제3회 전자구조계산 겨울학교

# 제1원리 양자수송 계산 – 강의 2

## *1st-Principles Quantum Transport Calculations*

**김 용 훈**

한국과학기술원 (KAIST)
전기및전자공학부
*y.h.kim@kaist.ac.kr*

2023년 1월 20일

KAIST EE

## 강의 계획



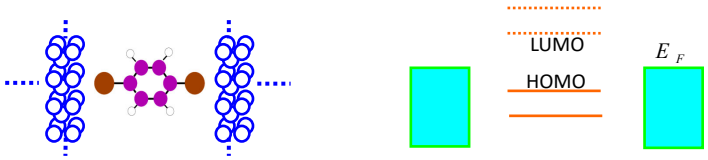
강의 1: 열린 양자계(Open quantum systems)

강의 2: 비평형 양자계(Non-equilibrium quantum systems)

강의 3: 양자수송의 새로운 관점 – 다공간 여기

## We need to describe …






- Size of junction << scattering lengths (e.g. de Broglie wavelength, mean free path, phase coherence length, *etc.*) → Needs fully quantum-mechanical descriptions



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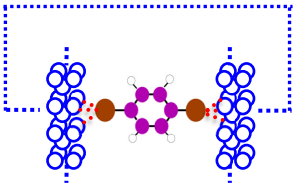
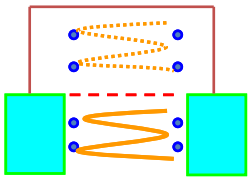
- Nature of channel + electrode interface (charge transfer & bonding)

## We need to describe …





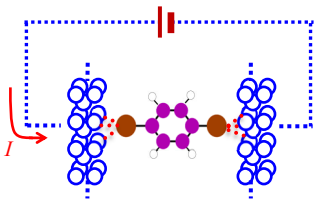


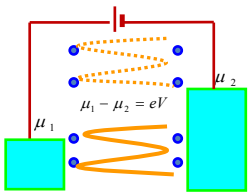
- Size of junction << scattering lengths (e.g. de Broglie wavelength, mean free path, phase coherence length, *etc.*) → Needs fully quantum-mechanical descriptions
- Nature of channel + electrode interface (charge transfer & bonding)
- Open boundary condition, i.e., semi-infinite electrodes @ equilibrium



## We need to describe …



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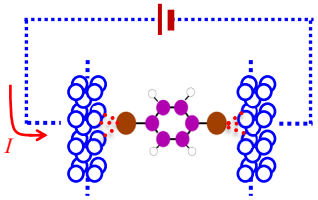
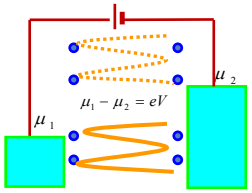
## We need to use …



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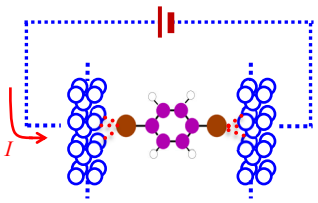
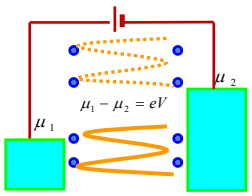
  ← Landauer picture + 1st-principles
- Nature of channel + electrode interface (charge flow & bonding)
  ← "large-scale" 1st-principles (includes many lead atoms within the junction model)
- Open boundary condition, i.e., semi-infinite electrodes
  ← MGF: self-energy $\Sigma = \tau g_s \tau^+$ & surface Green's function $g_s$
- Non-equilibrium density matrix @ finite bias
  ← NEGF: $[G^n] = [G][\Sigma^{in}][G]^+ = [A_1]f_1 + [A_2]f_2$



## We will learn …



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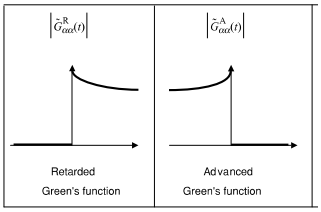
## 4 different types of GFs (see Lecture 1 Supplemental Material)



- Four interrelated GFs:
  *(N.B. more informative to consider the time domain)*
  - Retarded: $\mathbf{G}^R(t;\boldsymbol{r},\boldsymbol{r}') = -\frac{i}{\hbar}\Theta(t)\langle\{\psi(\boldsymbol{r},t),\psi^+(\boldsymbol{r}',0)\}\rangle$
  - Advanced: $\mathbf{G}^A(t;\boldsymbol{r},\boldsymbol{r}') = \frac{i}{\hbar}\Theta(-t)\langle\{\psi(\boldsymbol{r},t),\psi^+(\boldsymbol{r}',0)\}\rangle$
  - Lesser than: $\mathbf{G}^<(t;\boldsymbol{r},\boldsymbol{r}') = \frac{i}{\hbar}\langle\psi^+(\boldsymbol{r}',0)\psi(\boldsymbol{r},t)\rangle$
  - Greater than: $\mathbf{G}^>(t;\boldsymbol{r},\boldsymbol{r}') = -\frac{i}{\hbar}\langle\psi(\boldsymbol{r},t)\,\psi^+(\boldsymbol{r}',0)\rangle$

$$\mathbf{G}^A = \left[\mathbf{G}^R\right]^+$$




Here,

- $\langle\cdots\rangle = Tr\{\hat{\rho}(t=t_0\}$ : Trace over the initial statistical operator
- $\{A,B\} = AB + BA$ (fermions) → $[A,B] = AB - BA$ (bosons)
- Field operators: $\psi(\boldsymbol{r}) = \sum_{\boldsymbol{k}}\langle\boldsymbol{r}|\phi_k\rangle\hat{a}_k$ & $\psi^+(\boldsymbol{r}) = \sum_{\boldsymbol{k}}\langle\boldsymbol{r}|\phi_k\rangle\hat{a}_k^+$
  single-ptl. states, creation op., annihilation op.

