Transport Calculations with TranSIESTA

Pablo Ordejón

Instituto de Ciencia de Materiales de Barcelona - CSIC, Spain

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M. Brandbyge, K. Stokbro and J. Taylor

Mikroelectronik Centret - Technical Univ. Denmark

Jose L. Mozos and Frederico D. Novaes

Instituto de Ciencia de Materiales de Barcelona - CSIC, Spain

The SIESTA team

E. Anglada, E. Artacho, A. García, J. Gale. J. Junquera, D. Sánchez-Portal, J. M. Soler, ...

Outline

1. Electronic transport in the nanoscale: Basic theory
2. Modeling: the challenges and our approach:
    - The problem at equilibrium (zero voltage)
    - Non-equilibrium (finite voltage)
3. Practicalities

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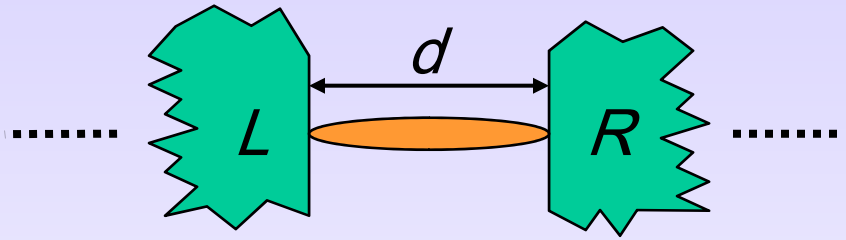
1. Electronic Transport in the Nanoscale: Basic Theory



- Scattering in nano-scale systems:
  - electron-electron interactions
  - phonons
  - impurities, defects
  - elastic scattering by the potential of the contact

$$d \ll L_m$$

$$d \approx \lambda$$

- Semiclassical theory breaks down – QM solution needed
- Landauer formulation: Conductance as transmission probability

S. Datta, *Electronic transport in mesoscopic systems* (Cambridge)

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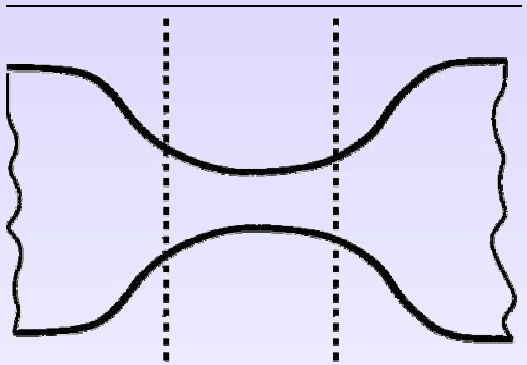
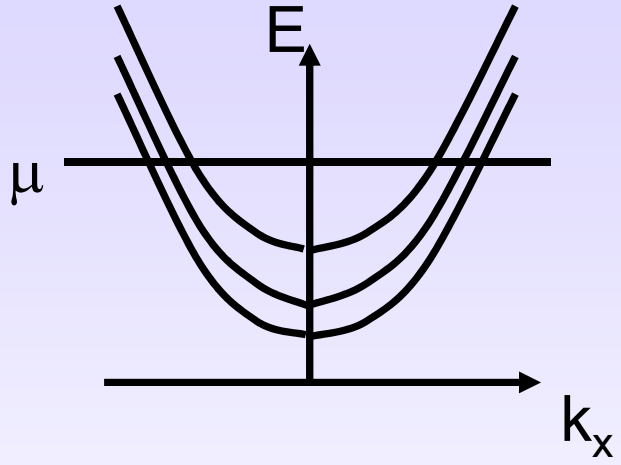
Narrow constriction (meso-nanoscopic)





Transversal confinement ⟹ QUANTIZATION

$$\psi(x,y) = \psi_m(y)\, e^{ik_x x} \qquad E_m(k_x) = \varepsilon_m + \frac{\hbar^2 k_x^2}{2m}$$

