Theory and DFT simulations of spin-polarized transport

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Outline

Theoretical background for electron transport

– Non-Equilibrium Green"s Functions (NEGF)

– relation of NEGF to the Landauer-Büttiker formalism: conductance=transmission

– NEGF for different system partitionings:
4-parts (STM, Cesar lecture)
5-parts (single molecule geometry)

– spin-polarized transport

– NEGF with Density functional theory (DFT)

Selected results on spin-polarized transport in molecular junctions

– perfect spin-filtering by orbital mismatch of wave functions

– efficient spin-filtering by quantum interference

– spin-orbit torque exerted on a magnetic molecule

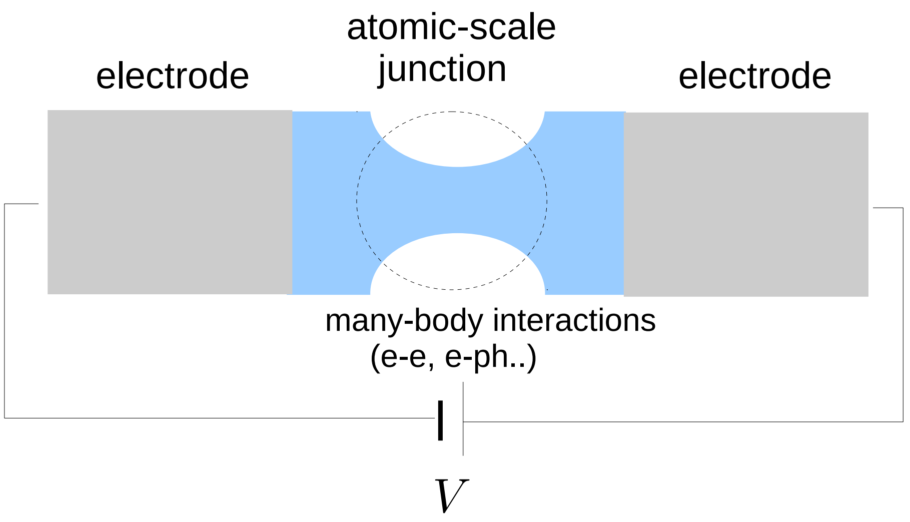
Bibliography

[1] Elke Scheer (Auteur), Juan Carlos Cuevas, «Molecular Electronics: An Introduction To Theory And Experiment», 2017

[2] Mahdi Pourfath, «The Non-Equilibrium Green's Function Method for Nanoscale Device Simulation», 2014

[3] Pier A. Mello, Narendra Kumar, «Quantum Transport in Mesoscopic Systems: Complexity and Statistical Fluctuations», 2004

Charge transport: set up the problem



Calculate current $I(V)$

General framework — Non-Equilibrium Green“s Functions (NEGF) **(Ref. [1,2])**

[Kadanoff and Baym, and independently by Keldysh in the early 1960’s]

↓ Elastic (coherent) regime, no many-body interactions

Landauer-Büttiker formula for elastic conductance:

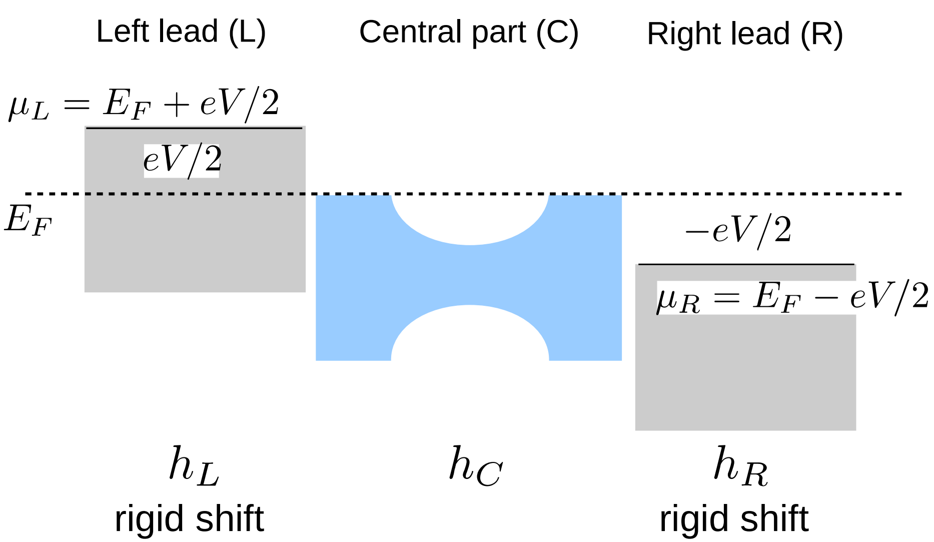
$$G = I/\delta V = G_0 T(E_F)$$

Where $T(E_F)$ is the total electron transmission at the Fermi energy;

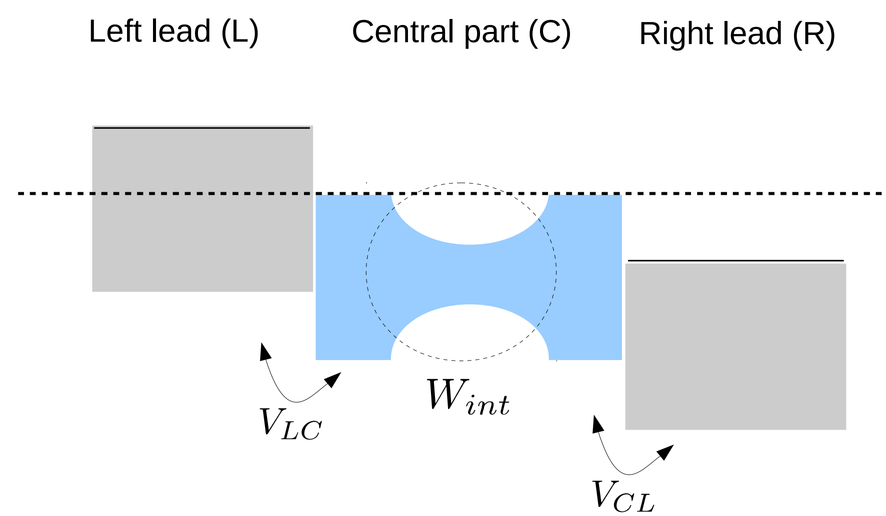
$G_0 = e^2/h$ is the conductance quantum (per spin).

NEGF: short motivation

Disconnected junction



Connected junction



Tight-binding like Hamiltonian:

$$h = h_L + h_C + h_R = \sum_{mn} V_{mn} c_m^\dagger c_n$$

$\{m, n\}$ are localized orbitals (like *s,p,d,..*)

$$\mathcal{H} = h + [V_{LC} + V_{RC} + h.c.] + W_{int}$$

perturbation $= V + W_{int}$

- V: couplings
- W_{int}: many-body interactions

Known state ψ → Unknown state Ψ

?

$$\psi = |\psi_L\rangle \otimes |\psi_C\rangle \otimes |\psi_R\rangle$$

- $|\psi_L\rangle$: Fermi see: μ_L
- $|\psi_C\rangle$: ground state
- $|\psi_R\rangle$: Fermi see: μ_R

NEGF: short motivation

Mean value of any observable $\hat{O}$: $\quad O(t) = \left\langle \Psi \left| \hat{O}_{\mathcal{H}}(t) \right| \Psi \right\rangle; \quad \hat{O}_{\mathcal{H}}(t) = e^{\frac{i}{\hbar}\hat{\mathcal{H}}t} \hat{O} e^{-\frac{i}{\hbar}\hat{\mathcal{H}}t}$

Adiabatic switching on interactions:

$$|\Psi\rangle = \hat{S}(0,-\infty)\,|\psi\rangle$$

with the evolution operator:

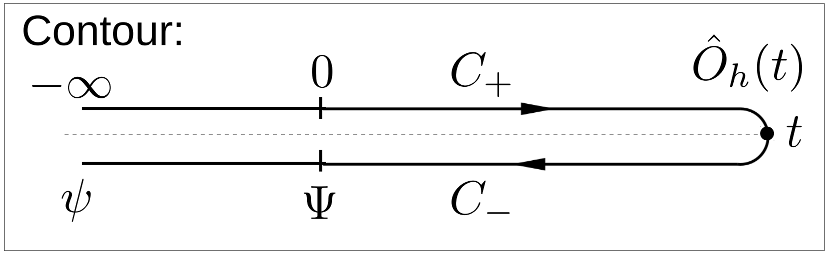
$$\hat{S}(t_2,t_1) = \mathcal{T}_t \left\{ \exp\left(-\frac{i}{\hbar} \int_{t_1}^{t_2} \delta\hat{h}_I(t)dt \right) \right\}$$

+

$$\hat{O}_{\mathcal{H}}(t) = \hat{S}(0,t)\hat{O}_h(t)\hat{S}(t,0)$$

$$\hat{O}_h(t) = e^{\frac{i}{\hbar}\hat{h}t} \hat{O} e^{-\frac{i}{\hbar}\hat{h}t}$$

$$O(t) = \left\langle \psi \left| \hat{S}(-\infty,t)\hat{O}_h(t)\hat{S}(t,-\infty) \right| \psi \right\rangle$$



Four main Green functions are needed due to two branches on the contour:

$$i\hbar G^{>}_{mn}(t_2,t_1) = \left\langle \Psi \left| \hat{c}_{m,\mathcal{H}}(t_2)\hat{c}^{\dagger}_{n,\mathcal{H}}(t_1) \right| \Psi \right\rangle, \quad \text{or } G^{-+}_{mn}(t_2,t_1)$$ – greater

$$i\hbar G^{<}_{mn}(t_2,t_1) = -\left\langle \Psi \left| \hat{c}^{\dagger}_{n,\mathcal{H}}(t_1)\hat{c}_{m,\mathcal{H}}(t_2) \right| \Psi \right\rangle, \quad \text{or } G^{+-}_{mn}(t_2,t_1)$$ – lesser

$$i\hbar G^{r}_{mn}(t_2,t_1) = \theta(t_2-t_1) \left\langle \Psi \left| \left[\hat{c}_{m,\mathcal{H}}(t_2)\hat{c}^{\dagger}_{n,\mathcal{H}}(t_1) \right]_+ \right| \Psi \right\rangle$$ – retarded

$$i\hbar G^{a}_{mn}(t_2,t_1) = -\theta(t_1-t_2) \left\langle \Psi \left| \left[\hat{c}_{m,\mathcal{H}}(t_2)\hat{c}^{\dagger}_{n,\mathcal{H}}(t_1) \right]_+ \right| \Psi \right\rangle$$ – advanced

Simple example: noninteracting electrode GFs

$$\hat{h} = \sum_{mn} t_{mn} \hat{c}_m^\dagger \hat{c}_n = \sum_k \epsilon_k \hat{c}_k^\dagger \hat{c}_k$$

$$\hat{c}_k(t) = \hat{c}_k e^{-\frac{i}{\hbar}\varepsilon_k t}$$

In time domain:

$$i\hbar g_k^>(t_2,t_1) = [1 - f(\varepsilon_k)] e^{\frac{i}{\hbar}\varepsilon_k(t_1-t_2)}$$

$$i\hbar g_k^<(t_2,t_1) = -f(\varepsilon_k)\ e^{\frac{i}{\hbar}\varepsilon_k(t_1-t_2)}$$

$$i\hbar g_k^r(t_2,t_1) = \theta(t_2 - t_1)\ e^{\frac{i}{\hbar}\varepsilon_k(t_1-t_2)}$$

$$i\hbar g_k^a(t_2,t_1) = -\theta(t_1 - t_2)\ e^{\frac{i}{\hbar}\varepsilon_k(t_1-t_2)}$$

Fourier transform to time domain:

$$g_k^>(E) = -2\pi i\ [1 - f(\varepsilon_k)]\delta(E - \varepsilon_k)$$

$$g_k^<(E) = 2\pi i\ f(\varepsilon_k)\delta(E - \varepsilon_k)$$

$$g_k^{r/a}(E) = \frac{1}{E \pm i\eta - \varepsilon_k}$$

where Fermi-Dirac distribution: $f(E) = [1 + e^{(E-\mu)/kT}]^{-1}$

In any basis too:

$$g^>(E) = [1 - f(E)]\{g^r(E) - g^a(E)\}$$

$$g^<(E) = -f(E)\{g^r(E) - g^a(E)\}$$

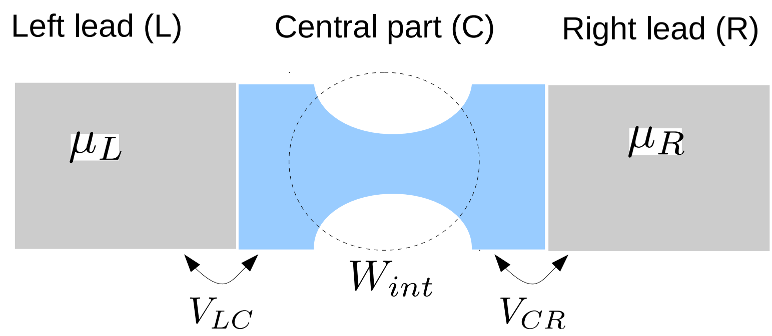
$$g^{r/a}(E) = \frac{1}{E \pm i\eta - h}$$

Well-known relation with Cauchy principal part:

$$\frac{1}{E \pm i\eta - \varepsilon_k} = \mathrm{P}\left(\frac{1}{E - \varepsilon_k}\right) \mp i\pi\delta(E - \varepsilon_k)$$

NEGF: general expression for electron current

$$\hat{N}_L = \sum_{j \in L} c_j^\dagger c_j$$



Starting with:
$$I_L = -e \frac{d \left\langle \hat{N}_{L,\mathcal{H}}(t) \right\rangle}{dt}$$

Final expression:

$$I_L = \frac{e}{h} \int dE \; \mathrm{Tr}[\Sigma_L^< G_{CC}^> - \Sigma_L^> G_{CC}^<]$$

$$G_{CC}^{</>} = G_{CC}^r [\Sigma_L^{</>} + \Sigma_R^{</>} + \Sigma_{int}^{</>}] G_{CC}^a$$
$$G_{CC}^{r/a} = [(E - H_{CC} - \Sigma_L^{r/a} - \Sigma_R^{r/a} - \Sigma_{int}^{r/a})]^{-1}$$

Dyson equation:
$$G^{r/a} = g^{r/a} + g^{r/a} V G^{r/a} + g^{r/a} \Sigma_{int}^{r/a} G^{r/a}$$

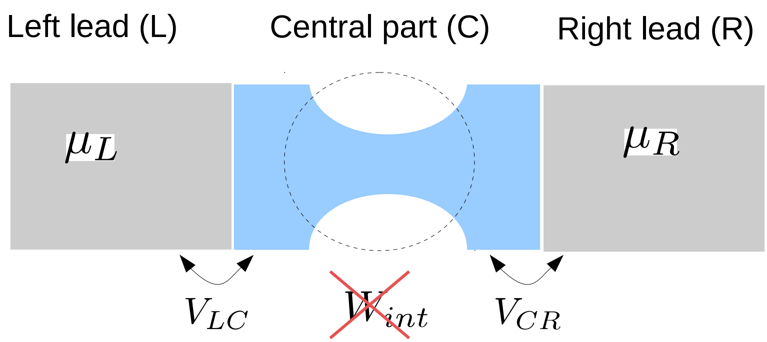
Contact self-energies:

$$\Sigma_L^{r/a}(E) = V_{CL} g_{LL}^{r/a}(E) V_{LC}$$
$$\Sigma_L^<(E) = V_{CL} g_{LL}^<(E) V_{LC} = -f_L(E) V_{CL} [g_{LL}^r(E) - g_{LL}^a(E)] V_{LC} = i f_L(E) \Gamma^L(E)$$
$$\Sigma_L^>(E) = V_{CL} g_{LL}^>(E) V_{LC} = -i[1 - f_L(E)] \Gamma^L(E)$$

