Electron transport in carbon nanotubes: contact effects

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1. Motivation
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3. Unpolarized Transport in Metall-CNT systems
4. GMR in FM-CNT systems (Prof. S. Krompiewski, Poznan)
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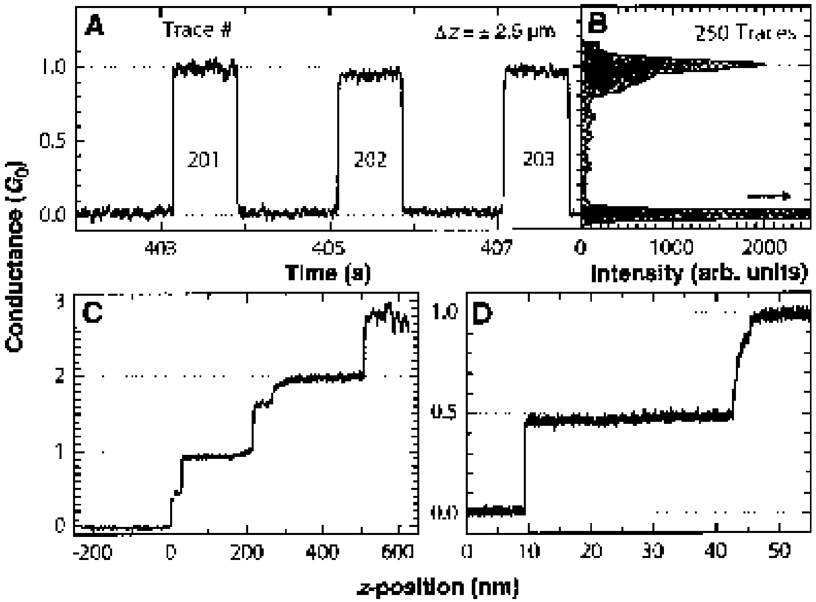
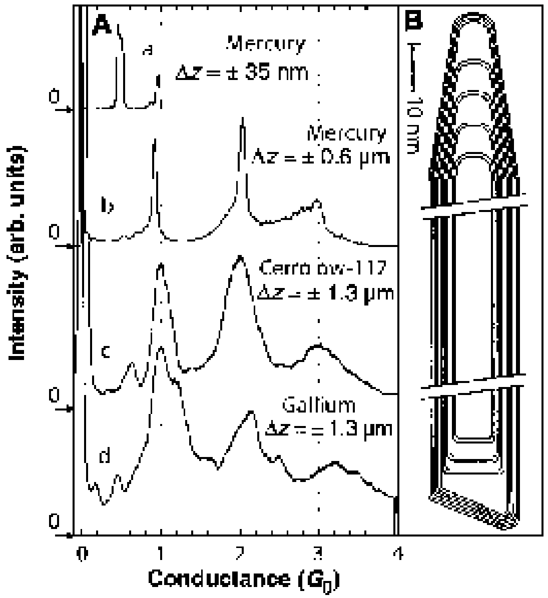
Motivation

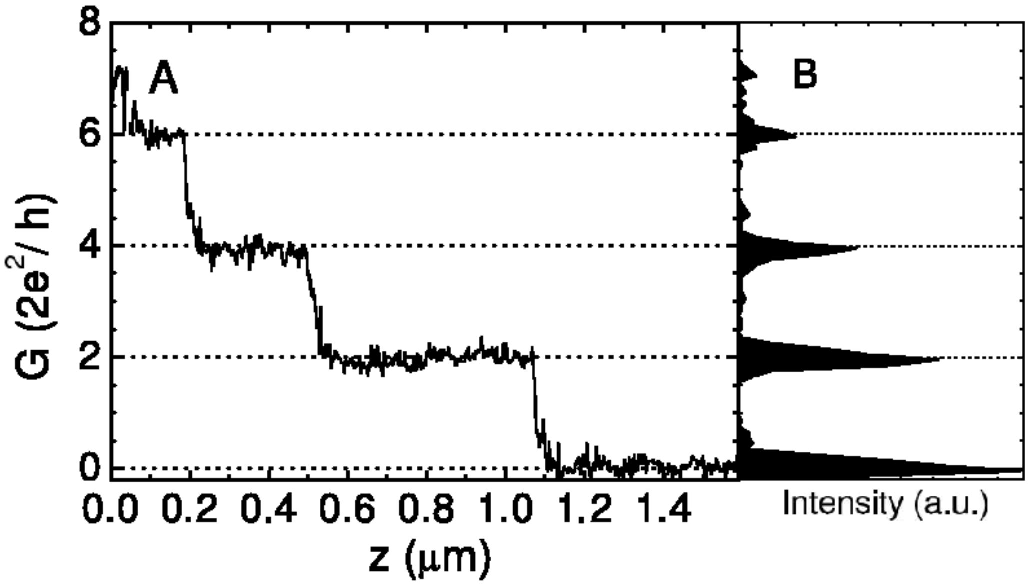
- Experiment: S. Frank et al. Science **280** 1744 (1998).
- MWCNTs ($d \sim 5-25\,nm,\ L \sim 1\mu m$) attached to a STM-tip are stepwise immersed into a liquid metal (Hg)





