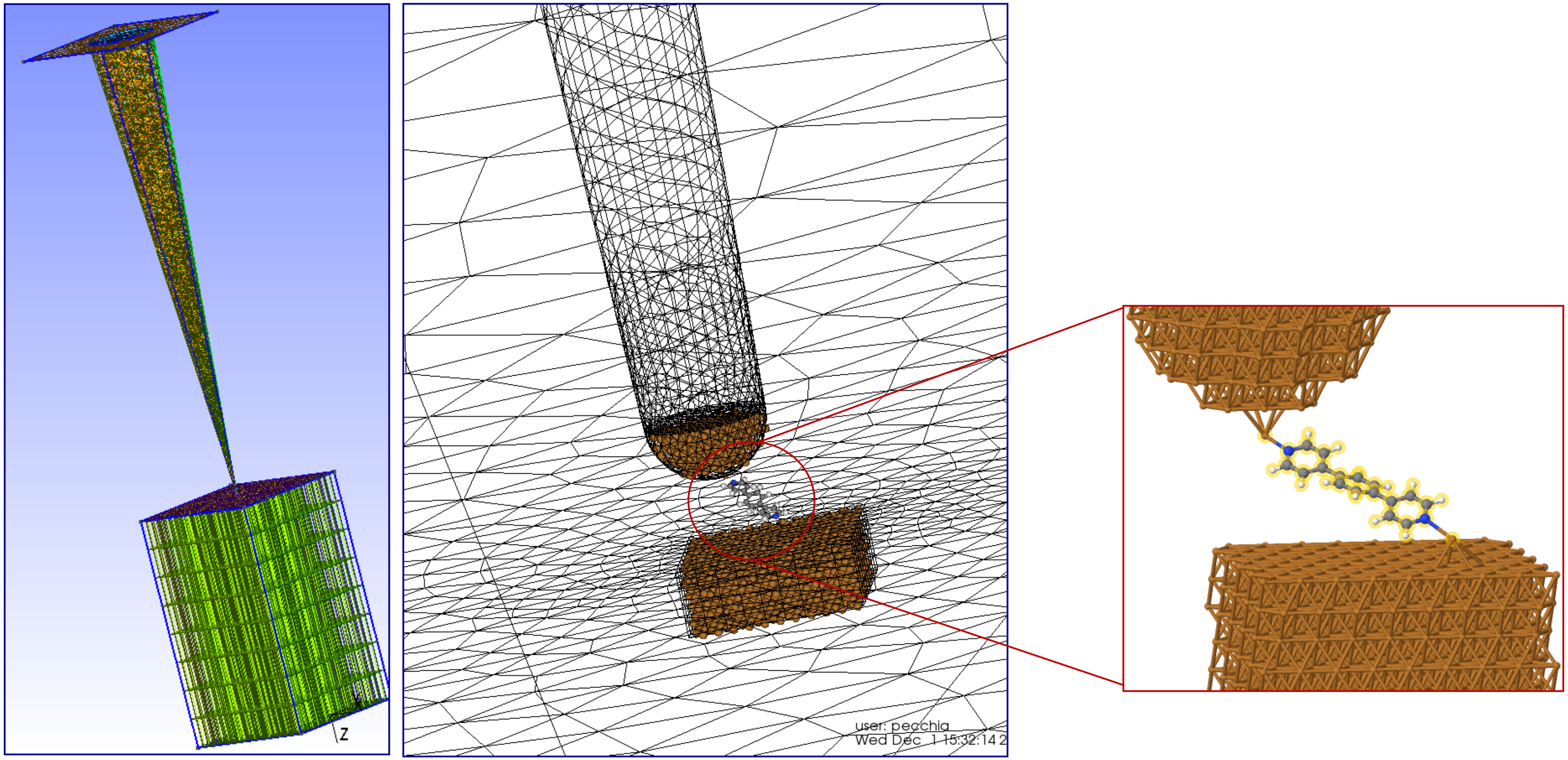
Modeling of STM junction





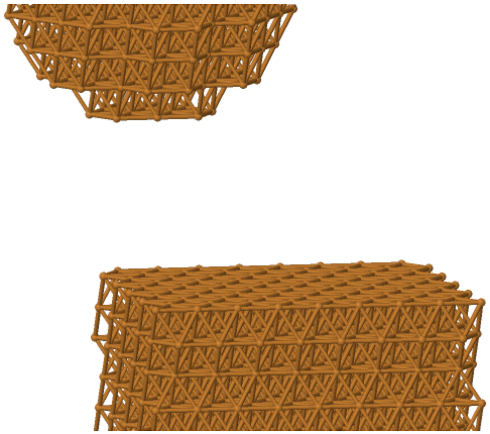
Construction of FEM – Atomistic Model of the STM junction



