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Ab initio studies of the transport properties of doped or functionalized nanotubes.

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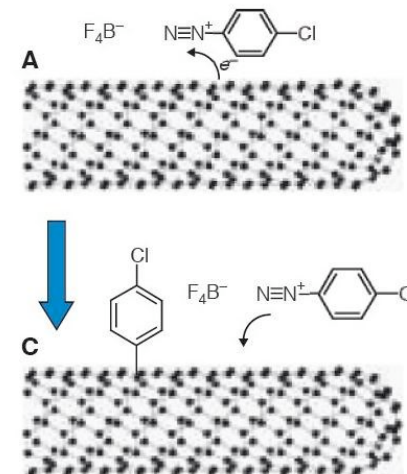
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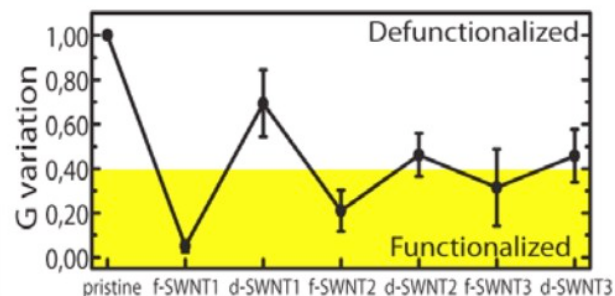
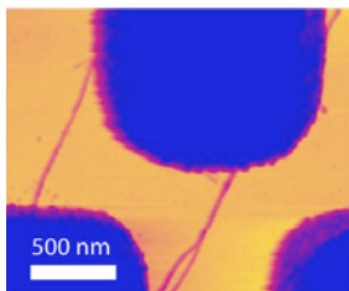
Outline: Conductance of doped/functionalized tubes/wires

Greatness and misery of doping and functionalization

Doping and/or functionalization allows to process, manipulate, assembly, select metal/semiconducting tubes, increase carrier density, add optical or sensing properties, act as a chemical gate, tune the contact resistance, etc. (too many references..)



However, in 1D systems in the small size limit, any disruption of the conducting channel is very efficient at destroying the conductance.

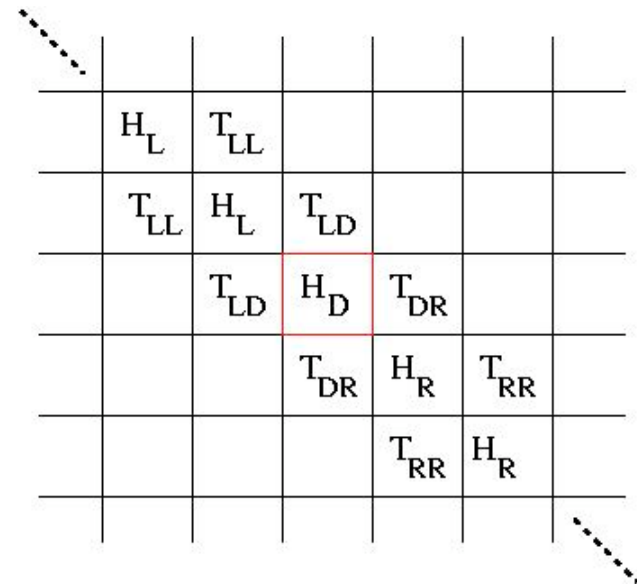
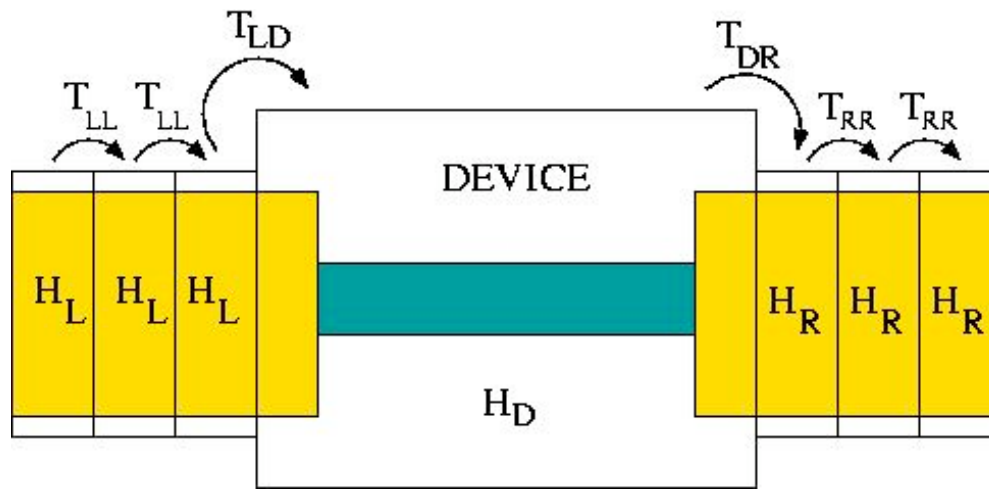


Martel, Cabana, JACS 2008.