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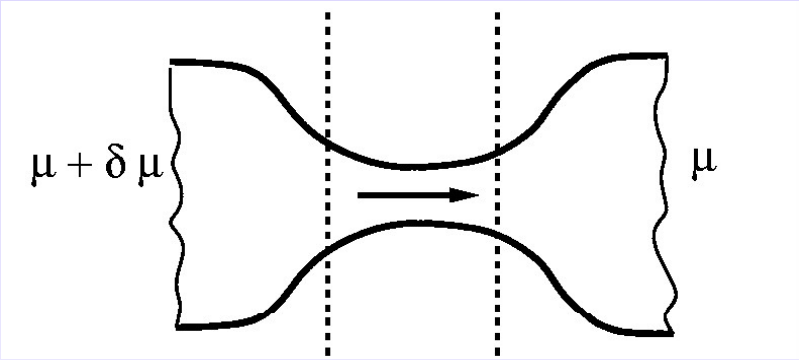
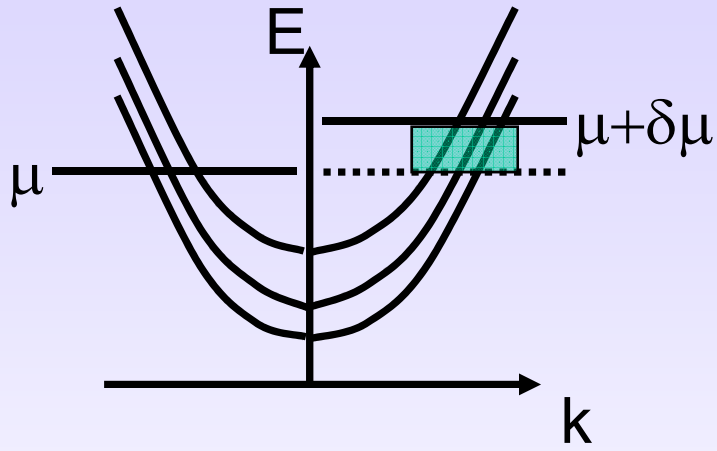
Landauer formulation - no scattering





$$\delta\mu = -eV$$

$$I = -2e \sum_{i}^{bands} \int_{BZ} d\mathbf{k}\, f(\mathbf{k}) v_g(\mathbf{k},i) \approx \frac{2e^2}{h} V N_{bands}(E_F)$$

$$G = \frac{I}{V} = N_{bands} \frac{2e^2}{h} = N_{bands} G_0$$

QUANTUM OF CONDUCTANCE

$$G_0 = \frac{2e^2}{h} \Rightarrow 12.9\ k\Omega$$

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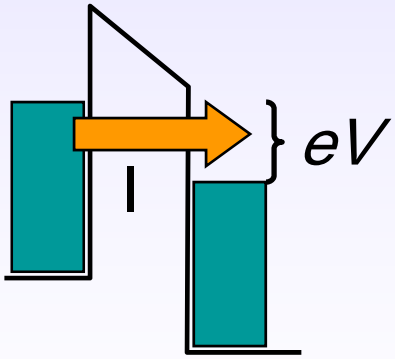
Landauer formulation - scattering

- Transmission probability of an incoming electron at energy ε:

$$T(\varepsilon) = Tr\left[t^{\dagger} t\right](\varepsilon)$$

transmission matrix: $\psi_{out} = t\,\psi_{in}$

- Current:



$$I = \frac{2e^2}{h} \int d\varepsilon \, \left(f_L(\varepsilon) - f_R(\varepsilon)\right) T(\varepsilon)$$

- Perfect conductance (one channel): T=1

$$G_0 = \frac{2e^2}{h}$$

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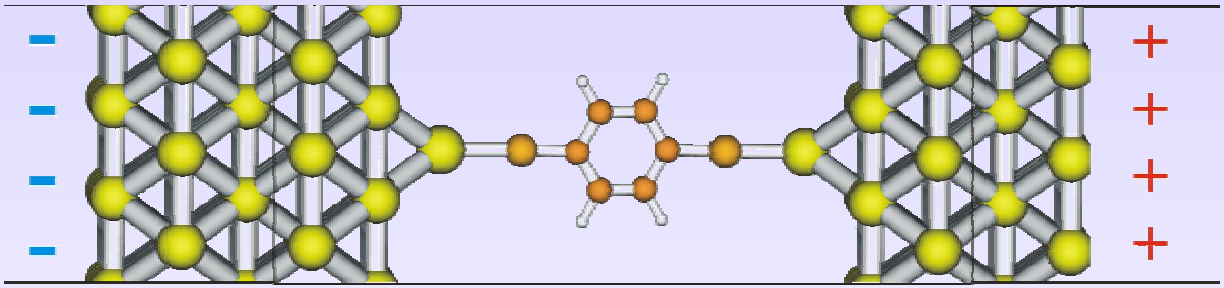
2. Modeling:
The Challenges and Our Approach