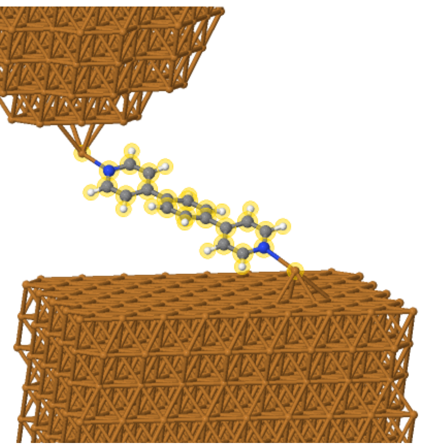

z
user: pecchia
Wed Dec 1 15:32:14 2

Geometry setup

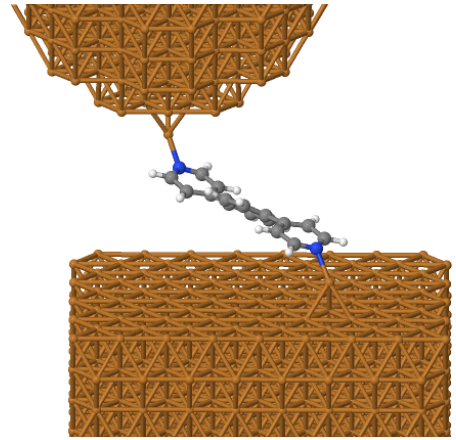
1. Atomistic Generator

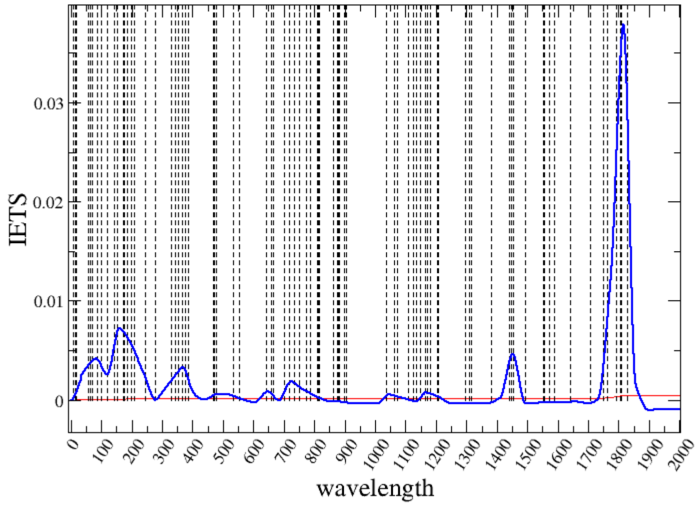


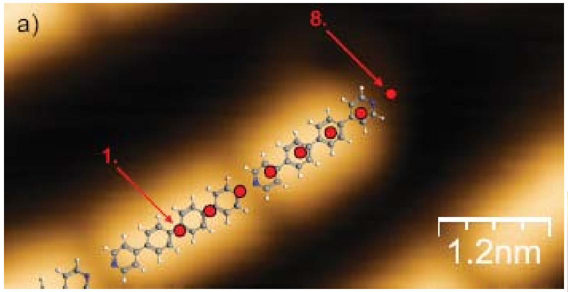
2. Starting guess

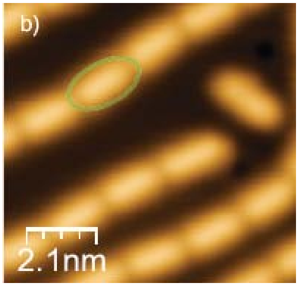


3. Geometry relaxations





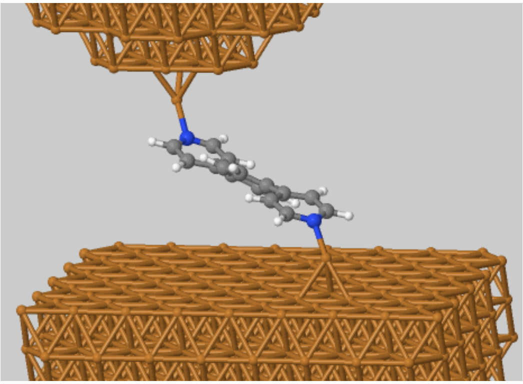




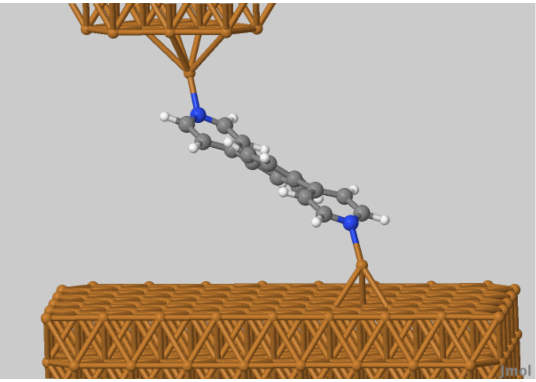
(Gunnar Shulze PhD Thesis)