Compromise to tune between number of functional groups and conductance.

Code **TABLIER**: Transport AB initio in Lyon in the Elastic Regime
 (Hamiltonian expressed on a strictly localized basis)

(Adessi, Blase,
2006)



$$G_D(\varepsilon) = (\varepsilon - H_D - \Sigma_R - \Sigma_L)^{-1}$$

$$\Sigma_L = T_{LD}^\dagger G_L T_{LD}$$

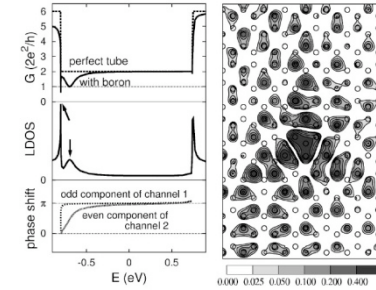
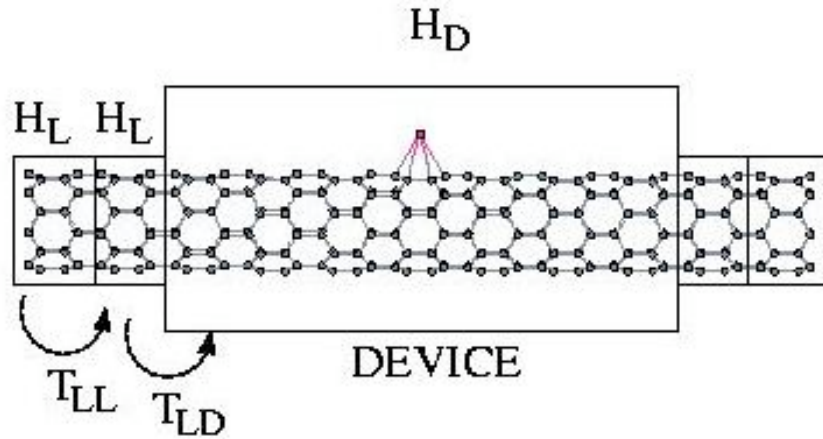
« left » self-energy

$$G_L^{(n+1)} = [(\varepsilon - H_L) - T_{LL}^\dagger G_L^{(n)} T_{LL}]^{-1} \quad \text{« left-electrode » } G$$

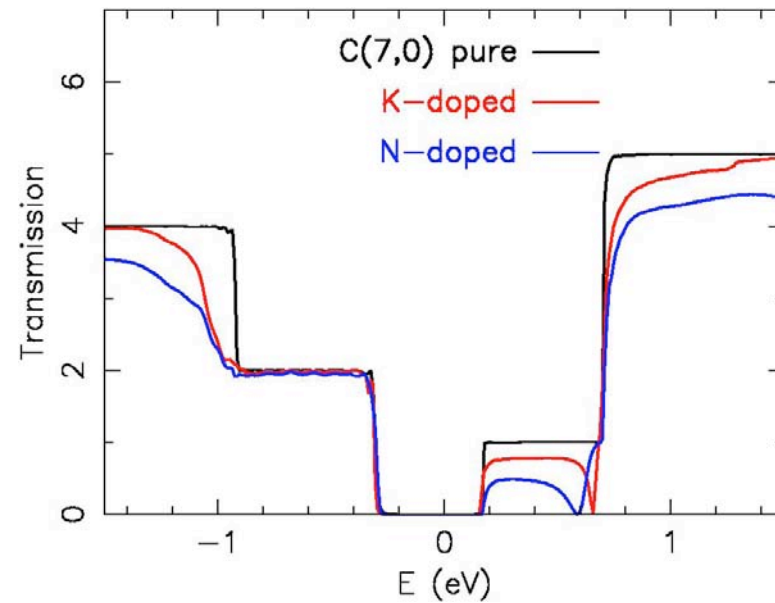
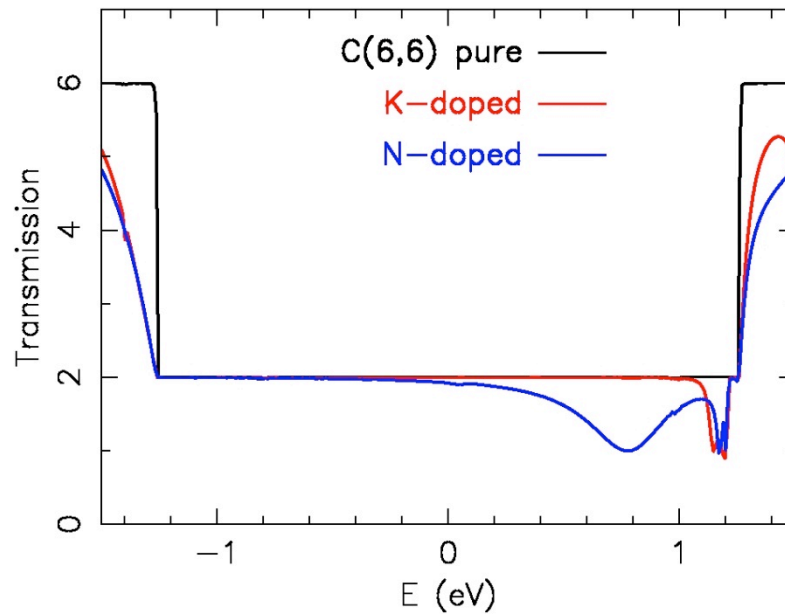
The conductance can be calculated (iteratively) within order-N