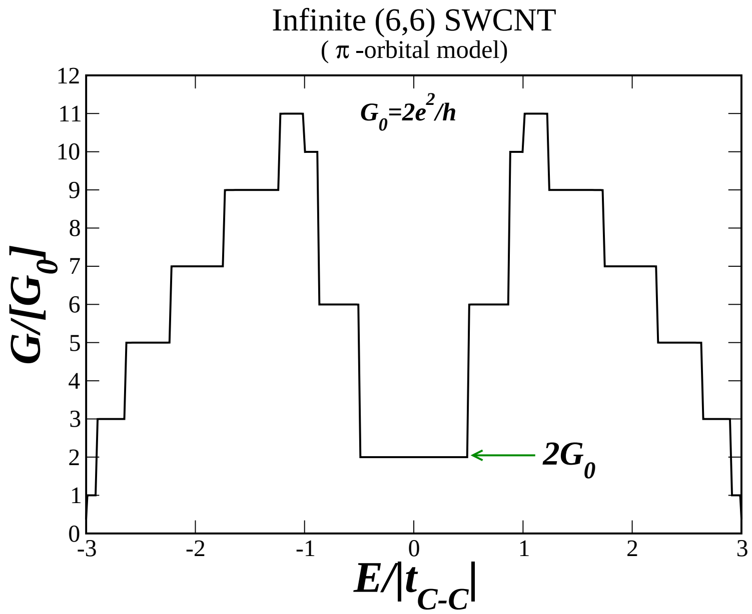
- Conductance quantization in MWCNTs observed, but starting with $\mathbf{1} \times G_0 (= 2\frac{e^2}{h})$.

- Experiment: Urbina et al. PRL **90** 106603 (2003)
- MWCNTs : STM-contacted MWCNTs deposited on a Gold-surface
- Conductance in multiples of $\mathbf{2}\times G_0 \rightsquigarrow$ only the outer shell is conducting



We know ···

- Conductance of an **infinite** metallic SWCNT $= 2 \times G_0 = 4\frac{e^2}{h} \rightsquigarrow$ two electronic bands π and π^* (each 2fold degenerate) crossing the Fermi level at k_F



- Can this be extrapolated to finite size tubes? What about MWCNTs?

- What is the effect of the contacts?

Green functions + Landauer theory

$$G = \frac{2e^2}{h} T(E_F) = G_0 T(E_F), \qquad k_B T = 0$$

How to calculate $T(E)$? Meir/Wingreen, PRL **68** , 2512 (1992)

$$H = \underbrace{\underbrace{-t_{\pi\pi} \sum_{l,j} c_l^\dagger c_j}_{H_{\mathcal{M}}}}_{\pi-\text{orbital model}} + \underbrace{\underbrace{\sum_{\mathbf{k}} \sum_{\alpha \in L,R} \epsilon_{\mathbf{k}}^{\alpha} d_{\mathbf{k}\alpha}^\dagger d_{\mathbf{k}\alpha}}_{H_\alpha}}_{\text{single}-\text{orbital model}} + \underbrace{\sum_{i,\mathbf{k}} \sum_{\alpha \in L,R} V_{i,\alpha} c_i^\dagger d_{\mathbf{k}\alpha} + c.c.}_{V_{\alpha,\mathcal{M}}}$$

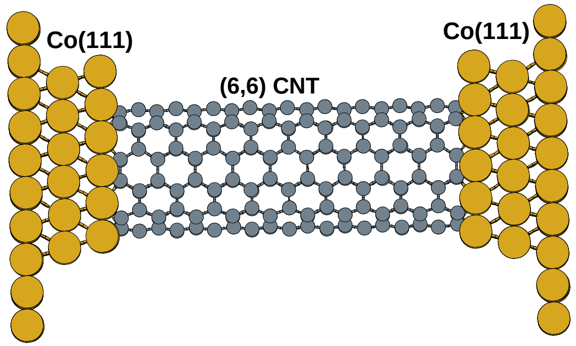
$$\boxed{T = Tr_{\mathcal{M}}[\mathbf{G}_{\mathcal{M}}^\dagger \mathbf{\Gamma}_{\mathbf{R}} \mathbf{G}_{\mathcal{M}} \mathbf{\Gamma}_{\mathbf{L}}]}$$

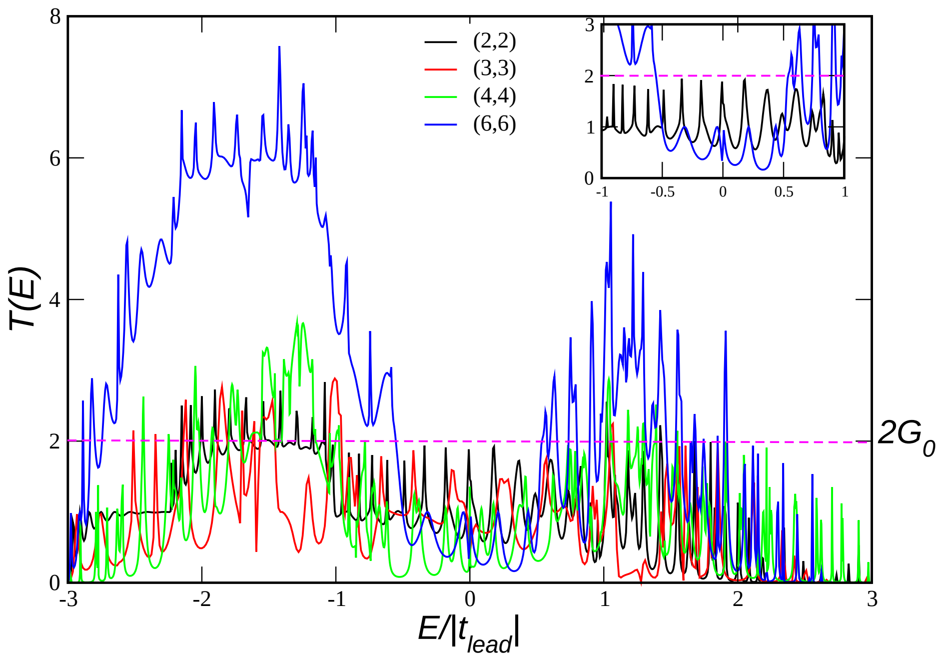
$\mathbf{G}_{\mathcal{M}} =$ molecular GF $\quad \Rightarrow [E\mathbf{1}_{\mathcal{M}} - \mathbf{H}_{\mathcal{M}} - \mathbf{\Sigma}_{\mathbf{L}} - \mathbf{\Sigma}_{\mathbf{R}}]\mathbf{G}_{\mathcal{M}} = \mathbf{1}$