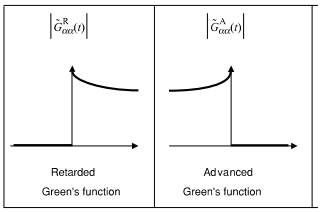


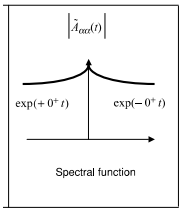
## 4 different types of GFs



- Four interrelated GFs:
  *(N.B. more informative to consider the time domain)*
  - Retarded: $\mathbf{G}^R(t;\boldsymbol{r},\boldsymbol{r}') = -\frac{i}{\hbar}\Theta(t)\langle\{\psi(\boldsymbol{r},t),\psi^+(\boldsymbol{r}',0)\}\rangle$
  - Advanced: $\mathbf{G}^A(t;\boldsymbol{r},\boldsymbol{r}') = \frac{i}{\hbar}\Theta(-t)\langle\{\psi(\boldsymbol{r},t),\psi^+(\boldsymbol{r}',0)\}\rangle$
  - Electron correlation function: $G^n \equiv -iG^<$
  - Hole correlation function : $G^p \equiv +iG^>$

$$\mathbf{G}^A = \left[\mathbf{G}^R\right]^+$$




- Spectral function (~ "DOS"): $\mathbf{A} = i\left[\mathbf{G}^R - \mathbf{G}^A\right] = \mathbf{G}^n + \mathbf{G}^p$
- Density matrix: $\mathbf{D} = \frac{1}{2\pi}\int_{-\infty}^{\infty}\mathbf{G}^n(E)dE$

## cf. Original NEGF

*Valid for: time-dependent Schrodinger eq. & closed quantum system*

*Di Ventra Ch. 4*

- Retarded (Advanced) GF:

$$\mathbf{G}^R(E) = [(E + i0^+)\mathbf{1} - \mathbf{H}]^{-1}$$

→ Spectral function (~"DOS")

$$\mathbf{A}(E) = i[\mathbf{G}(E) - \mathbf{G}^+(E)]$$


Leonid Keldysh (1931~2016)


Gordon Baym (1935~)


Leo Kadanoff (1937~2015)

- Correlation function ← "Keldysh eq.":

$$\underbrace{\mathbf{G}^n}_{\psi\psi^+} = \mathbf{G}^R \underbrace{\mathbf{\Sigma}^{in}}_{ss^+} \mathbf{G}^A$$

→ Density matrix

$$\mathbf{D} = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE\mathbf{G}^n(E)$$

or Density

$$n(\vec{r}) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE\mathbf{G}^n(\vec{r},E)$$

*Assumption 1:*
Closed quantum systems

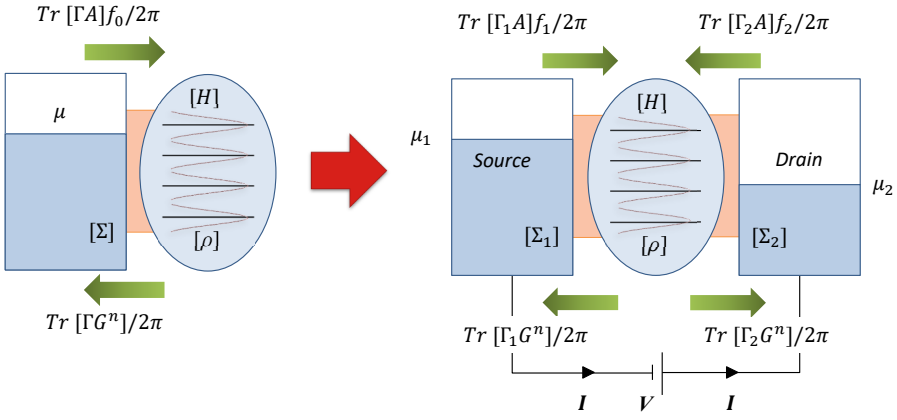
*Assumption 2:*
Initial global equilibrium

*Assumption 3:*
Adiabatic approximation



## Open + Non-equilibrium: Landauer + NEGF

- From 1-contact (eqm.) → 2-contact (non-eqm.) case:




$$[G^n] = [G][\Sigma^{in}][G]^+ \;;\; [G^n] \approx [A_1]f_1 + [A_2]f_2$$

## Assumptions within Landauer + NEGF



Di Ventra Ch. 4

*Assumption 1:*
Closed quantum systems

*Assumption 2:*
Initial global equilibrium

*Assumption 3:*
Adiabatic approximation

*Transport @ Steady State*

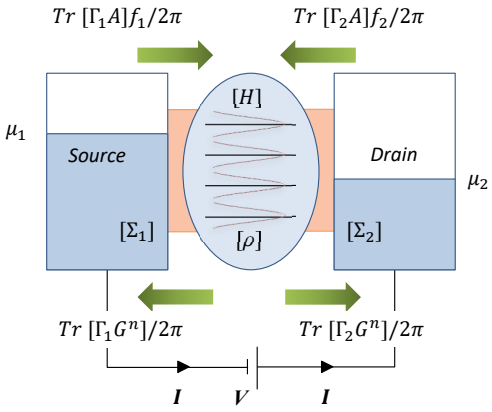
@ Landauer

*Approx. 1:*
Open quantum systems

*Approx. 2:*
Ideal steady state

*Approx. 3:*
"Openness" ↔ Boundary conditions

*Approx. 4:*
Mean-field approximation for the leads



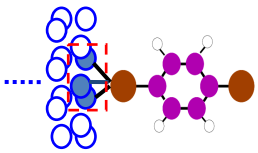



## 1. Open BC via "self-energy" Σ



cf. Datta Ch. 8

Semi-infinite problem → Partitioning



$$\begin{bmatrix} E\mathbb{I}_R - H_R + i0^+\mathbb{I}_R & -\tau^+ \\ -\tau & E\mathbb{I} - H \end{bmatrix} \begin{Bmatrix} \phi_R + \chi \\ \psi \end{Bmatrix} = \begin{Bmatrix} S_R \\ 0 \end{Bmatrix}$$

Finite problem via "Self-Energy"

$E\{\psi\} = [H + \Sigma(E)]\{\psi\} + \{S\}$ where $[\Sigma] = [\tau][G_R][\tau]^+$ & $\{S\} \equiv [\tau]\{\phi_R\}$

↳ *Energy level shift (real part) & broadening (imaginary part)*

or

$\{\psi\} = [G]\{S\}$ where $[G] = [E\mathbb{I} - H + \Sigma(E)]^{-1}$

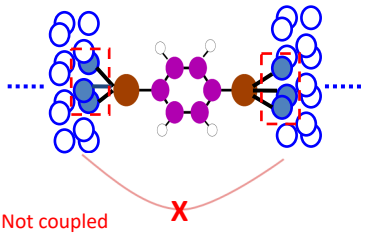
*Q: How to calculate $G_R$?*

## Open BC via Σ: 2 terminal case



*cf. Datta Ch. 9.2*

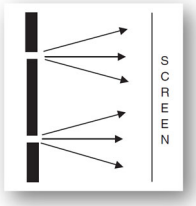
Semi-infinite problem → Partitioning



Not coupled *within localized basis sets*

$$G(E) = [(E + i0^+)\mathbb{I} - H]^{-1} = \begin{bmatrix} C_{11} & \tau_{m1}^+ & 0 \\ \tau_{m1} & E\mathbb{I}_m - H_m & \tau_{m2} \\ 0 & \tau_{m2}^+ & C_{22} \end{bmatrix}^{-1}$$

$$\boldsymbol{\Sigma} = \boldsymbol{\Sigma}_1 + \boldsymbol{\Sigma}_2$$




Finite problem via "Self-Energy"

$$H_{\mathrm{m}}$$
$$\Rightarrow H_{\mathrm{m}} + \Sigma_1(E) + \Sigma_2(E) \quad (\Sigma_{1/2} = \tau G_{1/2} \tau^+)$$
$$\Rightarrow G_{\mathrm{m}}(E) = [E_m \mathbb{I}_m - H_m - \Sigma_1(E) - \Sigma_2(E)]^{-1}$$