Molecular rising

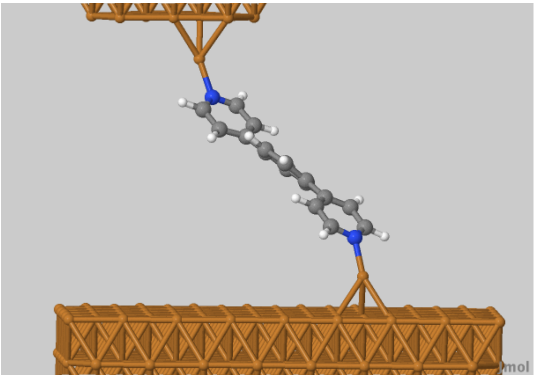
1 nm distance



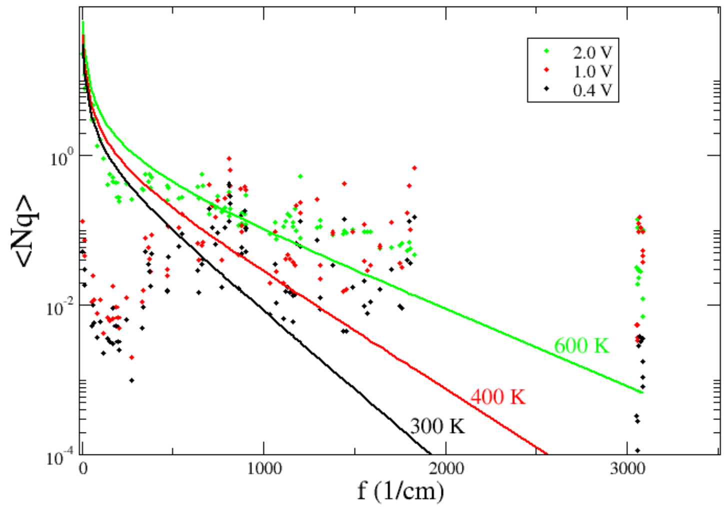
1.2 nm distance



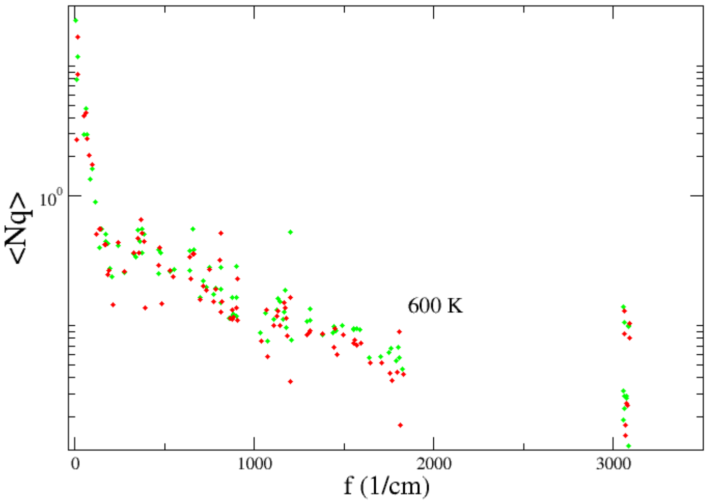
1.4 nm distance

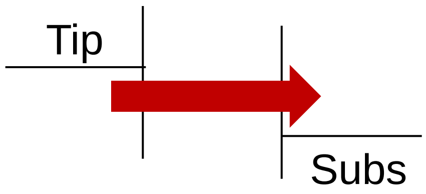


T vs V at 1.0 nm

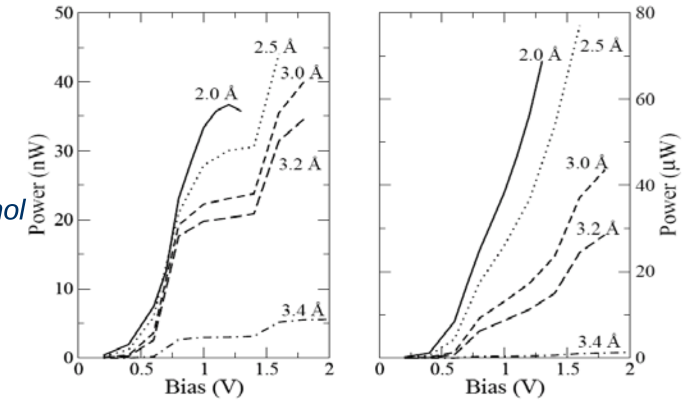


d=1.0, 1.4 nm at 2.0 V





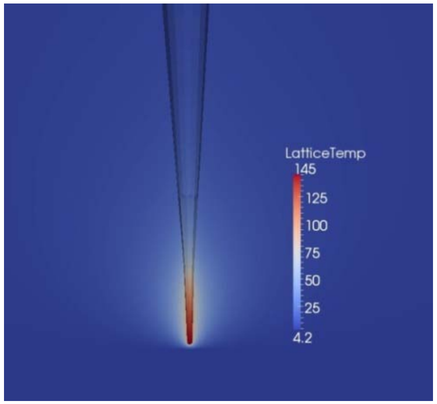
P_{mol}



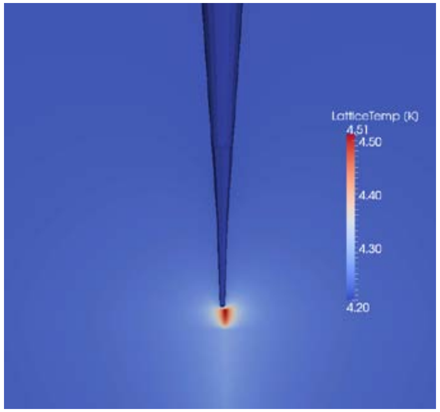
$P=I \cdot V$

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

Most of the power is dissipated in the contacts



Fourier dissipation



Large heating of the tip

reduced thermal conductance due to size effects

Low heating of the substrate

What we learned from gDFTB

DFTB is a versitile intermediate method bridging between ab-initio and empirical potentials for electronic calculatons, structural relaxations and transport (gDFTB).

- Consistent computational framework (geometries, electronics,…)
- Relatively fast but should improve SCC convergence

– Multigrid Poisson solver allows to study complicated device geometries

– Electron-phonon and heating effects can be included

– Electron-electron interactions (GW)… still in progress

outlooks

1. A forthcoming release of dftb+ will contain NEGF (+examples, documentation…)

2. We are working at a general libNEGF to compute electronic densities and current in equilibrium and under bias (FEM / Atomistic)

3. dftb+ has been also included in TiberCAD and we would like to develop QM/MM schemes for nanodevices (interfaces, defects, …)