$\mathbf{\Gamma}_\alpha =$ spectral densities $\quad \Rightarrow -i\mathbf{\Gamma}_\alpha = \mathbf{\Sigma}_\alpha - \mathbf{\Sigma}_\alpha^\dagger, \quad \mathbf{\Sigma}_\alpha = \mathbf{V}^\dagger \mathbf{G}_{surf}^\alpha \mathbf{V}$

(n,n) SWCNTs , π-orbital model , $t_{C-C} = t_{Co-Co}$=-2.66 eV

n.n. C-Co coupling = V = const.





- Strong oscillations related to finite size effects

- For (2,2) **both** (π, π^*) channels open, for (6,6) only **one** channel open ?

What happens?

$\rightsquigarrow$ Represent the Selfenergy $\Sigma = V^\dagger \mathcal{G}_{surf} V$ in the MO-basis of the CNT and uses WF of the lead's surface:

$$|\Phi_\sigma> = \sum_n c_{n,\sigma}|\phi_n> \, , |\phi_n> \in \{CNT\} \qquad |\mathbf{k}> = \sum_{\mathbf{l}_{||}} e^{i\mathbf{k}_{||}\mathbf{l}_{||}} f(\mathbf{k}_\perp)|\chi_{\mathbf{l}_{||}}>, \qquad |\chi_{\mathbf{l}_{||}}> = |\chi_{\mathbf{l}_{||},\mathbf{l}_\perp=\mathbf{1}}>$$

$$<\Phi_\sigma|\Sigma(E)|\Phi_{\sigma'}> = \Sigma_{\sigma\sigma'}(E) = \sum_{\mathbf{k}} <\Phi_\sigma|V^\dagger|\mathbf{k}> \mathcal{G}_{surf}(\mathbf{k},E) <\mathbf{k}|V|\Phi_{\sigma'}>$$

$$\Sigma_{\sigma\sigma'}(E) = \sum_{\mathbf{l}_{||},\mathbf{m}_{||}} \sum_{\mathbf{k}_{||}} [\sum_{\mathbf{k}_\perp} f^\dagger(\mathbf{k}_\perp) f(\mathbf{k}_\perp)\mathcal{G}_{surf}(\mathbf{k},E)] \, e^{i\mathbf{k}_{||}(\mathbf{l}_{||}-\mathbf{m}_{||})} \, V^\dagger_\sigma(\mathbf{m}_{||}) \quad \underbrace{V_{\sigma'}(\mathbf{l}_{||})}_{<\sigma'|V|\chi_{\mathbf{l}_{||},\mathbf{l}_\perp=\mathbf{1}}>}$$

$$= \sum_{\mathbf{k}_{||}} G_{\mathbf{k}_{||}}(E) \; [\sum_n c^\dagger_{n,\sigma} \sum_{\mathbf{m}_{||}} e^{-i\mathbf{k}_{||}\mathbf{m}_{||}} V^\dagger_n(\mathbf{m}_{||})] \; [\sum_m c_{m,\sigma'} \sum_{\mathbf{l}_{||}} e^{i\mathbf{k}_{||}\mathbf{l}_{||}} V_m(\mathbf{l}_{||})]$$

For constant n.n. C-metal atom coupling $n \in \{S_{CNT}\}$, $\mathbf{m}_{||}[n] \in \{S_l\} \rightsquigarrow$

$$\Sigma_{\sigma\sigma'}(E) = |V|^2 \sum_{\mathbf{k}_{||}} G_{\mathbf{k}_{||}}(E) \underbrace{[\sum_{\mathbf{m}_{||}[n]} \sum_n c^\dagger_{n,\sigma} e^{-i\mathbf{k}_{||}\mathbf{m}_{||}}]}_{\Lambda^\dagger_\sigma(\mathbf{k}_{||})} \; \underbrace{[\sum_{\mathbf{l}_{||}[m]} \sum_m c_{m,\sigma'} e^{i\mathbf{k}_{||}\mathbf{l}_{||}}]}_{\Lambda_{\sigma'}(\mathbf{k}_{||})}$$

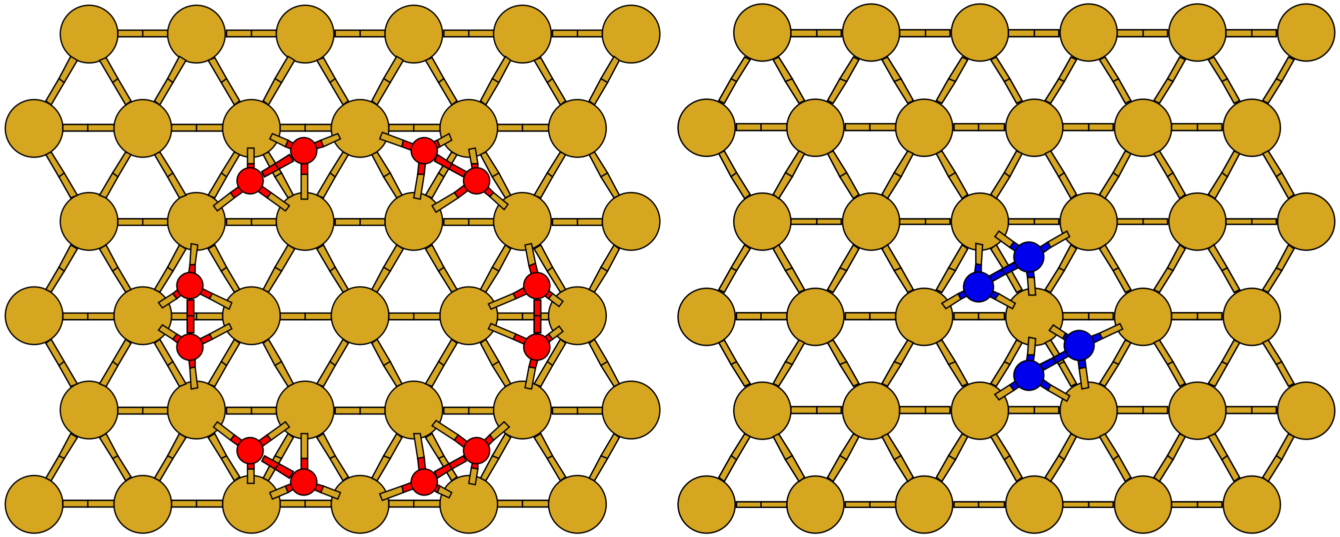
A channel may be blocked in dependence of the symmetries of $\Lambda_\sigma(\mathbf{k}_{||})$