## 2. EOM for electron correlation functions



*cf. Datta Ch. 9.2*

Within 1-particle picture :

- Schrodinger eq. + In/Out flow:

$$E\psi = \boldsymbol{H}\psi + \boldsymbol{\Sigma}\psi + s$$

$$\{\psi\} = \underbrace{[E\mathbb{I} - H_s - \Sigma]^{-1}}_{[G]} \{S\}$$

(out: $\Sigma$, in: $S$)

- NEGF equation (Keldysh, 1965):

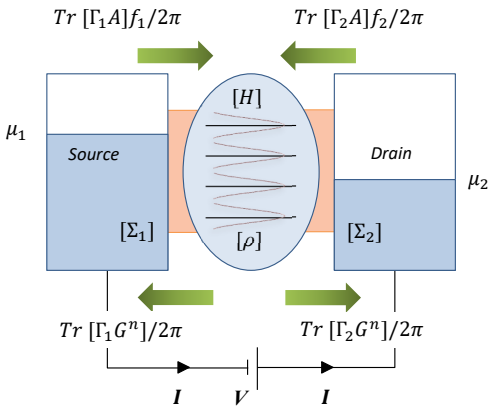
$$\mathbf{G}^n = \mathbf{G}^R \boldsymbol{\Sigma}^{in} \mathbf{G}^A$$

$$\mathbf{G}^n \equiv -i\mathbf{G}^< \; ; \; \boldsymbol{\Sigma}^n \equiv -i\boldsymbol{\Sigma}^<$$

← Devising the rule:

$$\{\phi_R\}\{\phi_R\}^+ \Rightarrow \int_{-\infty}^{\infty} \frac{dE}{2\pi} f_0(\varepsilon - \mu)\mathbf{A}_R(E)$$

$$\{\psi\}\{\psi\}^+ \Rightarrow \int_{-\infty}^{\infty} \frac{dE}{2\pi} \mathbf{G}^n(E)$$



## EOM for electron correlation functions



Within 1-particle picture :

- Schrodinger eq. + In/Out flow:

$$E\psi = \boldsymbol{H}\psi + \boldsymbol{\Sigma}\psi + s$$

$$\{\psi\} = \underbrace{[E\mathbb{I} - H_s - \Sigma]^{-1}}_{[G]}\{S\}$$

(out: $[G]$, in: $\{S\}$)

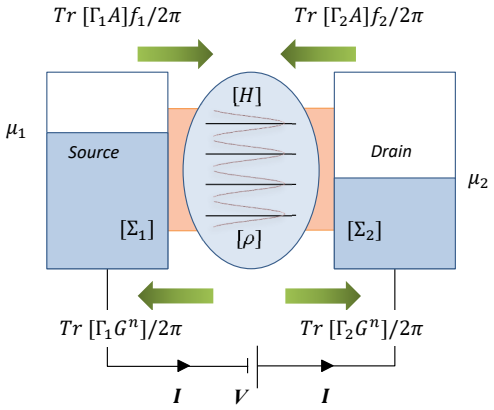
- NEGF equation (Keldysh, 1965):

$$\mathbf{G}^n = \mathbf{G}^R\boldsymbol{\Sigma}^{in}\mathbf{G}^A$$

$$\underbrace{\{\psi\}\{\psi\}^+}_{\mathbf{G}^n} = \mathbf{G}^R\underbrace{\{s\}\{s\}^+}_{\boldsymbol{\Sigma}^{in}}\mathbf{G}^A$$

or

$$\mathbf{G}^h = \mathbf{G}^R\boldsymbol{\Sigma}^{out}\mathbf{G}^A$$






## Non-equilibrium density matrix



- Density matrix (→ density):

$$\mathbf{D} = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE\,\mathbf{G}^{\mathrm{n}}(E)$$

$$n(\vec{r}) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE\;\mathbf{G}^{\mathrm{n}}(\vec{r},E)$$

- @ Independent-particle approximation:

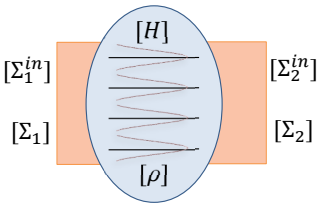
$$\boldsymbol{\Sigma}^{\mathrm{in}} \approx \boldsymbol{\Gamma}_1 f(E-\mu_1) + \boldsymbol{\Gamma}_2 f(E-\mu_2)$$

$$\mathbf{G}^{\mathbf{n}} = \mathbf{G}\boldsymbol{\Sigma}^{\mathrm{in}}\mathbf{G}^{+} \approx \underbrace{\mathbf{G}\boldsymbol{\Gamma}_1\mathbf{G}^{+}}_{\mathbf{A}_1} f_1 + \underbrace{\mathbf{G}\boldsymbol{\Gamma}_2\mathbf{G}^{+}}_{\mathbf{A}_2} f_2$$

$$n(\vec{r}) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE\,\mathbf{G}^{\mathrm{n}}(\vec{r},E) \approx \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE[f_1\mathbf{A}_1 + f_2\mathbf{A}_2]$$

$$\boldsymbol{\Sigma} = \boldsymbol{\Sigma}_1 + \boldsymbol{\Sigma}_2$$

$$\boldsymbol{\Sigma}^{\mathrm{in}} = \boldsymbol{\Sigma}_1^{\mathrm{in}} + \boldsymbol{\Sigma}_2^{\mathrm{in}}$$



## NEGF current (density)



*cf. Datta Ch. 9.3*

- Current operator:

$$I_{op}(E) = \frac{-e}{ih}\{[\mathbf{H}\mathbf{G}^{\mathrm{n}} - \mathbf{G}^{\mathrm{n}}\mathbf{H}] + [\mathbf{\Sigma}^{\mathrm{in}}\mathbf{G}^{+} - \mathbf{G}\mathbf{\Sigma}^{\mathrm{in}}] + [\mathbf{\Sigma}\mathbf{G}^{\mathrm{n}} - \mathbf{G}^{\mathrm{n}}\mathbf{\Sigma}^{+}]\}$$