With coupling matrices defined as: $\Gamma^L = i(\Sigma_L^r - \Sigma_L^a)$

NEGF: coherent (elastic) regime

$$W_{int} = 0$$



If no interactions are present in the Central part (elastic or coherent regime): $\Sigma_{int}^{<,>,r,a} = 0$

$$I_L = \frac{e}{h}\int dE \ \mathrm{Tr}\Big[i f_L(E)\Gamma^L(E)G^r(E)\Big\{ -i[1-f_L(E)]\Gamma^L(E) - i[1-f_R(E)]\Gamma^R(E)\Big\}G^a(E)$$

$$+ i[1-f_L(E)]\Gamma^L(E)G^r(E)\Big\{ i f_L(E)\Gamma^L(E) + i f_R(E)\Gamma^R(E)\Big\}G^a(E)\Big]$$

$$= \frac{e}{h}\int [f_L(E) - f_R(E)]\cdot \mathrm{Tr}[\Gamma^L(E)G^r_{CC}(E)\Gamma^R(E)G^a_{CC}(E)]dE$$

Denoting $\mathrm{Tr}[\Gamma^L(E)G^r_{CC}(E)\Gamma^R(E)G^a_{CC}(E)] = T(E)$

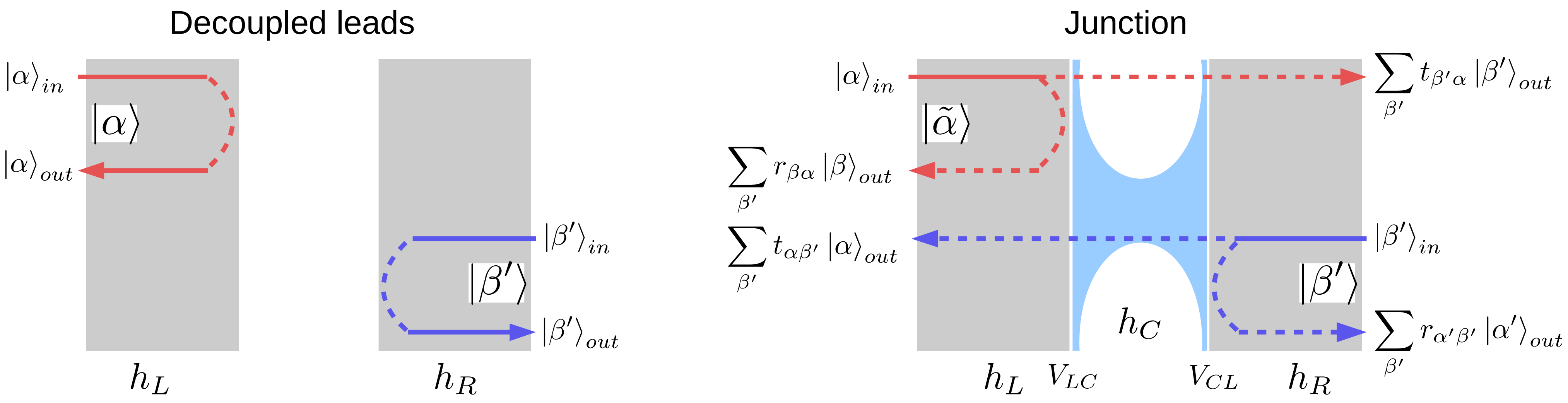
We arrive at:

$$\boxed{I_L = \frac{e}{h}\int [f_L(E) - f_R(E)]T(E)dE}$$

Transmission function ?

Scattering problem: Landauer-Büttiker formalism

At fixed energy *E*:



$\alpha = 1, ..N_L$ $\qquad$ $\beta' = 1, ..N_R$

$N_{L/R}$ – number of channels = bands crossing the energy *E*

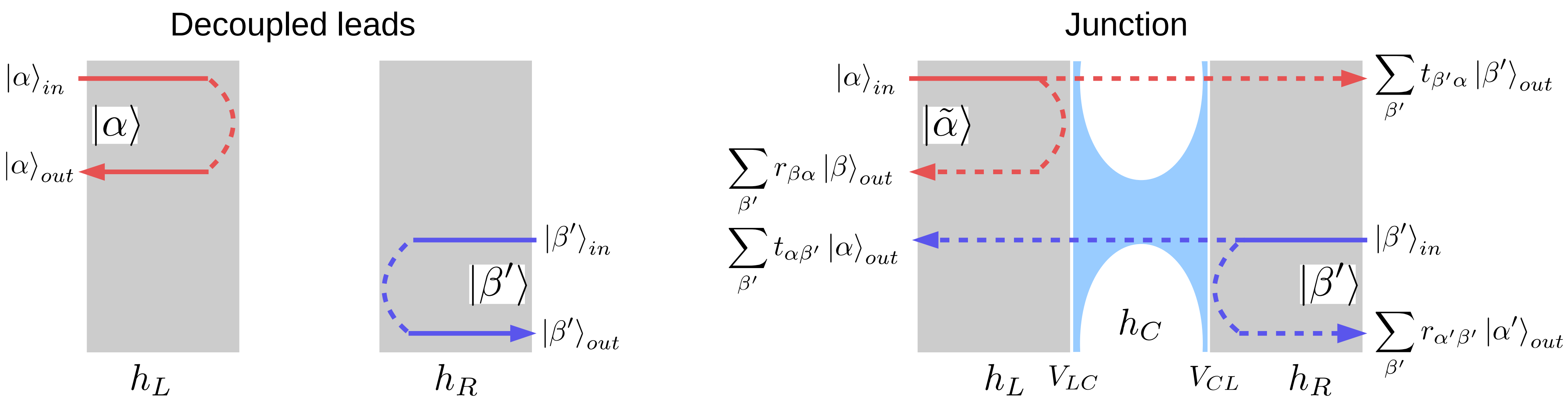
$|\rangle_{in/out}$ are normalized to carry the unit current

When the coupling is switched on:

$$\{\alpha, \beta'\} \rightarrow \{\tilde{\alpha}, \tilde{\beta}'\}$$

Energy-dependent transmission is defined by:

$$T_{L\to R} = \sum_{\beta'\alpha} |t_{\beta'\alpha}|^2 = \sum_{\alpha\beta'} |t_{\alpha\beta'}|^2 = T_{L\leftarrow R} = T(E)$$



$$\{\alpha, \beta'\} \Longrightarrow \{\tilde{\alpha}, \tilde{\beta}'\}$$

Lippmann-Schwinger equation **(Ref. [3])**:

$$|\tilde{\alpha}\rangle = |\alpha\rangle + g^r(E)V|\tilde{\alpha}\rangle\,; \quad V = V_{LC} + V_{RC} + h.c.$$

and GFs for decoupled system are:

$$g^r(E) = [E + i\delta - h]^{-1}; \quad h = h_L + h_C + h_R$$

Iterating:

$$|\tilde{\alpha}\rangle = |\alpha\rangle + g^r(E)V|\alpha\rangle + g^r(E)V\underbrace{\left[g^r(E) + g^r(E)Vg^r(E) + ...\right]}_{\text{full } G^r(E)}V|\alpha\rangle$$

and therefore:

$$|\tilde{\alpha}\rangle = |\alpha\rangle + g^r(E)V\,|\alpha\rangle + g^r(E)VG^r(E)V\,|\alpha\rangle$$

In the decoupled leads GFs can be coveniently written in terms of asymptotic states as **(Ref. [3])**:

$$g^r_L(E) = -2\pi i \sum_{\alpha=1}^{N_L} |\alpha\rangle_{out}\,\langle\alpha|\,; \quad g^r_R(E) = -2\pi i \sum_{\beta'=1}^{N_R} |\beta'\rangle_{out}\,\langle\beta'|$$

so that the scattering wave in Leads, originated from $|\alpha\rangle$, take the following form:

$$|\alpha\rangle \to |\alpha\rangle - 2\pi i \sum_{\beta} |\beta\rangle^{out} \sum_{m,n\in C} V^L_{\beta m} G^r_{mn} V^L_{n\alpha} - 2\pi i \sum_{\beta'} |\beta'\rangle^{out} \sum_{m.n\in C} V^R_{\beta' m} G^r_{mn} V^L_{n\alpha}$$

with $V^L_{\beta m} = \langle\beta|V_{LC}|m\rangle$ and $V^R_{\beta' m} = \langle\beta'|V_{RC}|m\rangle$

It follows therefore that reflection and transmission amplitudes are given by:

$$\boxed{r_{\beta\alpha} = \delta_{\beta\alpha} - 2\pi i \sum_{m.n\in C} V^L_{\beta m} G^r_{mn} V^L_{n\alpha}; \quad t_{\beta'\alpha} = -2\pi i \sum_{m,n\in C} V^R_{\beta' m} G^r_{mn} V^L_{n\alpha}}$$

hopping $C \to R$ — propagation $C \to C$ — hopping $L \to C$

Finally, the coupling matrices in the diagonal α, β' basis take form:

$$\Gamma^L_{mn} = 2\pi \sum_{\alpha} V^L_{m\alpha} V^L_{\alpha n}; \quad \Gamma^R_{mn} = 2\pi \sum_{\alpha'} V^R_{m\alpha'} V^R_{\alpha' n}$$

So we now arrive at the final expression for the Trace:

$$\mathrm{Tr}[\Gamma^L G^r_{CC}\Gamma^R G^a_{CC}] = 4\pi^2 \sum_{iklm,\alpha\alpha'} V^L_{i\alpha}V^L_{\alpha k}G^r_{kl}V^R_{l\alpha'}V^R_{\alpha' m}G^a_{mi}$$

$$= 4\pi^2 \sum_{iklm,\alpha\alpha'} V^L_{\alpha k}G^r_{kl}V^R_{l\alpha'}V^{L*}_{\alpha i}G^{r*}_{im}V^{R*}_{m\alpha'} = \sum_{\alpha\alpha'}|t_{\alpha\alpha'}|^2 = T_{L\leftarrow R} = T_{L\rightarrow R} = T$$

Keeping in mind:

$$t_{\beta'\leftarrow\alpha} = t_{\beta'\alpha} = -2\pi i \sum_{m,n\in C} V^R_{\beta' m}G^r_{mn}V^L_{n\alpha}; \qquad t_{\alpha\leftarrow\beta'} = t_{\alpha\beta'} = -2\pi i \sum_{m,n\in C} V^L_{\alpha m}G^r_{mn}V^R_{n\beta'}$$

$$\mathrm{Tr}[\Gamma^L G^r_{CC}\Gamma^R G^a_{CC}] = T$$

Landauer formula for electric current:

$$I = e/h \int T(E)[f_L - f_R]dE; \qquad f_{L/R}(E) = \frac{1}{1+e^{(E-\mu_{L/R})/kT}}$$

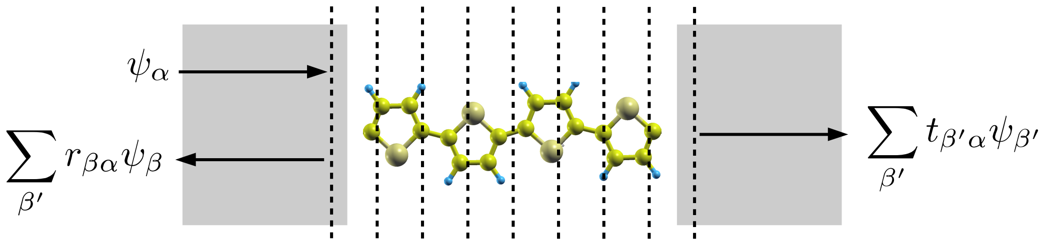
with otal transmission $T(E) = \sum_{\beta'\alpha}|t_{\beta'\alpha}|^2$

In the linear regime (small bias):

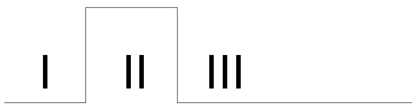
$$G = I/\delta V = G_0 T(E_F)$$

where $G_0 = e^2/h$ is the conductance quantum (per spin).

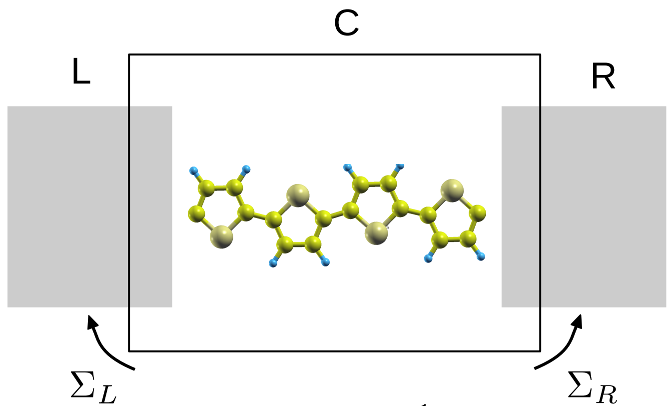
– Scattering approach, wave function matching



$$T = \sum_{\beta'\alpha} |t_{\beta'\alpha}|^2$$



– Non-Equilibrium Green Functions (NEGF)



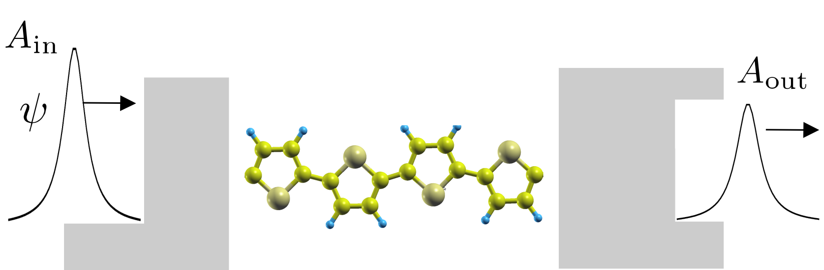
$$G_{CC} = \frac{1}{E - h_C - \Sigma_L - \Sigma_R}$$

$$T = \mathrm{Tr}[\Gamma^L G^r_{CC} \Gamma^R G^a_{CC}]$$

$$\Gamma^{L/R} = i(\Sigma^r_{L/R} - \Sigma^a_{L/R})$$

$\Sigma_{L/R}$ – contact self-energies

– Wave packet propagation

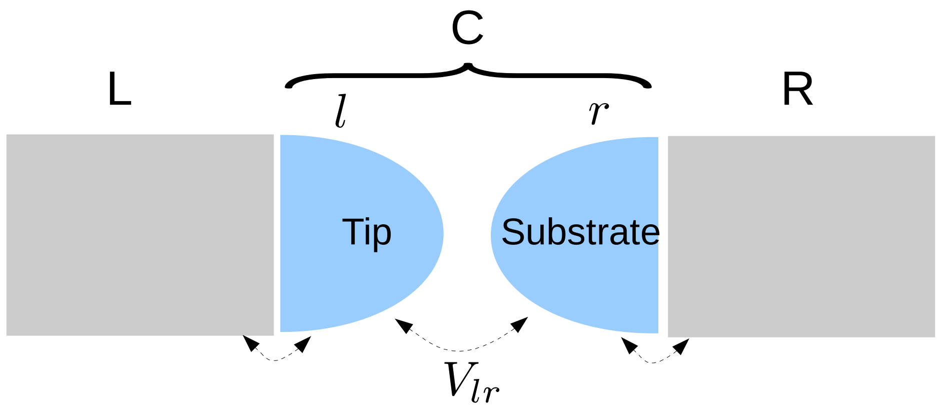


$$T \approx A_{out}/A_{in}$$

Time evolution with Schrodinger equation:

$$i\hbar\frac{\partial\psi}{\partial t} = \hat{H}\psi$$

few el. wave packets / fs $(\sim 1/h)$

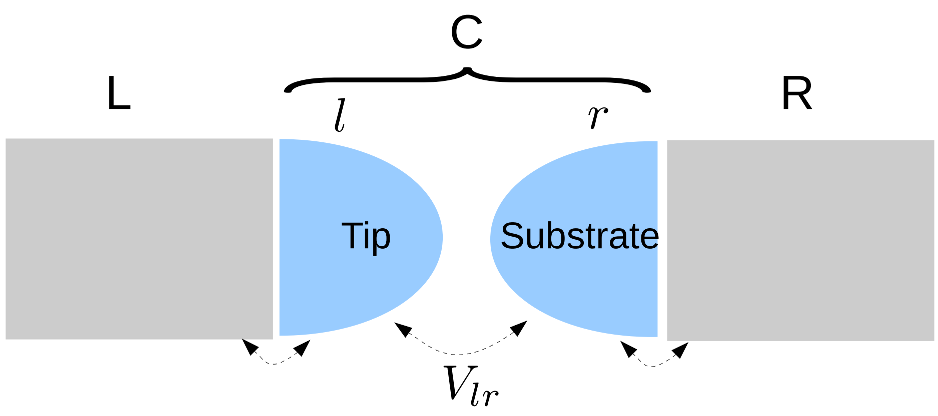


$$V_{Lr} = 0$$

$$V_{lR} = 0$$

Perturbation = V_{lr}

NEGF: 4 parts division, relevant for STM



$$V_{Lr} = 0$$

$$V_{lR} = 0$$

Perturbation = V_{lr}

$$T = \mathrm{Tr}\left[\Gamma^L G^r_{CC} \Gamma^R G^a_{CC}\right] = \mathrm{Tr}\left[\Gamma^L_l G^r_{lr} \Gamma^R_r G^a_{rl}\right]$$