Application : *n*-type doping for carbon nanotubes in FETs

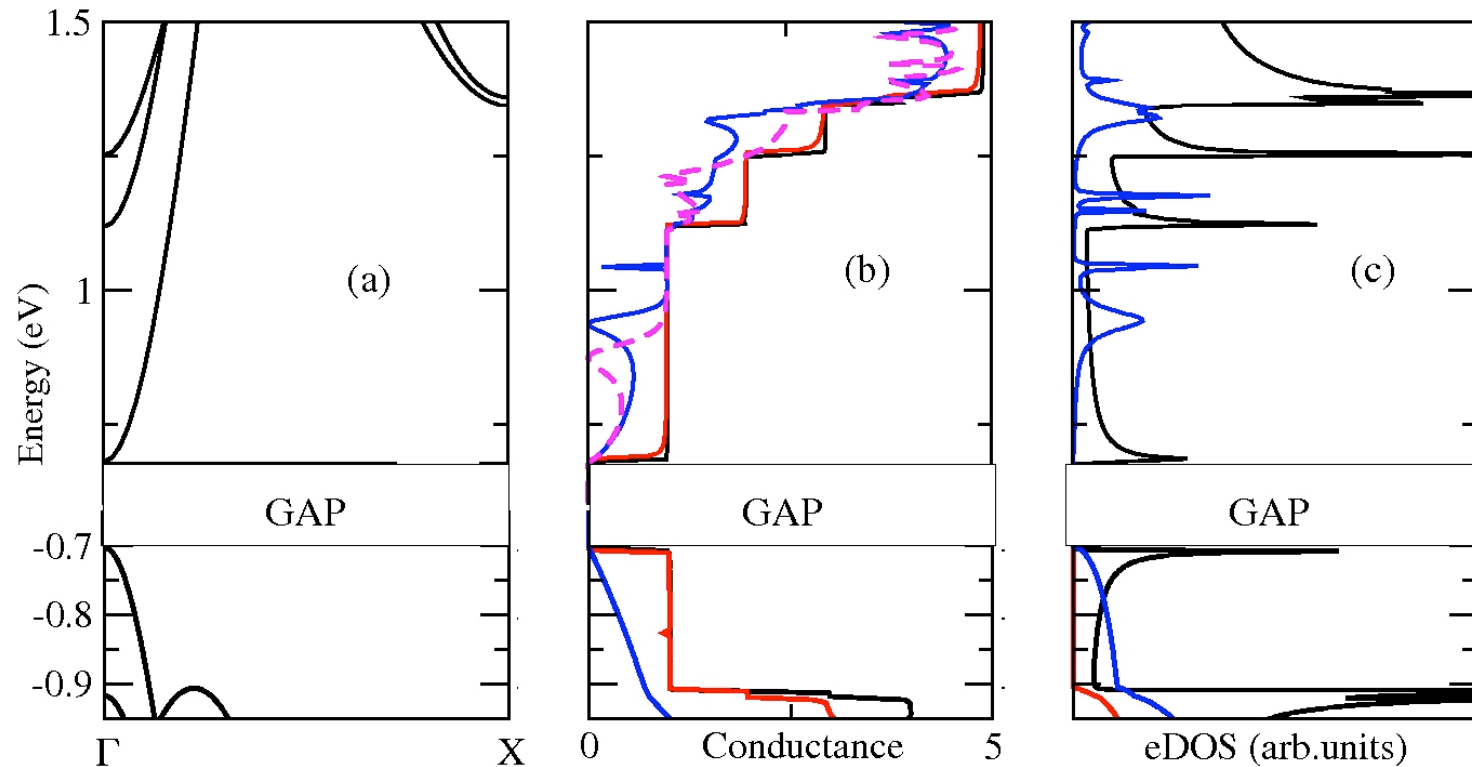
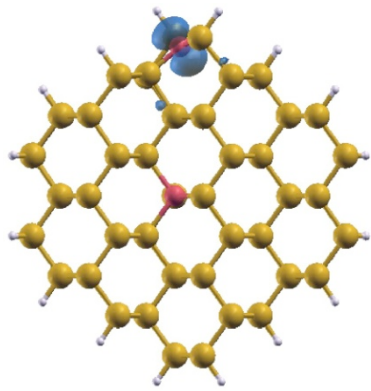
K doping (K doping: Javey, NanoLett. 2005; Appenzeller, PRL 2004; Radosavljevic, APL 2004)
Graphene: Chen *et al.*, Nature Physics 2008.