see also: G. Cuniberti, G. Fagas, and K. Richter, Chem. Phys. **281**, 465 (2002)

J. J. Palacios et al., PRL **90**, 10680 (2003)

$$\Lambda_\sigma(\mathbf{k}_{||}) = \sum_{\mathbf{l}_{||}[m]} \sum_{\mathbf{m}} \mathbf{c}_{\mathbf{m},\sigma}\, \mathbf{e}^{\mathbf{i}\mathbf{k}_{||}\mathbf{l}_{||}}$$

$$< S|\pi >: c_{n,\pi} = 1, \qquad < S|\pi^* >: c_{n,\pi^*} = (-1)^n$$

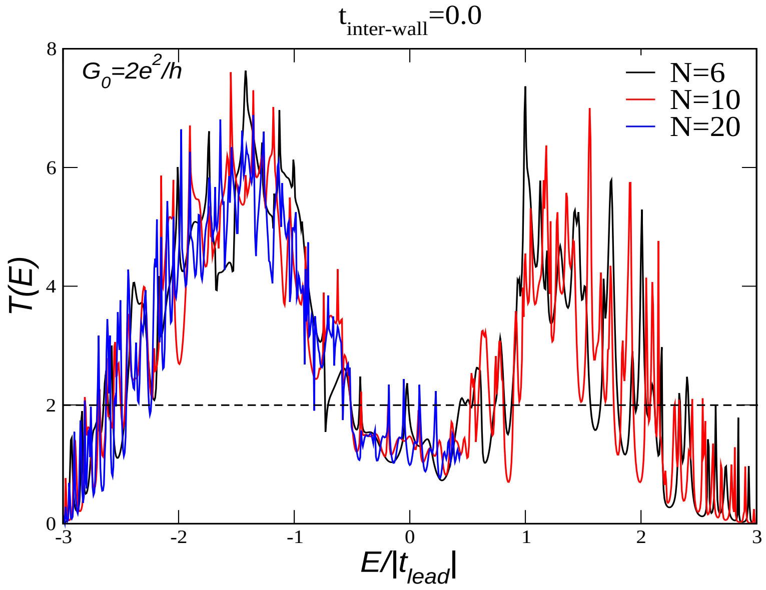


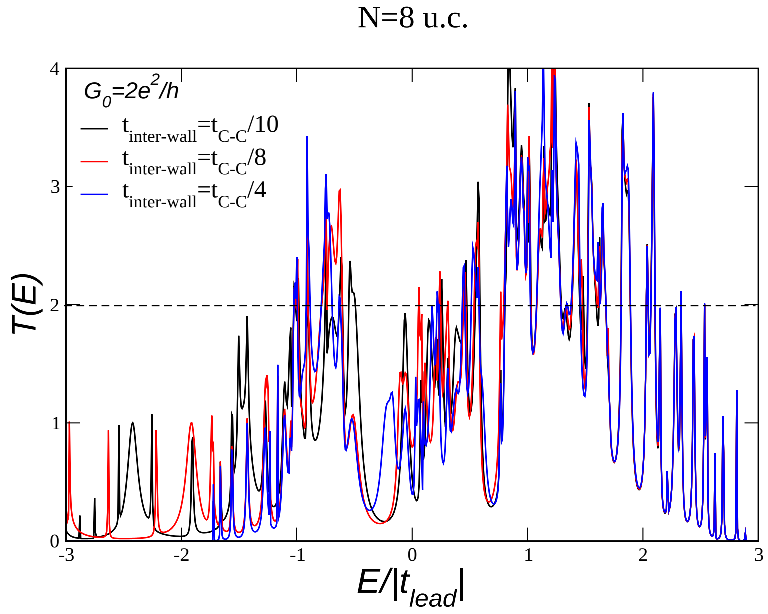
For (2,2) both channels open : $\pi, \pi^* \rightsquigarrow \Lambda_{\sigma=\pi,\pi^*}(\mathbf{k}_{||}) \neq 0$

For (6,6) the π^* **channel is closed** $\Lambda_{\sigma=\pi^*}(\mathbf{k}_{||}) = 0$

Can the channels conductances be simply added, e.g. $G_{(2,2)@(6,6)} = 3 \times G_0$**?**