- Model the molecule-electrode system from *first principles:*

  No parameters fitted to the particular system → DFT

- Model a **molecule** coupled to **bulk (infinite) electrodes**
- Electrons **out of equilibrium** (do not follow the thermal Fermi occupation)
- Include **finite bias** voltage/current and determine the **potential profile**
- Calculate the **conductance** (quantum transmission through the molecule)
- Determine **geometry**: Relax the atomic positions to an energy minimum

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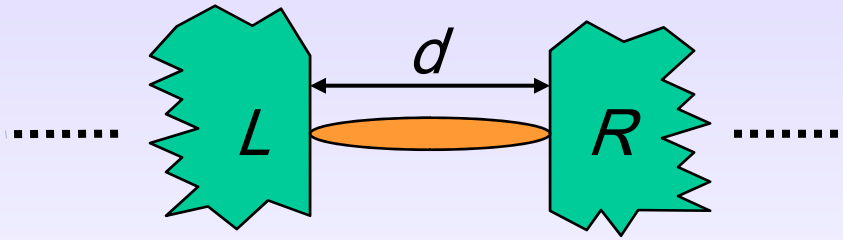
Restriction: Ballistic conduction

- Ballistic conduction: consider only the scattering of the incoming electrons by the potential created by the contact



- Two terminal devices (three terminals in progress)

- Effects not described: inelastic scattering
  - electron-electron interactions (Coulomb blockade)
  - phonons (current-induced phonon excitations)

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First Principles: DFT

- Many interacting-electrons problem mapped to a one particle problem in an external **effective potential** (Hohemberg-Kohn, Kohn-Sham)

$$V_{eff} = V_{ext} + V_{ps} + V_H + V_{XC}$$

- Charge density as basic variable:

$$V_{eff} = V_{eff}[\rho(\mathbf{r})]$$

- Self-consistency: $V_{eff} \leftrightarrow \rho$
- Ground state theory: V_{XC}

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SIESTA

http://www.uam.es/siesta

Soler, Artacho, Gale, García, Junquera, Ordejón and Sánchez-Portal

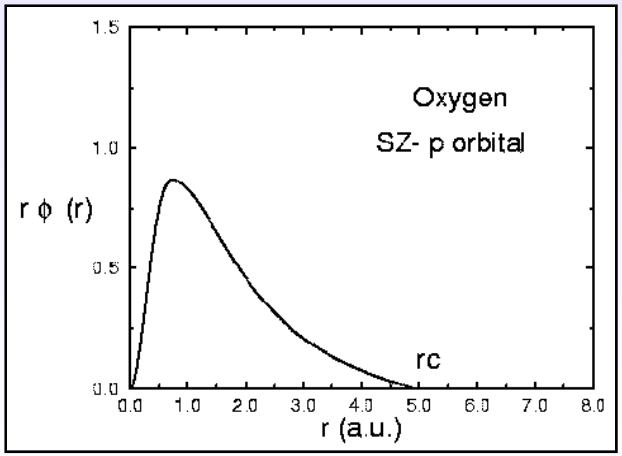
J. Phys.: Cond. Matt. **14**, 2745 (2002)

- Self-consistent DFT code (LDA, GGA, LSD)
- Pseudopotentials (Kleinman-Bylander)
- LCAO approximation:

  Basis set:

  Confined Numerical Atomic Orbitals

  $\varphi_\mu(\boldsymbol{r})$ Sankey's "fireballs"



- Order-N methodology (in the calculation and the solution of the DFT Hamiltonian)