Dyson equations: $G_{lr} = g_l V_{lr} G_{rr}; \quad G_{rr} = g_r + g_r V_{rl} G_{lr}$

$$G_{lr} = g_l V_{lr} g_r + g_l V_{lr} g_r V_{rl} G_{lr}$$

Expanding iterativly:

$$G_{lr} = g_l V_{lr} g_r + g_l V_{lr} g_r V_{rl} g_l V_{lr} g_r + \ldots = g_l \underbrace{[V_{lr} + V_{lr} g_r V_{rl} g_l V_{lr} + \ldots]}_{\mathcal{T}_{lr}} g_r$$

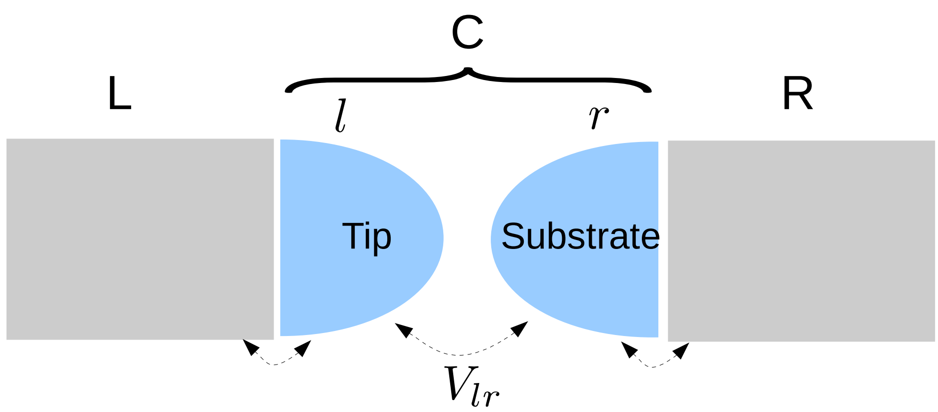
Introduced this way $\mathcal{T}$- matrix can be written as:

$$\mathcal{T}_{lr} = V_{lr} \cdot [1 - g_r V_{rl} g_l V_{lr}]^{-1} = V_{lr} \cdot D_r$$

So we get: $G_{lr} = g_l \mathcal{T}_{lr} g_r$ and similarly: $G_{rl} = g_r \mathcal{T}_{rl} g_l$

which describes propagation from one side to another with renormalized hopping elements

NEGF: 4 parts division (STM set-up)



$$T = \mathrm{Tr}\left[\Gamma_l^L \cdot g_l^r \mathcal{T}_{lr}^r g_r^r \cdot \Gamma_r^R \cdot g_r^a \mathcal{T}_{rl}^a g_l^a\right] = \mathrm{Tr}\left[g_l^a \Gamma_l^L g_l^r \cdot \mathcal{T}_{lr}^r \cdot g_r^r \Gamma_r^R g_r^a \cdot \mathcal{T}_{rl}^a\right]$$

Non-perturbed (by V_{lr}) Green functions are:

$$g_l^{r/a} = [E - h_l - \Sigma_L^{r/a}]^{-1} \longrightarrow i({g_l^a}^{-1} - {g_l^r}^{-1}) = i(\Sigma_L^r - \Sigma_L^a) = \Gamma_l^L$$

Multiply on both sides by $g_l^{a/r}$ and $g_l^{r/a}$:

$$2\pi\rho_l = i(g_l^r - g_l^a) = g_l^a \Gamma_l^L g_l^r = g_l^r \Gamma_l^L g_l^a$$

density of states

Similarly:

$$2\pi\rho_r = i(g_r^r - g_r^a) = g_r^a \Gamma_r^R g_r^r = g_r^r \Gamma_r^R g_r^a$$

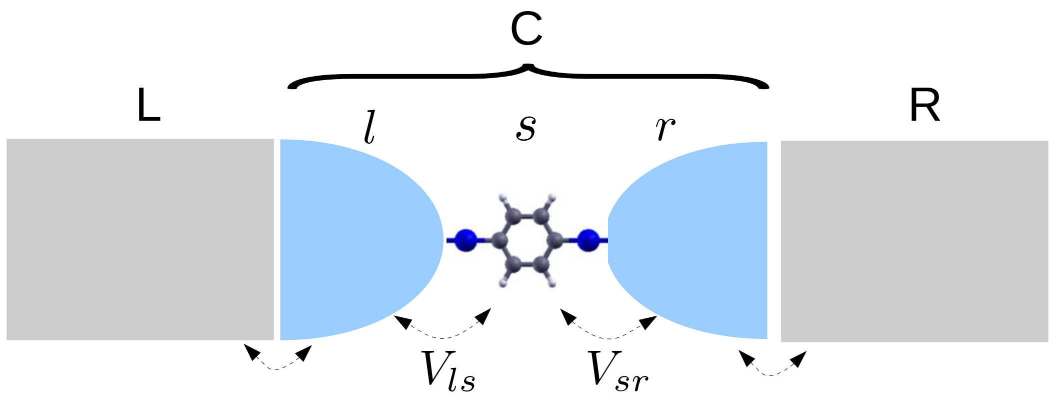
density of states

And finally:

$$T = 4\pi^2 \mathrm{Tr}\left[\rho_l \mathcal{T}_{lr}^r \rho_r \mathcal{T}_{rl}^a\right] = 4\pi^2 \mathrm{Tr}\left[\rho_l V_{lr} D_r^r \rho_r V_{rl} D_l^a\right]$$

In lowest order, $T_{lr}^r = V_{lr},\ T_{rl}^a = V_{rl},$ and we recover the well-known expression.

NEGF: 5 parts (single molecule junctions)

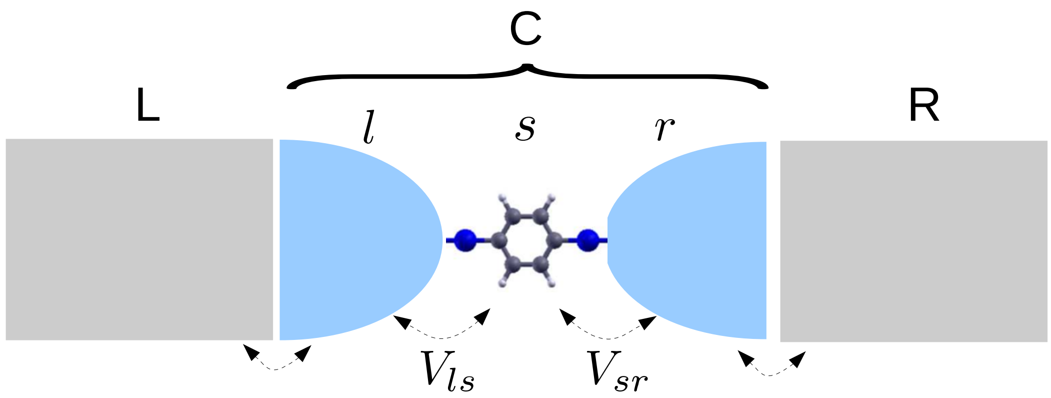


$V_{lr} = 0$

Perturbation = V_{ls}, V_{sr}

The transport goes fully through the active region (s), a molecule, for example

NEGF: 5 parts (single molecule junctions)



$$V_{lr} = 0$$

Perturbation = V_{ls}, V_{sr}

The transport goes fully through the active region (s), a molecule, for example

Again:

$$T = \mathrm{Tr}\left[\Gamma^L G^r_{CC} \Gamma^R G^a_{CC}\right] = \mathrm{Tr}\left[\Gamma^L_l G^r_{lr} \Gamma^R_r G^a_{rl}\right]$$

Dyson equations:

$$G_{lr} = g_l V_{ls} G_{sr}; \quad G_{sr} = G_{ss} V_{sr} g_r$$

$$\downarrow$$

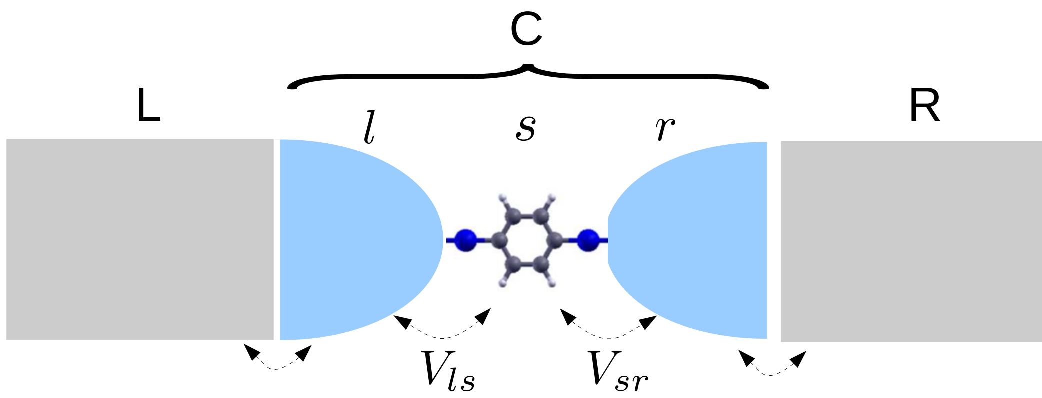
$$G_{lr} = g_l V_{ls} G_{ss} V_{sr} g_r$$

expressed in terms of the GF of the active region, G_{ss}

Therefore:

$$T = \mathrm{Tr}\left[\Gamma^L_l \cdot g^r_l V_{ls} G^r_{ss} V_{sr} g^r_r \cdot \Gamma^R_r \cdot g^a_r V_{rs} G^a_{ss} V_{sl} g^a_l\right]$$

NEGF: 5 parts (single molecule junctions)



$$V_{lr} = 0$$

$$\mathrm{Tr}\left[\Gamma_l^L \cdot g_l^r V_{ls} G_{ss}^r V_{sr} g_r^r \cdot \Gamma_r^R \cdot g_r^a V_{rs} G_{ss}^a V_{sl} g_l^a\right] = \mathrm{Tr}\left[\underbrace{V_{sl} g_l^a \Gamma_l^L g_l^r V_{ls}} \cdot G_{ss}^r \cdot \underbrace{V_{sr} g_r^r \Gamma_r^R g_r^a V_{rs}} \cdot G_{ss}^a\right]$$

Using derived above relations for 4-parts division:

$$V_{sl} g_l^a \Gamma_l^L g_l^r V_{ls} = V_{sl}[i(g_l^r - g_l^a)]V_{ls} = i[\Sigma_l^r - \Sigma_l^a] = \Gamma^l$$

$$V_{sr} g_r^r \Gamma_r^R g_r^a V_{rs} = V_{sr}[i(g_r^r - g_r^a)]V_{rs} = i[\Sigma_r^r - \Sigma_r^a] = \Gamma^r$$

Coupling matrices of the active region

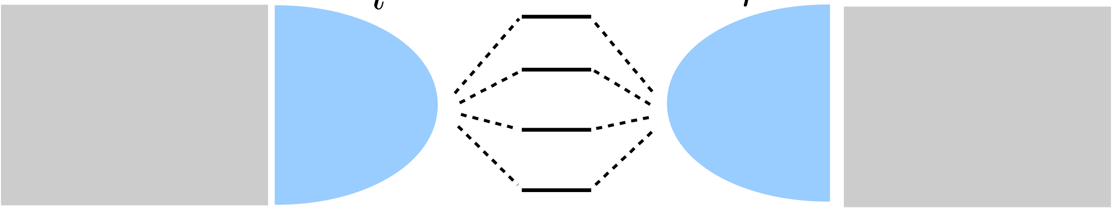
Its GFs are gived by:

$$G_{ss}^{r/a} = [E - h_s - \Sigma_l^{r/a} - \Sigma_r^{r/a}]^{-1}$$

And we arrive finally at:

$$T = \mathrm{Tr}\left[\Gamma^l G_{ss}^r \Gamma^r G_{ss}^a\right]$$

Resonant tunneling via molecular orbitals



$$T = \mathrm{Tr}\left[\Gamma^l G^r_{ss} \Gamma^r G^a_{ss}\right]; \quad G^{r/a}_{ss} = [E - h_s - \Sigma^{r/a}_l - \Sigma^{r/a}_r]^{-1}$$

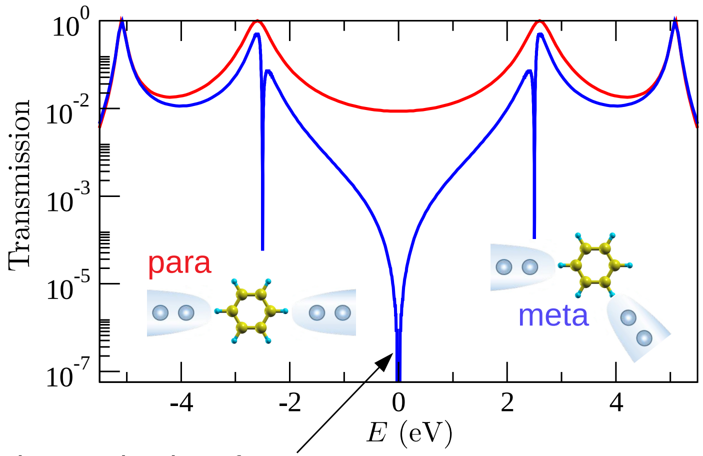
In the basis of molecular orbitals: $h_s = \mathrm{diag}\{\varepsilon_\alpha\}$

Independent coupling: $\Sigma_l = \mathrm{diag}\{\Sigma_{l,\alpha}\}$

Wide-band approximation: $\Sigma^{r/a}_{l,\alpha} = \mp \frac{i\Gamma_{l,\alpha}}{2}$

– similar for the right connection, r

Example: benzene, two connections:



In this case the total transmission takes the form:

$$T = \sum_\alpha T_\alpha; \quad T_\alpha = \frac{\Gamma^l_\alpha \Gamma^r_\alpha}{(E - \varepsilon_\alpha)^2 + (\Gamma^l_\alpha + \Gamma^r_\alpha)^2/4}$$

If symmetric coupling: $T_\alpha = \frac{\Gamma^2_\alpha}{(E - \varepsilon_\alpha)^2 + \Gamma^2_\alpha}$ → $T_\alpha \to 1$ at $E = \varepsilon_\alpha$

resonant tunneling

If non independent couplings, possible interference: → $T \neq \sum_\alpha T_\alpha; \quad T \to 0$ at some energies

Spin-polarized transport

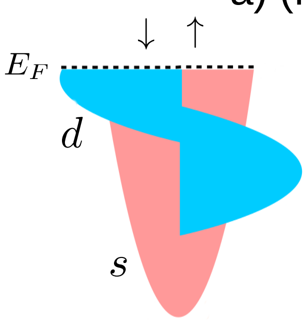
$$T = \mathrm{Tr}\left[\Gamma^l G^r_{ss} \Gamma^r G^a_{ss}\right]; \quad G^{r/a}_{ss} = [E - h_s - \Sigma^{r/a}_l - \Sigma^{r/a}_r]^{-1}$$

$$\Gamma^l = i[\Sigma^r_l - \Sigma^a_l] = V_{sl}[i(g^r_l - g^a_l)]V_{ls} \sim 2\pi |V_{sl}|^2 \rho_l$$ *– similar for* r

Spin polarized transmission, $T_\uparrow \neq T_\downarrow, \quad T = T_\uparrow + T_\downarrow$

Two possible mechanisms:

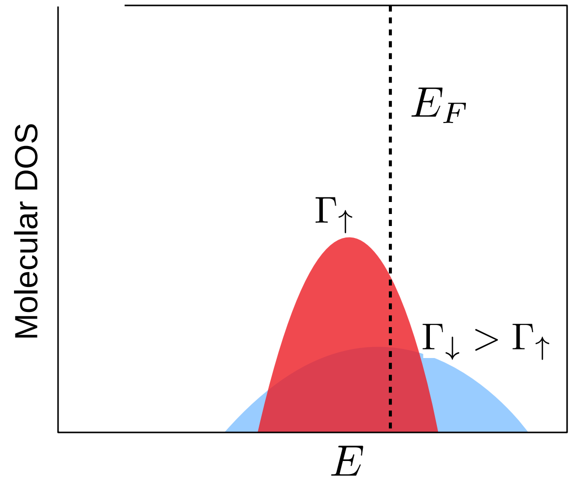
a) (ferro)magnetic electrodes



$\rho_\uparrow \neq \rho_\downarrow \rightarrow$ spin-dependent Γ

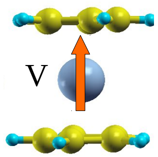
Spin-dependent hybridization

Spin-polarized molecular states

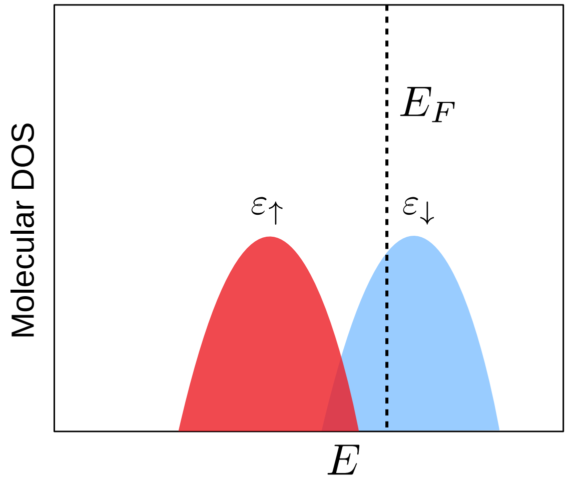


b) magnetic molecule

$h_{s\uparrow} \neq h_{s\downarrow}$



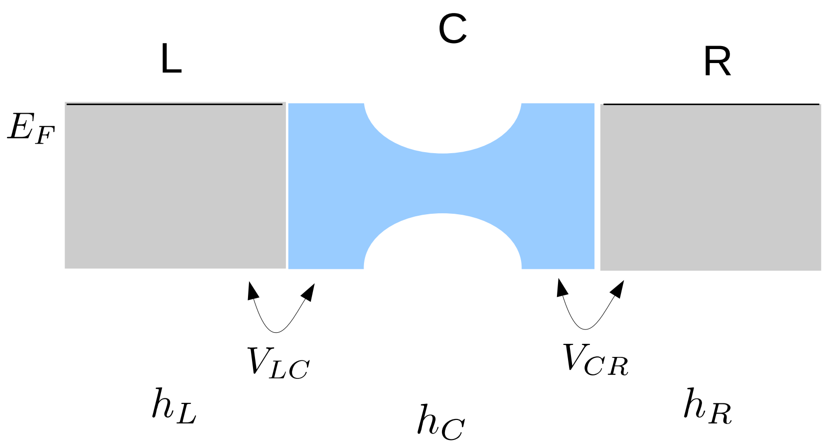
Spin-polarized molecular states



$$T_\uparrow(E_F) \neq T_\downarrow(E_F)$$

$$G = G_\uparrow + G_\downarrow = (e^2/h)[T_\uparrow(E_F) + T_\downarrow(E_F)]$$

DFT calculations for transport: construct Hamiltonians



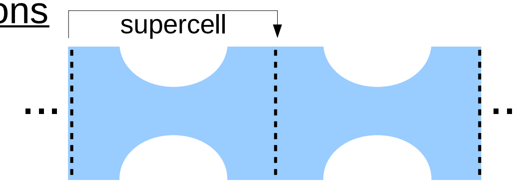
(Spin-dependent) $h_L, h_C, h_R, V_{LC}, V_{CR}$ $-?$

<u>DFT (Density functional theory) calculations in equilibrium state:</u>

– perfect lead calculations

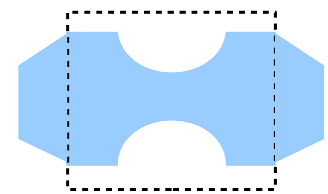
– central scattering region

<u>periodic conditions</u>



or

<u>finite cluster</u>



– Self-consistent Kohn-Sham (KS) equations of DFT:

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V_{\mathrm{KS}}(r)\right]\phi_i(r) = \epsilon_i \phi_i(r)$$

$$V_{\mathrm{KS}}(r) = V(r) + e^2 \int \frac{n(r')}{|r - r'|} d^3r' + V_{\mathrm{xc}}[n](r)$$

$$n_0(r) = \sum_i^{occ.} |\phi_i(r)|^2$$

Hartree potential

exchange-correlation potential of n

Codes for transport simulations

TranSiesta: DFT in localized basis set, NEGF

QuantumATK: DFT in localized basis set, NEGF

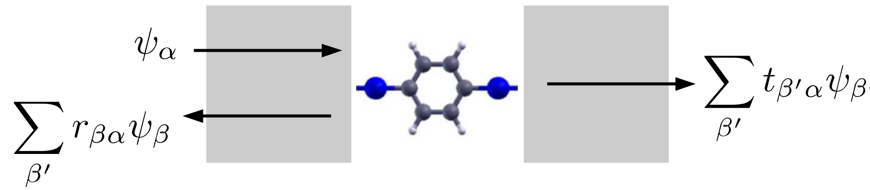
Fireball: DFT in localized basis set, NEGF

Kwant: large-scale tight-binding, scatt. approach and wave packets

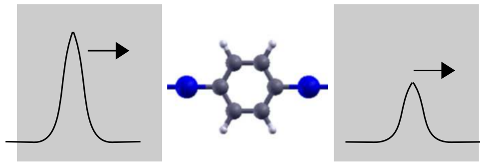
...

Electron transport with Quantum-ESPRESSO (QE) plane wave code:

– PWcond: plane-waves, scattering approach:



– TBcond: «tight-binding», NEGF or wave packets:



Wannier90 code: Hamiltonian

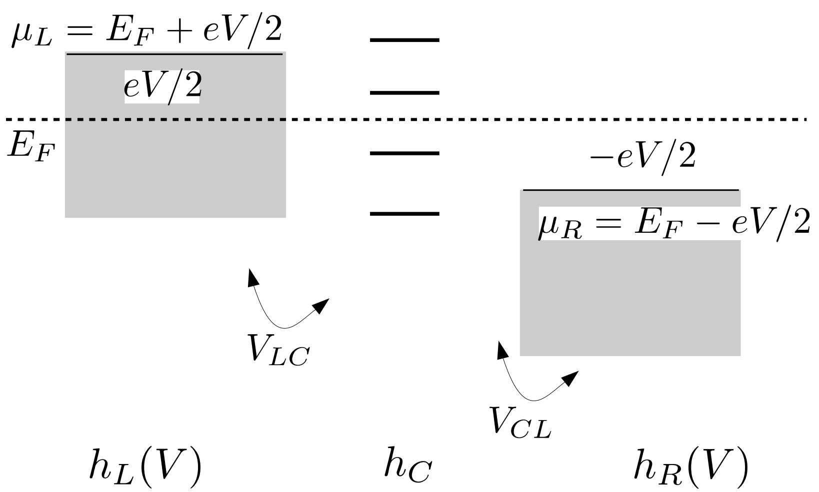
QE: DFT calculations $\xrightarrow[H_{\mathrm{DFT}}]{V_{\mathrm{KS}}}$ $h = \sum_{mn} V_{mn} c^\dagger_m c_n; \quad V_{mn} = \langle m|H_{\mathrm{DFT}}|n\rangle$ $\xrightarrow{V_{mn}}$ TBcond

in localized Wannier orbitals

$\{m, n\} = s, p, d, ...$ – atomic-like orbitals

Finite voltage: NEGF + DFT

At finite applied voltage:



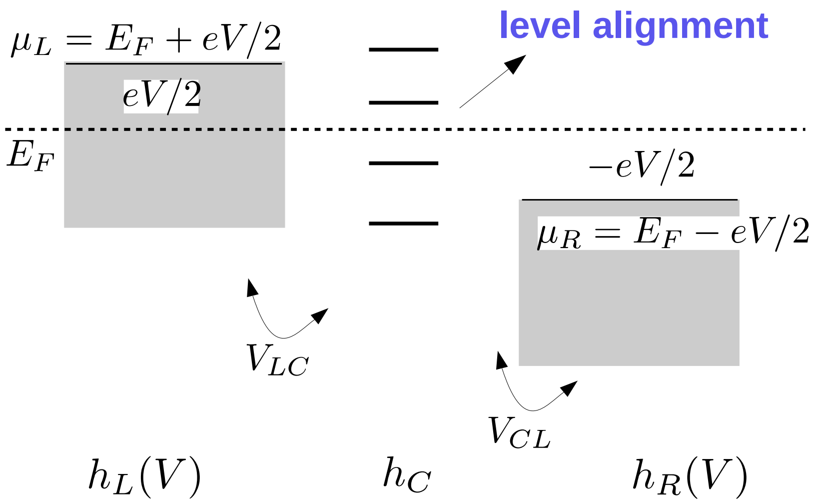
$$h_X(V) \to g_X(V) \to \Sigma_X(V) \to \Gamma_X(V); \quad X = L, R$$

$$\downarrow$$

$$T(E,V) = \mathrm{Tr}\left[\Gamma^L(E,V)G^r(E,V)\Gamma^R(E,V)G^a(E,V)\right]$$

Finite voltage: NEGF + DFT

At finite applied voltage:



$$h_X(V) \rightarrow g_X(V) \rightarrow \Sigma_X(V) \rightarrow \Gamma_X(V); \quad X = L, R$$

$$\downarrow$$

$$T(E,V) = \mathrm{Tr}\left[\Gamma^L(E,V) G^r(E,V) \Gamma^R(E,V) G^a(E,V)\right]$$

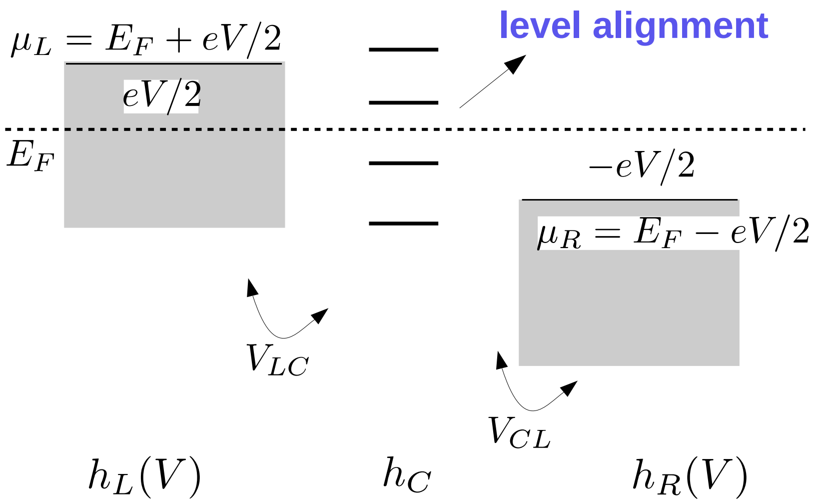
$$G^{r/a} = [(E - \boxed{h_C} - \Sigma_L^{r/a} - \Sigma_R^{r/a})]^{-1}$$

?

$$h_C[n_0] \rightarrow h_C[n]$$

$n = n_0 + \delta n$ – redistributed charge due to applied *V*

At finite applied voltage:



$$h_X(V) \to g_X(V) \to \Sigma_X(V) \to \Gamma_X(V); \quad X = L, R$$

$$T(E,V) = \mathrm{Tr}\left[\Gamma^L(E,V)G^r(E,V)\Gamma^R(E,V)G^a(E,V)\right]$$

$$G^{r/a} = [(E - h_C - \Sigma_L^{r/a} - \Sigma_R^{r/a})]^{-1}$$

?

$$h_C[n_0] \to h_C[n]$$

$n = n_0 + \delta n$ – redistributed charge due to applied *V*

Therefore a self-consistent calculations is required in principle:

$$G^{r/a} = [(E - h_C[n] - \Sigma_L^{r/a} - \Sigma_R^{r/a})]^{-1}$$

$$G^< = G^r[\Sigma_L^< + \Sigma_R^<]G^a$$

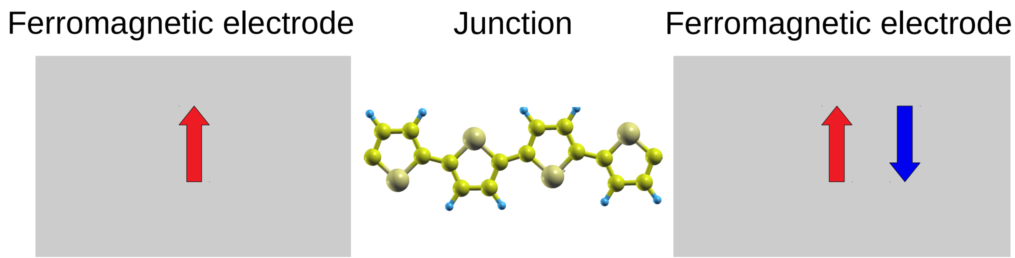
$$\rho_{mn} = \langle c_m^\dagger c_n \rangle = \frac{1}{2\pi i}\int dE G_{nm}^<(E)$$

$$i\hbar G^<(t_2, t_1) = -\langle \hat{c}^\dagger(t_1)\hat{c}(t_2)\rangle$$

$$n(r) = \left\langle \hat{\psi}^\dagger(r)\hat{\psi}(r)\right\rangle = \sum_{mn}\phi_m^*(r)\phi_n(r)\left\langle \hat{c}_m^\dagger \hat{c}_n\right\rangle = \sum_{mn}\phi_m^*(r)\phi_n(r)\rho_{mn}$$

$$\text{where } \hat{\psi}(r) = \sum_m \phi_m(r)\hat{c}_m; \quad \hat{\psi}^\dagger(r) = \sum_m \phi_m^*(r)\hat{c}_m^\dagger$$

Spin-polarized transport: basic effects



Parallel magnetic allignment (*P*):

Antiparallel magnetic allignment (*AP*):

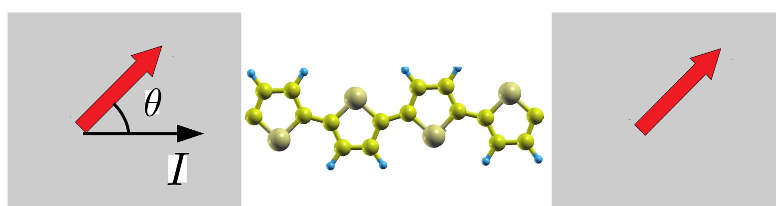
Spin-polarization, spin filtering (SP):

$$\mathrm{SP} = (G_{\downarrow} - G_{\uparrow})/(G_{\downarrow} + G_{\uparrow})$$

Magnetoresistance (MR):

$$\mathrm{MR} = (G_P - G_{AP})/G_{AP}$$

Aisotropic magnetoresistance (AMR):

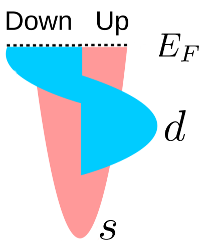


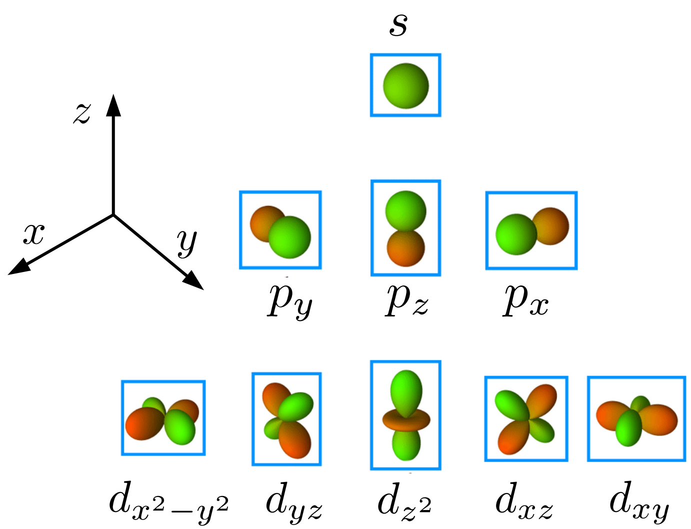
$$I(\theta) = ?$$

Need for spin-orbit coupling (SOC)