- Current density (internal current):

$$j(\vec{r},t) = \frac{-ie}{2}\lim_{t'\to t}(\nabla' - \nabla)\mathbf{G}^{n}(\vec{r},t;\vec{r}',t')$$

$$\boldsymbol{J}(E) = \frac{-e}{ih}[\mathbf{H}\mathbf{G}^{\mathrm{n}}(E) - \mathbf{G}^{\mathrm{n}}(E)\mathbf{H}]$$

- Terminal current:

$$I = I_1 = -I_2$$

$$I_\alpha = \frac{e}{h}\int dE\,\{Tr[\mathbf{\Gamma}_\alpha(E)\mathbf{A}(E)]f_\alpha - Tr[\mathbf{\Gamma}_\alpha(E)\mathbf{G}^{\mathrm{n}}(E)]\}$$

inflow – outflow



## NEGF current @ Independent-particle case



- Terminal current:

$$I_\alpha = \frac{e}{h}\int dE\,\{Tr[\mathbf{\Gamma}_\alpha(E)\mathbf{A}(E)]f_\alpha - Tr[\mathbf{\Gamma}_\alpha(E)\mathbf{G}^{\mathrm{n}}(E)]\}$$

$\mathbf{A}_\alpha + \mathbf{A}_\beta$ $\qquad$ $\mathbf{A}_\alpha f_\alpha + \mathbf{A}_\beta f_\beta$

$$I = I_1 = -I_2 \approx \frac{e}{h}\int dE\,\underbrace{Tr(\mathbf{\Gamma_1}\mathbf{G}\mathbf{\Gamma_2}\mathbf{G}^{+})(E)}_{T(E)}[f_1(E) - f_2(E)]$$

- Transmission

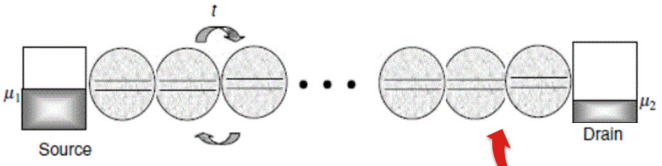
$$T(E) = Tr(\mathbf{\Gamma_1}\mathbf{G}\mathbf{\Gamma_2}\mathbf{G}^{+})$$

## Atom to transistor



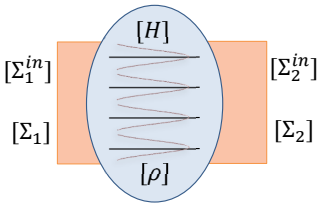
*cf. Datta Chs. 1 & 11*

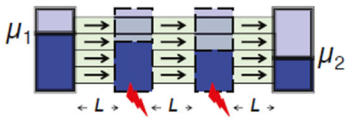
- Q. Ohm's law from Ladauer-NEGF?



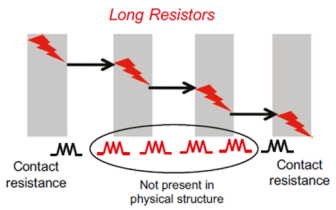

$$\mathbf{\Sigma} = \mathbf{\Sigma}_1 + \mathbf{\Sigma}_2 + \mathbf{\Sigma}_{diss}$$

$$\mathbf{\Sigma}^{\text{in}} = \mathbf{\Sigma}_1^{\text{in}} + \mathbf{\Sigma}_2^{\text{in}} + \mathbf{\Sigma}_{diss}^{\text{in}}$$




$$T(E) = \frac{\lambda(E)}{\lambda(E) + L}$$




$$G = \frac{2q^2M}{h}\frac{\lambda}{L+\lambda} = \frac{G_B}{\left(1+\frac{L}{\lambda}\right)}$$

$$R = \frac{h}{2q^2M}\left(1+\frac{L}{\lambda}\right) = R_B + R_B\frac{L}{\lambda}$$



## Summary: NEGF density matrix & current



- Key equations in NEGF:

① Open B.C.

$$[G] = \{E[\mathbb{I}] - [\mathrm{H}] - [\Sigma]\}^{-1} \;\; ; \;\; [A] = i([G] - [G]^{+})$$

$$[G^n(E)]_{eq} = [A(E)]f_0(E-\mu)$$

② Non-eqm. DM

$$[G^n] = [G][\Sigma^{in}][G]^{+} \qquad ; \; [G^n] \approx [A_1]f_1 + [A_2]f_2$$

③ Terminal current

$$I_\alpha = \frac{e}{h}\int dE\,\{Tr[\mathbf{\Gamma}_\alpha(E)\mathbf{A}(E)]f_\alpha - Tr[\mathbf{\Gamma}_\alpha(E)\mathbf{G}^{\mathrm{n}}(E)]\}$$

## DFT-NEGF: Landauer + NEGF (Landauer ≠ NEGF!)



$$i\hbar\frac{\partial\psi(t)}{\partial t} = H(t)\psi(t) + \int_0^t dt'\Sigma(t,t')\psi(t') + s(t)$$

- Retarded Green's function (~ DOS):

$$\mathbf{G}^R(E) = [E\mathbb{I} - \mathbf{H} - \mathbf{\Sigma}_1(E) + \mathbf{\Sigma}_2(E)]^{-1}$$

- Correlation function via "Keldysh eq.":

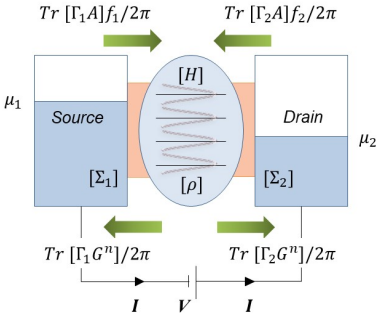
$$\mathbf{\Sigma}^{in} \approx \mathbf{\Gamma}_1 f_1 + \mathbf{\Gamma}_2 f_2 \quad \textit{"Landauer condition"}$$

$$\rightarrow\ n(\vec{r}) \approx \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE[f_1\mathbf{A}_1 + f_2\mathbf{A}_2]$$

- Terminal current:

$$I_\alpha = \frac{e}{h}\int dE\,\{Tr[\mathbf{\Gamma}_\alpha\mathbf{A}]f_\alpha - Tr[\mathbf{\Gamma}_\alpha\mathbf{G}^n]\}$$

$$I \approx \frac{e}{h}\int dE\,\underbrace{Tr(\mathbf{\Gamma_1 G \Gamma_2 G^+})}_{\textit{Transmission}}\,[f_1(E) - f_2(E)]$$




- *Time-dependent → Steady state transport*
- *Closed → Open quantum system via BC*

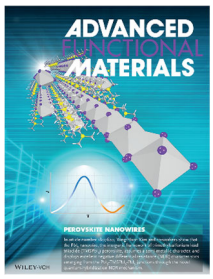
*"Electronic Transport in Mesoscopic Systems"* by S. Datta (1997)
*"Electrical Transport in Nanoscale Systems"* by M. Di Ventra (2008)



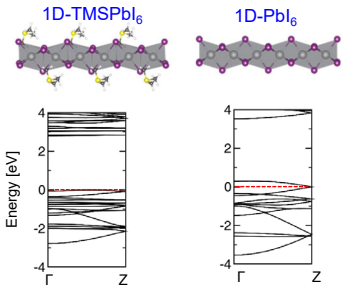
## Difficulties with DFT-NEGF: e.g.