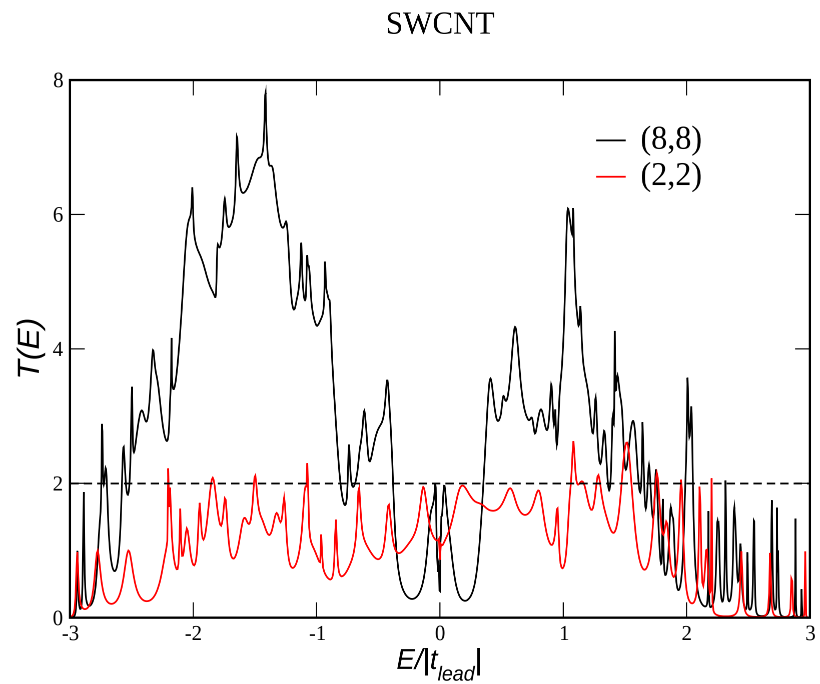
T(E) for DWCNT (2,2)@(6,6), t_{C-C}=-2.66 eV $t_{lj} = t_{inter-wall}\cos(\theta_{lj})\exp(-\delta\, r_{lj})$

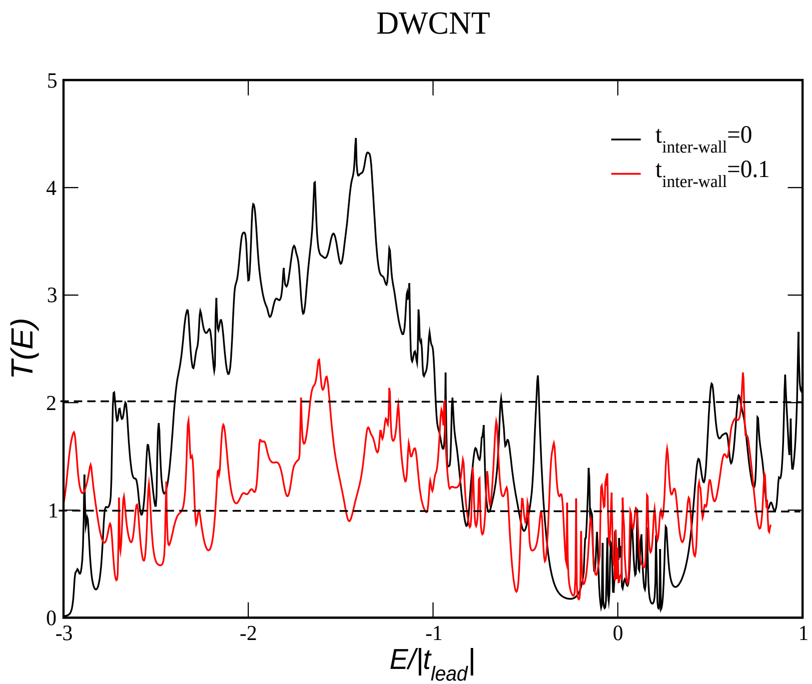




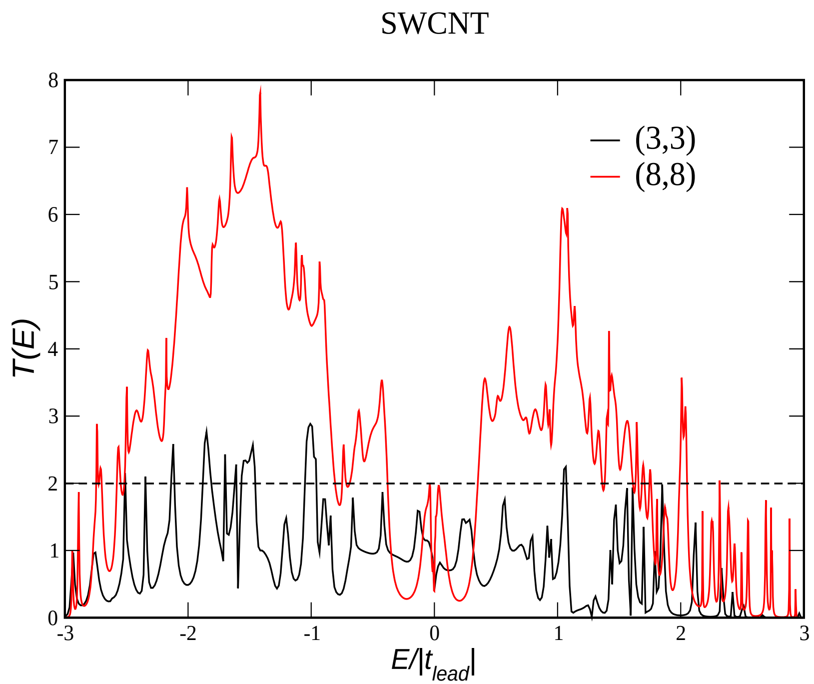
- For $(2,2)@(6,6)$ interference effects (non-diagonal $\Sigma_{\sigma,\sigma'}(E)$) $\leadsto$ at E_F $G_{(6,6)} + G_{(2,2)} \neq G_{(2,2)@(6,6)}$
- Inter-wall interactions modify $T(E)$ (mixing of CNT channels), although the behaviour $\sim E_F$ is less affected

