Ab Initio GW electron-electron interaction effects in Quantum Transport

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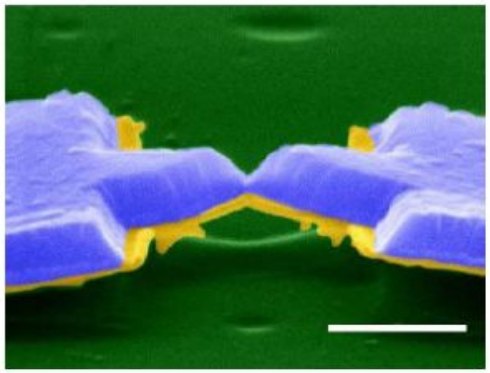
Outline:

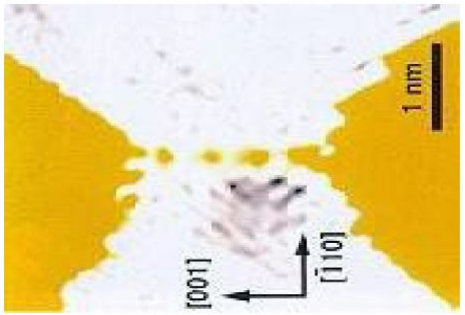
1. Quantum-Transport problem
2. DFT - Landauer-Buttiker Formalism
3. Non-Equilibrium Green's Functions (NEGF) Theory
4. e-e scattering effects and GW
5. Results on Gold Monoatomic chain vs EXP
6. Conclusions and Perspectives

NanoElectronics & Quantum Transport

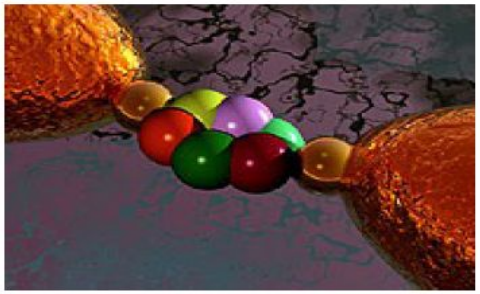
Nanoscale Electronics Devices



[1] N. Agrait et al., Phys. Rep. (2003)



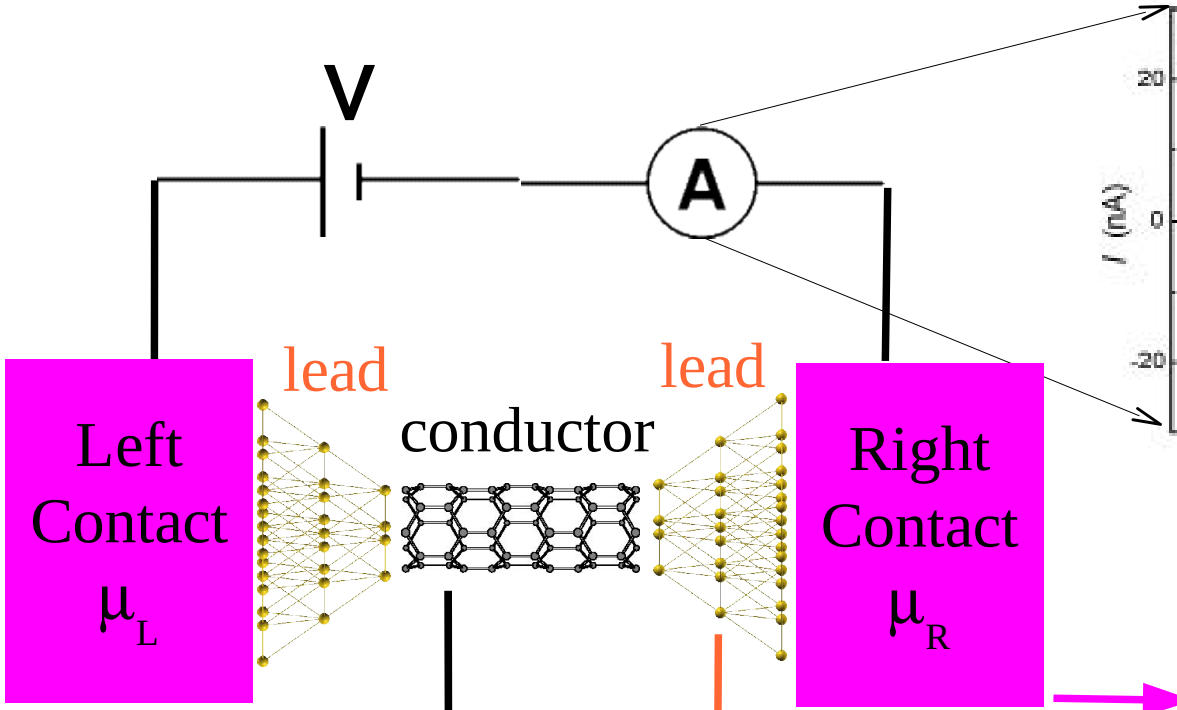
[2] H. Ohnishi et al., Nature (1998)

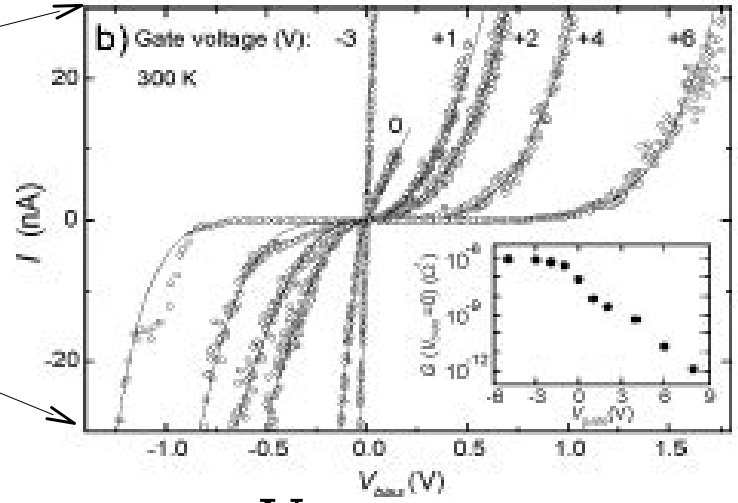


[3] M. Reed et al., Science (1997)

Both an Experimental, a Technological and a Theoretical Challenge!

Quantum Transport: The Working Bench





$$V = \mu_L - \mu_R$$

Macroscopic Reservoirs:
continuum of states,
thermodynamic equilibrium

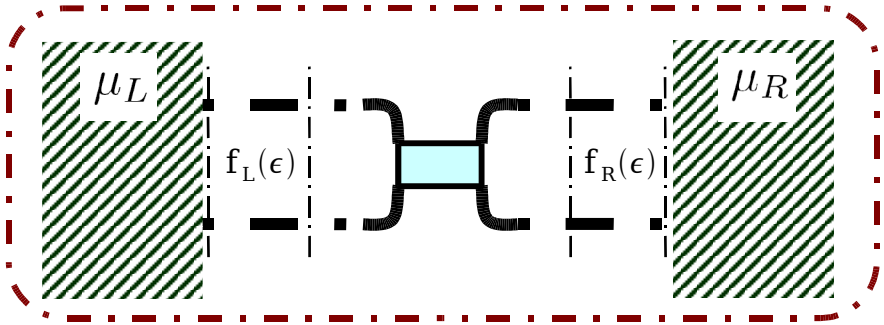
Nanoscale Conductor:
finite number of states,
out of equilibrium,
dissipative effects

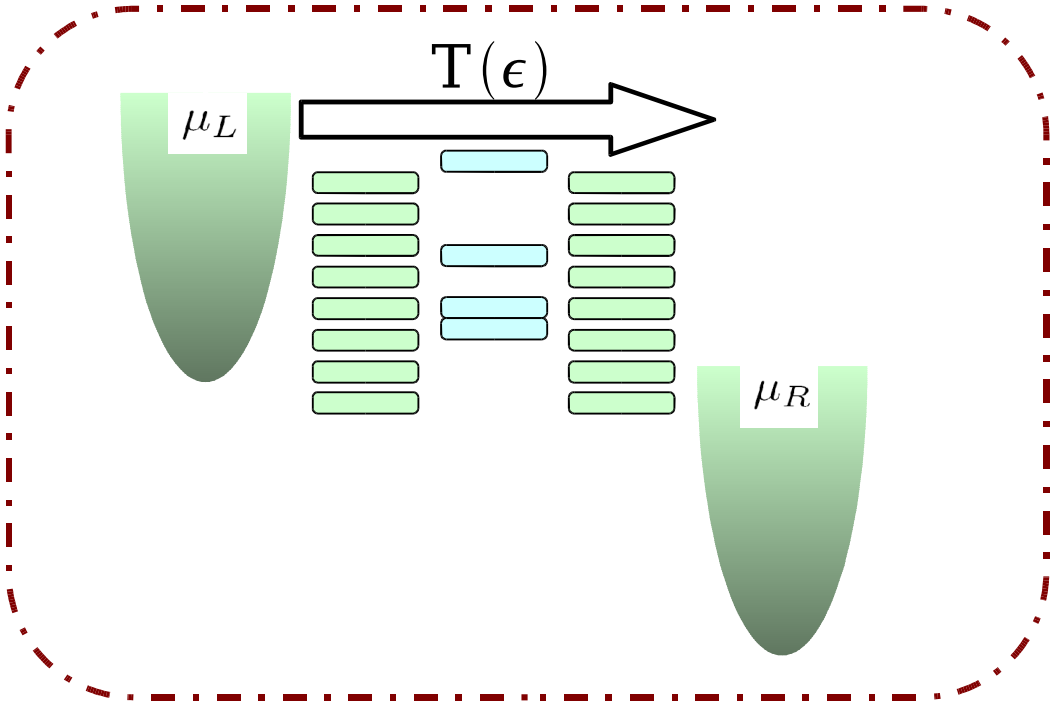
Mesoscopic Leads:
large but finite number of states,
partial equilibrium,
ballistic

We need:

- a First Principle description of the Electronic Structure
- for Finite Voltage: Open System and Out-of-Equilibrium description.