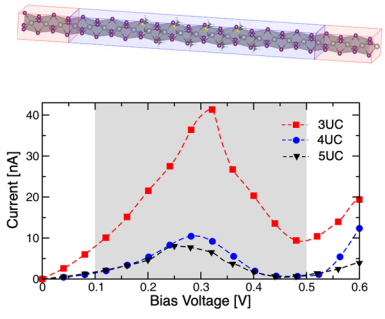
### Quantum-Hybridization (QH) Negative Differential Resistance (NDR)



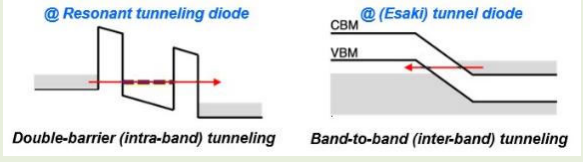

Adv. Funct. Mater. **29**, 1807620 (2019)




*Semi-metallic!*




*cf.*






Leo Esaki

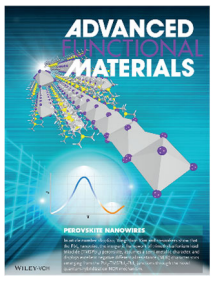
*Physics 1973*



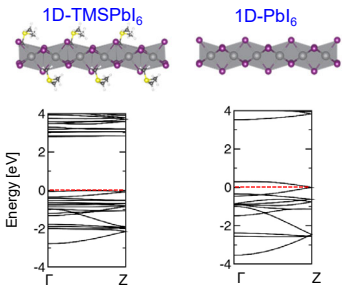
## Difficulties with DFT-NEGF: e.g.



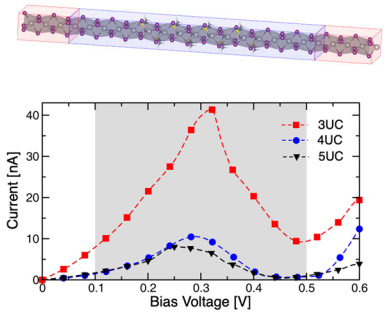
**Quantum-Hybridization (QH) Negative Differential Resistance (NDR)**

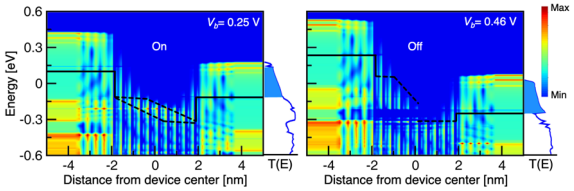


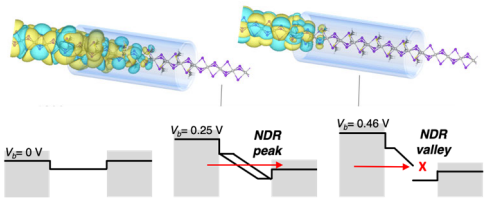

Adv. Funct. Mater. **29**, 1807620 (2019)




*Semi-metallic!*



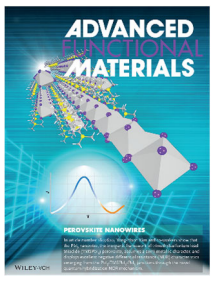





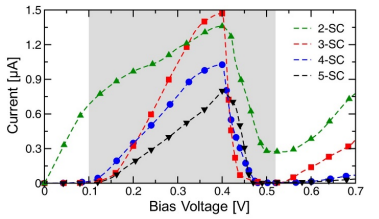

## Difficulties with DFT-NEGF: e.g.



**e.g. Quantum-Hybridization (QH) Negative Differential Resistance (NDR)**




Adv. Funct. Mater. **29**, 1807620 (2019)

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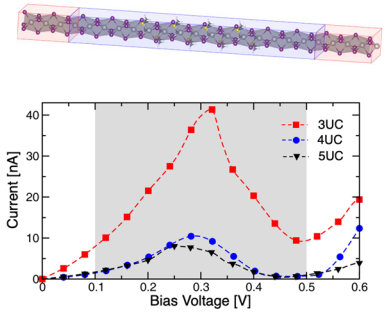
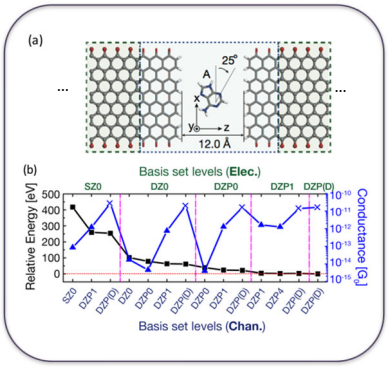


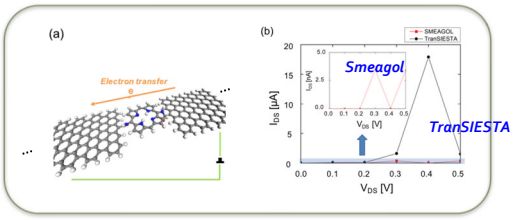

**Wrong BC** **Correct BC**

## Various difficulties with DFT-NEGF

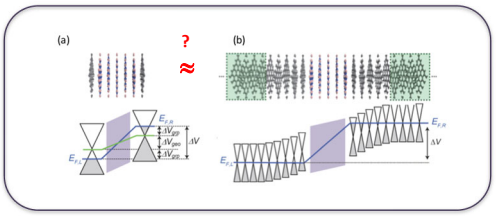





Nonvariational nature of "Landauer" current ($V_b = 0$ V)




Inequivalence between different codes ($V_b \neq 0$ V)




Restrictions from the requirement of semi-infinite electrodes ($V_b \neq 0$ V)