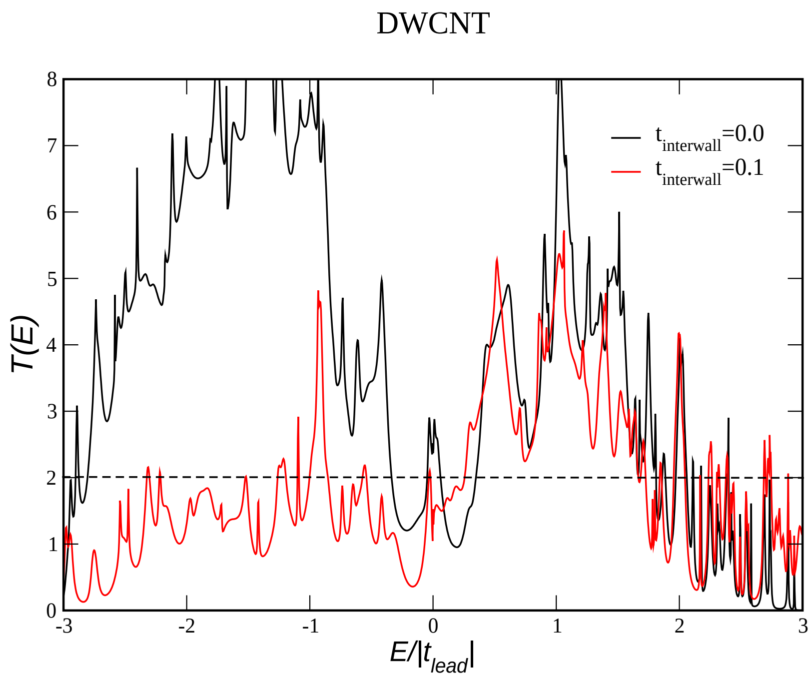
T(E) for DWCNT (2,2)@(8,8), t_{C-C}=-2.66 eV, N=6 u.c.





T(E) for DWCNT (3,3)@(8,8), t_{C-C}=-2.66 eV, N=6 u.c.

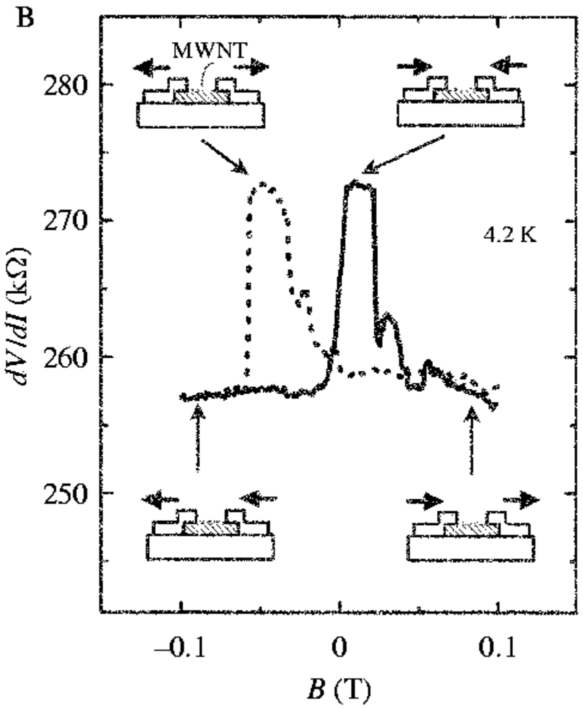




- Conductances cannot in general be simply added ⇝ mixing terms in the selfenergies ??

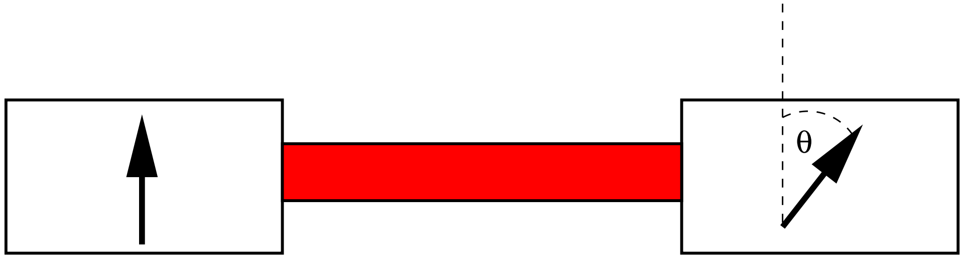
Spin-transport in DWCNTs

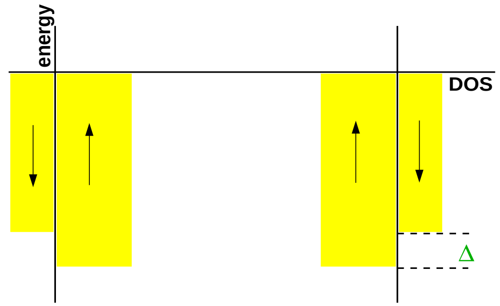
- Experiments: K. Tsukagoshi et al. Nature **401** 572 (1999), B. Zhao et al. APL **80** 3141 (2002), ibidem JAP **91** 7026 (2002), Kim et al. Physica E**18** 210 (2003)



- MR= $\frac{G_P - G_{AP}}{G_P}$
- Strong **B**-field dependence of MR $\rightsquigarrow$ spin-coherent transport in MWCNTs, spin-scattering length $l_s \sim 200\, nm - 1 \mu m$
- Negative MR observed $\sim -30\%$, sample dependent
- MR controlled by electronic band structure **AND** by interface topology (E. Y. Tsymbal et al. J. Phys:Condens. Matter **15** 109 (2003))