(Choi, Ihm, Louie, Cohen, PRL 2000)



Adessi, Roche, Blase, PRB 2006

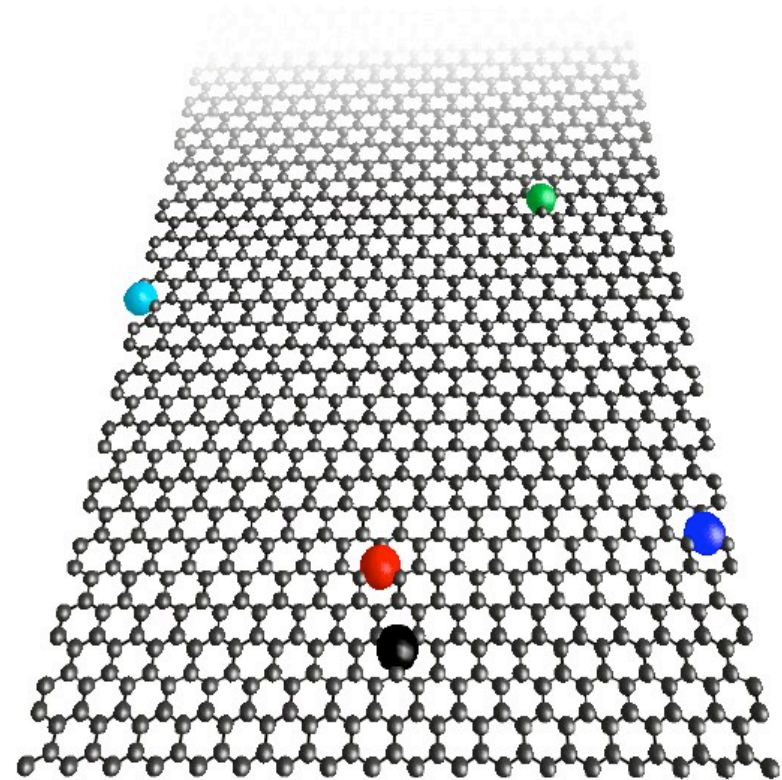
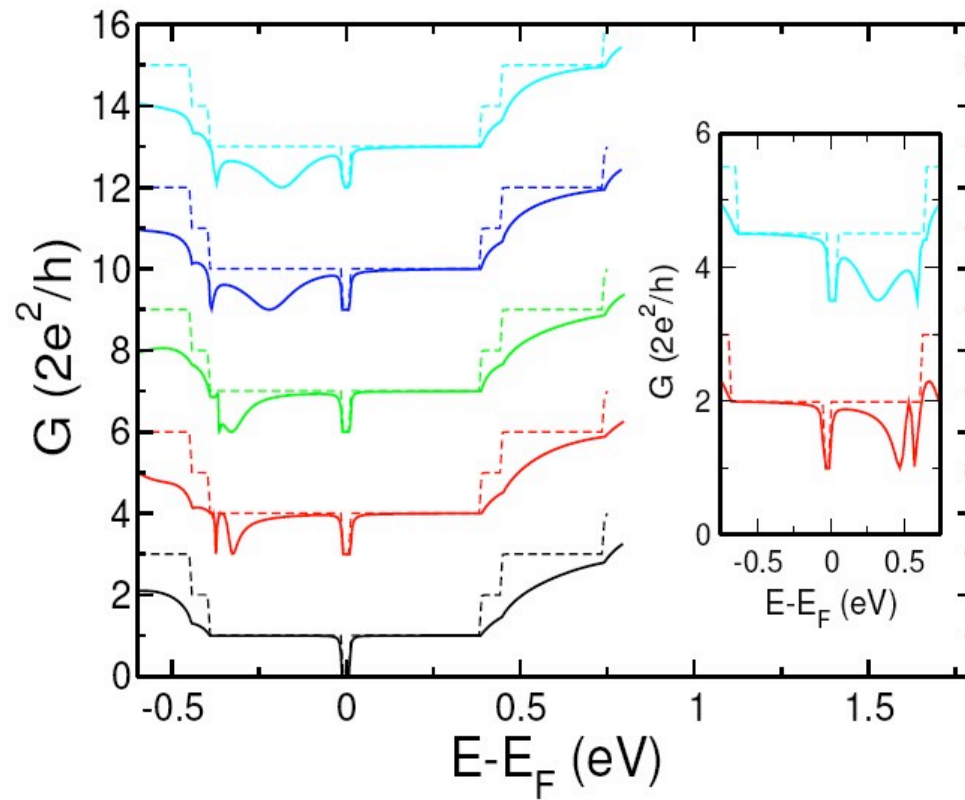


Blue: phosphorus at the core of the wire Violet dashed: phosphorus on surface Si atom

Strong resonant backscattering induced by dopants. Resonant energies depend on radial localization => importance of core-shell strategies.