4. Electron-phonon interactions in dftb+ (via libNEGF)

5. Applications to nanodevices

6. …

Acknowledgements

Prof. Aldo Di Carlo

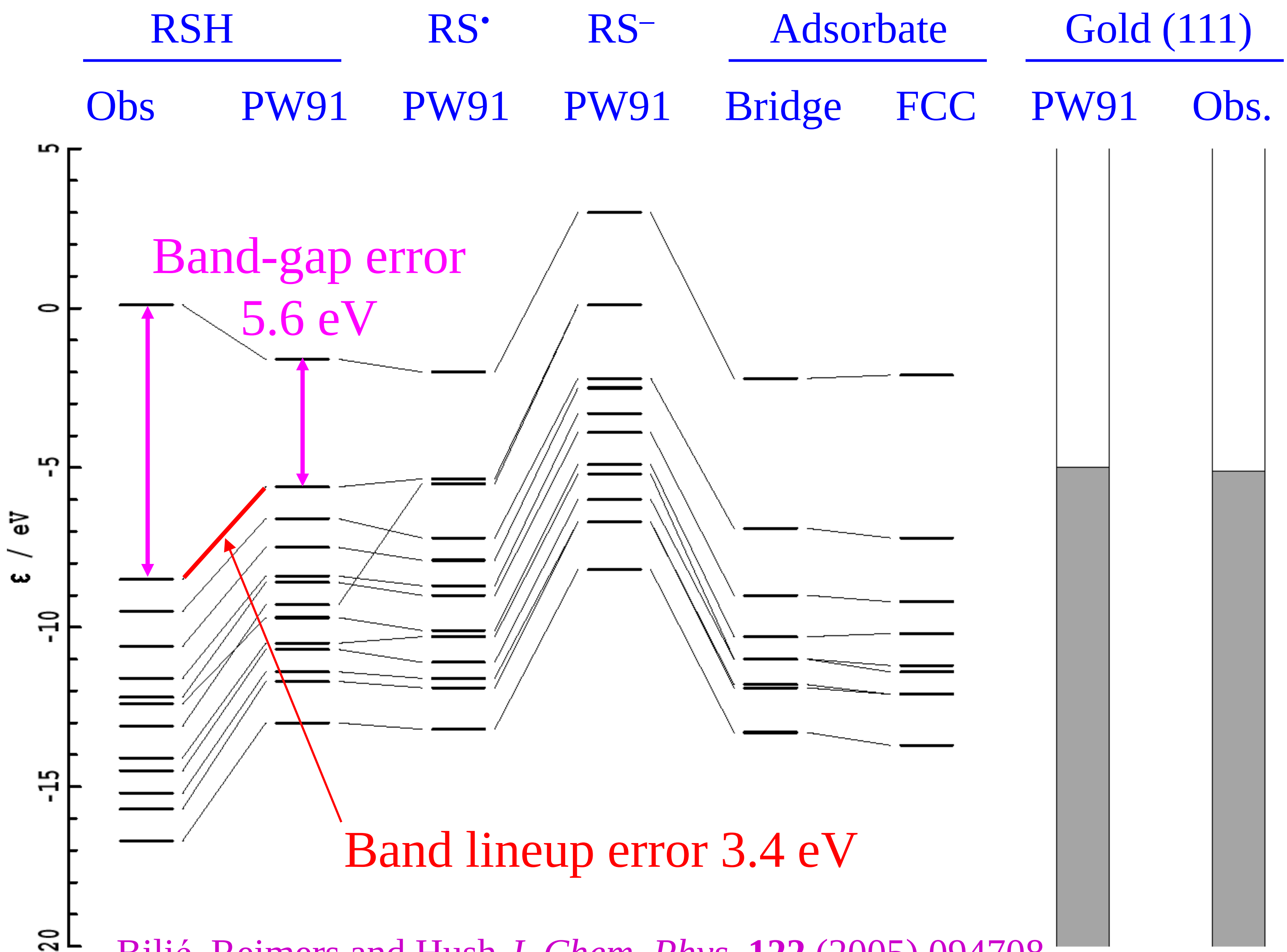
Matthias Auf Der Maur, Post Doc
Fabio Sacconi, Post Doc
Alessio Gagliardi, Post Doc
Michail Povolotsky, Post Doc

Giuseppe Romano, PhD
Gabirele Penazzi, PhD
Marco Pacini, Student
Luca Salvucci, Student

