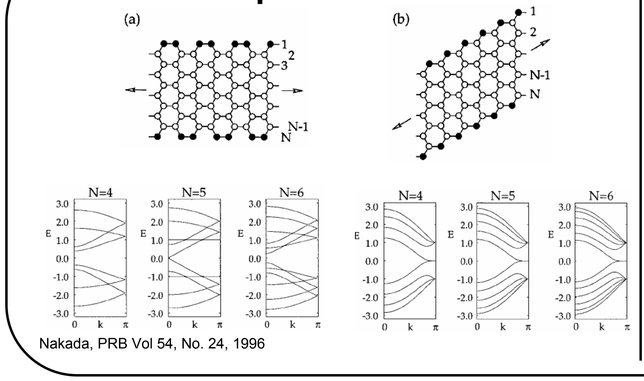
Ab initio calculation of a graphene-ribbon-based molecular switch

Luis Agapito, and Haiping Cheng
Department of Physics and Quantum Theory Project, University of Florida, Gainesville, Florida

Abstract

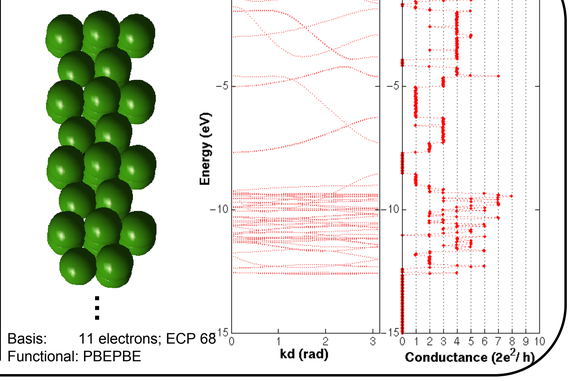
We perform ab initio calculations of electronic and transport properties of nano-electronic systems. The surface Green's function of semi-infinite ribbons made from graphene sheets is constructed from ab initio one-dimensional bulk calculations, and the energy dependent transmission coefficient through a molecular tunneling junction is computed from Landauer's formalism within a framework that combines Caroli's approach and density functional theory. By using the same level of theory and basis set to model both the molecule and the ribbons, and by an exact, analytical determination of surface Green's functions, we aim for a quantitative description of the configuration dependent electron transport through ribbon-molecule-ribbon switching devices.