Model Hamiltonian: no spin-scattering, no disorder, 2-channels model





$$\mathcal{H} = \sum_{l,j} \epsilon_{lj} d_l^\dagger d_j + \sum_{\mathbf{k},\sigma=\uparrow,\downarrow} (\epsilon_\sigma(\mathbf{k}) - \underbrace{\sigma \boldsymbol{\Delta/2}}_{\text{Stoner}}) c^\dagger_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma} + \sum_{\mathbf{k}\in L,\sigma} [V_{\mathbf{k},l,\sigma} c^\dagger_{\mathbf{k},\sigma} d_l + h.c]$$

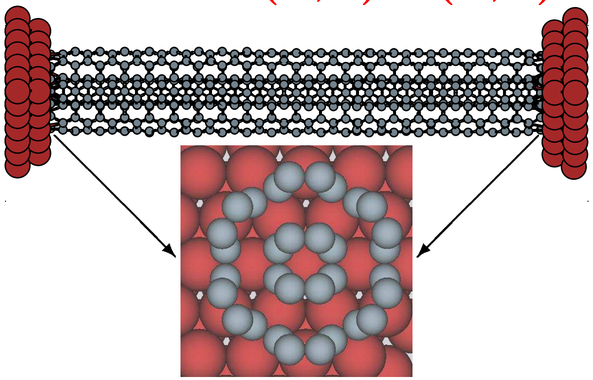
$$+ \sum_{\mathbf{k}\in R,\sigma} [V_{\mathbf{k},l,\sigma} (\sigma \cos(\frac{\theta}{2}) c^\dagger_{\mathbf{k},\sigma} + \sin(\frac{\theta}{2}) c^\dagger_{\mathbf{k},-\sigma}) d_l + V^\dagger_{\mathbf{k},l,\sigma} d_l^\dagger (\sigma \cos(\frac{\theta}{2}) c_{\mathbf{k},\sigma} + \sin(\frac{\theta}{2}) c_{\mathbf{k},-\sigma})]$$

$$\Sigma_{l=\mathrm{L(R)}} = \mathcal{R}_l(\theta) \begin{pmatrix} \Sigma^0_{l,\uparrow} & 0 \\ 0 & \Sigma^0_{l,\downarrow} \end{pmatrix} \mathcal{R}^\dagger_l(\theta), \quad \Gamma_l = \mathcal{R}_l(\theta) \begin{pmatrix} \Gamma^0_{l,\uparrow} & 0 \\ 0 & \Gamma^0_{l,\downarrow} \end{pmatrix} \mathcal{R}^\dagger_l(\theta), \quad \mathcal{R}_l(\theta) = \begin{pmatrix} \cos(\frac{\theta}{2}) & \sin(\frac{\theta}{2}) \\ -\sin(\frac{\theta}{2}) & \cos(\frac{\theta}{2}) \end{pmatrix}$$

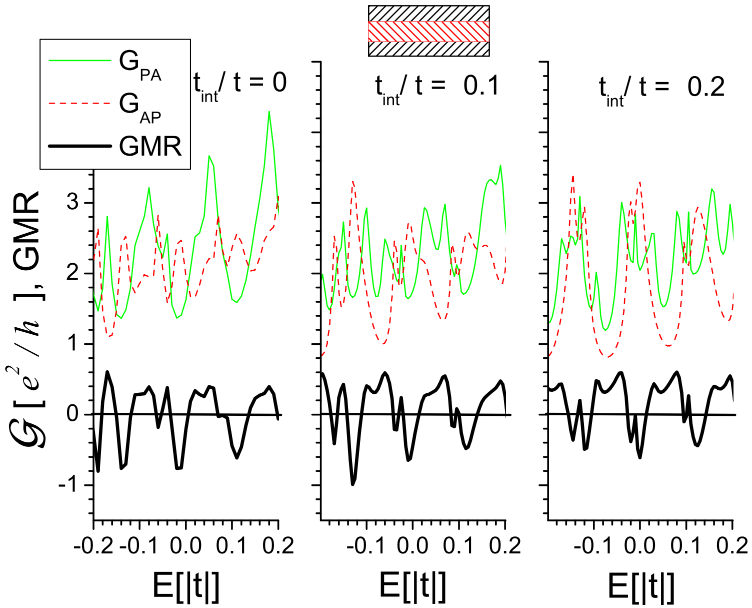
$$G(\theta) = \frac{\mathbf{e^2}}{\mathbf{h}} \sum_\sigma \mathbf{Tr}[\mathbf{G}^\dagger \boldsymbol{\Gamma}_\mathbf{R} \mathbf{G} \boldsymbol{\Gamma}_\mathbf{L}]_{\sigma,\sigma} \qquad n = \frac{1}{2\pi} \int dE\, [G f_\mathrm{L} \Gamma_L + f_R \Gamma_R G^\dagger] \rightsquigarrow \epsilon_{ll} \to \epsilon_{ll} \pm \delta\epsilon[n]$$

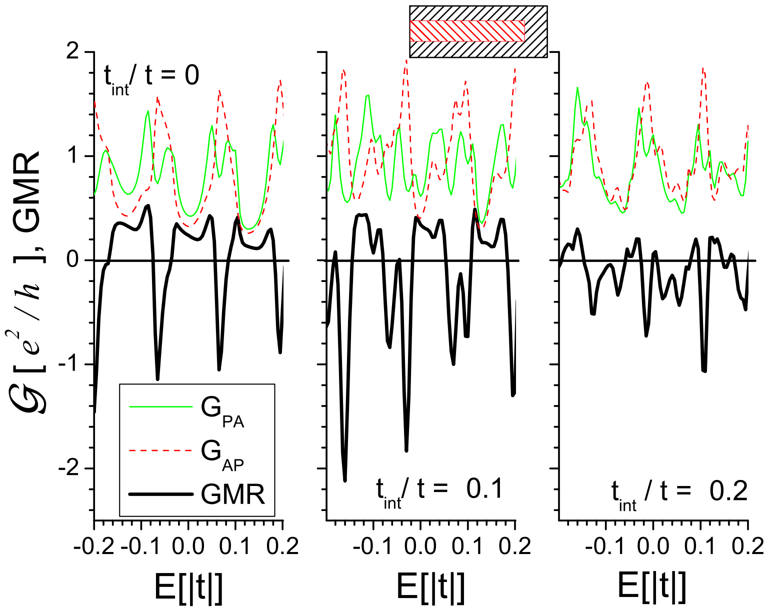
$$\mathbf{MR} = \frac{G(0) - G(\pi)}{G(0)} = \frac{G_{\uparrow,\uparrow} - G_{\uparrow,\downarrow}}{G_{\uparrow,\uparrow}}$$