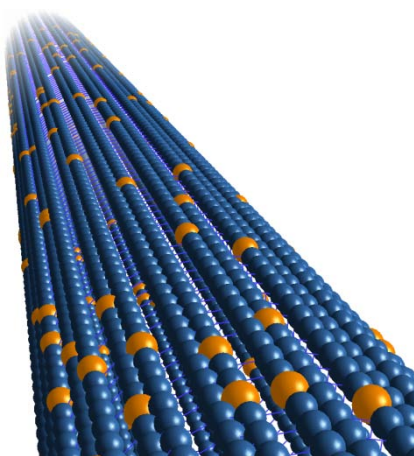
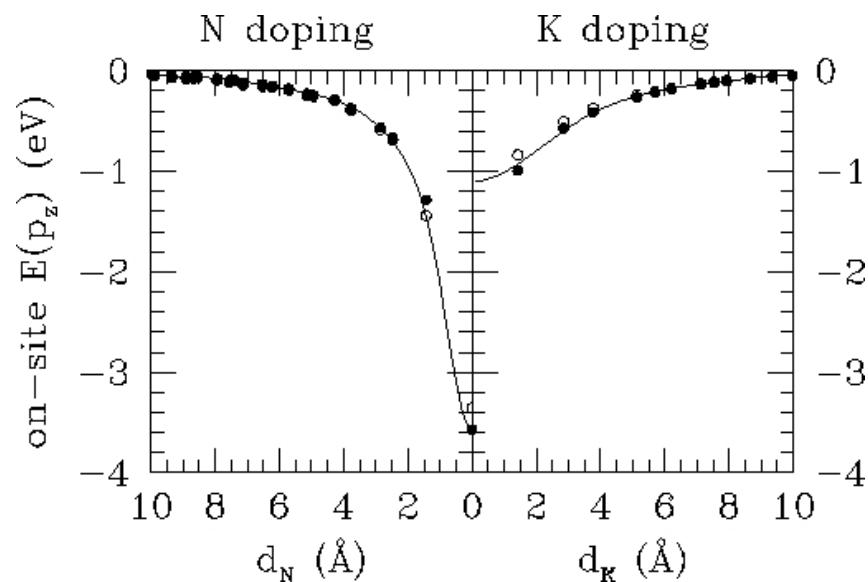
(see also: Markussen et al., PRL 2007)

Transport in doped nanoribbons



(B. Biel, X.B. F. Triozon, S. Roche, 2008)

Towards large scale simulations: coupling *ab initio* and TB

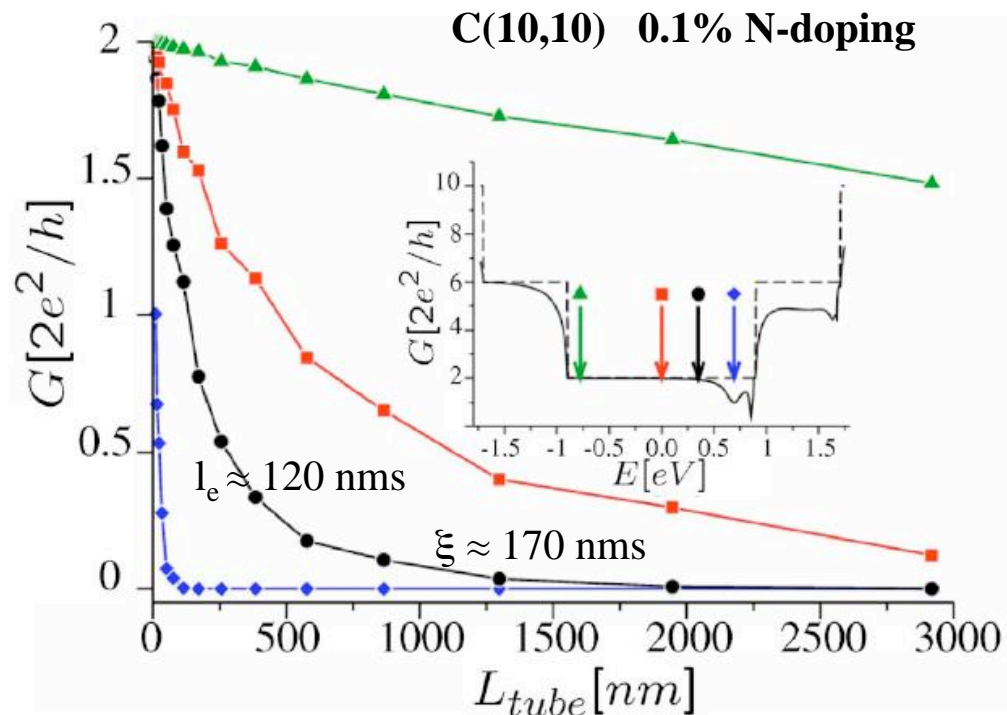


Avriller *et al.*, PRB RapComm 2006.