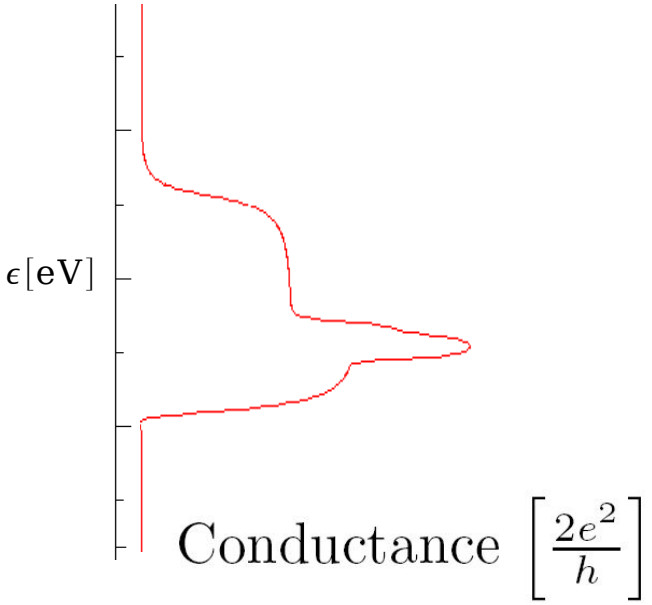
Landauer-Büttiker Theory





Landauer Formula

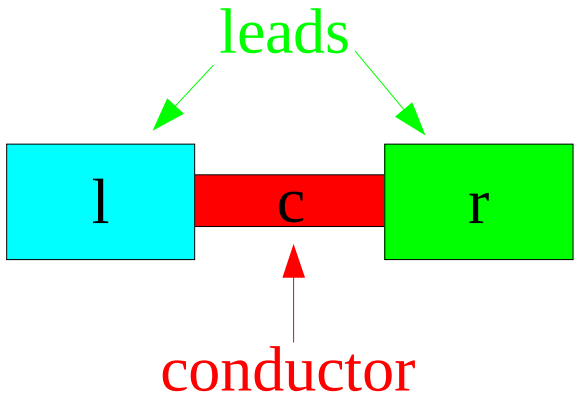
$$C(\epsilon) = \frac{2e^2}{h} M(\epsilon) T(\epsilon)$$



S. Datta, (1995)
R. Landauer, IBMJ. Res. Dev. (1957)
M. Büttiker, PRL (1986)

Landauer Formula in Green's functions

$$\begin{pmatrix} G_l & G_{lc} & G_{lcr} \\ G_{cl} & G_c & G_{cr} \\ G_{rcl} & G_{rc} & G_r \end{pmatrix} = \begin{pmatrix} (\epsilon - H_l) & -H_{lc} & 0 \\ -H_{lc}^{\dagger} & (\epsilon - H_c) & -H_{cr} \\ 0 & -H_{cr}^{\dagger} & (\epsilon - H_r) \end{pmatrix}^{-1}$$



$$G_c = (\epsilon - H_c - \Sigma_l - \Sigma_r)^{-1}$$

conductor Green's functions

$$\Sigma_l = H_{lc}^{\dagger} g_l H_{lc}$$

$$\Sigma_r = H_{cr} g_r H_{cr}^{\dagger}$$

lead self-energies

$$g_{l,r} = (\epsilon - H_{l,r})^{-1}$$

leads bulk Green's functions

$$\Gamma_{l,r} = i[\Sigma_{l,r}^{r} - \Sigma_{l,r}^{a}]$$

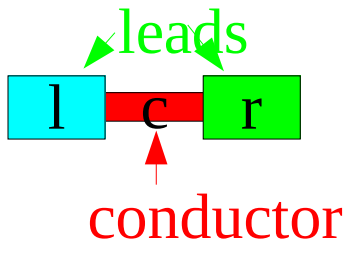
conductor-lead coupling

$$\bar{T} = \mathrm{tr}[\Gamma_l G_c^{r} \Gamma_r G_c^{a}]$$

Fisher-Lee relation

Landauer on top of DFT

What to take for the hamiltonian? $H = \begin{pmatrix} H_l & H_{lc} & 0 \\ H_{lc}^\dagger & H_c & H_{cr} \\ 0 & H_{cr}^\dagger & H_r \end{pmatrix}$



DFT Kohn-Sham hamiltonian! But projected in Real Space!

Convenient basis: **Maximally Localized Wannier functions (MLWF)**:

$$w_{n\mathbf{R}}(\mathbf{r}) = \frac{V}{(2\pi)^3} \int_{BZ} \Psi_{n\mathbf{k}}(\mathbf{r}) e^{-i\mathbf{k}.\mathbf{r}} d^3k$$

N. Marzari and D. Vanderbilt, PRB (1997)
I. Souza et *al.*, PRB (2001)

PWSCF or **ABINIT**: DFT on PW basis set, projections on MLWF

WanT: LB Conductance through Green's functions on MLWF

http://www.wannier-transport.org

A. Calzolari et *al.*, PRB (2004)

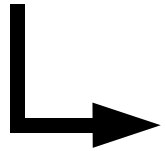
Landauer-Büttiker approach

Correctly describes:

- Contact Resistance
- Scattering on Defects, Impurities
- Non-commensurability patterns

DFT-Landauer drawbacks

- The Kohn-Sham electronic structure is in principle unphysical;
- DFT, no Open Systems, no Out-of-Equilibrium Theory -> only linear response, small bias;
- Non interacting quasiparticles -> only coherent part of transport.



Need to go Beyond!

Beyond LB-DFT: 2 Possibilities

- TDDFT for Quantum Transport
  - promising possibility
  - need a suitable approx for xc

G. Stefanucci and C.-O. Almbladh, Europhys Lett. (2005)

- NEGF (NonEquilibrium Green's Function theory)
  - full access to all observables
  - it has maybe a more intuitive physical meaning

Non-Equilibrium Green's Function Theory (NEGF)

(improperly called Keldysh)

Much more complete framework, allows to deal with:

- **Many-Body** description of **incoherent** transport (electron-electron interaction, electronic correlations and also electron-phonon);
- **Out-of-Equilibrium** situation;
- Access to **Transient** response (beyond Steady-State);
- Reduces to Landauer-Buttiker for coherent transport.

The theory is due to the works of Schwinger, Baym, Kadanoff and Keldysh

Many-Body Finite-Temperature formalism

$$\hat{H} = \hat{T} + \hat{V} + \hat{W}$$

hamiltonian

many-body

$$\bar{o} = \frac{\sum_i e^{-\beta E_i} \langle \Psi_i | \hat{o} | \Psi_i \rangle}{\sum_i e^{-\beta E_i}} = \mathrm{tr}\left[\hat{\rho}(\hat{H})\hat{o}\right]$$

observable

$$\hat{\rho}(\hat{H}) = \frac{e^{-\beta \hat{H}}}{\mathrm{tr}\left[e^{-\beta \hat{H}}\right]}$$

statistical weight

NEGF formalism

$$\hat{H}(t) = \hat{H} + \hat{U}(t) = \hat{T} + \hat{V} + \hat{W} + \hat{U}(t)$$ hamiltonian

many-body + time-dependence

$$\bar{o}(t) = tr\left[\hat{\rho}(\hat{H})\,\hat{o}_H(t)\right] \qquad t > t_0$$ observable

$$\hat{\rho}(\hat{H}) = \frac{e^{-\beta\hat{H}}}{tr\left[e^{-\beta\hat{H}}\right]}$$

statistical weight referred to
the unperturbed Hamiltonian and
the equilibrium situation before t_0

Time Contour

$$\hat{o}_H(t) = \hat{s}(t_0, t)\,\hat{o}(t)\,\hat{s}(t, t_0)$$

Heisenberg representation

$$\hat{s}(t, t_0) = T\left\{\exp\left(-i\int_{t_0}^{t} dt'\,\hat{H}(t')\right)\right\}$$

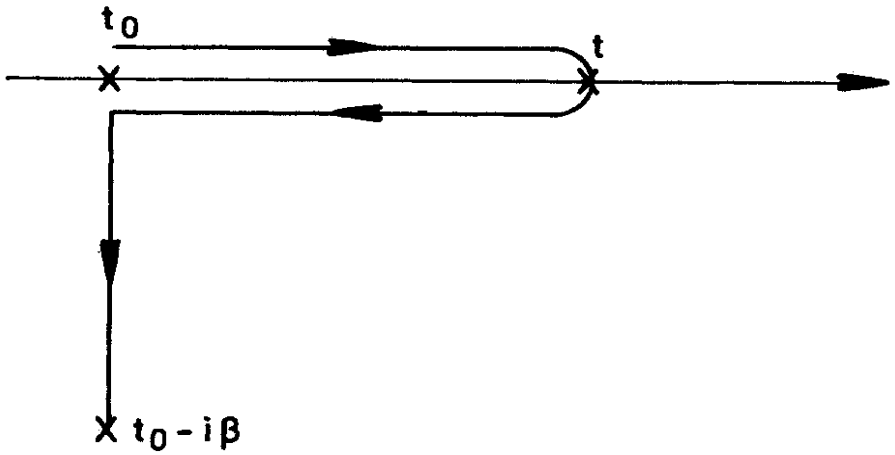
evolution operator

$$\hat{s}(t_0 - i\beta, t_0) = e^{-\beta \hat{H}}$$

trick to put the equilibrium weight into the evolution

$$\bar{o}(t) = \frac{\mathrm{tr}\,[\hat{s}(t_0 - i\beta, t_0)\,\hat{s}(t_0, t)\,\hat{o}(t)\,\hat{s}(t, t_0)]}{\mathrm{tr}\,[\hat{s}(t_0 - i\beta, t_0)]}$$

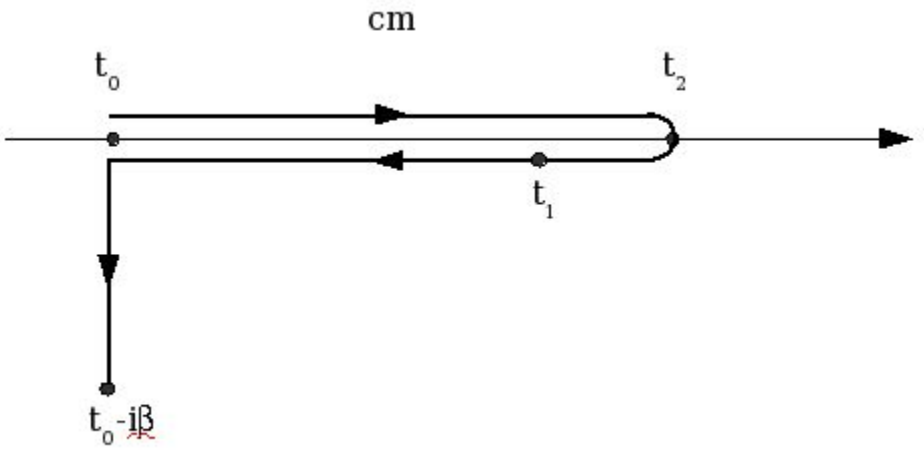
$$\bar{o}(t) = \frac{\mathrm{tr}\,[T_C\,[\exp(-i\int_C dt'\,\hat{H}(t'))\,\hat{o}(t)]]}{\mathrm{tr}\,[T_C\,[\exp(-i\int_C dt'\,\hat{H}(t'))]]}$$