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TranSiesta

Implementation of non-equilibrium electronic transport in SIESTA

- **Atomistic description** (both contact and electrodes)
- **Infinite electrodes**
- Electrons **out of equilibrium**
- Include **finite bias** and determine the **potential profile**
- Calculates the **conductance** (both linear and non-linear)
- **Forces** and **geometry**

Brandbyge, Mozos, Ordejón, Taylor and Stokbro
Phys. Rev. B. **65**, 165401 (2002)

Mozos, Ordejón, Brandbyge, Taylor and Stokbro
Nanotechnology **13**, 346 (2002)

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The problem at Equilibrium (Zero Bias)

Challenge:

Coupling the finite contact to infinite electrodes

Solution:

Green's Functions

$$G(z) = (z - H)^{-1}$$

$$\rho(\varepsilon) = -\frac{1}{\pi} Im\, G(\varepsilon + i\delta)$$

$$\rho(\boldsymbol{r}) = \sum_{\mu\nu} \underbrace{\left[\int_{-\infty}^{\infty} d\varepsilon\, \rho_{\mu\nu}(\varepsilon) n_F(\varepsilon - \mu_F) \right]}_{D_{\mu\nu}} \varphi_\mu(\boldsymbol{r}) \varphi_\nu(\boldsymbol{r})$$

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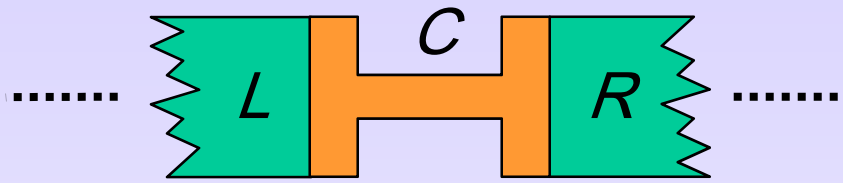
Setup (zero bias)

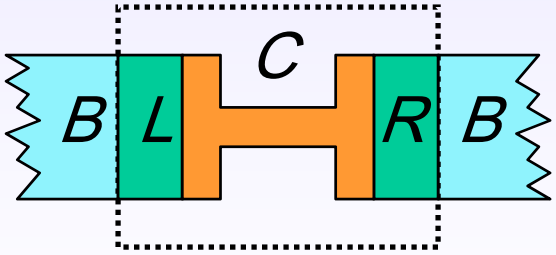


$$G(\varepsilon)=(\varepsilon-H)^{-1}=$$

$$\begin{bmatrix} \ldots & -V_L^{+} & & & \\ -V_L^{-} & \varepsilon-H_L & -V_{LC} & & \\ & -V_{CL} & \varepsilon-H_C & -V_{CR} & \\ & & -V_{RC} & \varepsilon-H_R & -V_R^{+} \\ & & & -V_R^{-} & \ldots \end{bmatrix}^{-1}$$

Contact:

- Contains the molecule, and part of the Right and Left electrodes
- Sufficiently large to include the screening



Solution in finite system:

$$G(\varepsilon)=\begin{bmatrix} \varepsilon-H_L-\Sigma_L & -V_{LC} & \\ -V_{CL} & \varepsilon-H_C & -V_{CR} \\ & -V_{RC} & \varepsilon-H_R-\Sigma_R \end{bmatrix}^{-1}$$

$\Sigma(\varepsilon)$ = Selfenergies. Can be obtained from the bulk Greens functions

Lopez-Sancho et al. J. Phys. F **14**, 1205 (1984)

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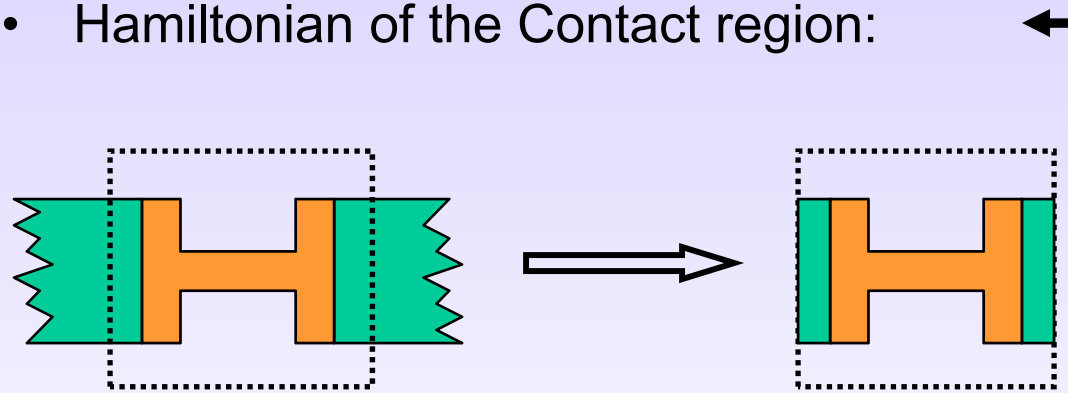
Calculations (zero bias):