## Origin of difficulties with DFT-NEGF: Open-ness

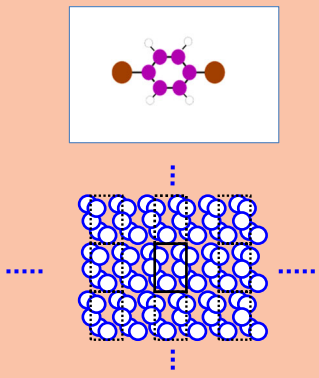


**DFT (Equilibrium)**

*Closed or Periodic B. C.*

$H\psi_i = \varepsilon_i \psi_i$

$f_{FD}(E-\mu)$

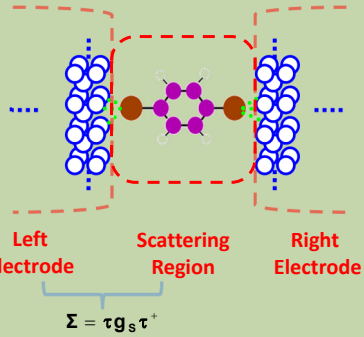


**NEGF (Non-equilibrium)**

*Open B. C.*

$G^R = (E1 - H - \Sigma_1 - \Sigma_2)^{-1}$

$G^n(E) \approx f_1 A_1(E) + f_2 A_2(E)$




$\Sigma = \tau g_s \tau^+$

## Origin of difficulties with DFT-NEGF: Open-ness

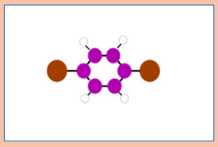


**DFT (Equilibrium)**

*Closed or Periodic B. C.*

$H\psi_i = \varepsilon_i\psi_i$

$f_{FD}(E-\mu)$



☺ **High-reliability of simulations!!**

$E[\chi] = \frac{\langle\chi|H|\chi\rangle}{\langle\chi|\chi\rangle} \rightarrow E[\chi] \geq E_{GS} = \langle\psi_{GS}|H|\psi_{GS}\rangle$

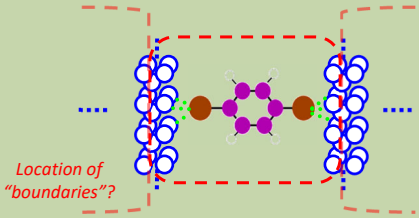
- Total energy ← Variational principle

**NEGF (Non-equilibrium)**

*Open B. C.*

$G^R = (E1 - H - \Sigma_1 - \Sigma_2)^{-1}$

$G^n(E) \approx f_1 A_1(E) + f_2 A_2(E)$




☹ **Ambiguities??**

$I(V) \approx \frac{e}{h}\int dE\, Tr(\mathbf{\Gamma_1 G \Gamma_2 G^+})\,[f_1 - f_2]$

- Current: Non-variational quantity





# Supplemental Notes

## Review: Two key physical quantities



Datta Ch. 8

- Spectral funciton ← density of states (DOS)

$$A(\vec{r},\vec{r}';E) = 2\pi \sum_{\alpha} \delta(E-\varepsilon_\alpha)\,\psi_\alpha(\vec{r})\psi_\alpha^*(\vec{r}')$$

$$\Updownarrow$$

$$[A](E) = \sum_{\alpha} \delta(E-\varepsilon_\alpha)\,\{\psi_\alpha\}\{\psi_\beta\}^+$$

real space basis ↕ eigenstate basis (unitary transformation)

- Density matrix $\varrho$ ← density

$$\varrho(\vec{r},\vec{r}') = \sum_{\alpha} f_0(\varepsilon_\alpha-\mu)\,\psi_\alpha(\vec{r})\psi_\alpha^*(\vec{r}')$$

$$\Updownarrow$$

$$\boxed{[\varrho] = f_0([H[\varrho]]-\mu[\mathbb{I}])}$$

$$[\varrho] = \sum_{\alpha} f_0(\varepsilon_\alpha-\mu)\,\{\phi_\alpha\}\{\phi_\beta\}^+$$

real space basis ↕ eigenstate basis (unitary transformation)



## Review: Self-energy matrix



Datta Ch. 8

- We want to calculate:

$$G \equiv \begin{bmatrix} G_S & G_{sR} \\ G_{Rs} & G_R \end{bmatrix} = \begin{bmatrix} (E+i0^+)\mathbb{I}_S - H_S & -\tau \\ -\tau^+ & (E+i0^+)\mathbb{I}_R - H_R \end{bmatrix}^{-1}$$

- Then, using $a = (A-BD^{-1}C)^{-1}$ :

$$G_s(E) = \{(E+i0^+)\mathbb{I}_s - \mathrm{H_s} - \tau G_R(E)\,\tau^+\}^{-1}$$

- Self-energy matrix: $\boxed{\Sigma_s(\mathrm{E}) = \tau\ \ G_R(E)\ \tau^+}$

$(s\times s)$ $(s\times r)$ $(r\times r)$ $(r\times s)$

- Advantage of GFs: Can evaluate the $(s\times s)$ subsection of $G$ exactly from

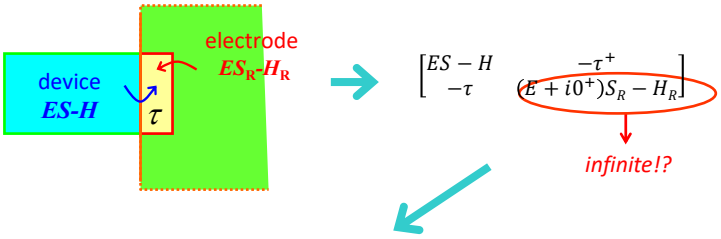
$$G_s(E) = \{(E+i0^+)\mathbb{I}_s - H_s - \Sigma_S(\mathrm{E})\}^{-1}$$

→ Dyson equation:

$$\boxed{G_s = [G_0{}^{-1} - \Sigma]^{-1}} \text{ or } \boxed{\Sigma = G_0{}^{-1} - G_s{}^{-1}} \text{ or } \boxed{G_s = G_0 + G_0\,\Sigma\,G_s}$$

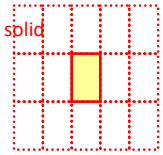
## How to calculate $G_R$ ? ← "surface GF" $g_s$






**Algorithm for $g_s$ evaluation:**

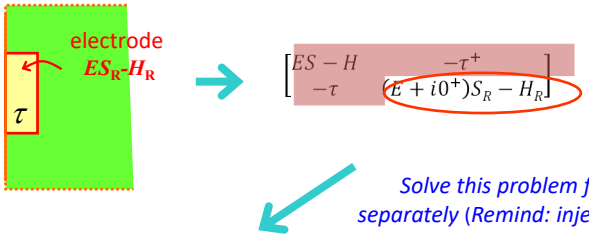
1. P.B.C. calculation of bulk solid to obtain $S^{\vec{R}}$ & $H^{\vec{R}}$