Contour and Perturbation Theory

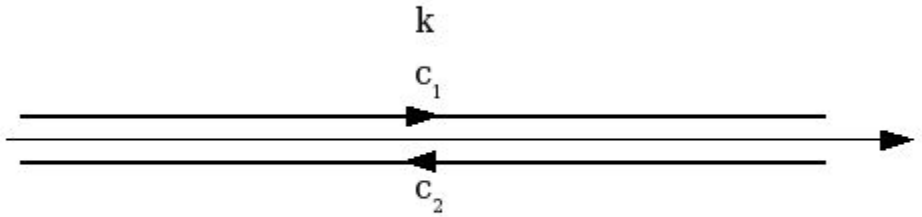
$$G^{co}(x_1, x_2) = (-i)\overline{T_c\{\psi_H(x_1)\psi_H^{\dagger}(x_2)\}}$$ contour ordered Green's function

- To recover perturbation theory (Wick's theorem, Feynman diagrams, etc.) you have to declare the Green's function and all the quantities on the Closed Contour.



$$G^{co}(x_1, x_2) = \begin{cases} G^{to}(x_1, x_2) & t_1, t_2 \in C_{\rightarrow} \\ G^{<}(x_1, x_2) & t_1 \in C_{\rightarrow}, t_2 \in C_{\leftarrow} \\ G^{>}(x_1, x_2) & t_1 \in C_{\leftarrow}, t_2 \in C_{\rightarrow} \\ G^{ato}(x_1, x_2) & t_1, t_2 \in C_{\leftarrow} \end{cases}$$

Keldysh Formulation



Schwinger-Keldysh contour

$$G^{ko} \rightarrow \mathbf{G} = \begin{pmatrix} \mathbf{G_{11}} & \mathbf{G_{12}} \\ \mathbf{G_{21}} & \mathbf{G_{22}} \end{pmatrix}$$

$$\mathbf{G_{11}}(x_1, x_2) = G^{to}(x_1, x_2)$$
$$\mathbf{G_{12}}(x_1, x_2) = G^{<}(x_1, x_2)$$
$$\mathbf{G_{21}}(x_1, x_2) = G^{>}(x_1, x_2)$$
$$\mathbf{G_{22}}(x_1, x_2) = G^{ato}(x_1, x_2)$$

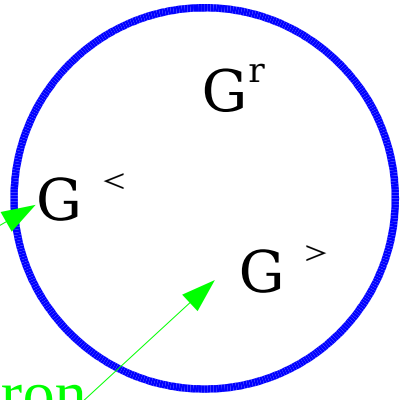
$$\mathbf{G'} = \begin{pmatrix} 0 & G^a \\ G^r & G^k \end{pmatrix}$$ Keldysh formulation

$$G^k = G^{>} + G^{<}$$

Keldysh Green's function

$$\mathbf{G''} = \begin{pmatrix} G^r & G^k \\ 0 & G^a \end{pmatrix}$$ Larkin-Ovchinnikov formulation

Green's and Correlation Functions



hole and electron distribution functions

Out of Equilibrium
we need to introduce
at least three unrelated
"Green's" functions.

Once we know the Green's and the Correlation functions, the problem is solved!
They contain all the physics!

$$G^{to} = G^r + G^<$$

$$A = i(G^r - G^a)$$ Spectral Function

$$-iG^<(\omega) = f_{FD}(\omega)A(\omega)$$

$$iG^>(\omega) = [1 - f_{FD}(\omega)]A(\omega)$$

At Equilibrium the Correlation functions are related to the Green's function through the Fermi-Dirac distribution.

NEGF Fundamental Kinetic Equations

$$G^{r}=[\omega-H_{c}-\Sigma^{r}]^{-1}$$

$$G^{<}=G^{r}\Sigma^{<}G^{a}$$

$$G^{>}=G^{r}\Sigma^{>}G^{a}$$

Caveat!: in case we want to consider also the transient,
then we should add another term to these equations:

$$G^{<}=G^{r}\Sigma^{<}G^{a}+(1+G^{r}\Sigma^{r})G^{0<}(1+\Sigma^{a}G^{a})$$ Keldysh equation

Self-Energy and Scattering Functions

Σ^{r} Self-Energy

$\Sigma^{<}$ In-scattering function
(represent the rate at which the electrons come in)

$\Sigma^{>}$ Out-scattering function

$\Gamma = \mathrm{i}(\Sigma^{\mathrm{r}} - \Sigma^{\mathrm{a}})$ Decay Rate

$$-\mathrm{i}\Sigma^{<}(\omega) = \mathrm{f}_{\mathrm{FD}}(\omega)\Gamma(\omega)$$

$$\mathrm{i}\Sigma^{>}(\omega) = [1 - \mathrm{f}_{\mathrm{FD}}(\omega)]\Gamma(\omega)$$

At Equilibrium

Quantum Transport: composition of the Self-energy

$$\Sigma^{r<>} = \sum_p \Sigma_p^{r<>} + \Sigma_{e-ph}^{r<>} + \Sigma_{e-e}^{r<>}$$

interaction with the leads

electron-phonon interaction
-> SCBA (Frederiksen et al. PRL 2004)

electron-electron interaction
-> ?

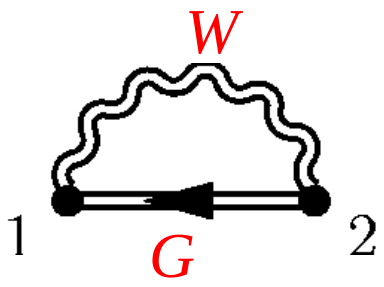
Critical point :

- Choice of relevant approximations for the Self-Energy and the in/out scattering functions

e-e interactions, our choice: the GW Approximation

$$\Sigma_{GW}(x_1, x_2) = iG(x_1, x_2)W(x_1, x_2)$$ GW Self-Energy

$$\Sigma_M^{GW}(x_1, x_2) =$$



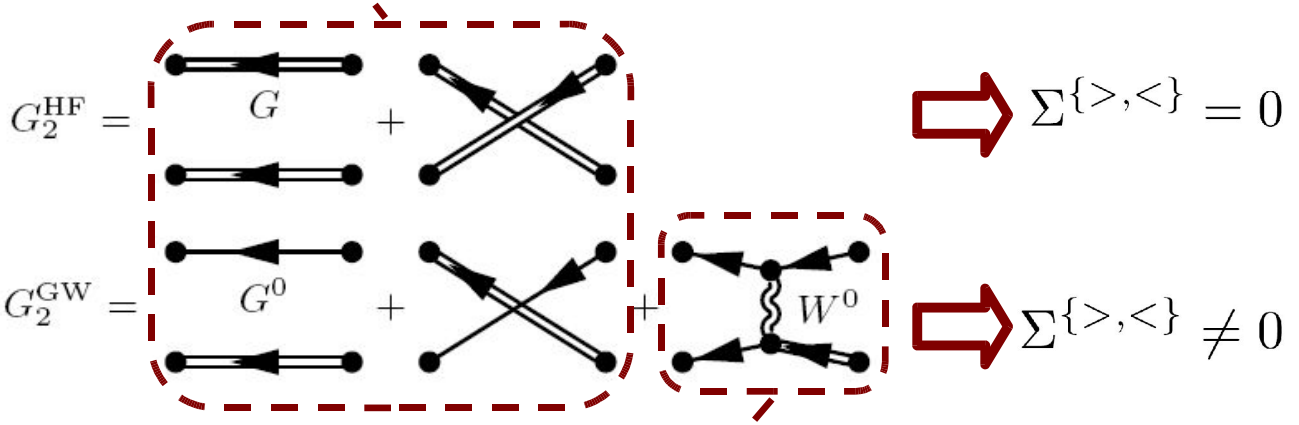
Why GW?

Direct and Exchange terms:

Band Structure Renormalization

Selfconsistent Hartree Fock

G^0W^0



Collisional Term:

Band structure renormalization for Electronic Correlations +

e-e Scattering ->

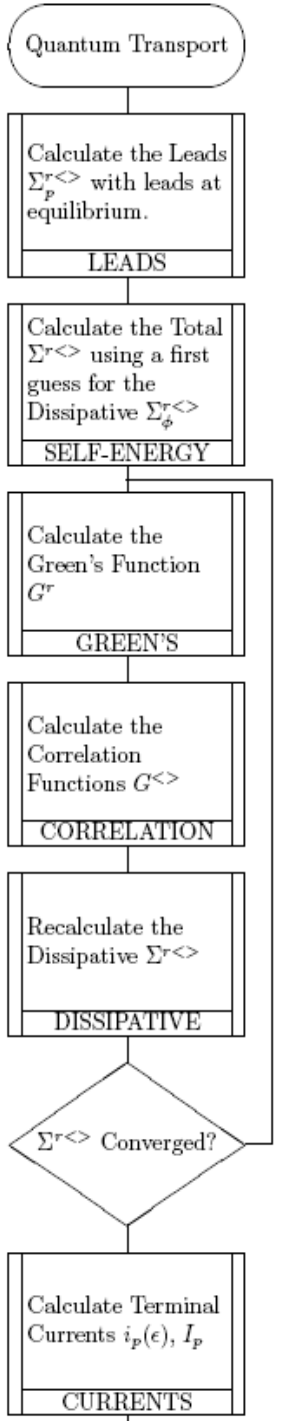
Conductance Degrading Mechanisms, Resistance, non-coherent transport