DWCNT(6,6)@(2,2)



- Spin polarization $P = 0.5$
- full contact : MR < 0 (weakly dependent on t_{IW})
- Positive MR with partial contact
- Essential: charge neutrality $\rightsquigarrow$ band rearrangment at the interface



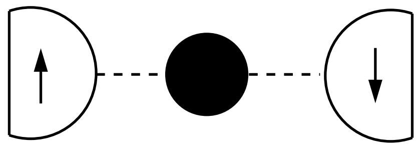


- Negative GMR $\leftrightarrow$ charge neutrality ? Toy model :

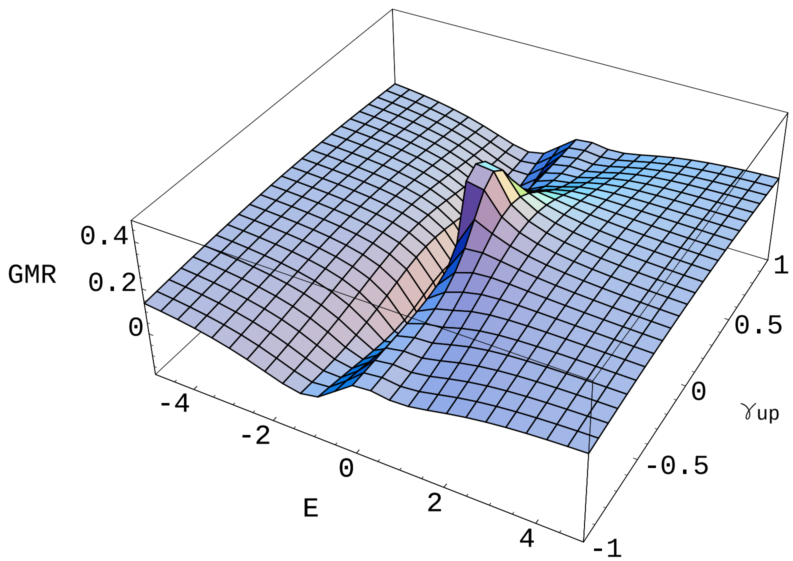
$$\Sigma_\sigma(E) = -i\gamma_\sigma, \qquad \gamma_\uparrow/\gamma_\downarrow \approx 2.0$$

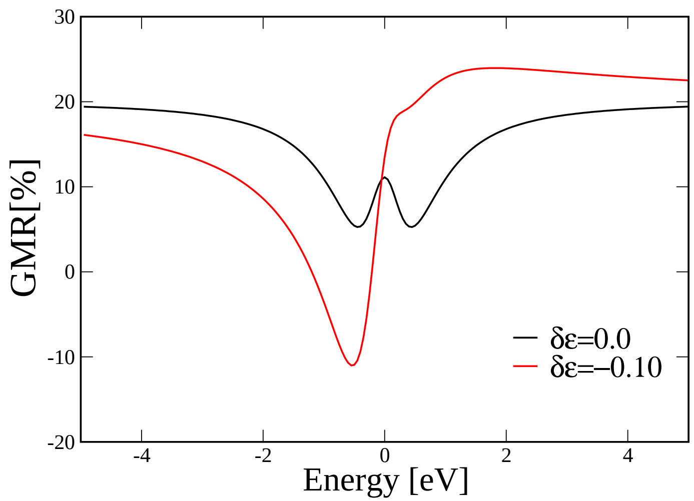
$$G(0) = G_{\uparrow\uparrow} = \frac{4e^2}{h}\left[\frac{\gamma_\uparrow^2}{(E-\epsilon_0)^2+4\gamma_\uparrow^2} + \frac{\gamma_\downarrow^2}{(E-\epsilon_0)^2+4\gamma_\downarrow^2}\right]$$

$$G(\pi) = G_{\uparrow\downarrow} = \frac{8e^2}{h}\frac{\gamma_\uparrow\gamma_\downarrow}{(E-[\epsilon_0+\delta\epsilon])^2+(\gamma_\uparrow+\gamma\downarrow)^2}$$



$\delta\epsilon$ mimics charge transfer effects





- **Modifications of the charge distribution (mainly at the metall–molecule interface) might change the sign of the GMR**

Conclusions + Perspectives

- Contact symmetries strongly modify the device conductance $\rightsquigarrow$ channel blocking
- Influence of inter-wall interactions quite strong
- How do conductances of SW combine in DWCNTs ?
- Sign of GMR closely related to charge neutrality condition $\rightsquigarrow$ band mismatch
- Stability issues $\leftrightarrow$ conductance
- What happens with $\cdots$ disorder, (n,0) CNTs , incommensurability , spin valve $G(\theta)$
- Realistic description of leads+interface electronic structure $\rightsquigarrow$ 4s,4p,3d-bands

Thanks to N. Ranjan, G. Fagas and G. Cuniberti

GMR: Prof. S. Krompiewski (IfMP Poznan)