## Surface GF $g_s$



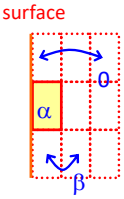



**Algorithm for $g_s$ evaluation:**

1. P.B.C. calculation of bulk (∞-size) solid to obtain $S/H^{\vec{R}}$
2. Divide inter- & intra-layer parts
   (define surface-|| & ⊥ directions)
3. Non-0 coupling of only $H/S^{\{\vec{k}_{||},\vec{R}_{\perp}\}}$ in the neighboring layers
4. Iteratively solve for finite-size surface:

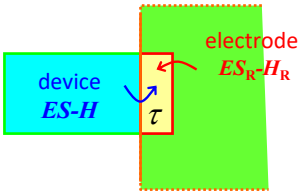
$$\begin{bmatrix} \alpha & \beta & 0 & \cdots \\ \beta^+ & \alpha & \beta & \\ 0 & \beta^+ & \alpha & \\ \vdots & & & \ddots \end{bmatrix}^{-1}$$

$$g_s = (\alpha - \beta g_s^{-1} \beta^+)^{-1}$$



## Q. Numerical issues? (for 1st-principles)






*We assumed:*

$$\begin{bmatrix} ES-H & \tau & 0 & 0 & \\ \tau^{+} & \alpha & \beta & 0 & \\ 0 & \beta^{+} & \alpha & \beta & \cdots \\ 0 & 0 & \beta^{+} & \alpha & \\ & & \vdots & & \ddots \end{bmatrix}$$

*In fact:*

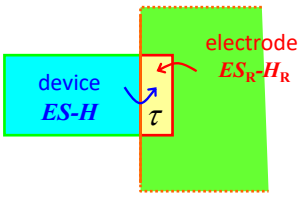
$$\begin{bmatrix} ES-H & \tau' & 0 & 0 & \\ \tau'^{+} & \alpha' & \beta' & 0 & \\ 0 & \beta'^{+} & \alpha & \beta & \cdots \\ 0 & 0 & \beta^{+} & \alpha & \\ & & \vdots & & \ddots \end{bmatrix}$$

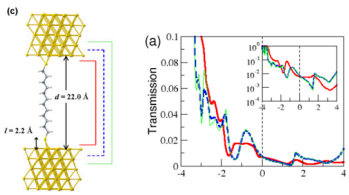
- Note $[S]$, $[0]$ blocks, etc. → Best choice of basis?
- For $\tau$, we need only the part that has overlap with device (# of non-0 $\tau$ elements is finite)
- Include large enough electrode part for the reliable estimation of $\tau$ & $E_F$ (large-scale calculation) → Check the convergence by moving the channel-electrode boundary outward
- Should $\tau$ & $g_S$ treated consistently (same level of basis set, etc.)
- Can treat slab geometry via $\vec{k}_{||}$ -sampling



## Q. Numerical issues? (for 1st-principles)

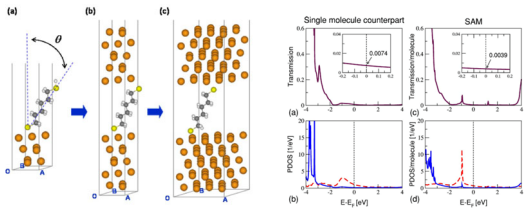





YHK, J. Kor. Phys. **52**, 1181 (2008).

- Include large enough electrode part for the reliable estimation of $\tau$ & $E_F$ (large-scale calculation) → Check the convergence by moving the channel-electrode boundary outward
- Treat slab geometry via $\vec{k}_{||}$ -sampling.

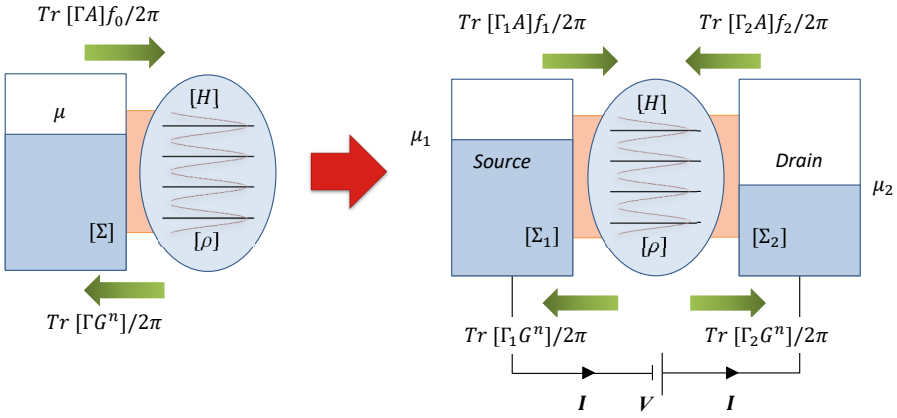



YHK *et al.*
Phys. Rev. B **73**, 235419 (2006)

## Open + Non-equilibrium: Advanced (NEGF) formalism

전자구조계산 겨울학교 2023. 1. 20

- From 1-contact (eqm.) → 2-contact (non-eqm.):




$$[G^n] \approx [A_1]f_1 + [A_2]f_2 \;\; ; \;\; I \approx \frac{e}{h}\int dE\, Tr(\mathbf{\Gamma_1 G \Gamma_2 G^+})(E)[f_1(E) - f_2(E)]$$

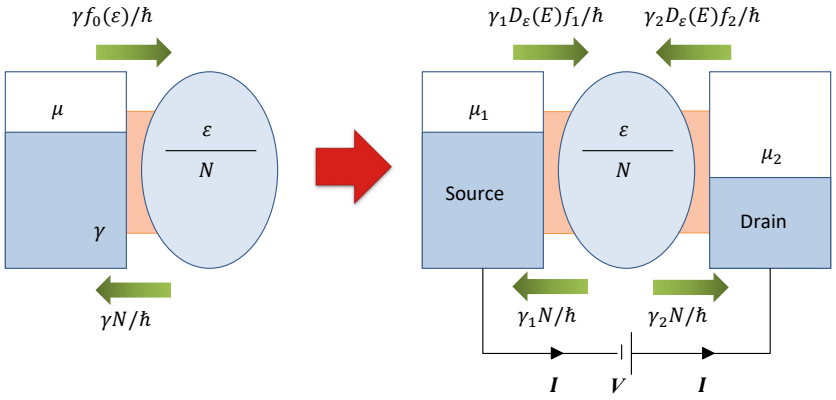


## Open + Non-equilibrium: Physical picture

전자구조계산 겨울학교 2023. 1. 20

- From 1-contact (eqm.) → 2-contact (non-eqm.):