Use *ab initio* on-site/hopping p_z terms to build TB π - π^* model

Tight-Binding

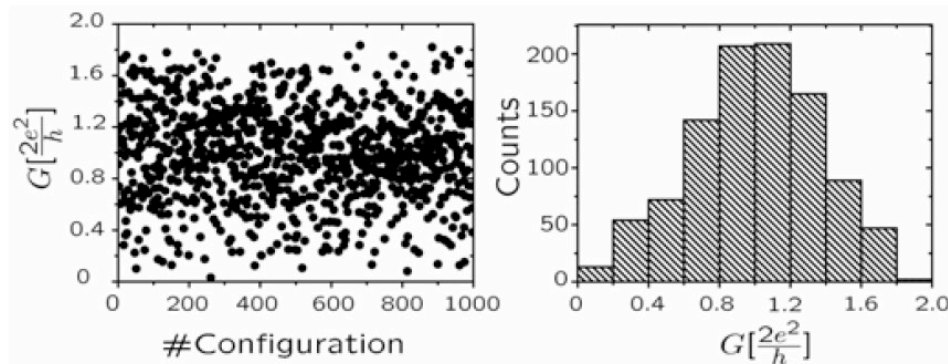
Study random doping of long tubes



Mesoscopic physics analysis: sample-to-sample fluctuations

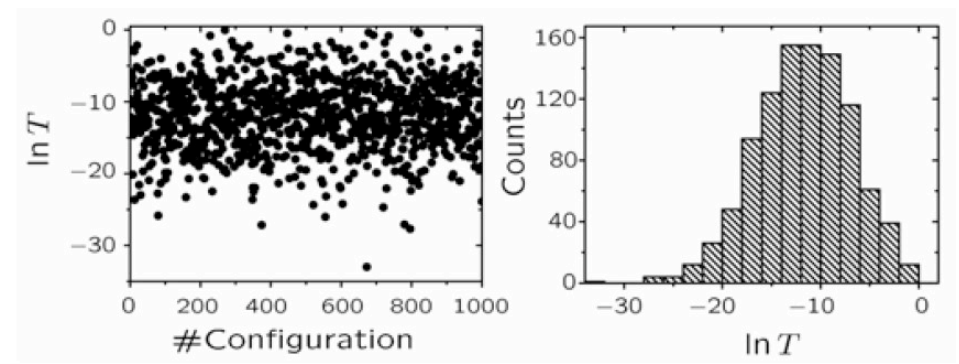
The analysis of the transport properties requires an average over dozens of doping configurations and an analysis of the conductance fluctuations.

(0.1% N-doping of (10,10) tube, black line previous graph: $l_e \sim 120$ nms and $\xi \sim 170$ nms)



Diffusive regime ($L = 120$ nms)

$$\bar{T} = N_C \frac{1}{1 + L/l_e}$$

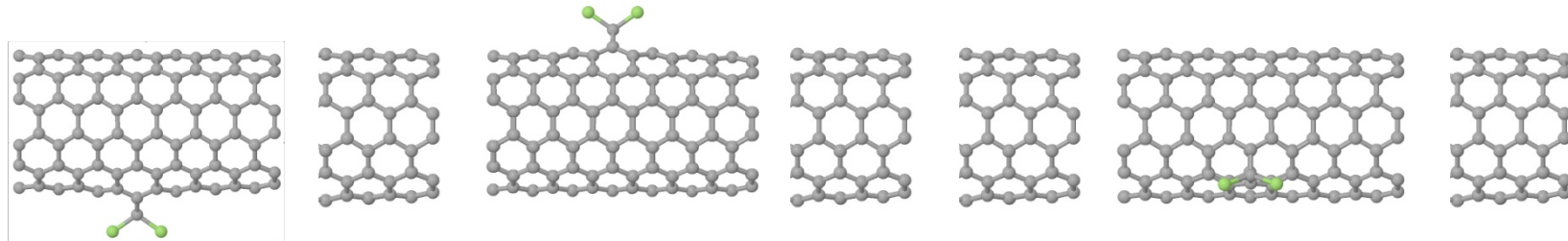


Localisation regime ($L = 400$ nms)

$$\ln T = -L/\xi$$

(large fluctuations of T,
Gaussian-like for $\ln T$)

Towards mesoscopic physics with *ab initio* calculations:



In the linear regime, recursive techniques on sparse Hamiltonian matrix allows to calculate within $O(N)$ the (off-diagonal) Green's function matrix elements connecting left and right leads.

(code Tbsim developed in Grenoble: Triozon, Niquet, Roche et al.)

μ -meter long tubes (a few 10^4 atoms) can be dealt with *ab initio*

See also:

- Gomez-Navarro *et al.*, Nature Mater. 4, 534 (2005).
- Lee, Nardelli, Marzari, PRL (2005, 2006).
- A.R. Rocha, M. Rossi, A. Fazzio *et al.*, PRL (2008).
- Markussen, T; Rurali, R; Jauho, AP, *et al.*, PRL (2008).