Goal: find systems with enhanced effects

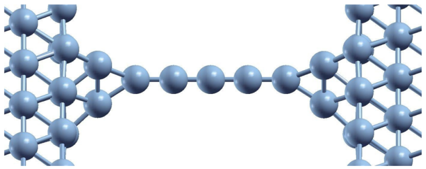
Partial spin filtering in ferromagnetic nanocontacts



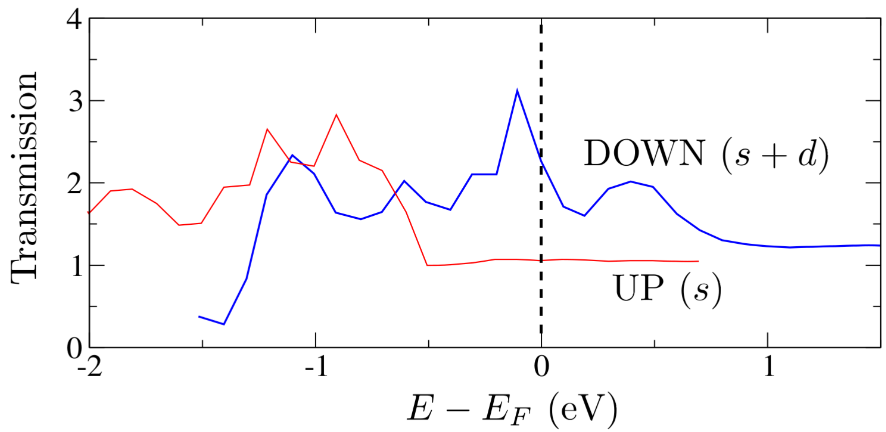


$$G_\sigma = e^2/h \sum_\alpha T_{\sigma\alpha}(E_F)$$ – sum over all orbital channels, *s* and *d*

Ni nanocontact

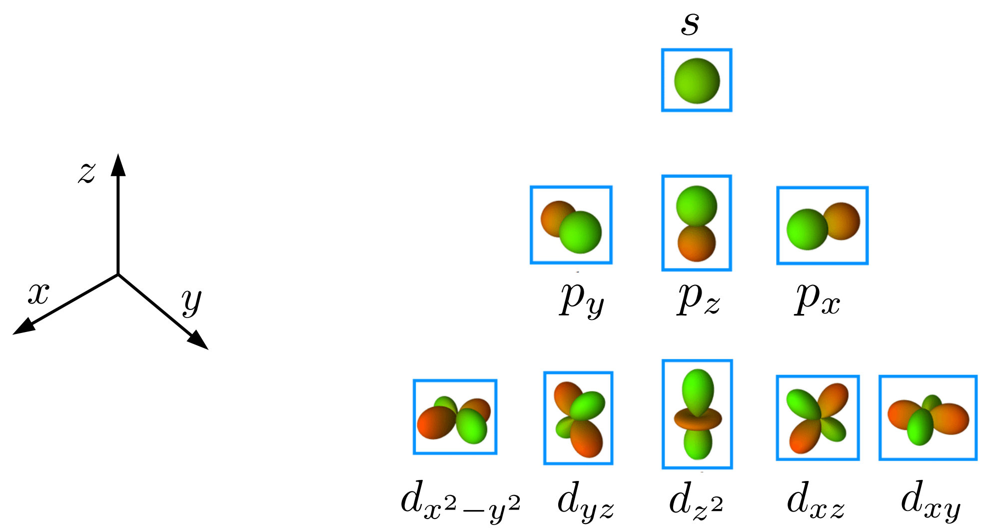


[PRB **73**, 075418 (2006)]

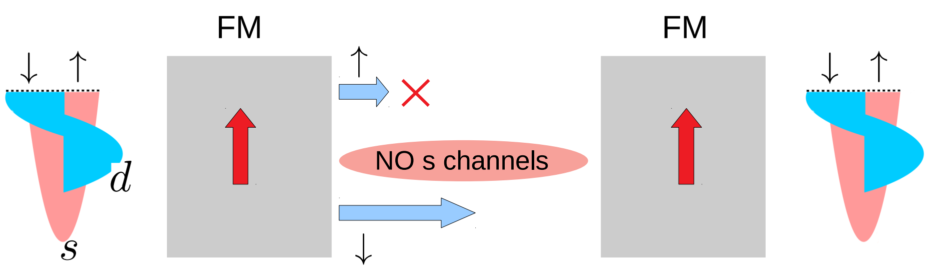


$$\mathrm{SP} = (G_\downarrow - G_\uparrow)/(G_\downarrow + G_\uparrow) \approx 33\%$$

Perfect spin filtering by symmetry mismatch

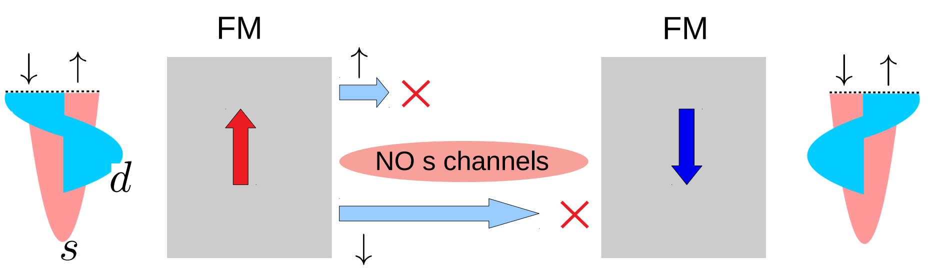


Parallel magnetic allignment:



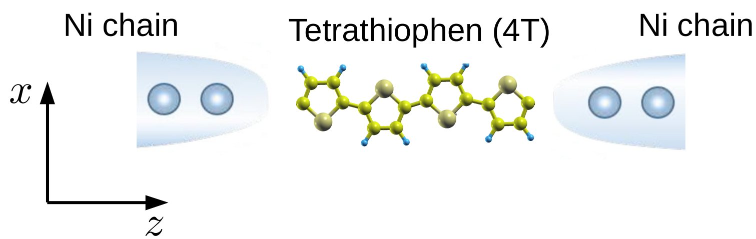
$$G_\uparrow = 0,\ G_\downarrow \neq 0 \ \rightarrow\ \mathrm{SP} = 100\%$$

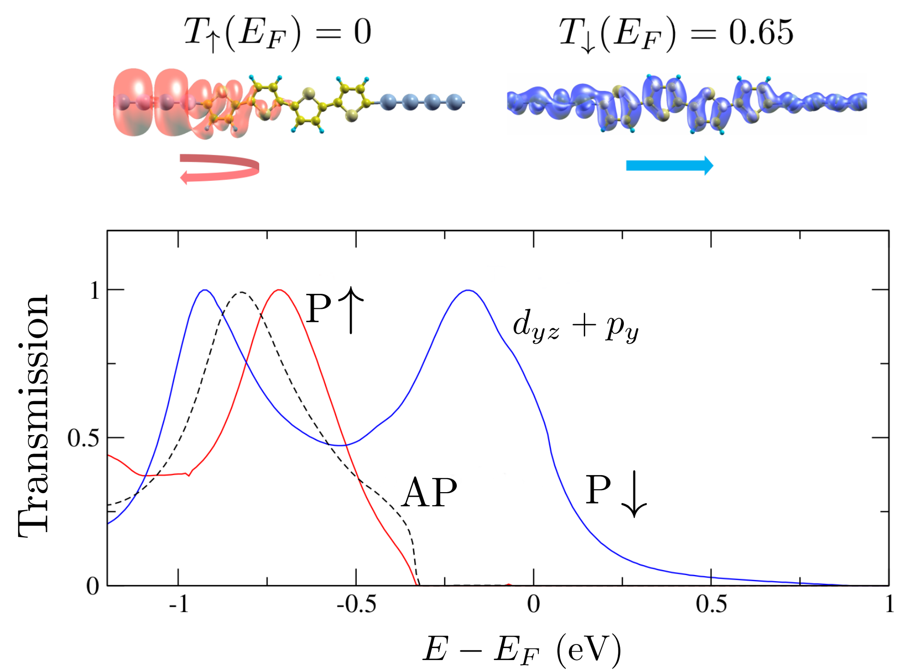
Antiparallel magnetic allignment:



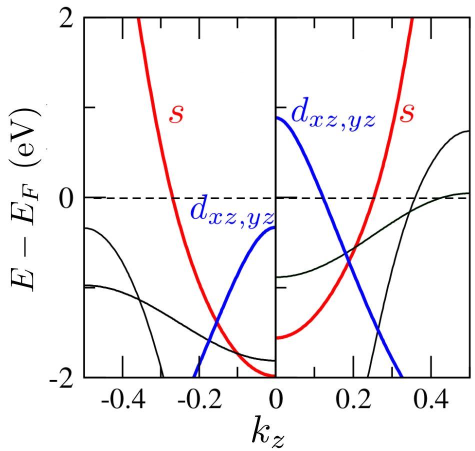
$$G_\uparrow = 0,\ G_\downarrow = 0 \ \rightarrow\ \text{infinite MR}$$

Example #1: Molecular junction, model electrodes

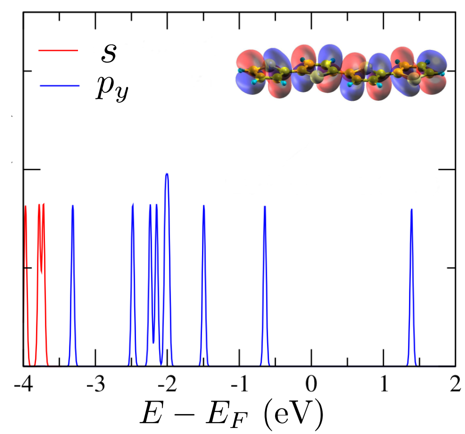


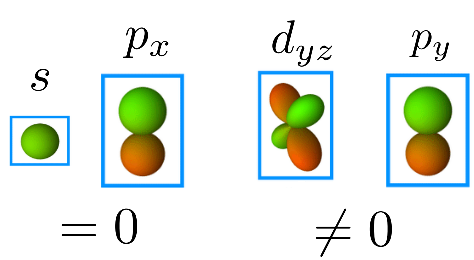


Ni bands



4T levels



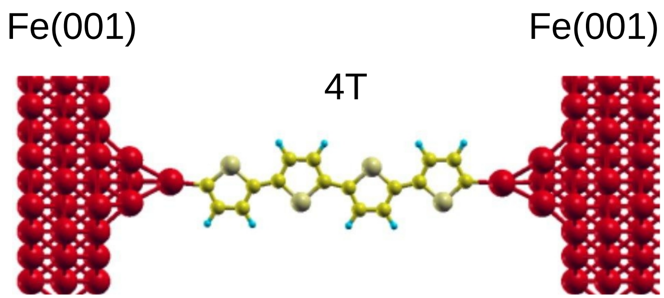


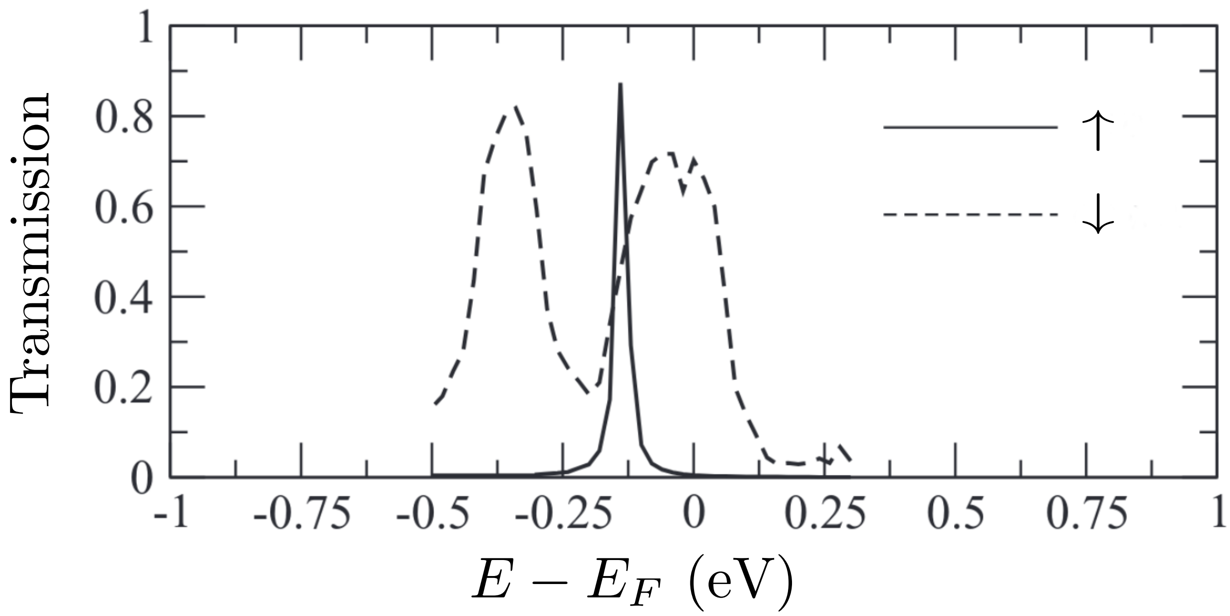
$\text{SP} = 100\%$

infinite MR

[Nano Lett. **15**, 3552 (2015)]

Example #1: Molecular junction, realistic electrodes

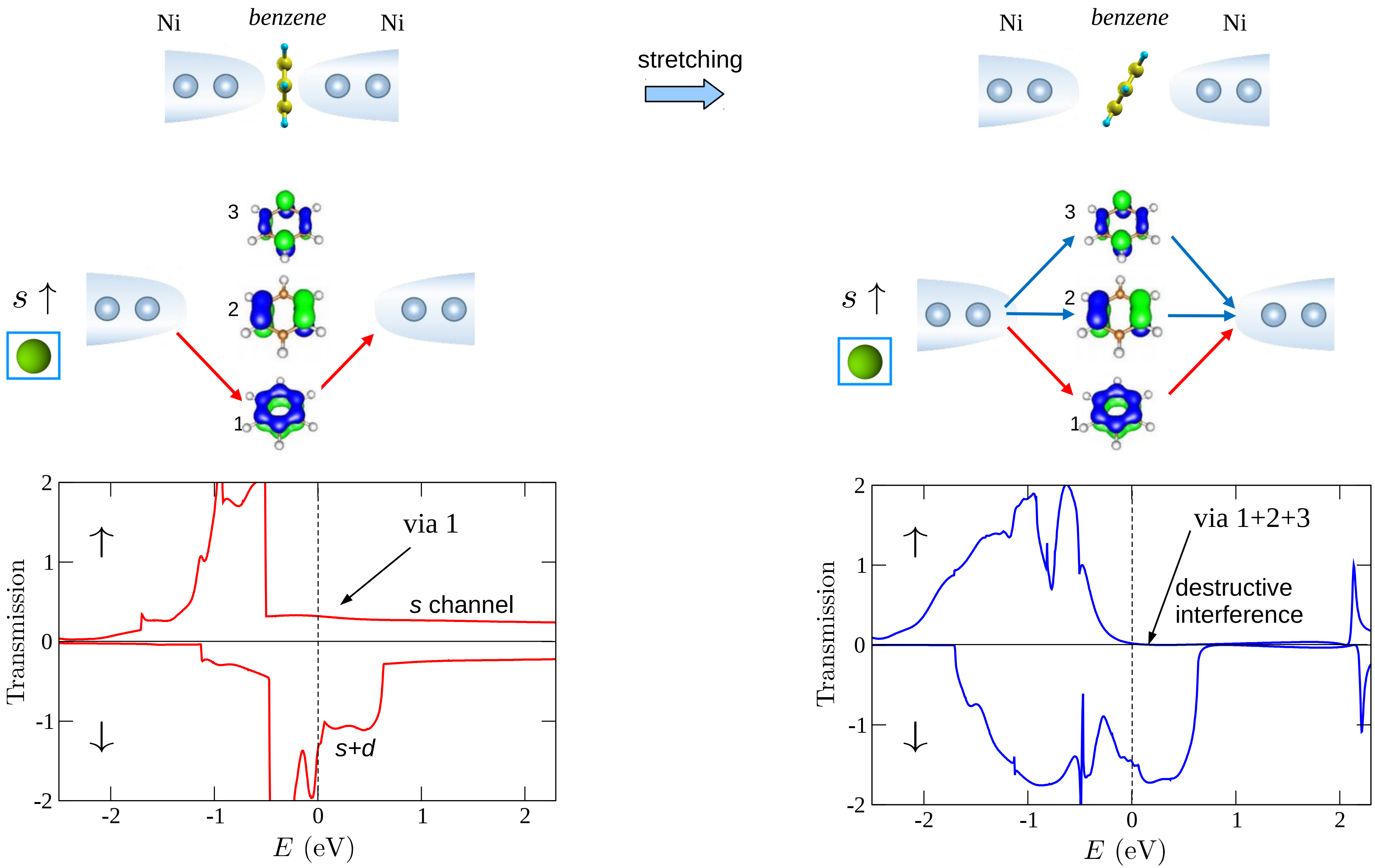




$$G_\uparrow \approx 0.004 G_0$$
$$G_\downarrow \approx 0.7 G_0$$

[PRB **93**, 201403 (2016)]