Thank you

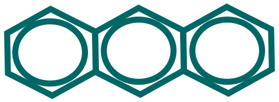
e-e correlations

Level Alignment problem



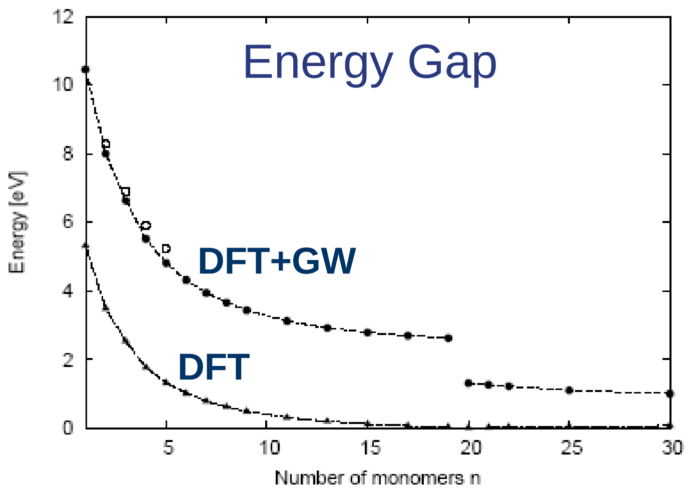
Bilić, Reimers and Hush *J. Chem. Phys.* **122** (2005) 094708

GW on DFTB



T.A. Niehaus et al., Phys. Rev. A, (2005)

State	Sym.	ϵ^i GTO	ϵ^i DFTB	v^i_{xc} GTO	v^i_{xc} DFTB	Σ^i_x GTO	Σ^i_x DFTB	Σ^i_c GTO	Σ^i_c DFTB	ϵ^i_{QP} GTO	ϵ^i_{QP} DFTB
Anthracene											
B_{3u}	π	-7.97	-7.62	-12.98	-12.25	-16.32	-15.61	1.98	2.27	-9.33	-8.71
B_{2g}	σ	-7.85	-7.28	-13.07	-12.17	-16.40	-15.32	2.01	1.90	-9.15	-8.53
A_u	π	-6.82	-6.78	-13.12	-12.24	-15.81	-15.11	1.38	1.66	-8.12	-7.98
B_{1g}	π	-6.51	-6.40	-13.30	-12.10	-15.27	-14.18	1.01	1.06	-7.47	-7.42
B_{2g}	π	-5.30	-5.51	-13.28	-12.18	-14.90	-14.37	0.58	0.88	-6.34	-6.82
B_{3u}	π^*	-2.86	-2.97	-13.08	-11.86	-8.78	-8.17	-1.82	-0.90	-0.37	-0.19
A_u	π^*	-1.58	-1.59	-13.18	-11.62	-8.49	-7.92	-2.21	-1.38	0.89	0.74
B_{1g}	π^*	-1.25	-1.28	-12.89	-11.63	-7.63	-7.34	-2.54	-1.83	1.47	1.18
B_{3u}	π^*	-0.52	0.01	-11.90	-11.41	-6.47	-6.94	-2.84	-2.45	2.07	2.03