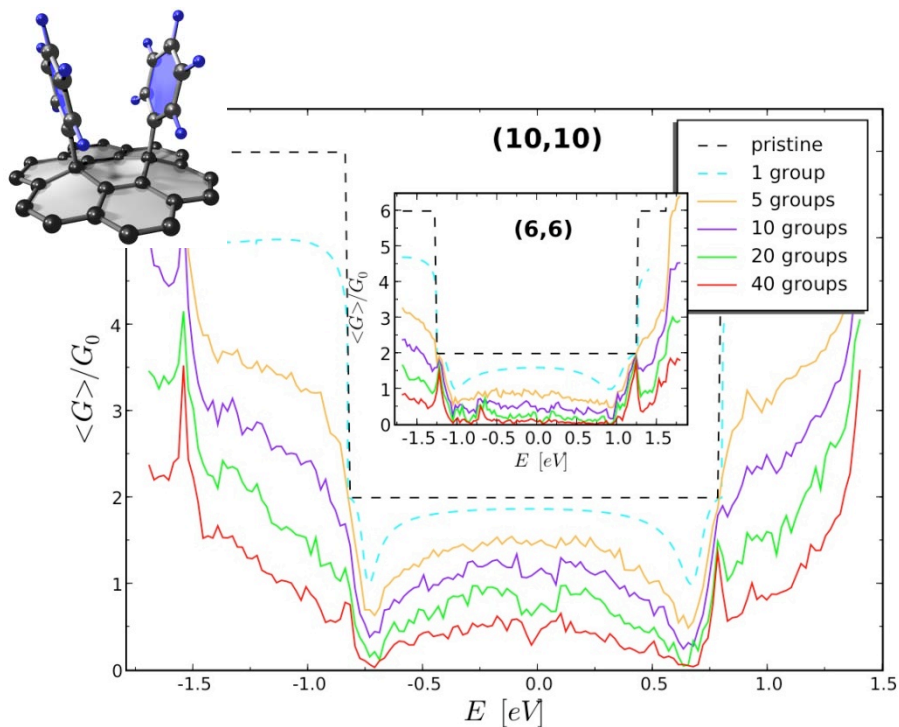
Chemistry matters: on the right choice of functional groups for tubes.



Lee, Nardelli, Marzari, PRL 2005, 2006.

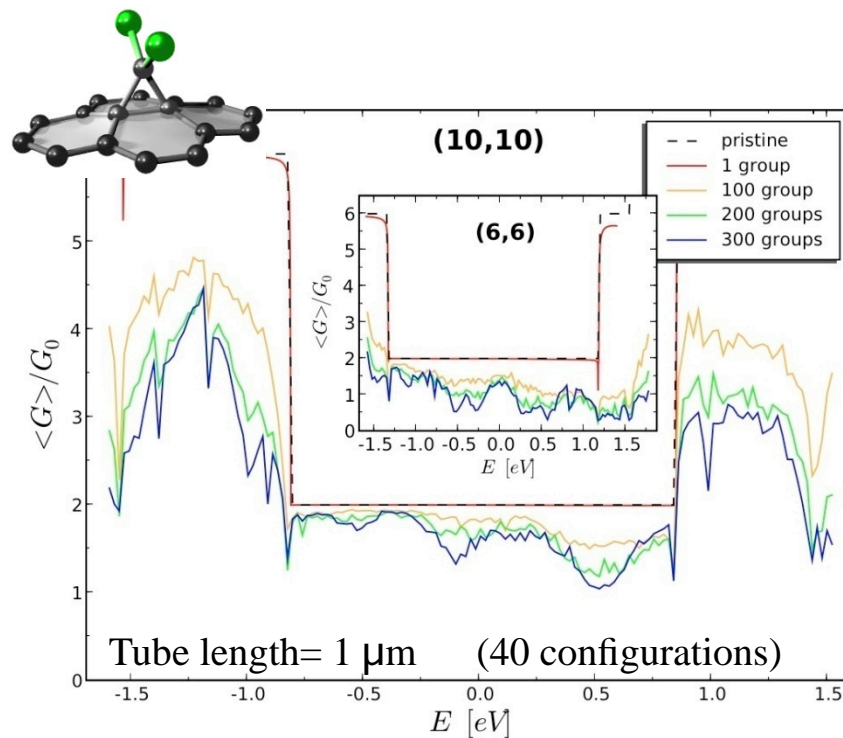
Phenyls

Synthesis: Strano et al. Science 2003; Bahr et al., JACS 2001; Cabana, Martel, JACS 2007; Campidelli et al., Small 2007; etc.