NEGF Quantum Transport resolution scheme

Iterating the Kinetic Equations

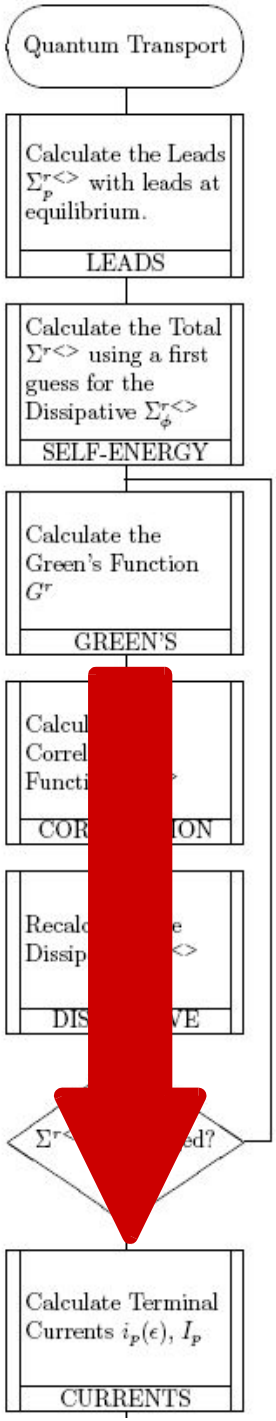
(G and Σ have to be recalculated at each iteration):

- Highly Time-consuming
- So far applied to model Hamiltonians (Anderson, Kondo)
  But very few applications for real systems



Our Scheme

- Approximations:
  1) G^0W^0 Non Self-Consistently
  2) Equilibrium (linear response, small bias)
  3) Neglect Transient (Steady-State)
- We take into account:
  1) Many-Body Correlations (Renormalization of the electronic structure)
  2) e-e Scattering (appearance of Resistance and Loss-of-Coherence)
  3) Finite Lifetime and Dynamical effects (beyond Plasmon-Pole GW)



Formula for the Current: From Landauer to Meir-Wingreen

$$G_{0C}^{r/a}(\omega) = [\omega - H_C - \Sigma_L^{r/a}(\omega) - \Sigma_R^{r/a}(\omega)]^{-1}$$

$$G_{C}^{r/a}(\omega) = [\omega - H_C - \Sigma_L^{r/a}(\omega) - \Sigma_R^{r/a}(\omega) - \Sigma_{corr}^{r/a}(\omega)]^{-1}$$

$$I = \frac{e}{h}\int \frac{d\omega}{2\pi}\,(f_L(\omega) - f_R(\omega))\,Tr\left\{G_{0C}^{a}(\omega)\Gamma_L(\omega)G_{0C}^{r}(\omega)\Gamma_R(\omega)\right\}$$

Landauer

$$I = \frac{e}{h}\int \frac{d\omega}{2\pi}\,(f_L(\omega) - f_R(\omega))\,Tr\left\{G_{C}^{a}(\omega)\Gamma_L(\omega)G_{C}^{r}(\omega)\Gamma_R(\omega)\Lambda(\omega)\right\}$$

Meir-Wingreen
(Landauer-like formula)

$$\Lambda(\omega) = [\Gamma_L(\omega) + \Gamma_R(\omega)]^{-1}\,[\Gamma_L(\omega) + \Gamma_R(\omega) + \Gamma_{corr}(\omega)]$$

$$\Gamma_{L/R/corr}(\omega) = i[\Sigma_{L/R/corr}^{r}(\omega) - \Sigma_{L/R/corr}^{a}(\omega)]$$

Allows to calculate the current through an interacting region at equilibrium

Our Scheme

DFT-LDA (PWSCF)

Leads ϕ^{KS}_{Lead} ϵ^{KS}_{Lead}

Conductor ϕ^{KS}_{Cond} ϵ^{KS}_{Cond}

Correlated Conductor ϕ^{KS}_{Cond} ϵ^{KS}_{Cond}

GW (Modified Abinit GW)

$$\Sigma^{GW}(\omega) = \frac{i}{2\pi}\int_{-\infty}^{\infty} d\omega' G^0(\omega-\omega') W^{RPA}(\omega')$$

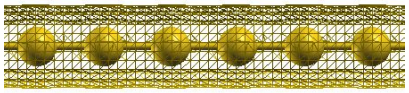
MLWF (WanT)

$U(\mathbf{k})\, w_{n\mathbf{R}}(\mathbf{r})$ $U(\mathbf{k})\, w_{n\mathbf{R}}(\mathbf{r})$ $\langle w_{n\mathbf{R}} | \Sigma(\omega) | w_{n\mathbf{R}} \rangle$

Transport Calculation

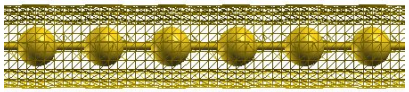
Meir-Wingreen $I = \frac{e}{h}\int \frac{d\omega}{2\pi}\left(f_L(\omega) - f_R(\omega)\right) Tr\left\{\Gamma_L(\omega) G^r_C(\omega) \Gamma_R(\omega) \Lambda(\omega) G^a_C(\omega)\right\}$