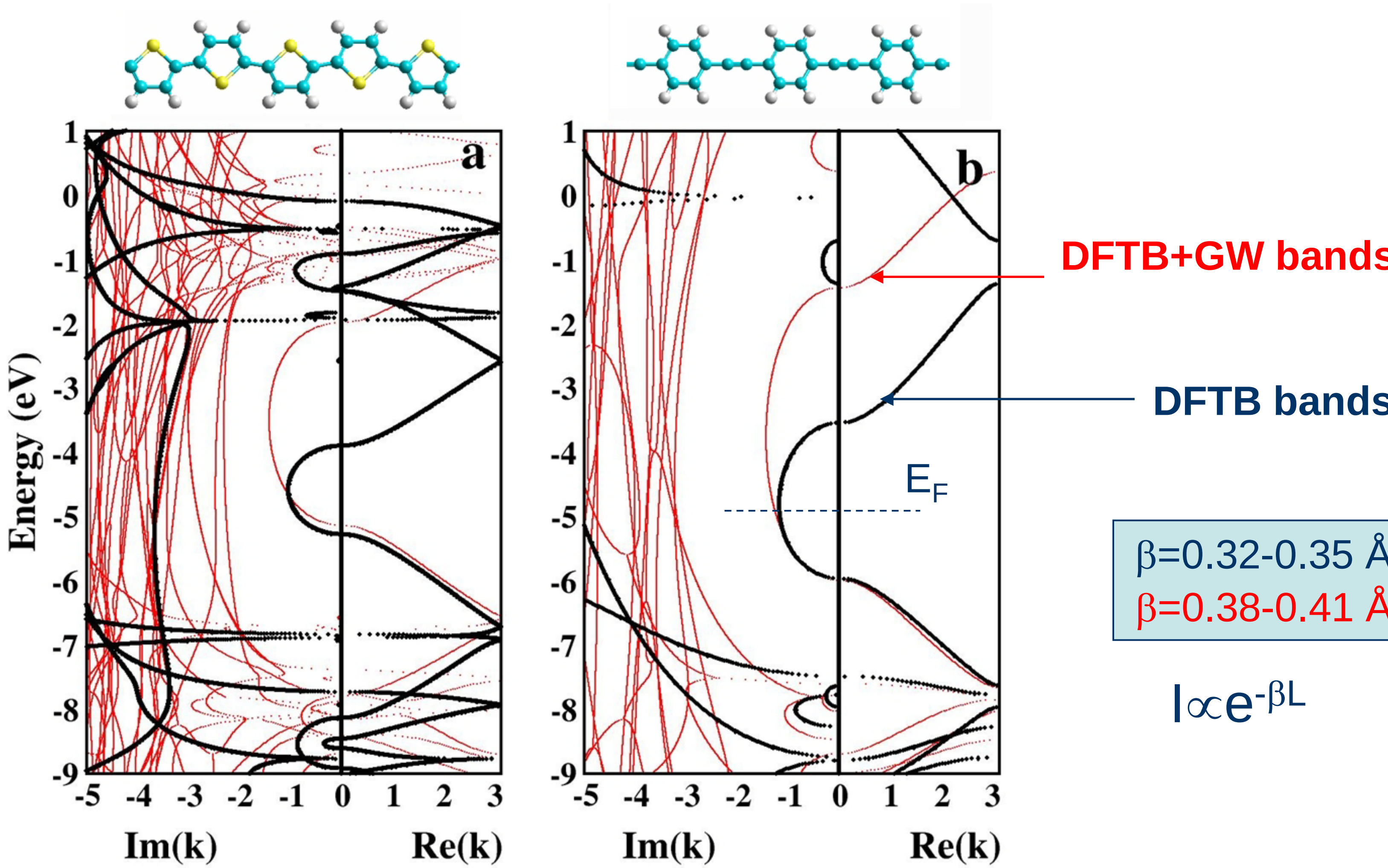


Very Good agreement on π-π*

Worse on σ orbitals

Efficient GW:
Can compute more than 30 rings

Complex bandstructures

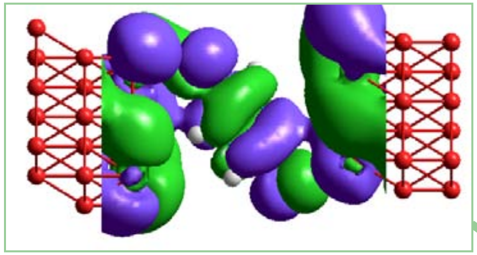


β=0.32-0.35 $Å^{-1}$

β=0.38-0.41 $Å^{-1}$

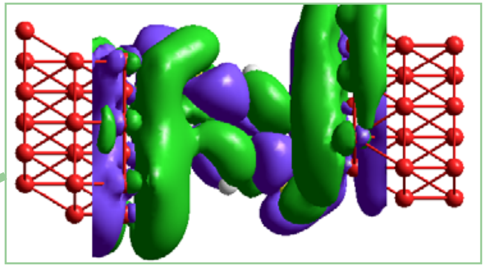
$I \propto e^{-\beta L}$

GW on BDT junction

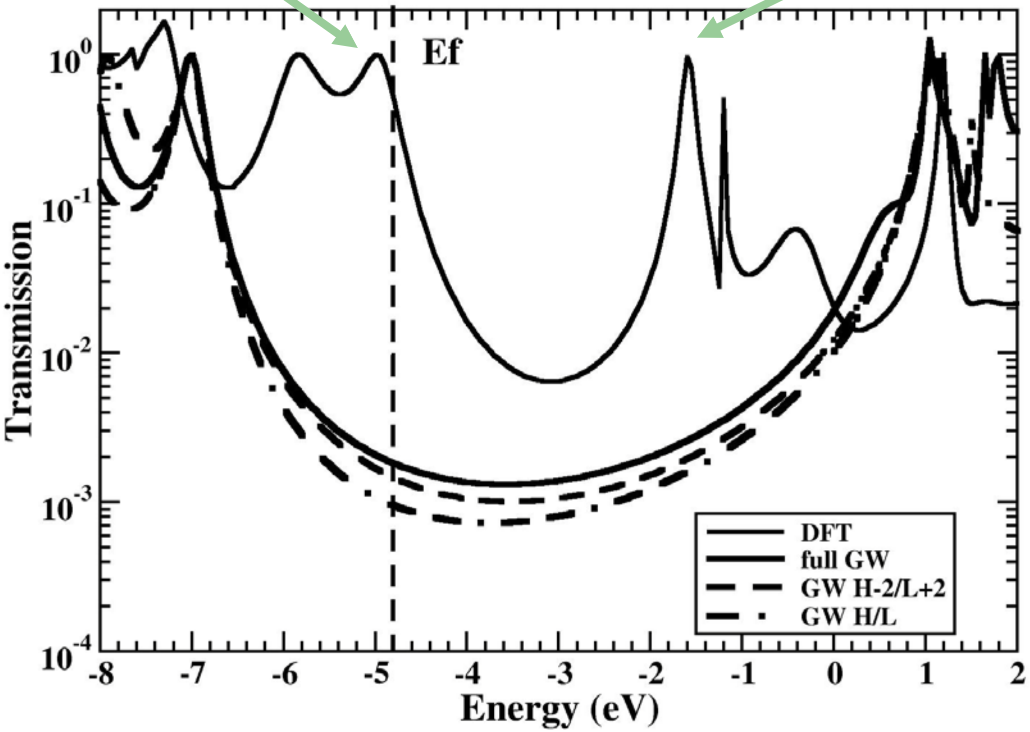


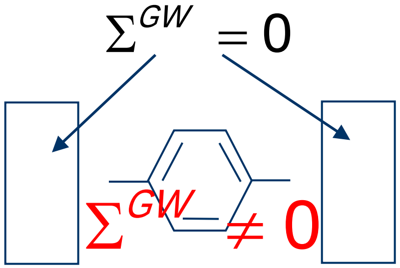