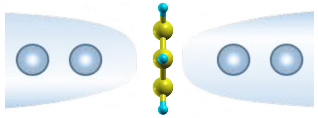
Tuning spin-filtering by quantum interference

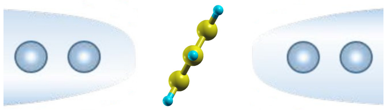


$G_{\uparrow} = 0.3 G_0$ SP $\approx 60\%$ SP $\approx 100\%$ $G_{\uparrow} \approx 0$

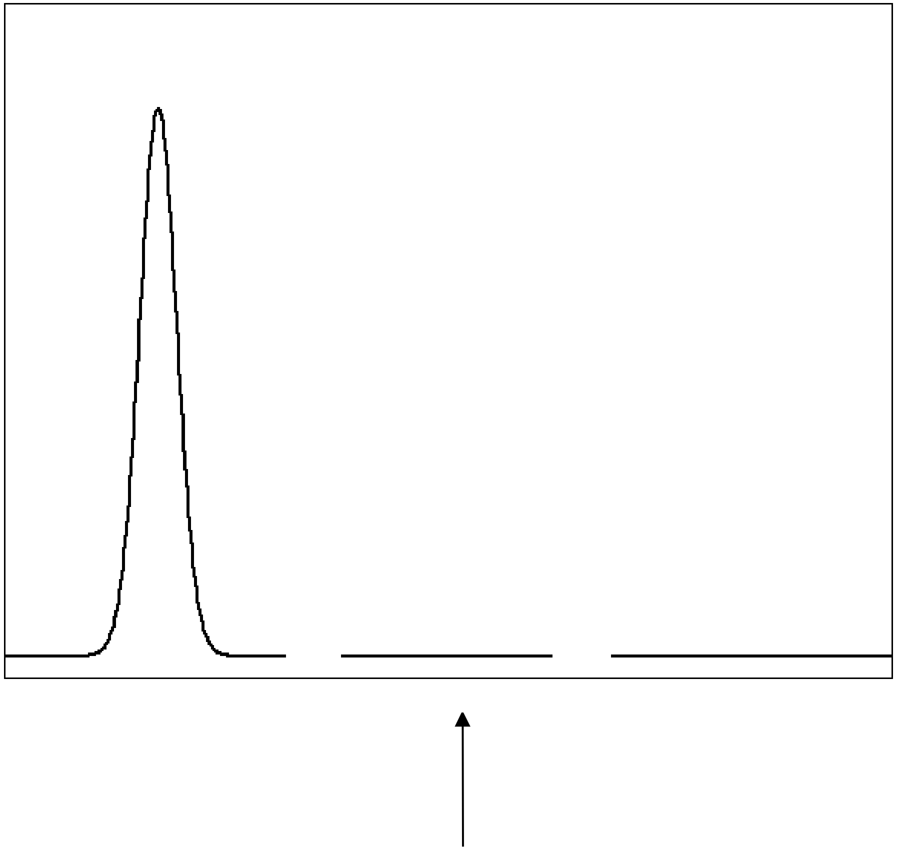
[PRB **99**, 115403 (2019)]

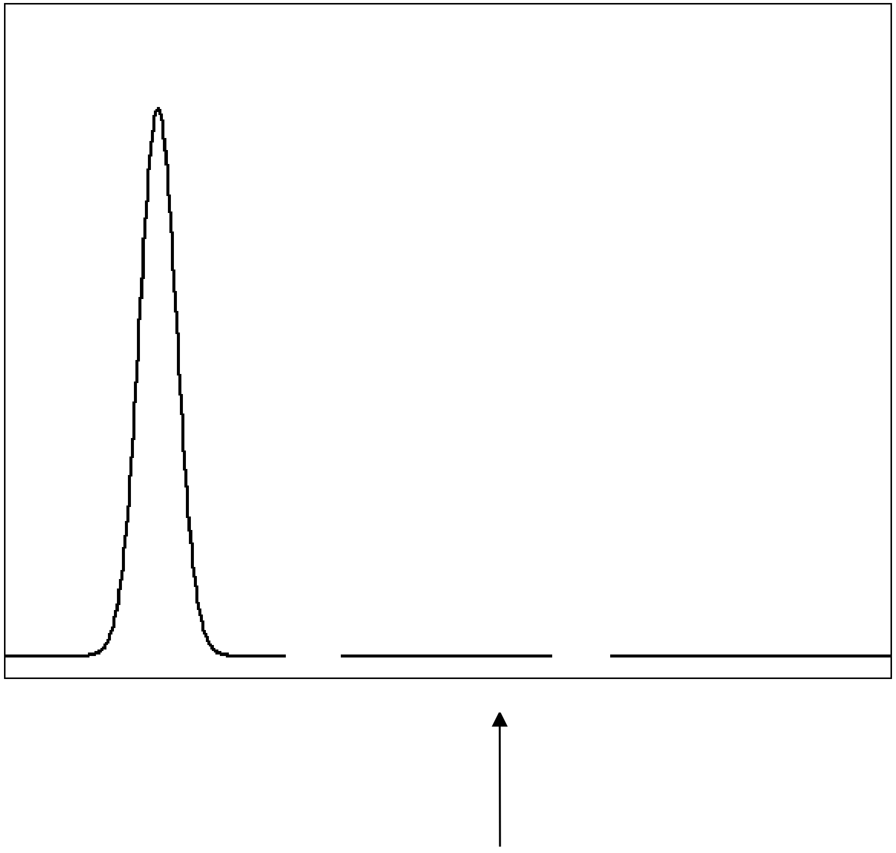
Tuning spin-filtering by quantum interference



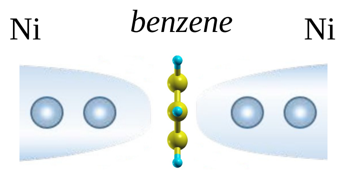


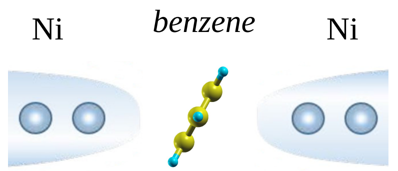
Wave packets propagation at the Fermi energy:



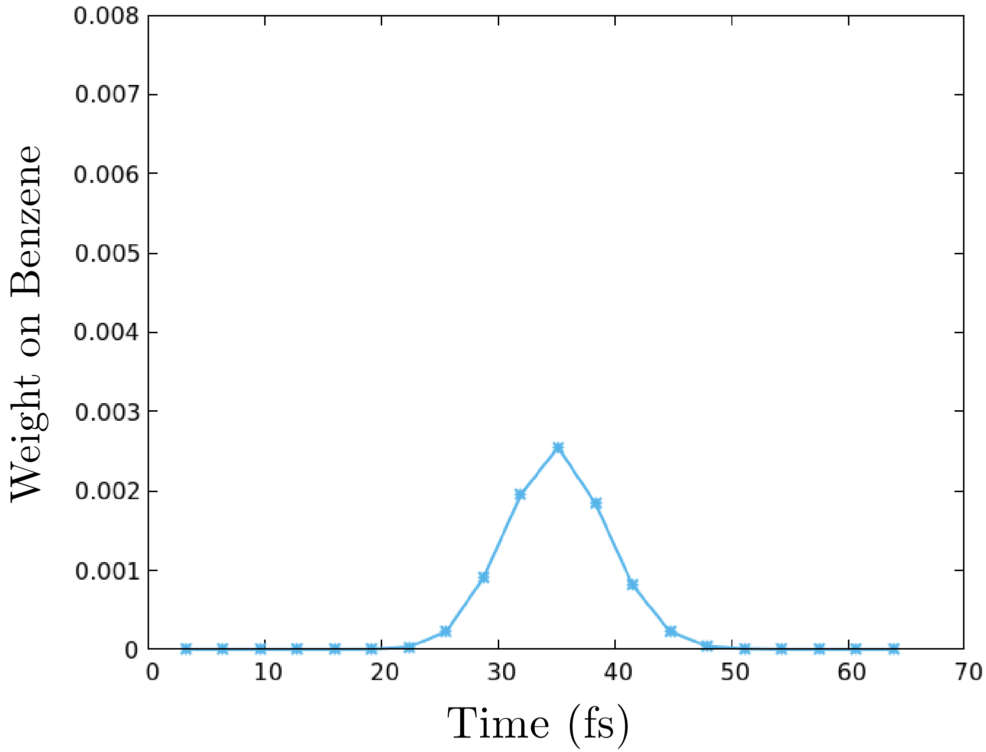


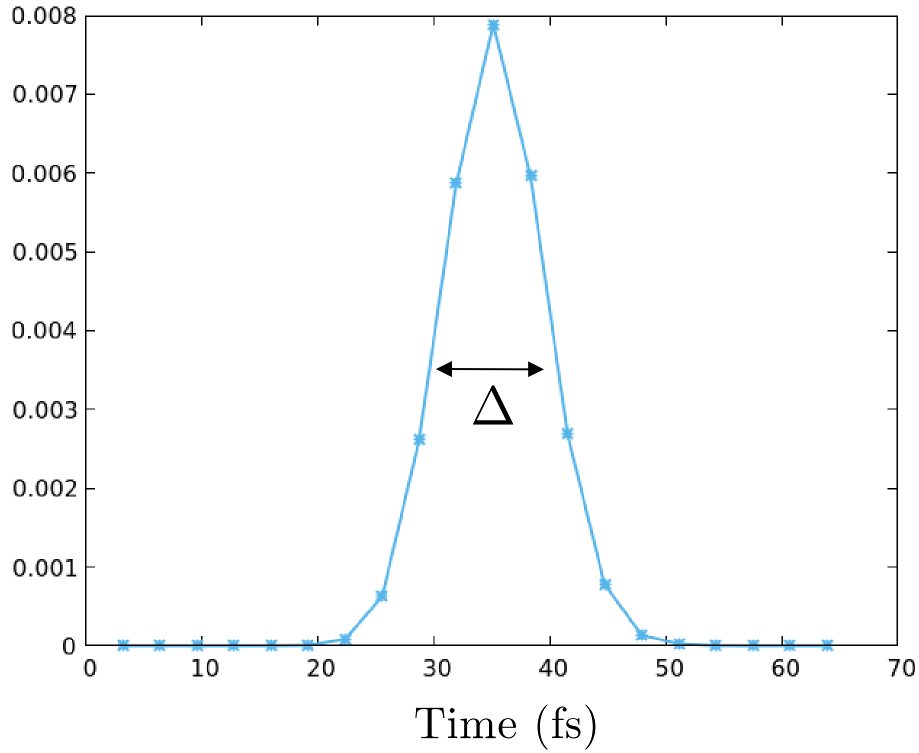
Tuning spin-filtering by quantum interference





Wave packets propagation time across the molecule:

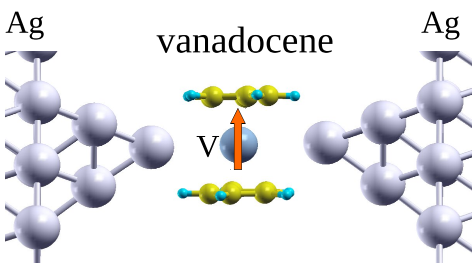




$$\Delta \approx 10 \text{ fs}$$

Tuning spin-filtering by quantum interference

Molecular junction

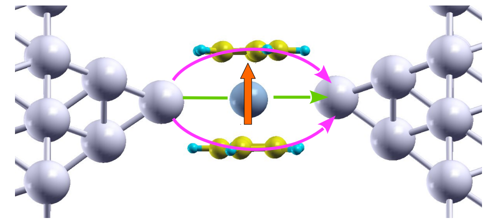


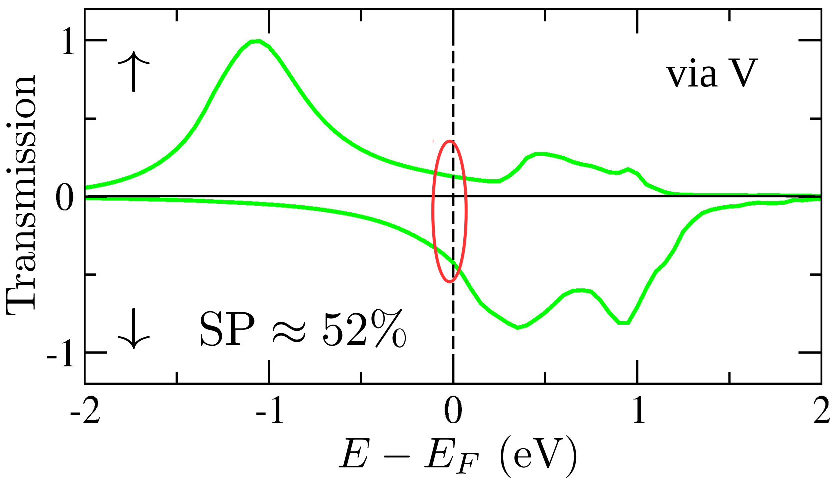
Exp: $G \approx 1G_0$
(Oren Tal's group, Israel)

↓

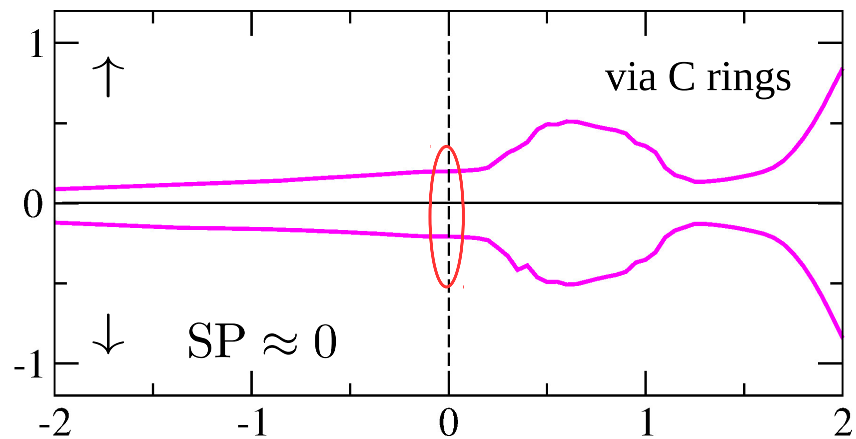
One fully polarized channel

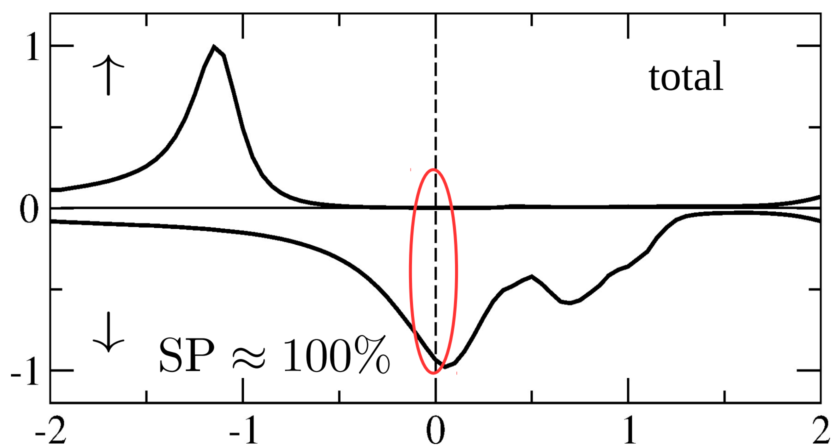
Propagation paths:





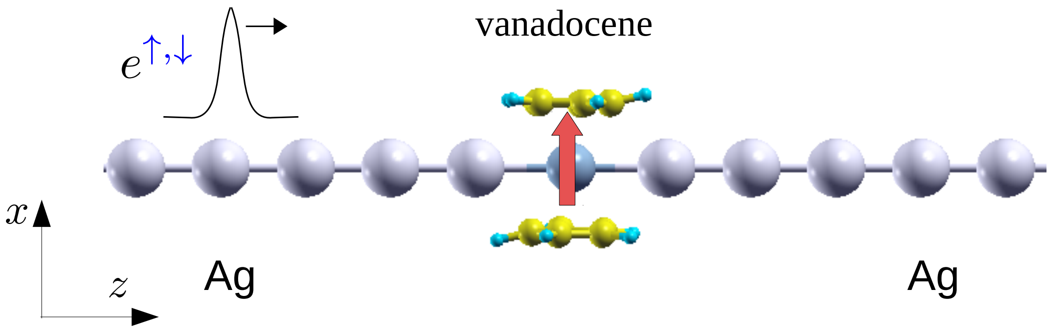
+



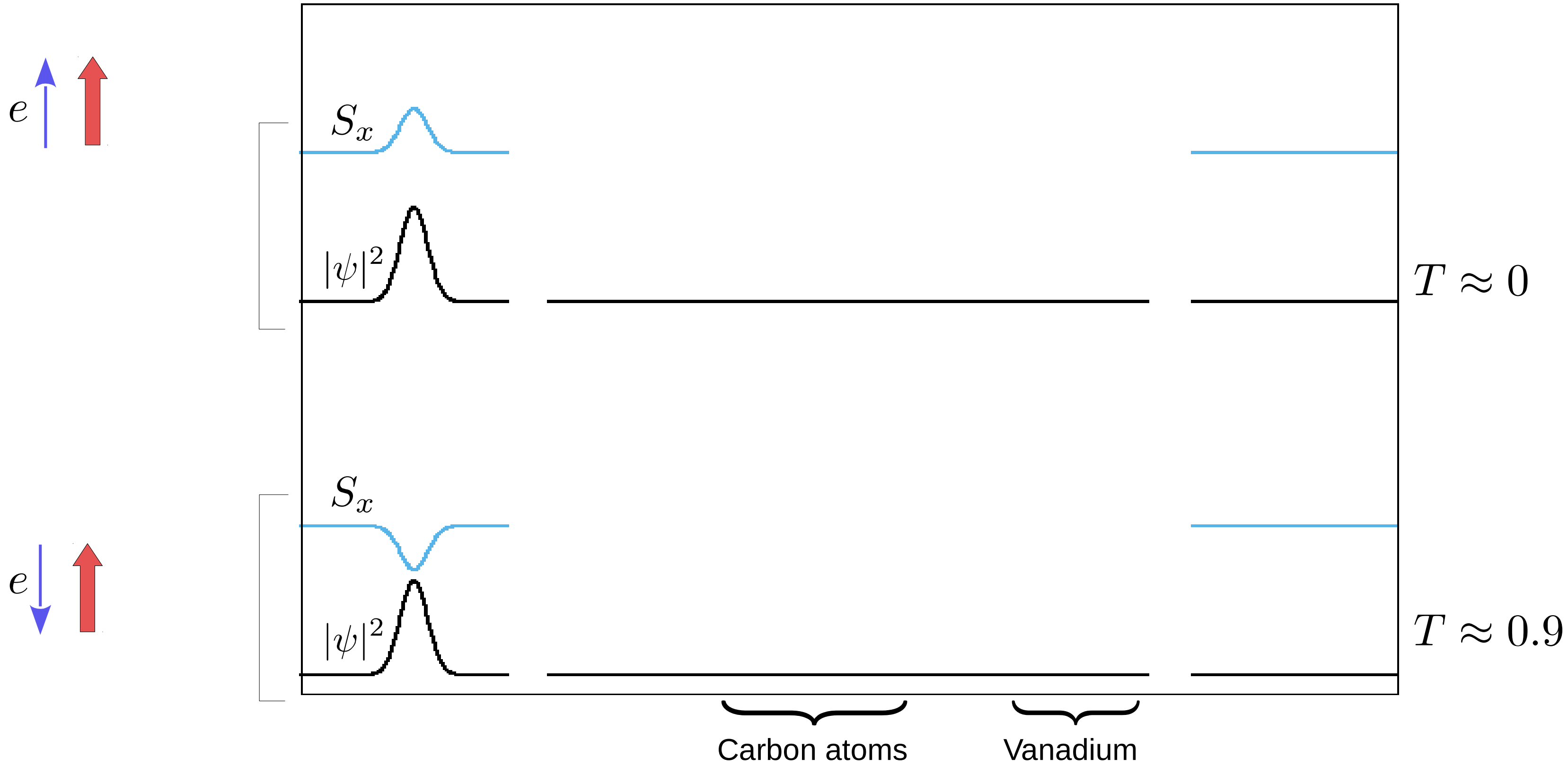


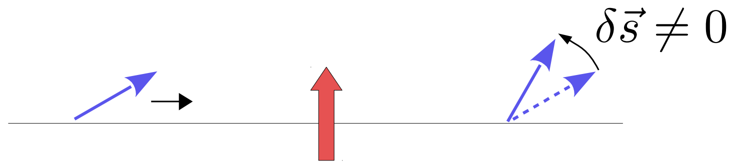
[Mature Comm. **10**, 5565 (2019)]

Tuning spin-filtering by quantum interference

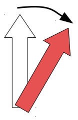


Wave packets at the Fermi energy:



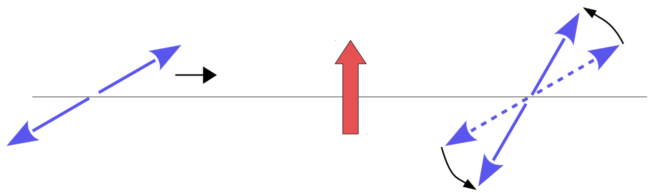


Spin-transfer torque



$$\delta\vec{S} = -\delta\vec{s}$$

No effect expected for non-polarized current

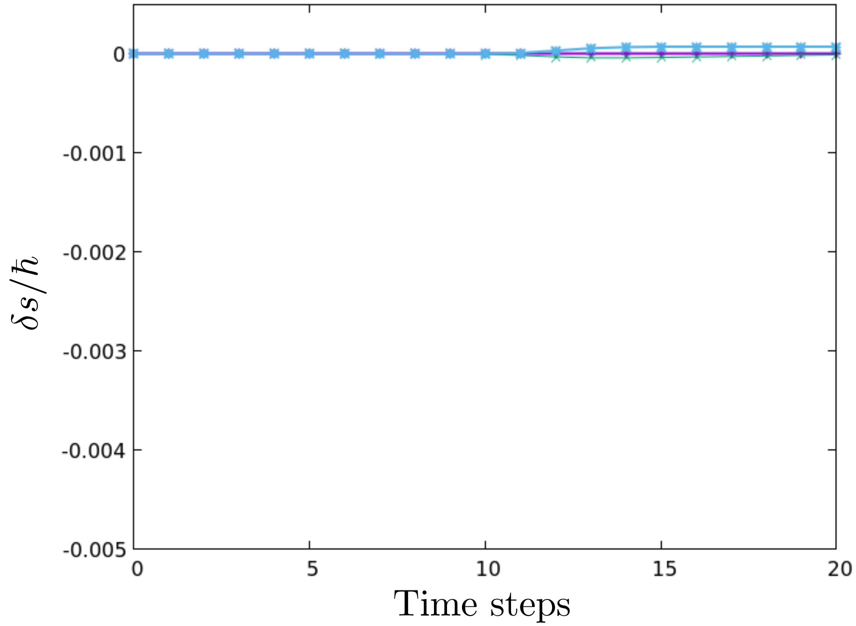


$$\delta\vec{s} = 0$$

No spin-transfer torque

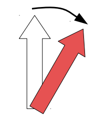
$$\delta\vec{S} = 0$$

No spin-orbit coupling



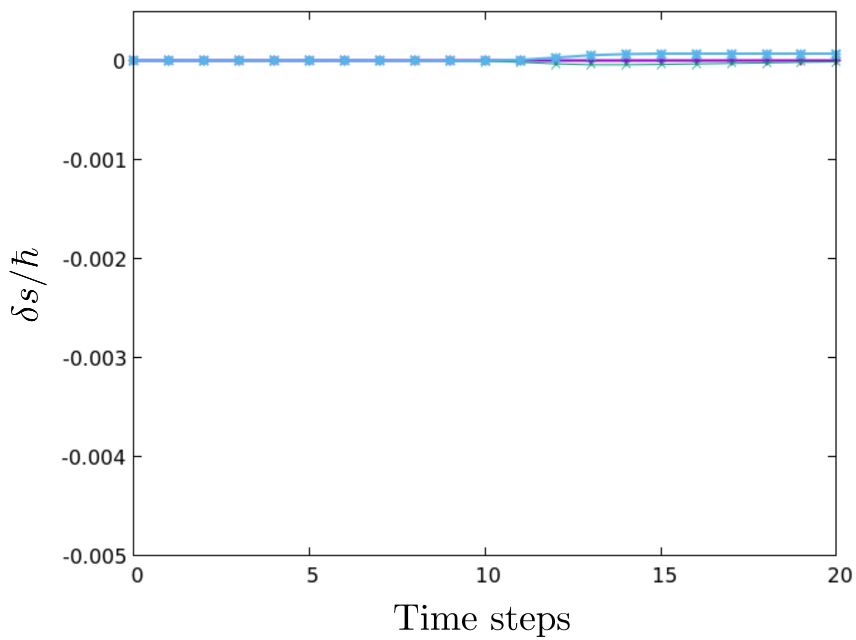
Spin-orbit torque (SOT) driven by spin-orbit interactions

Spin-orbit torque

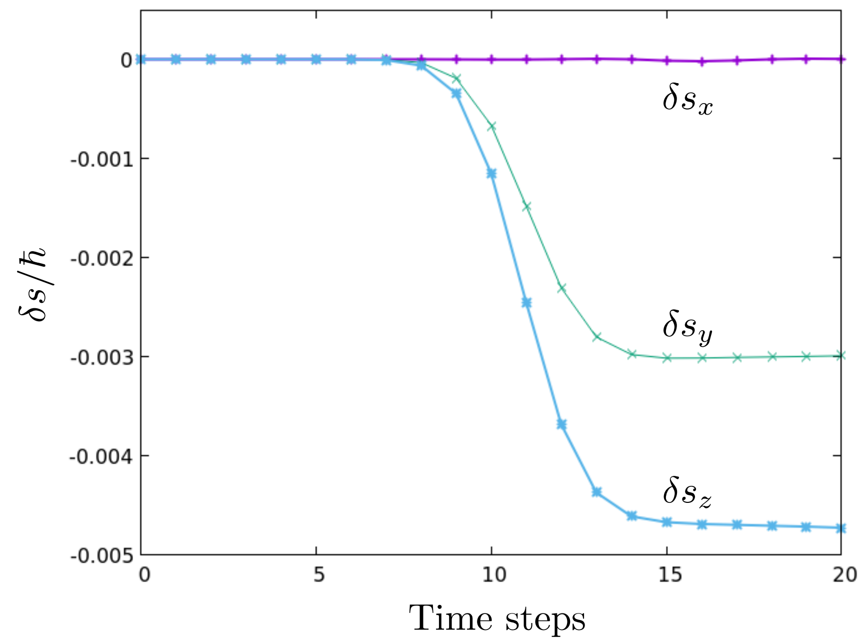


$$\delta\vec{S} \neq 0$$

No spin-orbit coupling

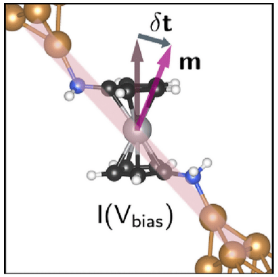


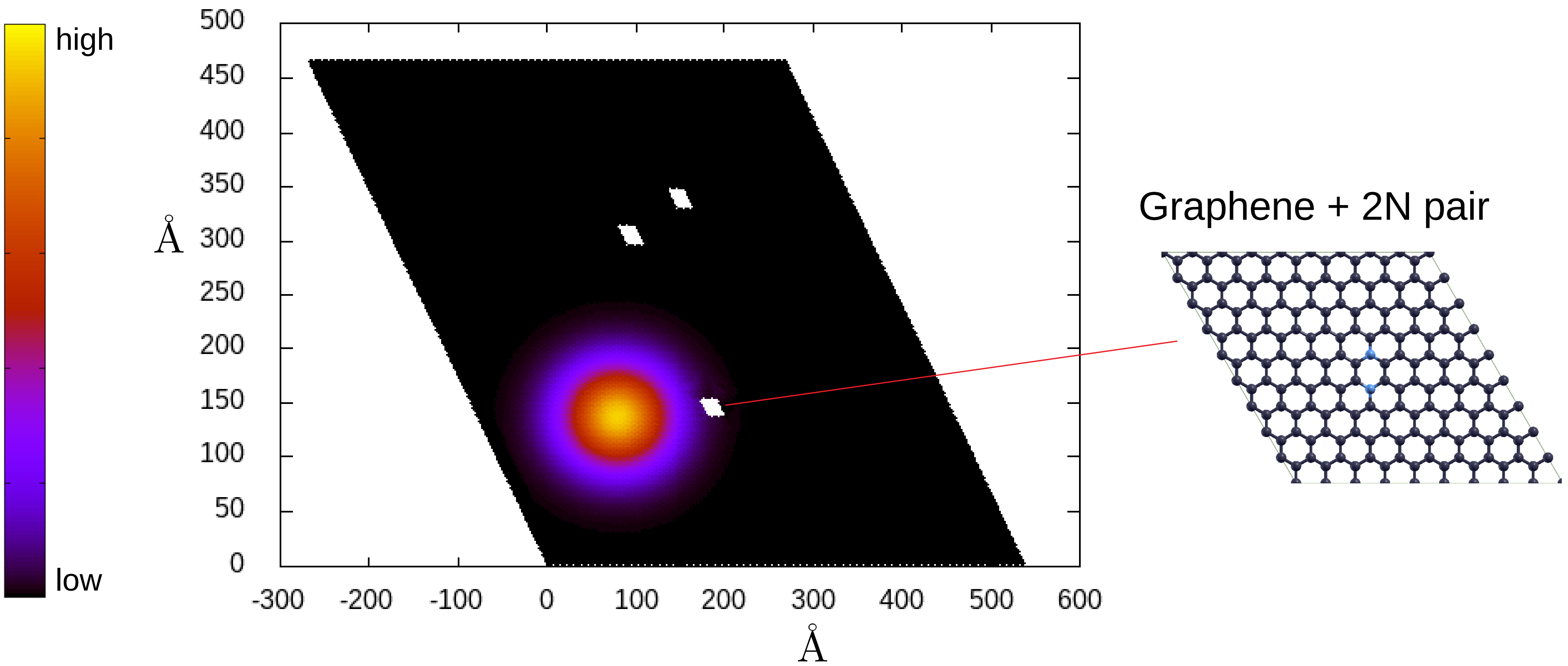
With spin-orbit coupling



Nice paper on the subject by M. Camarasa-Gómez, D. Hernangómez-Pérez, F. Evers

[J. Phys. Chem. Lett. **15**, 5747 (2024)]




high
low
500
450
400
350
300
250
200
150
100
50
0
Å
-300
-200
-100
0
100
200
300
400
500
600
Å
Graphene + 2N pair