Results on
Au Monoatomic Chain

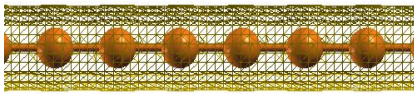


GW on **Au Monoatomic chain**: renormalization of the energies

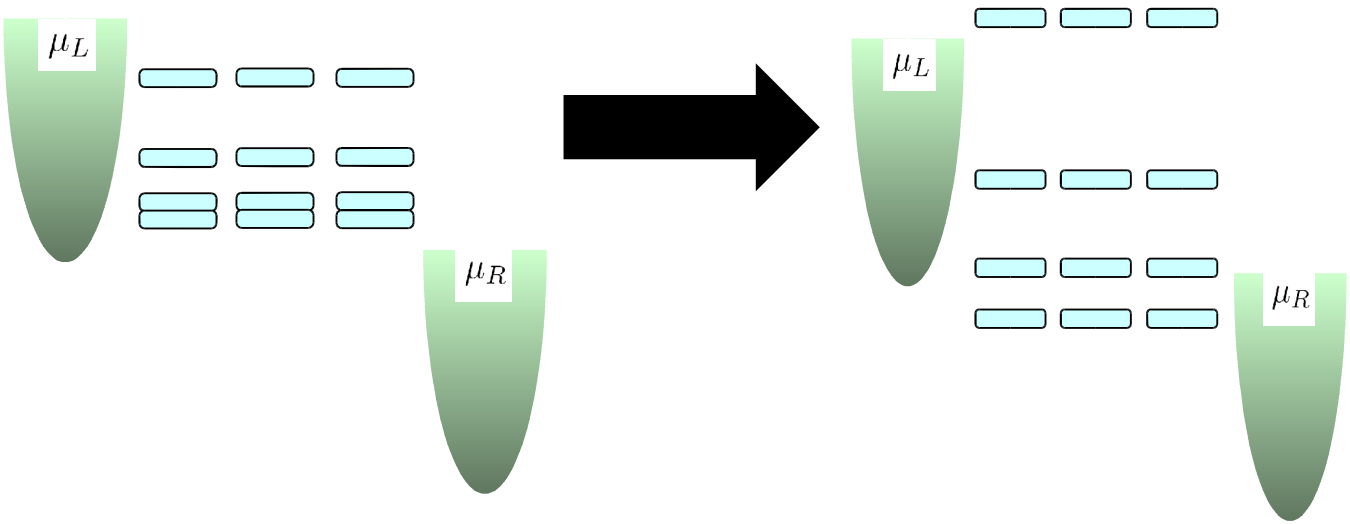
Kohn-Sham Au chain



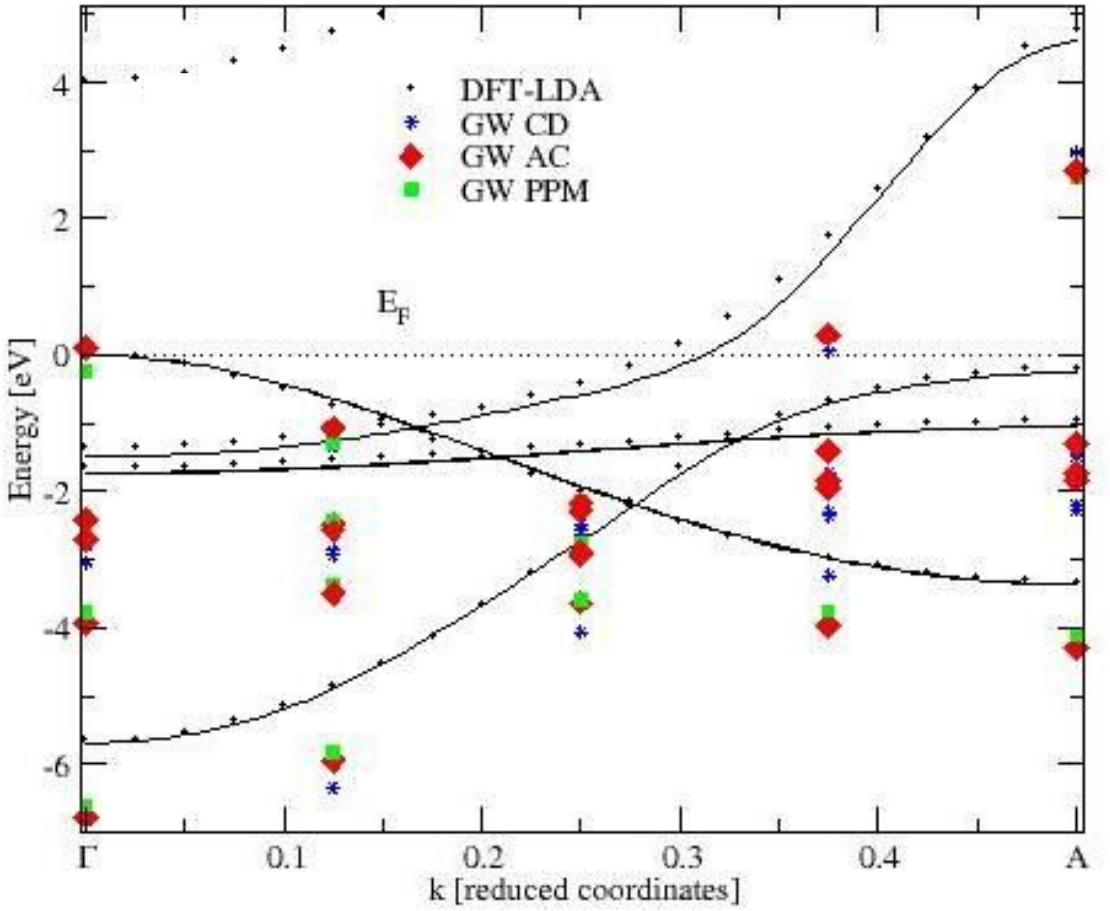
GW Au chain



$$\epsilon^{KS} \Rightarrow \epsilon^{GW}$$



Monoatomic Gold chain: GW vs DFT bandplot



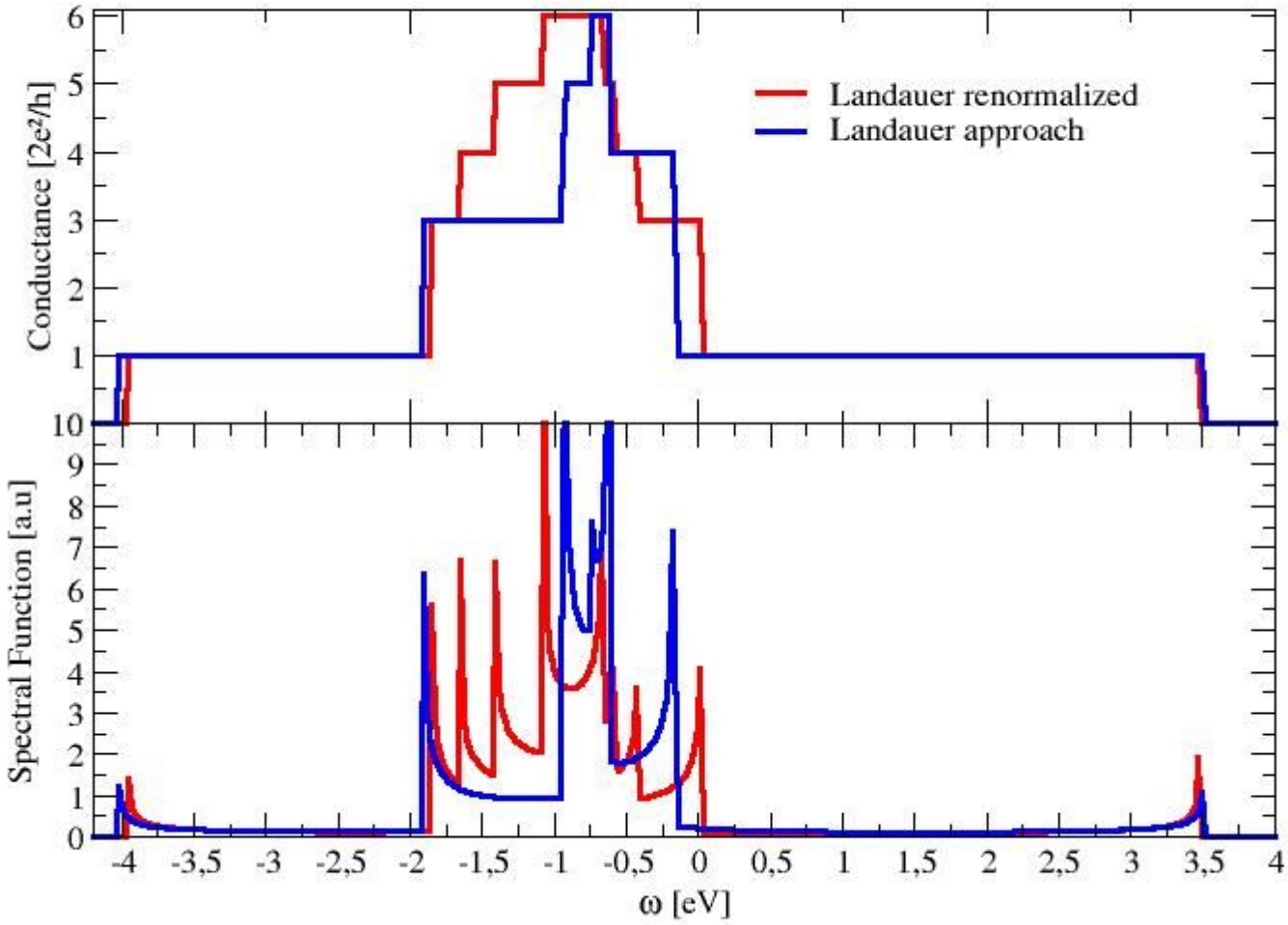
- PPM Plasmon-Pole Model
- AC Analytic Continuation
- CD Contour Deformation

(**more or less the same**)

- DFT :
  Cut-off : 15 Ha,
  Nk 5 in IBZ,
  Smearing 0.015 Ha
- GW :
  W : 150 bands, WF
  1505 PW, Eps 49 PW
  Sigma : 100 bands,
  PW 2503, freq 16000

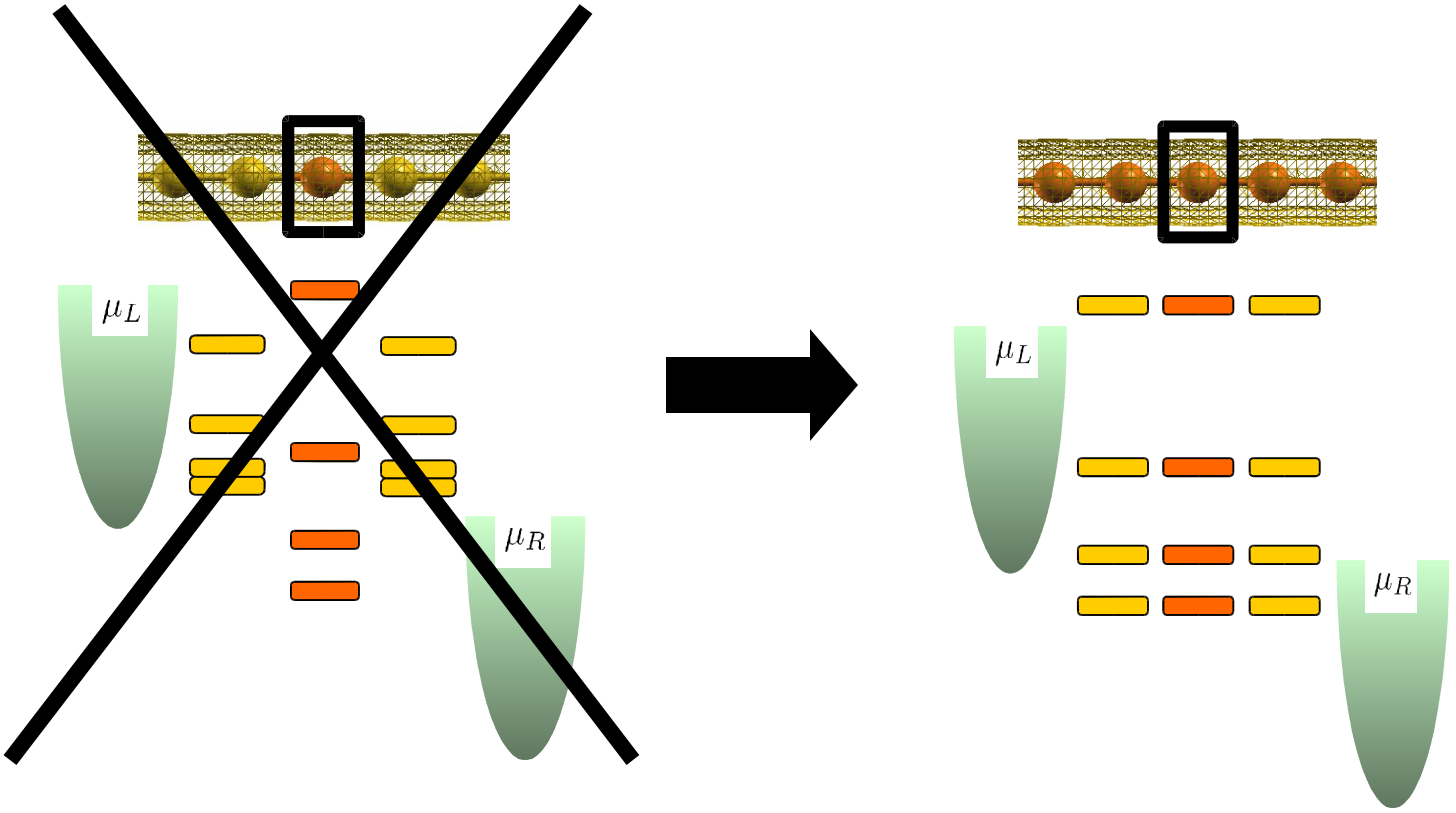
P. Darancet, A. Ferretti, D. Mayou, et V. Olevano, *submitted* (2006)