HOMO

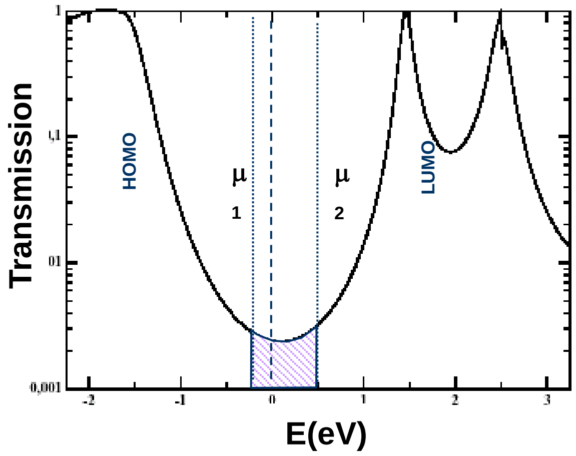
Cu-S-Benzene-S-Cu



LUMO





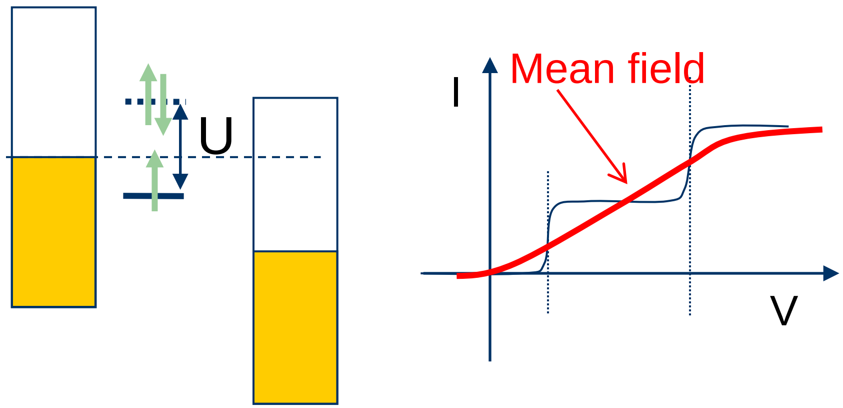


Landauer Formula for coherent transport:

$$I(V) = \frac{2e^2}{h} \int_{-\infty}^{+\infty} T(E,V)[f(E-\mu_1) - f(E-\mu_2)]dE$$

What this is NOT good at

Coulomb Blockades



$$H = \varepsilon n_{\uparrow} + \varepsilon n_{\downarrow} + U n_{\uparrow} n_{\downarrow}$$