1-D lead: Graphene ribbons



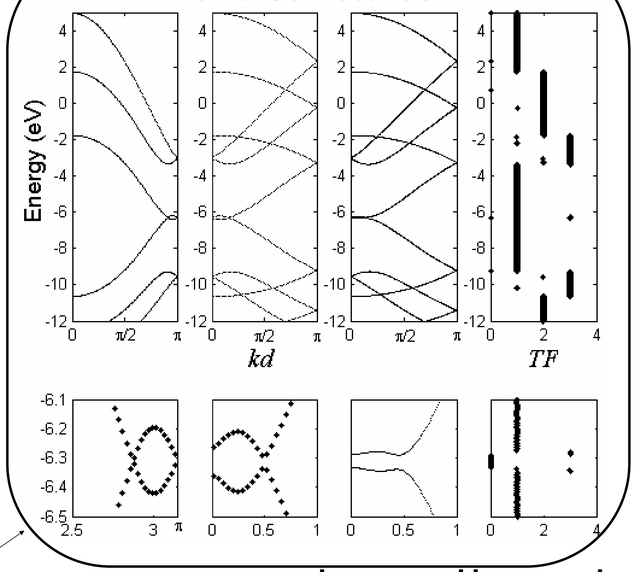
Nakada, PRB Vol 54, No. 24, 1996

Gold wires



Basis: 11 electrons; ECP 68
Functional: PBEPBE

Band Structures

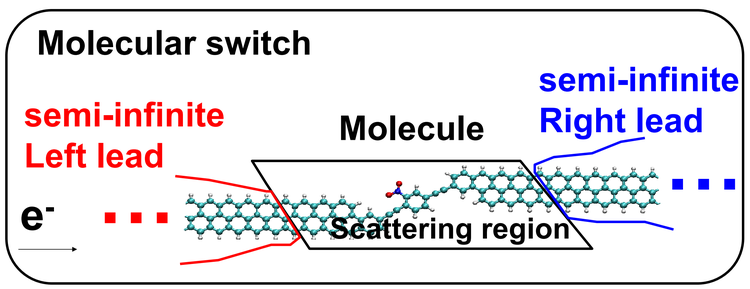


Full DFT/PBC

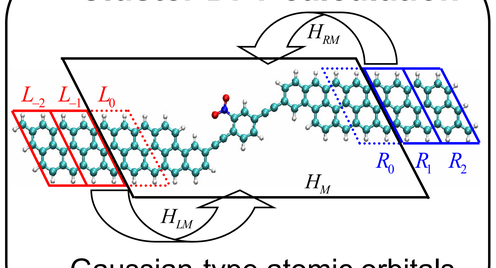
Recreated from H_{00} and H_{10}

Transmission through the infinite wire

Molecular switch

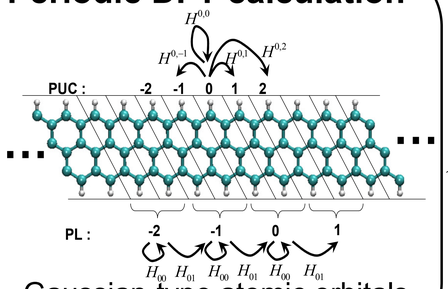


Cluster DFT calculation



Gaussian-type atomic orbitals
Gaussian 03

Periodic DFT calculation



Gaussian-type atomic orbitals
Gaussian 03

Lead's self-energies

The Green's function ($\mathcal{G}$) of the infinite system is defined from:

$$\begin{bmatrix} \varepsilon\mathcal{S}_L-\mathcal{H}_L & \varepsilon\mathcal{S}_{LM}-\mathcal{H}_{LM} & 0 \\ \varepsilon\mathcal{S}_{ML}-\mathcal{H}_{ML} & \varepsilon S_M-H_M & \varepsilon\mathcal{S}_{MR}-\mathcal{H}_{MR} \\ 0 & \varepsilon\mathcal{S}_{RM}-\mathcal{H}_{RM} & \varepsilon\mathcal{S}_R-\mathcal{H}_R \end{bmatrix}\begin{bmatrix} \mathcal{G}_L & \mathcal{G}_{LM} & \mathcal{G}_{LR} \\ \mathcal{G}_{ML} & G_M & \mathcal{G}_{MR} \\ \mathcal{G}_{RL} & \mathcal{G}_{RM} & \mathcal{G}_R \end{bmatrix}=\begin{bmatrix} \mathcal{I} & 0 & 0 \\ 0 & I_M & 0 \\ 0 & 0 & \mathcal{I} \end{bmatrix}$$

The Green's function of the Molecule is: $G_M(\varepsilon)=[\varepsilon S_M-H_M-\Sigma_L-\Sigma_R]^{-1}$

The self-energy for the Left lead is:

$$\Sigma_L=[\varepsilon\mathcal{S}_{ML}-\mathcal{H}_{ML}][\varepsilon\mathcal{S}_L-\mathcal{H}_L]^{-1}[\varepsilon\mathcal{S}_{LM}-\mathcal{H}_{LM}]$$
$$=[\varepsilon S_{ML}-H_{ML}][G_L^0][\varepsilon S_{LM}-H_{LM}]$$

Surface Green's function (SGF)

How to calculate the SGF ?

It is the last block of the inverse of a semi-infinite matrix

$$[\varepsilon\mathcal{S}_{L/R}-\mathcal{H}_{L/R}]^{-1}=\begin{bmatrix} \ddots & \vdots & \vdots & \vdots \\ \cdots & \times & \times & \times \\ \cdots & \times & \times & \times \\ \cdots & \times & \times & G_{L/R}^0 \end{bmatrix}$$

Lee et al., PRB 23, 4997 (1981).
Sanvito et al., PRB 59, 11936 (1999).
Krstic et al., PRB 66 205319 (2002).