Bibliography

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ANNEXE

Different pictures

$$\mathcal{H} = h + [V_{LC} + V_{RC} + h.c.] + W_{int}$$

perturbation $\delta\hat{h}$

$$\text{Mean value of any observable } \hat{O}(\text{current too}),\ t = 0 \text{ reference time :}$$

$$O(t) = \left\langle \Psi_S(t) \middle| \hat{O} \middle| \Psi_S(t) \right\rangle;\quad \Psi_S(t) = e^{\frac{-i}{\hbar}\hat{\mathcal{H}}t}\Psi_S(0);\quad \hat{O} \text{ is time independent}$$ – Schrödinger picture

$$O(t) = \left\langle \Psi_{\mathcal{H}} \middle| \hat{O}_{\mathcal{H}}(t) \middle| \Psi_{\mathcal{H}} \right\rangle;\quad \Psi_{\mathcal{H}} \text{ is time independent};\ \hat{O}_{\mathcal{H}}(t) = e^{\frac{i}{\hbar}\hat{\mathcal{H}}t}\hat{O}e^{-\frac{i}{\hbar}\hat{\mathcal{H}}t}$$ – Heisenberg picture

$$O(t) = \left\langle \Psi_I(t) \middle| \hat{O}_I(t) \middle| \Psi_I(t) \right\rangle;\quad \Psi_I(t) = e^{\frac{i}{\hbar}\hat{h}t}\Psi_S(t);\quad \hat{O}_I(t) = e^{\frac{i}{\hbar}\hat{h}t}\hat{O}e^{-\frac{i}{\hbar}\hat{h}t}$$ – Interaction picture

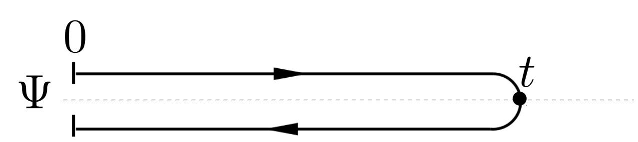
At reference time t = 0 states are the same in all pictures: $\Psi_S(0) = \Psi_{\mathcal{H}} = \Psi_I(0) = \Psi$

Time evolution of the state:

$$\Psi_I(t) = \hat{S}(t,0)\Psi$$

with evolution operator: $$\hat{S}(t_2,t_1) = \mathcal{T}_t\left\{\exp\left(-\frac{i}{\hbar}\int_{t_1}^{t_2}\delta\hat{h}_I(t)dt\right)\right\}$$

$$O(t) = \left\langle \Psi \middle| \hat{S}(0,t)\hat{O}_I(t)\hat{S}(t,0) \middle| \Psi \right\rangle$$



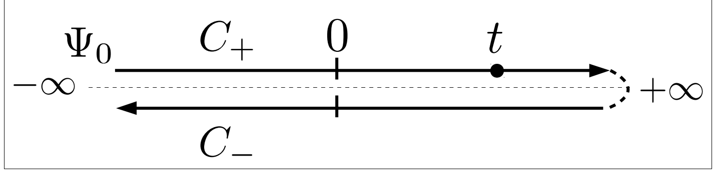
NEGF Contour

Assuming adiabatic switching on the perturbation:

$$|\Psi\rangle = \hat{S}(0,-\infty)\ |\Psi_0\rangle$$

$$O(t) = \left\langle \Psi_0 \left| \hat{S}(-\infty,0)\cdot\hat{S}(0,t)\hat{O}_I(t)\hat{S}(t,0)\cdot\hat{S}(0,-\infty) \right| \Psi_0 \right\rangle$$

Keldysh contour $C = C_+ + C_-$:



$$O(t) = \left\langle \Psi_0 \left| \mathcal{T}_C \left\{ \hat{S}_C \hat{O}_I(t) \right\} \right| \Psi_0 \right\rangle; \quad \hat{S}_C = \mathcal{T}_C \left\{ \exp\left(-\frac{i}{\hbar} \int_C \delta h_I(\tau) d\tau \right) \right\}$$

Contour Green functions:

$$i\hbar G_{mn}(\tau_2,\tau_1) = \left\langle \Psi \left| \mathcal{T}_C \left\{ \hat{c}_{m,\mathcal{H}}(\tau_2)\hat{c}^\dagger_{n,\mathcal{H}}(\tau_1) \right\} \right| \Psi \right\rangle$$

$$i\hbar G_{mn}(\tau_2,\tau_1) = \left\langle \Psi_0 \left| \mathcal{T}_C \left\{ \hat{S}_C \hat{c}_{m,I}(\tau_2)\hat{c}^\dagger_{n,I}(\tau_1) \right\} \right| \Psi_0 \right\rangle$$

Dyson equantion for contour GFs:

$$G(1',1) = g(1',1) + \int_C d\tau_2 g(1',2)V(2)G(2,1) + \int_C d\tau_2 \int_C d\tau_3\ g(1',2)\Sigma_{int}(2,3)G(3,1)$$

$$G(1',1) = g(1',1) + \int_C d\tau_2 G(1',2)V(2)g(2,1) + \int_C d\tau_2 \int_C d\tau_3\ G(1',2)\Sigma_{int}(2,3)g(3,1)$$

Green functions in the time domain

Produces four Green functions in the time domain:

$$G(\tau_2,\tau_1) = \begin{cases} G^{++}(t_2,t_1) & \tau_2,\tau_1 \in C_+ \\ G^{--}(t_2,t_1) & \tau_2,\tau_1 \in C_- \\ G^{<}(t_2,t_1) & \tau_2 \in C_+, \tau_1 \in C_- \\ G^{>}(t_2,t_1) & \tau_2 \in C_-, \tau_1 \in C_+ \end{cases}$$

$$G^r = G^{++} - G^<; \quad G^a = G^{--} - G^>$$

$$i\hbar G^<_{mn}(t_2,t_1) = -\left\langle \Psi \left| \hat{c}^\dagger_{n,\mathcal{H}}(t_1)\hat{c}_{m,\mathcal{H}}(t_2) \right| \Psi \right\rangle$$

$$i\hbar G^>_{mn}(t_2,t_1) = \left\langle \Psi \left| \hat{c}_{m,\mathcal{H}}(t_2)\hat{c}^\dagger_{n,\mathcal{H}}(t_1) \right| \Psi \right\rangle$$

$$i\hbar G^r_{mn}(t_2,t_1) = \theta(t_2-t_1)\left\langle \Psi \left| \left[\hat{c}_{m,\mathcal{H}}(t_2)\hat{c}^\dagger_{n,\mathcal{H}}(t_1)\right]_+ \right| \Psi \right\rangle$$

$$i\hbar G^a_{mn}(t_2,t_1) = -\theta(t_1-t_2)\left\langle \Psi \left| \left[\hat{c}_{m,\mathcal{H}}(t_2)\hat{c}^\dagger_{n,\mathcal{H}}(t_1)\right]_+ \right| \Psi \right\rangle$$

$$G^{++} + G^{--} = G^> + G^<; \quad G^r - G^a = G^> - G^<$$

Dyson equation in the time and frequency

Once again Dyson equantion for contour GFs :

$$G(1',1) = g(1',1) + \int_C d\tau_2 g(1',2)V(2)G(2,1) + \int_C d\tau_2 \int_C d\tau_3 \; g(1',2)\Sigma_{int}(2,3)G(3,1)$$

$$G(1',1) = g(1',1) + \int_C d\tau_2 G(1',2)V(2)g(2,1) + \int_C d\tau_2 \int_C d\tau_3 \; G(1',2)\Sigma_{int}(2,3)g(3,1)$$

Using real time GFs, going to time integrals and performing Fourrier transform:

$$G^{r/a}(E) = g^{r/a}(E) + g^{r/a}(E)VG^{r/a}(E) + g^{r/a}(E)\Sigma^{r/a}(E)G^{r/a}(E)$$

$$G^{</>}(E) = g^{</>}(E) + g^{</>}(E)VG^{a}(E) + g^{r}(E)VG^{</>}(E)$$
$$+g^{</>}(E)\Sigma^{a}(E)G^{a}(E) + g^{r}(E)\Sigma^{</>}(E)G^{a}(E) + g^{r}(E)\Sigma^{r}(E)G^{</>}(E)$$

Electric current within NEGF

$$I_L(t) = -e\frac{d\left\langle \hat{N}_{L,\mathcal{H}}(t)\right\rangle}{dt} = -\frac{ie}{\hbar}\left\langle [H, \hat{N}_{L,\mathcal{H}}(t)]\right\rangle \qquad \hat{N}_L = \sum_k c_k^\dagger c_k = \sum_j c_j^\dagger c_j$$

$$I_L = \frac{ie}{\hbar}\sum_{j\in L, n\in C}\left\langle V_{jn}c_j^\dagger(t)c_n(t) - V_{nj}c_n^\dagger(t)c_j(t)\right\rangle = \frac{e}{h}\int dE\ [G_{CL}^<(E)V_{LC} - V_{CL}G_{LC}^<(E)]$$

$$I_L = \frac{e}{h}\int dE \mathrm{Tr}[(G_{CC}^< V_{CL}g_{LL}^a + G_{CC}^r V_{CL}g_{LL}^<)V_{LC} - V_{CL}(g_{LL}^< V_{LC}G_{CC}^a + g_{LL}^r V_{LC}G_{CC}^<)]$$

$$= \frac{e}{h}\int dE \mathrm{Tr}[(G_{CC}^r - G_{CC}^a)V_{CL}g_{LL}^< V_{LC} - V_{CL}(g_{LL}^r - g_{LL}^a)V_{LC}G_{CC}^<]$$

$$I_L = \frac{e}{h}\int dE\ \mathrm{Tr}[V_{CL}\ g_{LL}^< V_{LC}(G_{CC}^> - G_{CC}^<) - V_{CL}(g_{LL}^> - g_{LL}^<)V_{LC}G_{CC}^<]$$

$$= \frac{e}{h}\int dE\ \mathrm{Tr}[\Sigma_L^< G_{CC}^> - \Sigma_L^> G_{CC}^<]$$

Slide 7

Derivation of the Keldysh formula

Equation on Page 7: $G_{CC}^{</>} = G_{CC}^{r}[\Sigma_L^{</>} + \Sigma_R^{</>} + \Sigma_{int}^{</>}]G_{CC}^{a}$

Dyson:

$$G^{<} = g^{<} + g^{<}VG^{a} + g^{r}VG^{<} + g^{<}\Sigma^{a}G^{a} + g^{r}\Sigma^{<}G^{a} + g^{r}\Sigma^{r}G^{<}$$

$$\Big[1 - g^{r}(V + \Sigma^{r})\Big]G^{<} = g^{<}\Big[1 + (V + \Sigma^{a})G^{a}\Big] + g^{r}\Sigma^{<}G^{a}$$

$$G^{<} = \underbrace{\Big[1 - g^{r}V - g^{r}\Sigma^{r}\Big]^{-1}}_{\textbf{1}} g^{<}\Big[1 + VG^{a} + \Sigma^{a}G^{a}\Big] + \underbrace{\Big[1 - g^{r}V - g^{r}\Sigma^{r}\Big]^{-1}}_{\textbf{1“}} g^{r}\Sigma^{<}G^{a}$$

Dyson:

$$G^{r} = g^{r} + g^{r}VG^{r} + g^{r}\Sigma^{r}G^{r} \longrightarrow G^{r} = \underbrace{\Big(1 - g^{r}V - g^{r}\Sigma^{r}\Big)^{-1}}_{\textbf{2}} g^{r}$$

$$G^{r} = g^{r} + G^{r}Vg^{r} + G^{r}\Sigma^{r}g^{r} \longrightarrow G^{r} = \underbrace{\Big(1 + G^{r}V + G^{r}\Sigma^{r}\Big)^{-1}}_{\textbf{3}} g^{r}$$

$$G^{<} = \Big[\overbrace{1 + G^{r}V + G^{r}\Sigma^{r}}^{\textbf{1-2-3}}\Big]g^{<}\Big[1 + VG^{a} + \Sigma^{a}G^{a}\Big] + \overbrace{G^{r}}^{\textbf{1“-2}}\Sigma^{<}G^{a}$$

valid in the whole region (L+C+R)

Derivation of the Keldysh formula (Central region)

$$G^< = \Big[1 + G^r V + G^r \Sigma^r\Big] g^< \Big[1 + V G^a + \Sigma^a G^a\Big] + G^r \Sigma^< G^a$$

Taking CC block:

$$X = L, R, \ldots$$

$$C^<_{CC} = g^<_C + \sum_X g^<_C V_{CX} G^a_{XC} + g^<_C \Sigma^a_{CC} G^a_{CC} +$$ 1-1,2,3

$$+ \sum_X G^r_{CX} V_{XC} g^<_C + \sum_X G^r_{CC} V_{CX} g^<_X V_{XC} G^a_{CC} + \sum_{XX'} G^r_{CX'} V_{X'C} g^<_C V_{CX} G^a_{XC} +$$ 2-1,2,2“

$$+ \sum_X G^r_{CX} V_{XC} g^<_C \Sigma^a_{CC} G^a_{CC} +$$ 2-3

$$+ G^r_{CC} \Sigma^r_{CC} g^<_C + \sum_X G^r_{CC} \Sigma^r_{CC} g^<_C V_{CX} G^a_{XC} + G^r_{CC} \Sigma^r_{CC} g^<_C \Sigma^a_{CC} G^a_{CC} +$$ 3-1,2,3

$$+ G^r_{CC} \Sigma^<_{CC} G^a_{CC}$$

$$C^<_{CC} = g^<_C \Big(1 + \sum_X \Sigma^a_X G^a_{CC} + \Sigma^a_{CC} G^a_{CC}\Big) + \Big(\sum_X G^r_{CC} \Sigma^r_X + G^r_{CC} \Sigma^r_{CC}\Big) g^<_C +$$ 1-1,2,3
2-1,3-1

$$+ G^r_{CC} \Big(\sum_{X'} \Sigma^r_{X'} + \Sigma^r_{CC}\Big) g^<_C \Big(\sum_X \Sigma^r_X + \Sigma^a_{CC}\Big) G^a_{CC}$$

$$+ G^r_{CC} \Big(\sum_X \Sigma^<_X + \Sigma^<_{CC}\Big) G^a_{CC}$$ 2-2

Derivation of the Keldysh formula (Central region)

$$C^{<}_{CC} = g^{<}_{C}\Big(1 + \sum_{X} \Sigma^{a}_{X} G^{a}_{CC} + \Sigma^{a}_{CC} G^{a}_{CC}\Big) + \Big(\sum_{X} G^{r}_{CC}\Sigma^{r}_{X} + G^{r}_{CC}\Sigma^{r}_{CC}\Big) g^{<}_{C} +$$

$$+ G^{r}_{CC}\Big(\sum_{X'} \Sigma^{r}_{X'} + \Sigma^{r}_{CC}\Big) g^{<}_{C} \Big(\sum_{X} \Sigma^{r}_{X} + \Sigma^{a}_{CC}\Big) G^{a}_{CC}$$

$$+ G^{r}_{CC}\Big(\sum_{X} \Sigma^{<}_{X} + \Sigma^{<}_{CC}\Big) G^{a}_{CC}$$

$$G^{<}_{CC} = \Big(1 + G^{r}_{CC}\tilde{\Sigma}^{r}_{CC}\Big) g^{<}_{C} \Big(1 + \tilde{\Sigma}^{a}_{CC} G^{a}_{CC}\Big) + G^{r}_{CC}\tilde{\Sigma}^{<}_{CC} G^{a}_{CC}$$

with $$\tilde{\Sigma}^{r/a/<}_{CC} = \sum_{X} \Sigma^{r/a/<}_{X} + \Sigma^{r/a/<}_{CC}$$

Often first term is disregarded (?):

$$G^{<}_{CC} = G^{r}_{CC}\tilde{\Sigma}^{<}_{CC} G^{a}_{CC}$$