- Bulk Greens functions and self-energies (unit cell calculation)
- Hamiltonian of the Contact region:



SCF

- Solution of GF's equations $\Rightarrow$ $\rho(r)$
- Landauer-Büttiker: transmission probability: $T(\varepsilon) = Tr\left[t^{\dagger} t\right](\varepsilon)$

$$t(\varepsilon) = \left(Im\left[\Sigma_R(\varepsilon)\right]\right)^{1/2} G(\varepsilon) \left(Im\left[\Sigma_L(\varepsilon)\right]\right)^{1/2}$$

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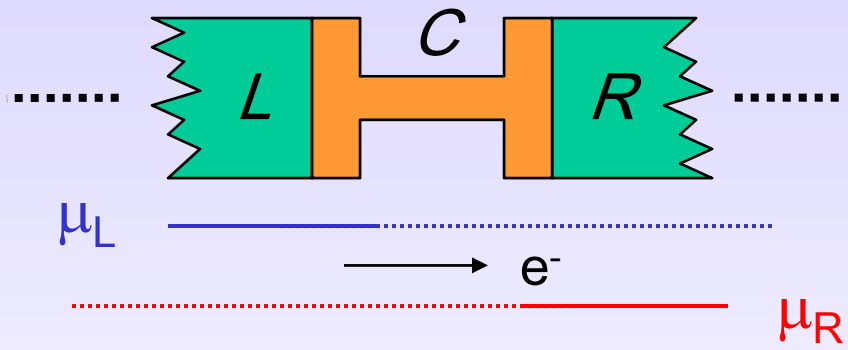
The problem at Non-Equilibrium (Finite Bias)



2 additional problems:

- Non-equilibrium situation:
  - current flow
  - two different chemical potentials

- Electrostatic potential with boundary conditions

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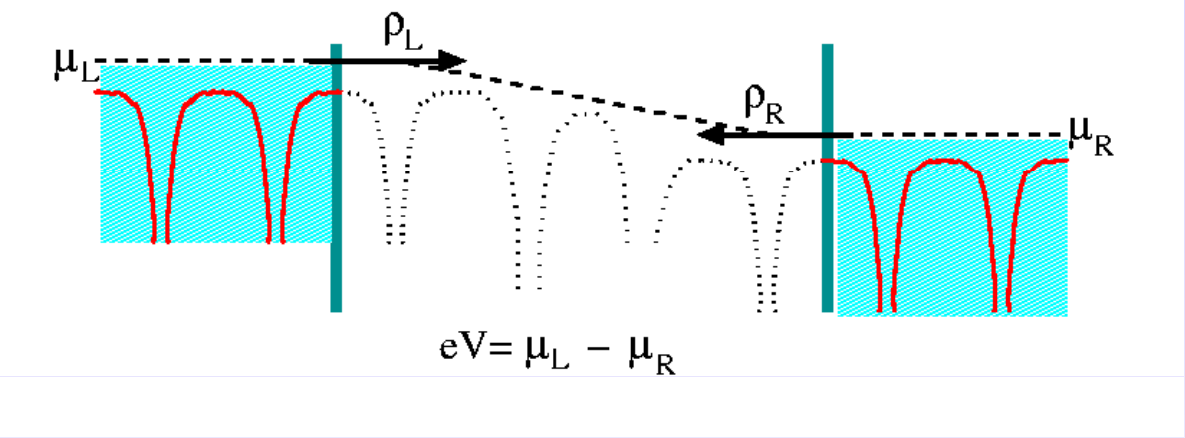
Non-equilibrium formulation:



- Scattering states (from the left)

  Lippmann-Schwinger Eqs.: $\psi_l = \psi_l^o + G(\varepsilon_o)\, V_L\, \psi_l^o$

- Non-equilibrium Density Matrix:

$$D_{\mu\nu} = \int_{-\infty}^{\infty} d\varepsilon \left[\rho^L_{\mu\nu}(\varepsilon) n_F(\varepsilon - \mu_L) + \rho^R_{\mu\nu}(\varepsilon) n_F(\varepsilon - \mu_R)\right]$$

$$\rho^L_{\mu\nu}(\varepsilon) = \frac{1}{\pi}\left(G(\varepsilon + i\delta)\, Im\left[\Sigma_L(\varepsilon + i\delta)\right] G^{\dagger}(\varepsilon + i\delta)\right)_{\mu\nu}$$

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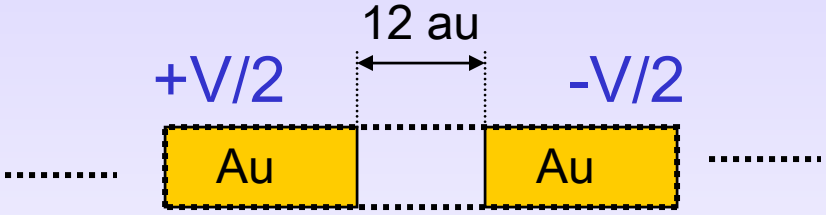
Electrostatic Potential

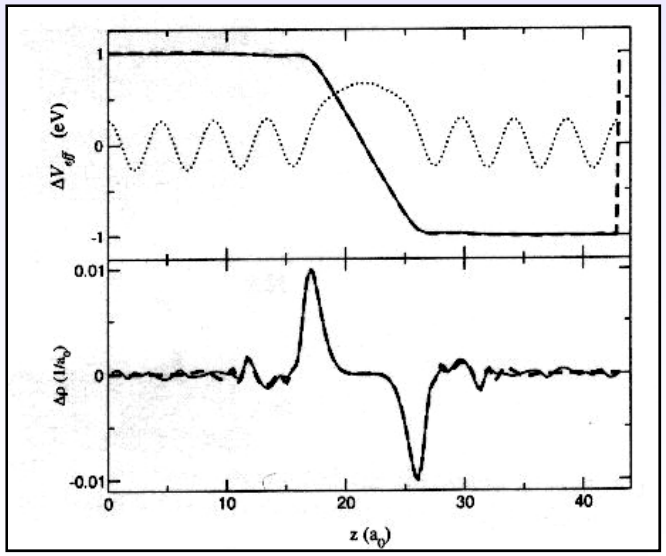
Given $\rho(r)$, $V_H(r)$ is determined except up to a linear term:

$$V_H(\mathbf{r}) = \phi(\mathbf{r}) + \mathbf{a}\cdot\mathbf{r} + \mathbf{b}$$

$\phi(\mathbf{r})$: particular solution of Poisson's equation

a and **b**: determined imposing BC: the shift V between electrodes





- ϕ (r) computed using FFT's
- Linear term: $-\frac{V}{L}\left(z-\frac{L}{2}\right)$

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3. TranSIESTA Practicalities

3 Step process:

1. SIESTA calculation of the bulk electrodes, to get H, ρ, and Self-energies
2. SIESTA calculation for the open system
   - reads the electrode data
   - builds H from ρ
   - solves the open problem using Green's Functions (TranSIESTA)
   - builds new ρ
3. Postprocessing: compute T(E), I, ...