Infinite molecular junction ab initio Hamiltonian

$$\mathcal{H}^{junction}=\begin{bmatrix} \mathcal{H}_L & \mathcal{H}_{LM} & & \\ \mathcal{H}_{ML} & H_M & \mathcal{H}_{MR} & \\ & \mathcal{H}_{RM} & \mathcal{H}_R & \end{bmatrix}$$

$$\mathcal{H}_L=\begin{bmatrix} H_{00} & H_{01} & 0 \\ H_{10} & H_{00} & H_{01} \\ 0 & H_{10} & H_{00} \end{bmatrix},\quad \mathcal{H}_{LM}=\begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ H_{LM} & 0 & 0 & 0 \end{bmatrix}$$

$$\mathcal{H}_{ML}=\begin{bmatrix} 0 & 0 & H_{ML} \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix},\quad H_M=\begin{bmatrix} H_M & H_{MR} & 0 & 0 \\ H_{RM} & H_{00} & H_{01} & 0 \\ 0 & H_{10} & H_{00} & H_{01} \\ 0 & 0 & H_{10} & H_{00} \end{bmatrix},\quad \mathcal{H}_{MR}=\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ H_{MR} & 0 & 0 \end{bmatrix}$$

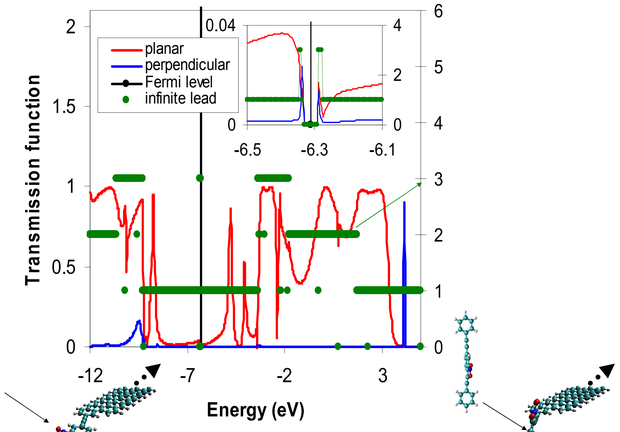
$$\mathcal{H}_{RM}=\begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & H_{RM} \end{bmatrix},\quad \mathcal{H}_R=\begin{bmatrix} H_{00} & H_{01} & 0 \\ H_{10} & H_{00} & H_{01} \\ 0 & H_{10} & H_{00} \end{bmatrix}$$

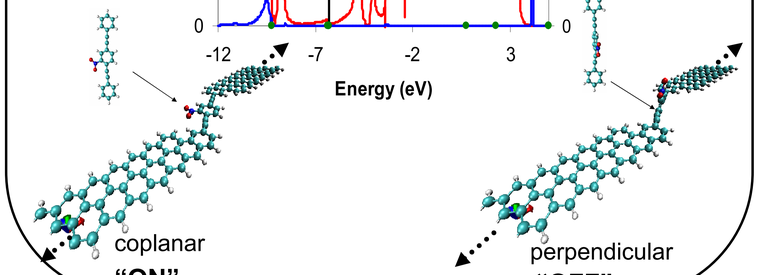
Transport calculation

- Analytical, exact surface Green's functions $G_M(\varepsilon)=[\varepsilon S_M-H_M-\Sigma_L-\Sigma_R]^{-1}$
- Junction's Green's function
- Transport $\quad \mathrm{TF}=trace(\Gamma_L G_M \Gamma_R G_M^{\dagger})$

Molecular switch calculation

$$\text{Transmission Function}=trace(\Gamma_L G_M \Gamma_R G_M^{\dagger})$$





coplanar
"ON"

perpendicular
"OFF"