GW vs DFT
Landauer Conductance

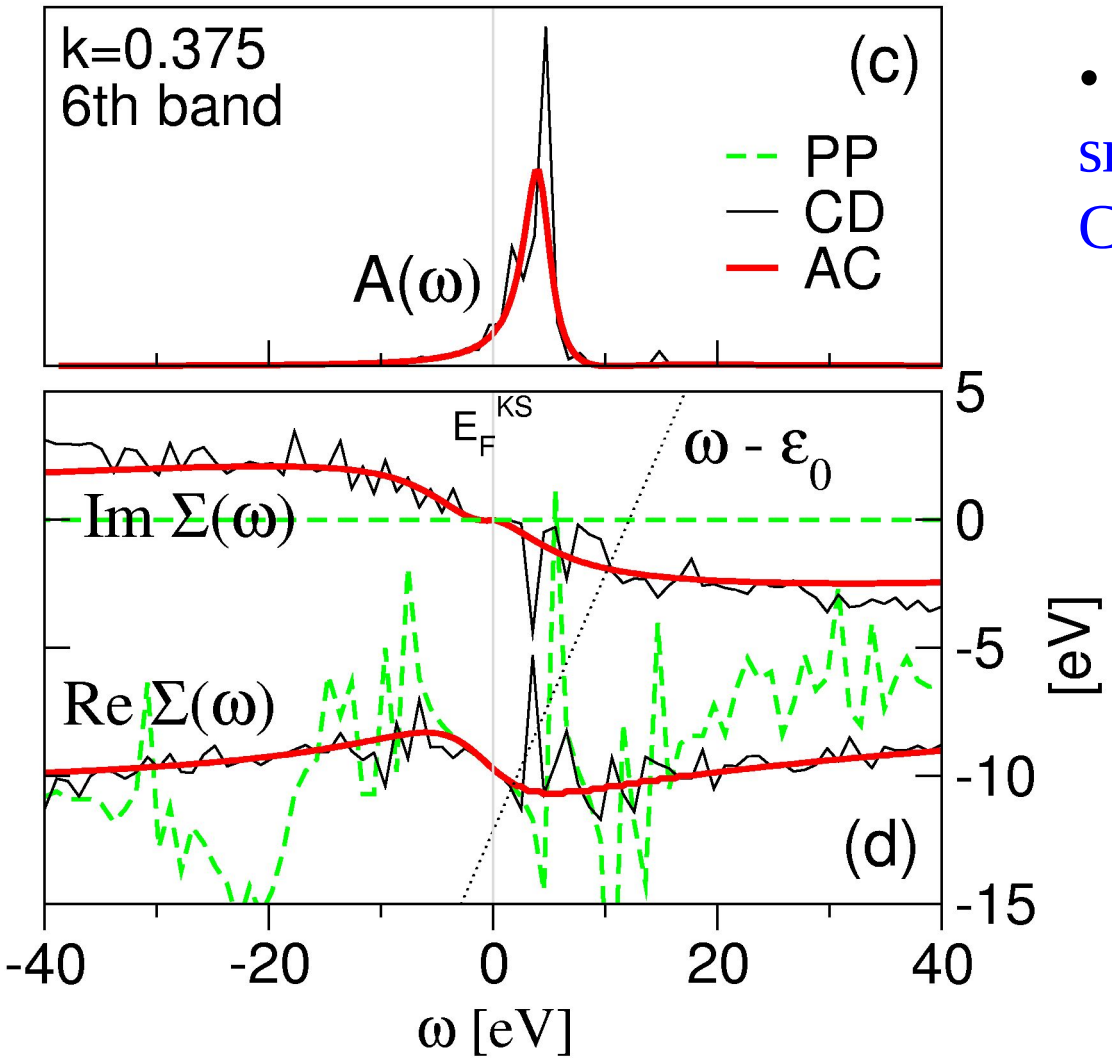


- non-negligible rearrangement of the conductance channels
- Still **ballistic** conductor (flat plateaus)

e-e scattering switched on in the Conductor

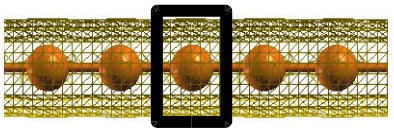


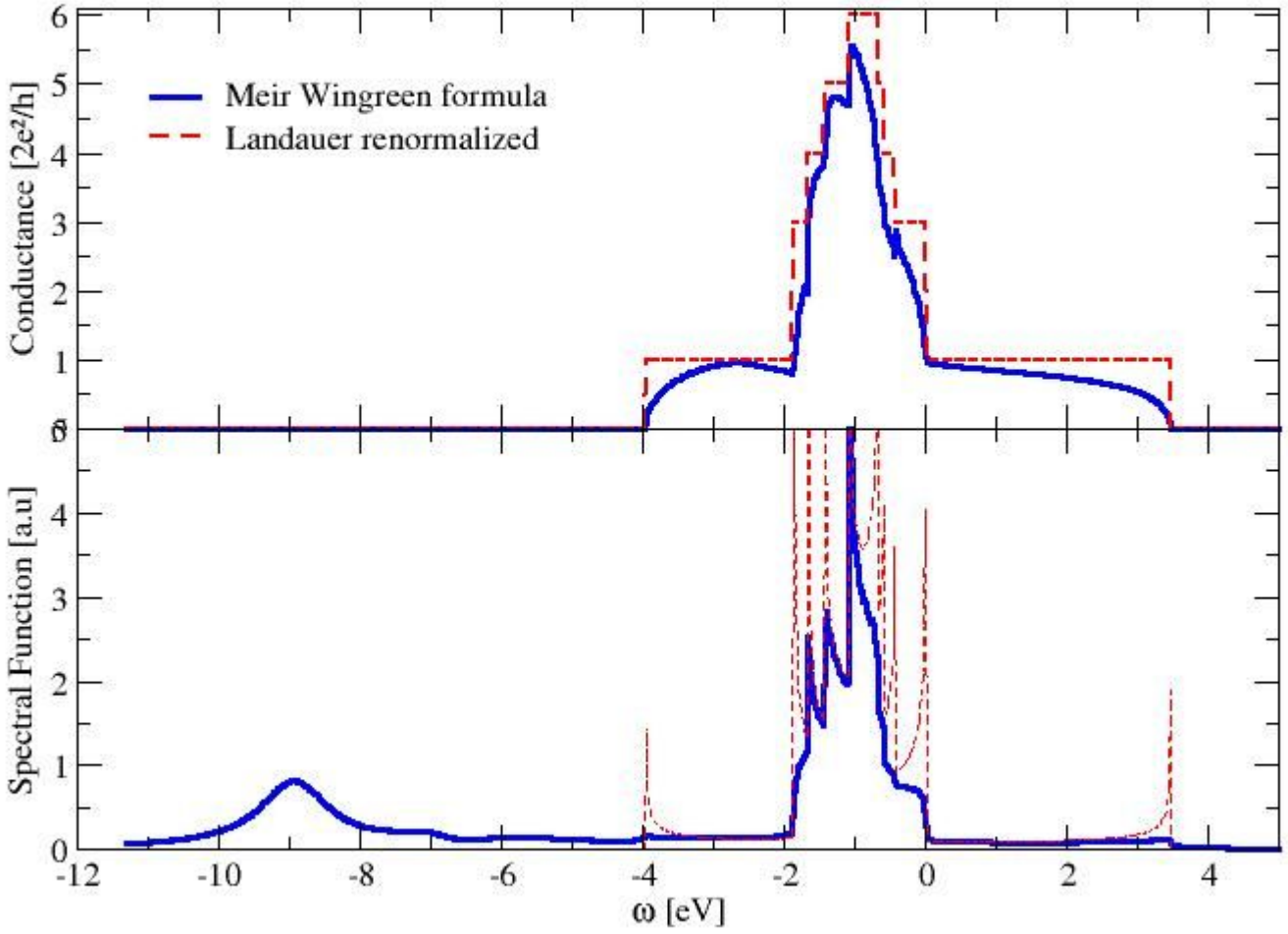
Self-Energy and Spectral Function



- The Analytic Continuation smooths the more accurate Contour Deformation

e-e scattering only in the Conductor





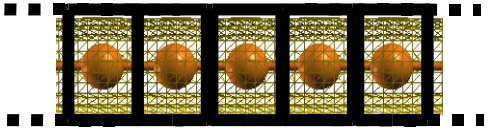
Loss of Conductance:

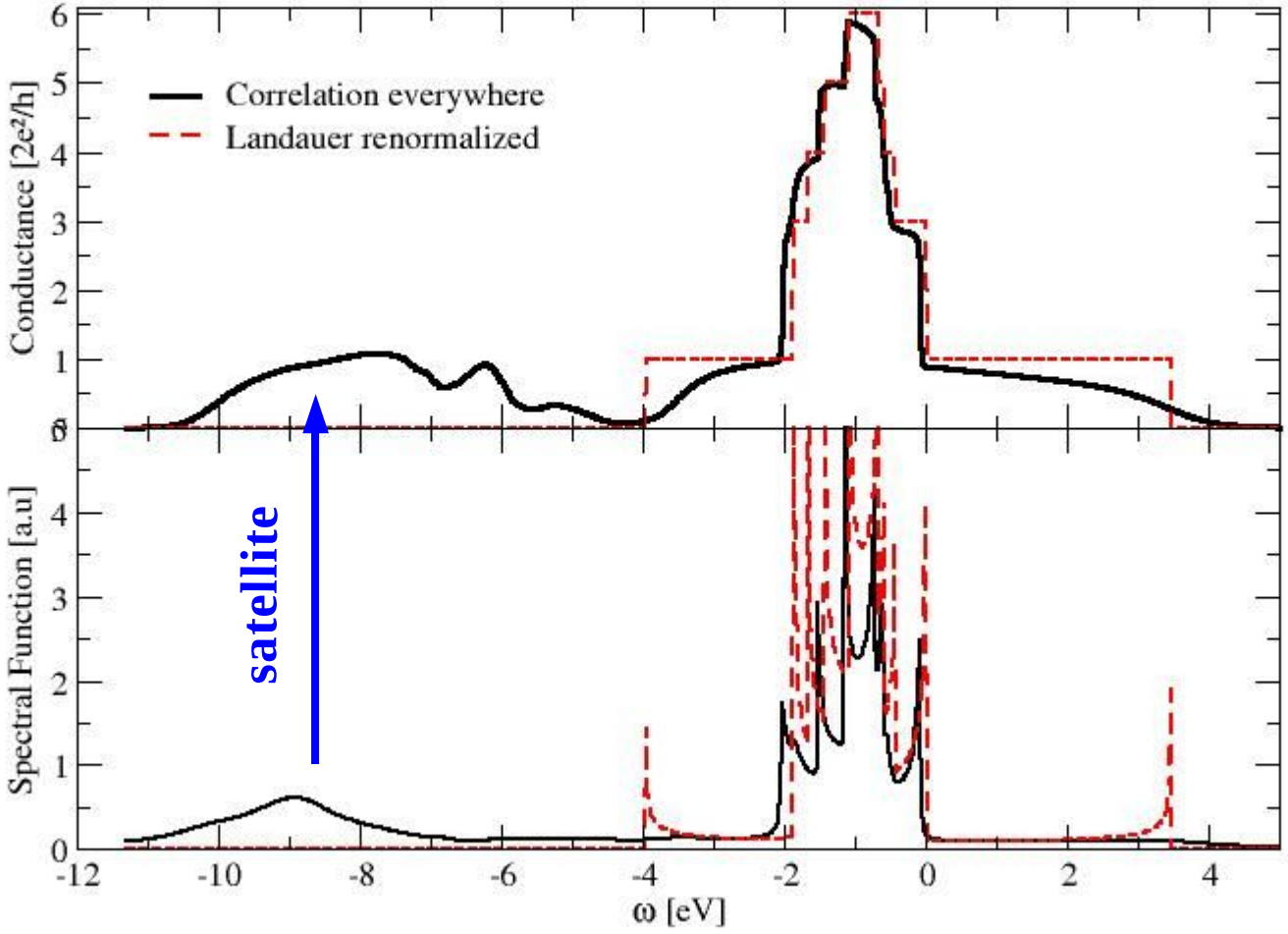
➔**Appearance of Resistance**

Broadening of the peaks:

➔ **QP lifetime**

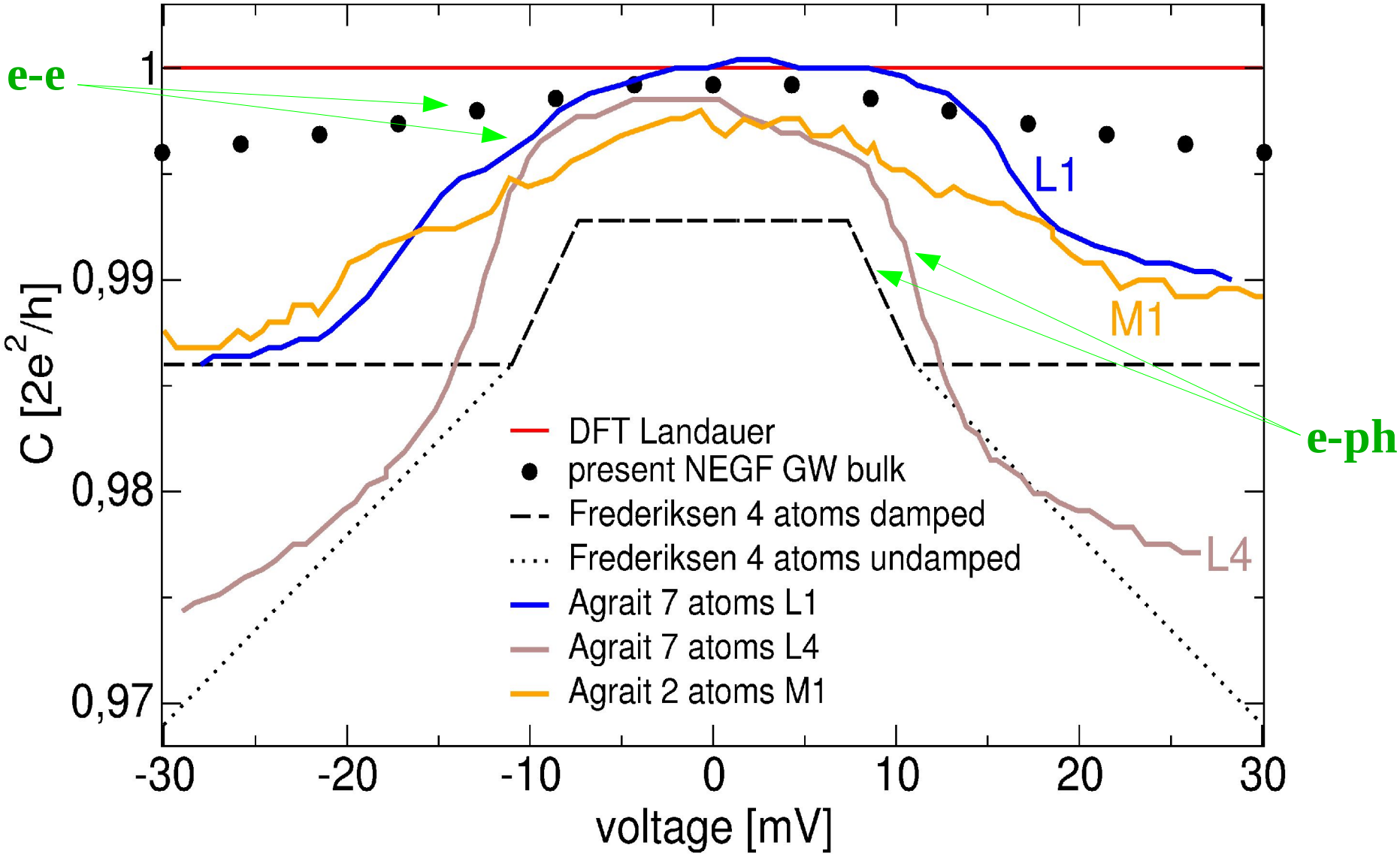
e-e scattering everywhere (in conductor and leads)





- No contact resistance (small increase in the central part)
- **Appearance of Satellite Conductance Channels**

C / V characteristics: GW vs EXP



Conclusions & Perspectives

GW electron-electron correlations in Quantum transport on Au chain

Effects :

- Static real part of the Self-energy (DFT+GW+LB) $\Rightarrow$ Modification of the Conductance profile
- Non-hermitean part of the Self-energy (GW-NEGF) $\Rightarrow$ Loss-of-coherence, dissipative effects
- Full dynamical Self-energy (GW-NEGF) $\Rightarrow$ Appearance of satellites

Au chain: Equilibrium G^0W^0 in good agreement with the smooth drop observed in the Experiment

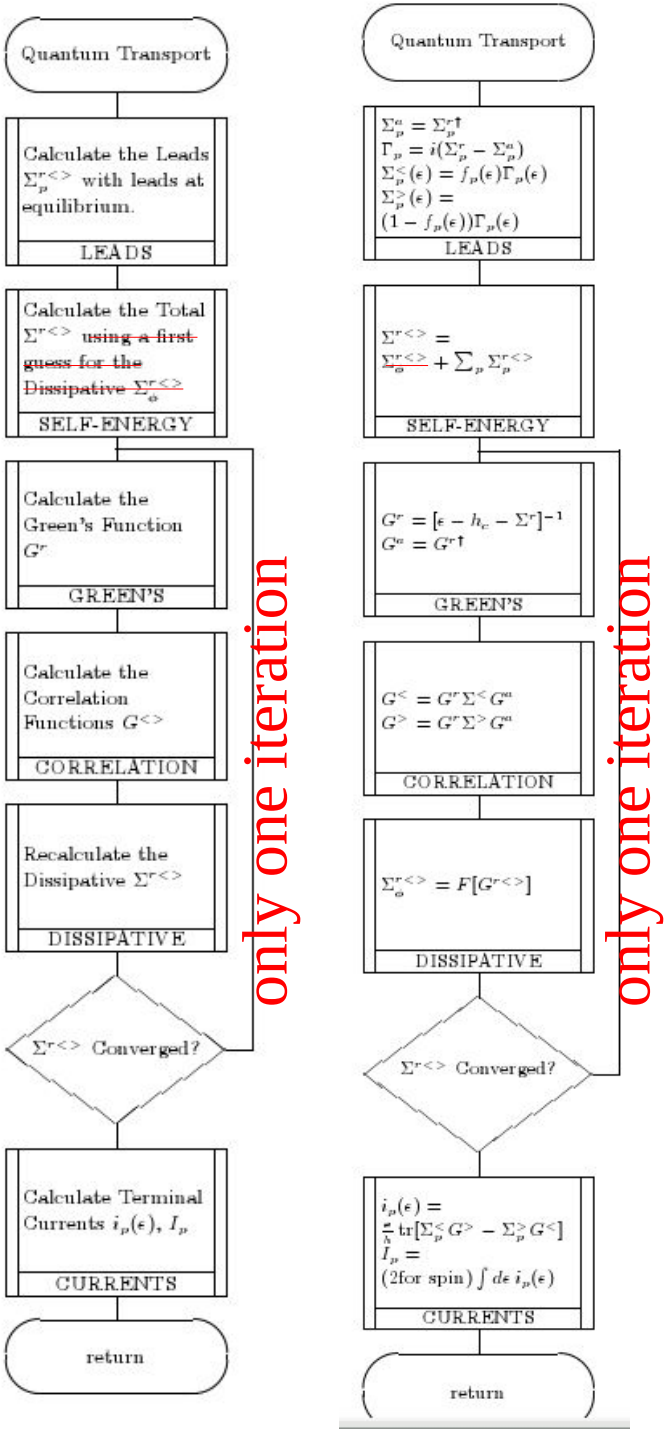
Perspectives

=>Self-consistent calculation of $G^<$ $G^>$

=>Out of Equilibrium (Finite bias)

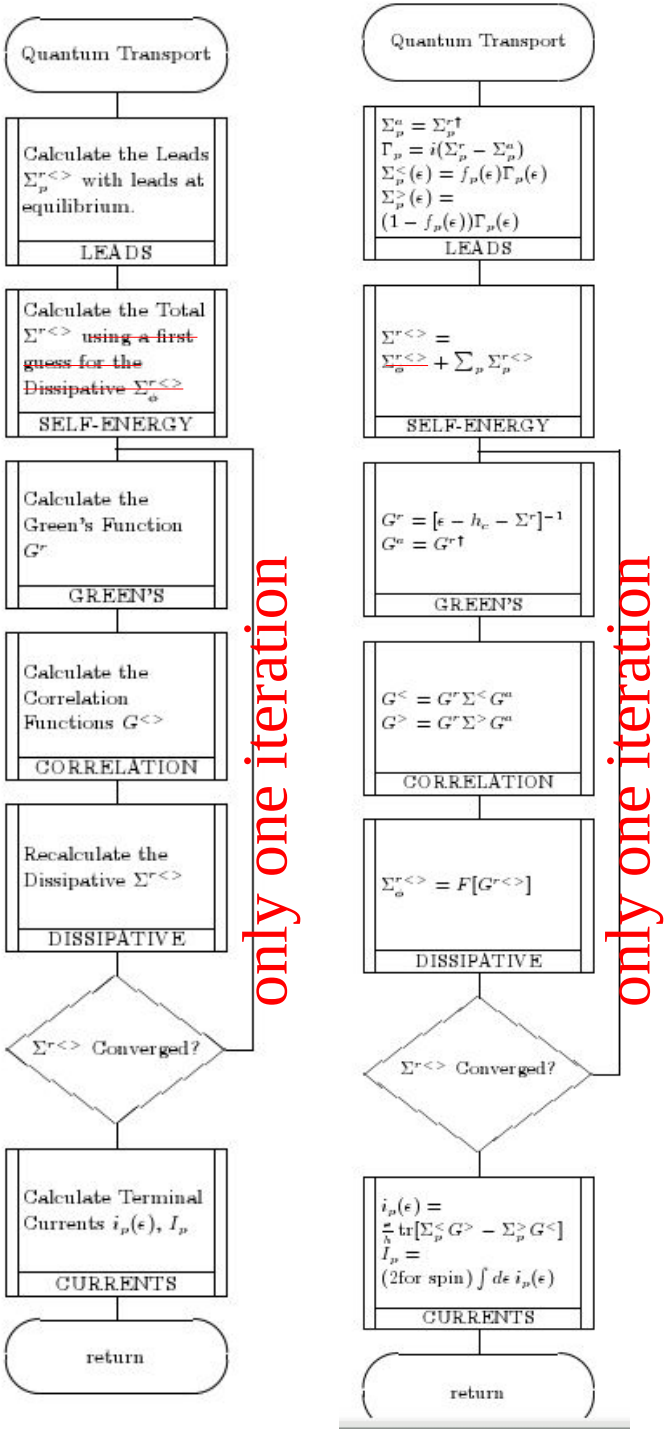
Next Attempts

- We first solve the fundamental NEGF equations, going out of equilibrium;
- Then we calculate the GW self-energy and scattering functions;
- We solve then directly the NEGF current equation (not a modified Landauer-Buttiker equation).