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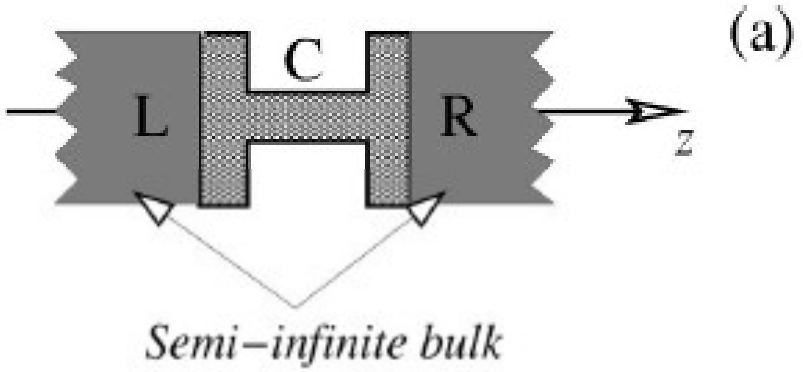
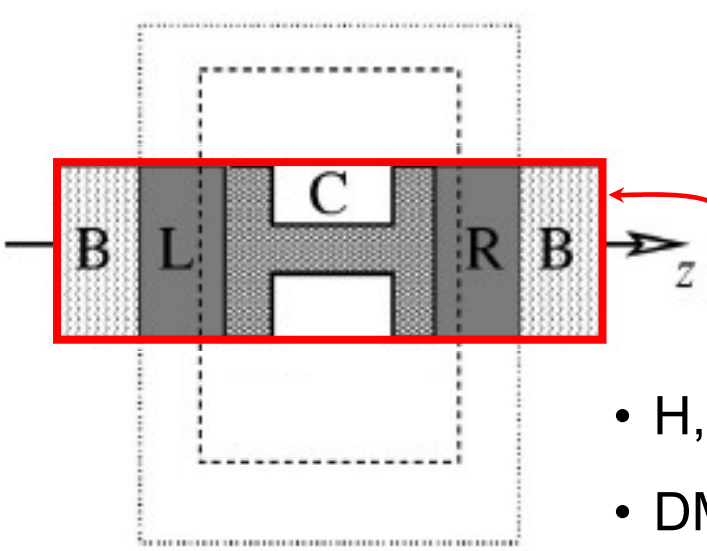


$$G(\varepsilon) = \begin{bmatrix} \varepsilon - H_L - \Sigma_L & -V_{LC} & \\ -V_{CL} & \varepsilon - H_C & -V_{CR} \\ & -V_{RC} & \varepsilon - H_R - \Sigma_R \end{bmatrix}^{-1}$$



Supercell - PBC

- H, DM fixed to bulk in L and R
- DM computed in C from Green's functions
- H_C, V_{LC} and V_{CR} computed in a supercell approach (with potential ramp)
- B (buffer) does not enter directly in the calculation (only in the SC calc. for $V_{Hartree}$)

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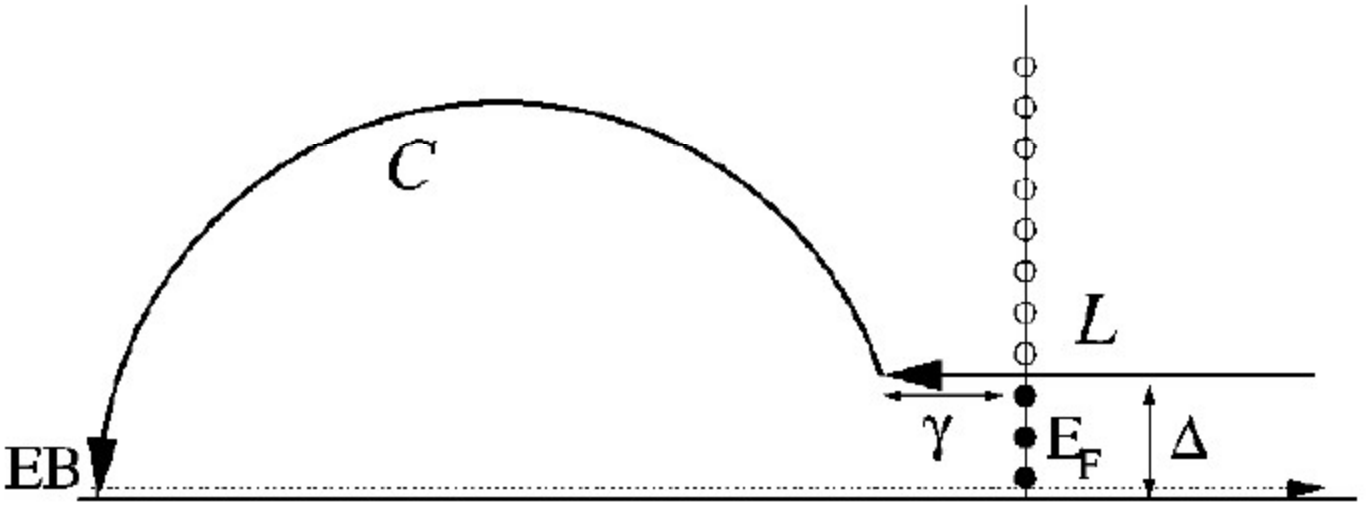
Contour integration