Carbene

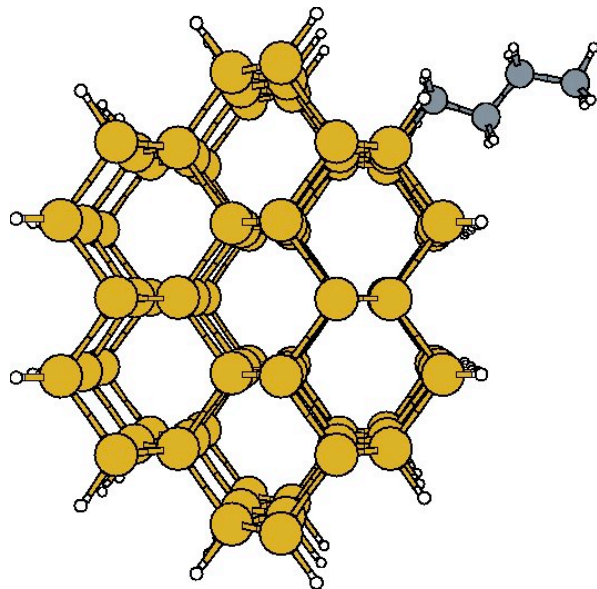
Synthesis: Kamaras et al., Science 2003; Hu et al., JACS 2003; Coleman et al., JACS 2003; Chen et al. Science 1998; etc.



(Alejandro Lopez-Bezanilla, F. Triozon, S. Latil, XB, S. Roche, 2008)

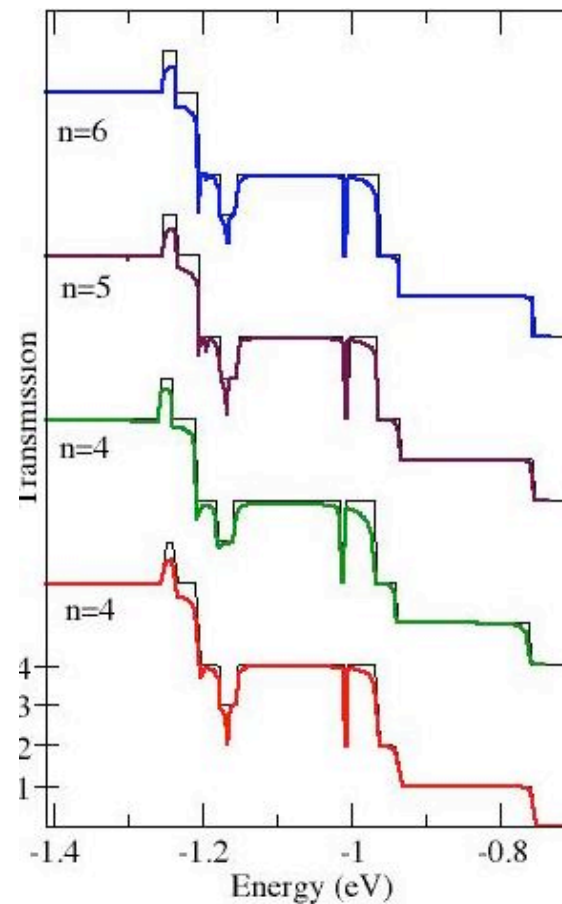
Conductance of functionalized nanowires

(Fernández-Serra, Blase, PRL 2008)

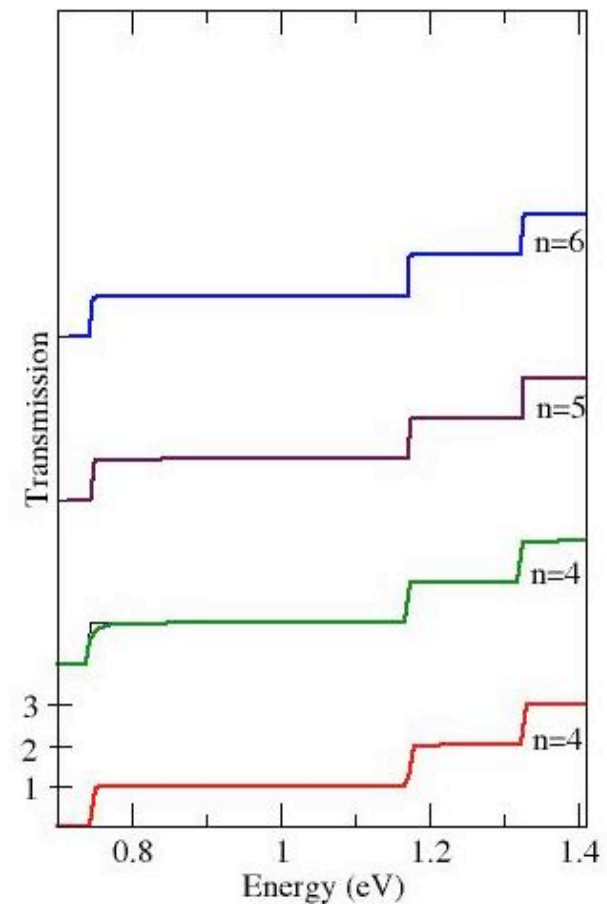


$\langle 110 \rangle$ SiNW

Valence bands



Conduction bands



Covalent functionalization of SiNWs by common “linkers” or “spacers” does not affect the conductance even in the small diameter limit.

Conclusions

Ab initio study of transport within the Landauer approach:

- **Ab initio calculations can help in optimizing doping or functionalization strategies**
- **Mesoscopic physics on realistic tubes (μms et randomly doped or functionalized) can be tackled with first-principle approaches.**

Thanks.