Future Attempts

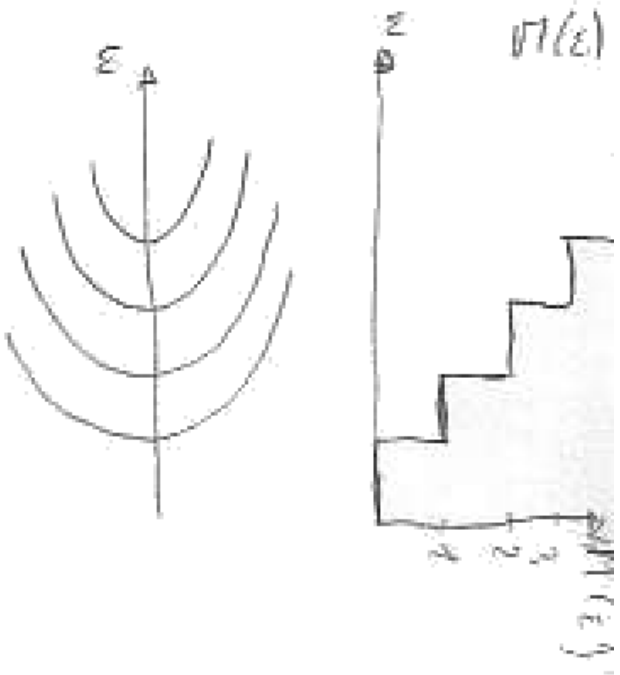
- Full NEGF self-consistent code, implementing GW or even beyond.



Future Projects and Perspectives

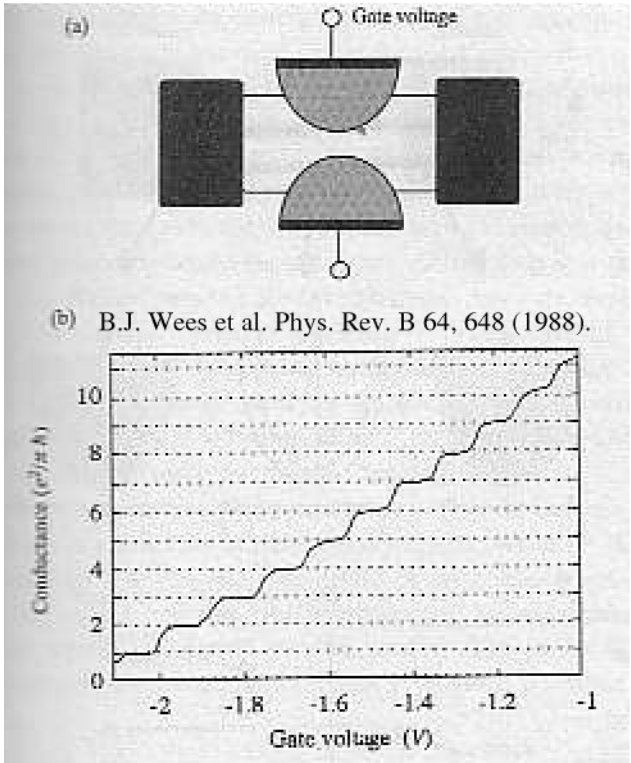
- NEGF seems to us to be the only framework for QT.
- Provided a well established localized basis is provided once and for all, next NEGF steps should be not such time-consuming and straightforward.
- Non-Equilibrium as first iterations should be feasible cheaply;
- In going toward Self-Consistent NEGF, DFT can be really considered as first guess or to provide the atomic structure. And we can even start from H0, neglecting exchange and correlation at the beginning.
- A fully NEGF self-consistent code, implementing the provided flow diagram, would be a valuable tool for QT.

The Number of Modes *M*

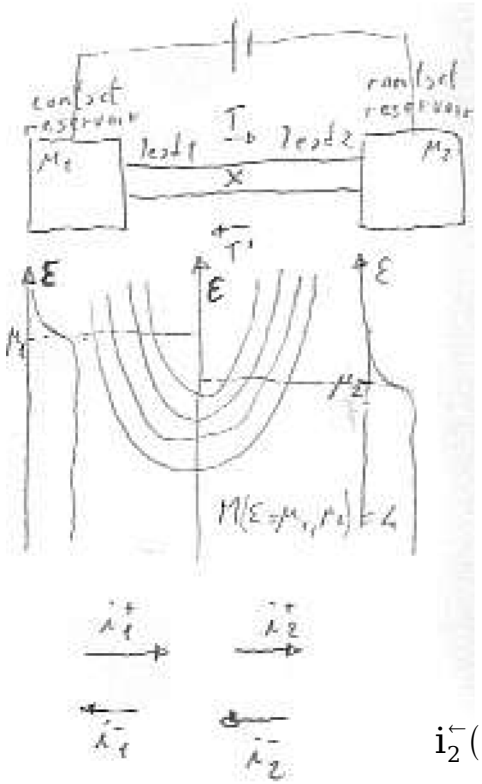


M(ε) counts the number of Modes available at a given energy.

The conductance somehow depends on *M(ε)*, and this is demonstrated by the famous devil-stair experiment showing the quantization of *C* when reducing the number of modes by a gate voltage.



Landauer Formula



$$i_1^{\rightarrow}(\epsilon)=\frac{2e}{h}M(\epsilon)f_1(\epsilon)$$

$$i_2^{\leftarrow}(\epsilon)=\frac{2e}{h}M(\epsilon)f_2(\epsilon)$$

$$i_1^{\leftarrow}(\epsilon)=\frac{2e}{h}[1-T(\epsilon)]i_1^{\rightarrow}(\epsilon)+T(\epsilon)i_2^{\leftarrow}(\epsilon)$$

$$i_2^{\rightarrow}(\epsilon)=\frac{2e}{h}T(\epsilon)i_1^{\rightarrow}(\epsilon)+[1-T(\epsilon)]i_2^{\leftarrow}(\epsilon)$$

$$i(\epsilon)=i_1^{\rightarrow}(\epsilon)-i_1^{\leftarrow}(\epsilon)=\frac{2e}{h}M(\epsilon)T(\epsilon)[f_1(\epsilon)-f_2(\epsilon)]$$

Total Current

Transmission Function

$$I=\int d\epsilon\, i(\epsilon)=\frac{2e}{h}\int d\epsilon\, M(\epsilon)T(\epsilon)[f_1(\epsilon)-f_2(\epsilon)]$$ Landauer Formula

Landauer Formula in Linear Response

1) T and M are constant in the bias range
2) The Bias is small

$$I = \frac{2e}{h} M T [\mu_1 - \mu_2]$$ The Current is linear in the bias

Conductance

$$C(\epsilon) = \frac{2e^2}{h} \bar{T}(\epsilon)$$ Landauer Formula

$$\bar{T}(\epsilon) = M(\epsilon) T(\epsilon)$$ ← How to calculate T?
Quantum Mechanics
scattering matrix approach or ...

Landauer Formula in Green's functions

Conductance

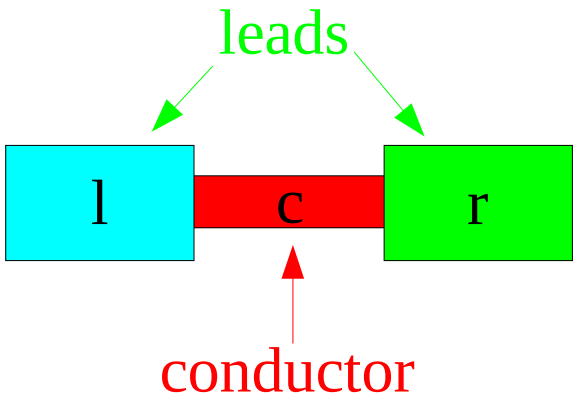
$$C(\epsilon) = 2\frac{e^2}{h}\bar{T}(\epsilon)$$ Landauer Formula

conductor Green's functions
(**a**dvanced and **r**etarded)

$$\bar{T} = \mathrm{tr}\left[\Gamma_l G_c^r \Gamma_r G_c^a\right]$$ Transmittance

conductor-lead
(**l**eft and **r**ight) coupling

The three-partitioned model



$$H=\begin{pmatrix} H_l & -H_{lc} & 0 \\ -H_{lc}^{\dagger} & H_c & -H_{cr} \\ 0 & -H_{cr}^{\dagger} & H_r \end{pmatrix}$$

$$H(r)=-\frac{\hbar^2}{2m}\partial_r+u(r)$$

$$t=\frac{\hbar^2}{2ma^2}$$

Real Space Representation
or on localized basis

$$H_{ij}=\begin{pmatrix} \ddots & -t & 0 & 0 & 0 \\ -t^{\dagger} & u_{-1}+2t & -t & 0 & 0 \\ 0 & -t^{\dagger} & u_0+2t & -t & 0 \\ 0 & 0 & -t^{\dagger} & u_1+2t & -t \\ 0 & 0 & 0 & -t^{\dagger} & \ddots \end{pmatrix}$$

Electron-Phonon Self-Consistent Born Approximation

$$\Sigma^{<}_{e-ph}(r,r',\epsilon)=\int d\omega D(r,r',\omega)G^{<}(r,r',\epsilon-\omega)$$

$$\Sigma^{>}_{e-ph}(r,r',\epsilon)=\int d\omega D(r,r',\omega)G^{<}(r,r',\epsilon+\omega)$$

$$D(r,r',\omega)=\sum_q U_q^2[N_q e^{-iq(r-r')}\delta(\omega-\omega_q)+(N_q+1)e^{+iq(r-r')}\delta(\omega+\omega_q)]$$

potential felt by an electron due to a single phonon at q

number of phonons at q (if at equilibrium, then Bose-Einstein)

Current Flow

$$i_p(\epsilon) = \frac{e}{h} \mathrm{tr}\left[\Sigma_p^{<} G^{>} - \Sigma_p^{>} G^{<}\right]$$

current flowing through lead p

rate at which electrons are scattered from lead p **in** i

probability that a state i is empty

This reduces to the ordinary Landauer-Buttiker formula when there is no e-e or e-ph scattering, that is the flow is coherent.

$$i_p^{coh}(\epsilon) = \frac{e}{h} \mathrm{tr}\left[\Gamma_p G^r \Gamma_q G^a - \Gamma_p G^r \Gamma_q G^a\right](f_p - f_q)$$