$$\mathbf{D} = -\frac{1}{\pi}\int_{-\infty}^{\infty} d\varepsilon\ \mathrm{Im}[\mathbf{G}(\varepsilon + i\delta)]\, n_F(\varepsilon - \mu)$$

$$= -\frac{1}{\pi}\mathrm{Im}\left[\int_{-\infty}^{\infty} d\varepsilon\ \mathbf{G}(\varepsilon + i\delta)\, n_F(\varepsilon - \mu)\right].$$

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Contour Integration

$$\mathbf{D}_{\mu\nu} = \mathbf{D}^{L}_{\mu\nu} + \Delta^{R}_{\mu\nu}, \tag{28}$$

$$\mathbf{D}^{L}_{\mu\nu} = -\frac{1}{\pi}\mathrm{Im}\left[\int_{EB}^{\infty} d\varepsilon\, \mathbf{G}(\varepsilon + i\delta)\, n_F(\varepsilon - \mu_L)\right], \tag{29}$$

$$\Delta^{R}_{\mu\nu} = \int_{-\infty}^{\infty} d\varepsilon\, \rho^{R}_{\mu\nu}(\varepsilon)[n_F(\varepsilon - \mu_R) - n_F(\varepsilon - \mu_L)], \tag{30}$$

or equivalently

$$\mathbf{D}_{\mu\nu} = \mathbf{D}^{R}_{\mu\nu} + \Delta^{L}_{\mu\nu}, \tag{31}$$

$$\mathbf{D}^{R}_{\mu\nu} = -\frac{1}{\pi}\mathrm{Im}\left[\int_{EB}^{\infty} d\varepsilon\, \mathbf{G}(\varepsilon + i\delta)\, n_F(\varepsilon - \mu_R)\right], \tag{32}$$

$$\Delta^{L}_{\mu,\nu} = \int_{-\infty}^{\infty} d\varepsilon\, \rho^{L}_{\mu\nu}(\varepsilon)[n_F(\varepsilon - \mu_L) - n_F(\varepsilon - \mu_R)]. \tag{33}$$

$$\mathbf{D}_{\mu\nu} = w_{\mu\nu}(\mathbf{D}^{L}_{\mu\nu} + \Delta^{R}_{\mu\nu}) + (1 - w_{\mu\nu})(\mathbf{D}^{R}_{\mu\nu} + \Delta^{L}_{\mu\nu}), \tag{36}$$

$$w_{\mu\nu} = \frac{(\Delta^{L}_{\mu\nu})^2}{(\Delta^{L}_{\mu\nu})^2 + (\Delta^{R}_{\mu\nu})^2}. \tag{37}$$

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```
SolutionMethod Transiesta
```

```
#       GENGF OPTIONS
TS.ComplexContour.Emin    -3.0 Ry
TS.ComplexContour.NPoles        6
TS.ComplexContour.NCircle      20
TS.ComplexContour.NLine         3
TS.RealContour.Emin    -3.0 Ry
TS.RealContour.Emax     2.d0 Ry
TS.TBT.Npoints          100
```

```
# TS OPTIONS
TS.Voltage 1.000000 eV
TS.UseBulkInElectrodes .True.
TS.BufferAtomsLeft  0
TS.BufferAtomsRight 0
```

```
# TBT OPTIONS
TS.TBT.Emin -5.5 eV
TS.TBT.Emax +0.5 eV
TS.TBT.NPoints 100
TS.TBT.NEigen 3
```