*(Improper) 1-level model*



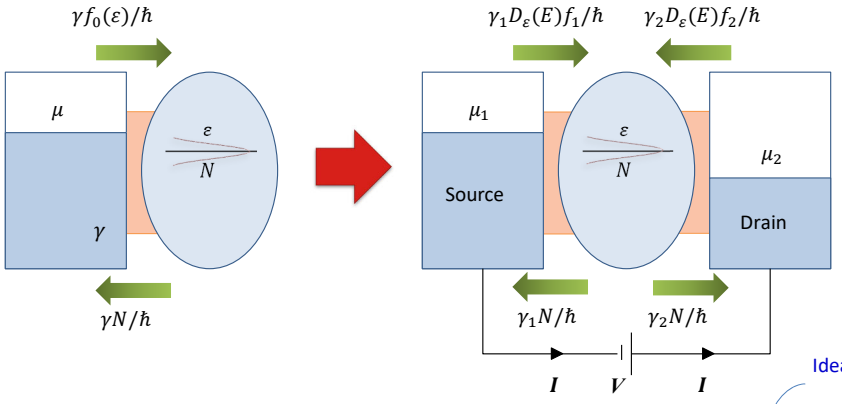

$$N = \frac{\gamma_1 f_1(\varepsilon) + \gamma_2 f_2(\varepsilon)}{\gamma_1 + \gamma_2} \;\; ; \;\; I = \frac{e}{\hbar}\frac{\gamma_1\gamma_2}{\gamma_1 + \gamma_2}[f_1(\varepsilon) - f_2(\varepsilon)] \underset{\gamma \Uparrow}{\Rightarrow} \infty \;?$$

## Open + Non-equilibrium: Physical picture



- From 1-contact (eqm.) → 2-contact (non-eqm.):

*(Proper) 1-level model → quantum of conductance*




Ideal 1-level with $\gamma_1 = \gamma_2$

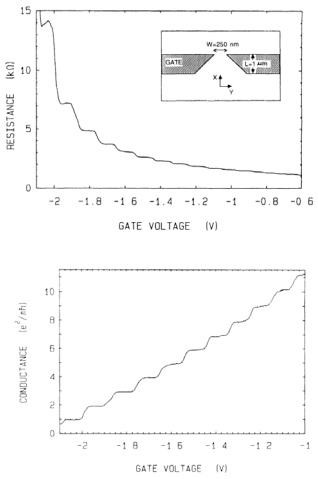
$$N = \int_{-\infty}^{\infty} dE \frac{\gamma_1 f_1(E) + \gamma_2 f_2(E)}{\gamma_1 + \gamma_2} \quad ; \quad I = \frac{e}{\hbar} \int_{-\infty}^{\infty} dE\, D_\varepsilon(E) \frac{\gamma_1 \gamma_2}{\gamma_1 + \gamma_2} [f_1(E) - f_2(E)] \Rightarrow \frac{q^2}{h}$$



## Quantum of conductance: Experiment

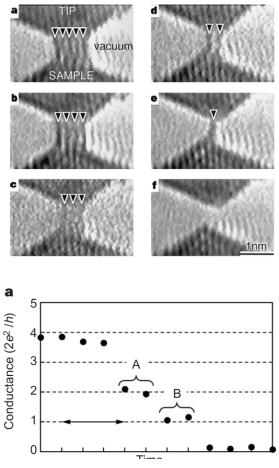


GaAs-AlGaAs




B. van Wees *et al. Phys. Rev. Lett.* **60,** 848 (1988).

Au chain




H. Ohnishi et al. *Nature* **395,** 780 (1998)

# Algorithm of LCAO KS-DFT



*Linear Combination Atomic Orbitals*

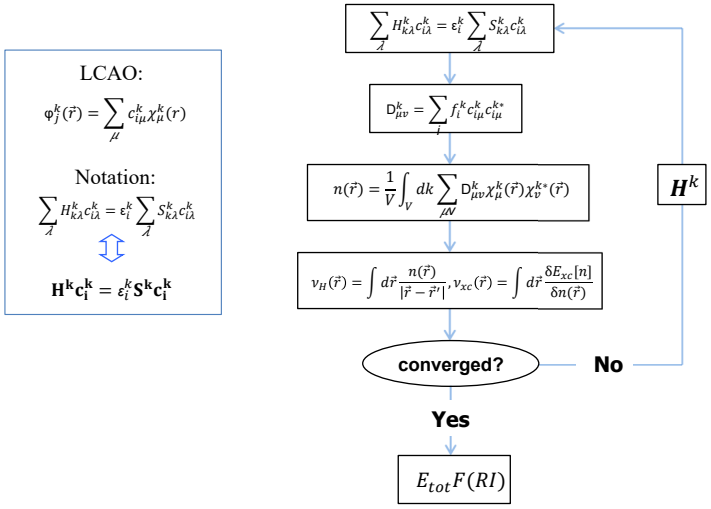
LCAO:

$$\varphi_j^k(\vec{r}) = \sum_\mu c_{i\mu}^k \chi_\mu^k(r)$$

Notation:

$$\sum_\lambda H_{k\lambda}^k c_{i\lambda}^k = \varepsilon_i^k \sum_\lambda S_{k\lambda}^k c_{i\lambda}^k$$

$$\mathbf{H^k c_i^k} = \varepsilon_i^k \mathbf{S^k c_i^k}$$

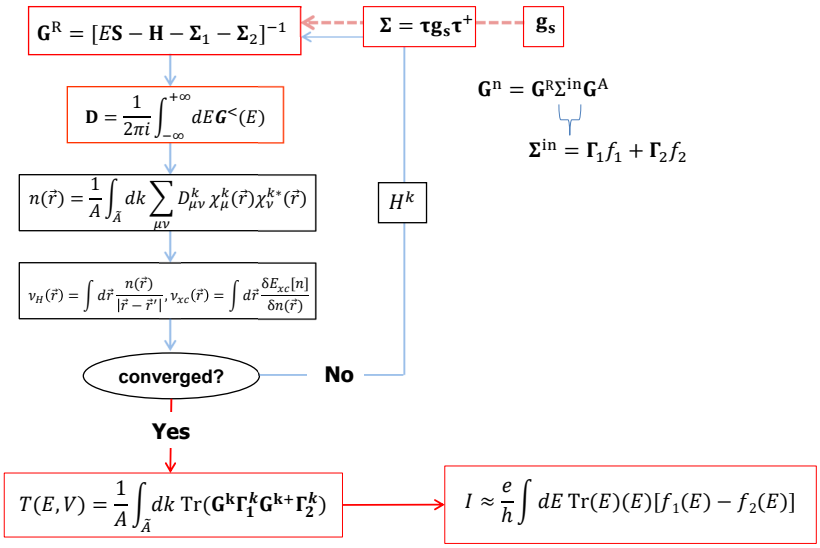



- Postprocessing: bandstructure, (L/P)DOS, populations, *etc.*



# Algorithm of LCAO DFT-NEGF






- Postprocessing: I-V curve, (L/P)DOS, etc.