Bigger and better: Large Scale NEGF Calculations and Coupling DFT with TB

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Outline

1 Motivation

2 Non-equilibrium Green functions (TRANSIESTA)
   • Block-Tri-Diagonal inversion
   • Why pivoting matters?
   • Scalability
   • Optimizing the integration

3 DFT coupled to tight-binding – graphene
   • DFT $\rightarrow$ TB
Motivation

Non-equilibrium Green functions (TRANSiesta)
- Block-Tri-Diagonal inversion
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DFT coupled to tight-binding – graphene
- DFT $\rightarrow$ TB
Motivation

Graphene

- Larger and larger devices
- Multiple electrodes
- Non-equilibrium effects
Non-equilibrium Green functions (TRANSIESTA)

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3 DFT coupled to tight-binding – graphene
   - DFT → TB
Non-equilibrium Green function
Basic equations — and problems herein

Equations for calculating the density matrix

\[ G(z) = \left[ zS - H - \sum \Sigma_c (z - \mu_c) \right]^{-1} \], with \( z = \epsilon + i\eta \)

\[ \Gamma_c(z) = \imath \left( \Sigma_c(z - \mu_c) - \Sigma_c^\dagger(z - \mu_c) \right) \]

\[ \rho(z) \sim -\int [G(z) - G^\dagger(z)] n_{F,c} + \int G(z) \Gamma_c'(z) G^\dagger(z) [n_{F,c'} - n_{F,c}] \]
Non-equilibrium Green function

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Sparse elements

- Sparse inversion techniques, MUMPS, PEXSI, ...
- Sparsity depends on basis set
Non-equilibrium Green function

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\rho(z) \sim -\int [G(z) - G^{\dagger}(z)] n_{F,\epsilon} + \int \frac{G(z)\Gamma_{\epsilon}(z)G^{\dagger}(z)[n_{F,\epsilon'} - n_{F,\epsilon}]}{n_{F,\epsilon'}}
\]

Sparse elements
- Sparse inversion techniques, MUMPS, PEXSI, ...
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Dense column matrix multiplications
- Sparse inversion techniques are less optimal
- Dense matrix operations may be more efficient → Block-Tri-Diagonal (BTD)
Inversion — Block-Tri-Diagonal

Green function

Utilise sparsity of Hamiltonian

- Reduce complexity using zeroes
- Same as the recursive Green function algorithm

\[
G^{-1} = \begin{pmatrix}
A_1 & C_2 & 0 & \cdots & \cdots & \cdots \\
B_1 & A_2 & C_3 & 0 & \cdots & \cdots \\
0 & B_2 & \ddots & \ddots & \ddots & \cdots \\
\vdots & 0 & \ddots & \ddots & \ddots & C_p \\
\vdots & 0 & \ddots & \ddots & \ddots & B_{p-1} \\
& & & & & A_p
\end{pmatrix}
\]

\[
\tilde{Y}_n = [A_{n-1} - Y_{n-1}]^{-1} C_n, \quad Y_1 = 0
\]

\[
Y_n = B_{n-1} \tilde{Y}_n
\]

\[
\tilde{X}_n = [A_{n+1} - X_{n+1}]^{-1} B_n, \quad X_p = 0
\]

\[
X_n = C_{n+1} \tilde{X}_n
\]

\[
G_{n,n} = [A_n - X_n - Y_n]^{-1}
\]

\[
G_{m-1,n} = -\tilde{Y}_m G_{m,n}, \quad m \leq n
\]

\[
G_{m+1,n} = -\tilde{X}_m G_{m,n}, \quad m \geq n
\]

Reuter et al.: 10.1088/1749-4699/5/1/014009
Papior et al.: 10.1016/j.cpc.2016.09.022
Enforcing BTD — going quasi 1D
Pivoting matrix elements, order from dis-order

- 2,400 atoms
- 21,600 orbitals

\[
\begin{pmatrix}
A_1 & C_2 & 0 & \cdots & & \cdots & \cdots \\
B_1 & A_2 & C_3 & 0 & \cdots & \cdots & \cdots \\
0 & B_2 & \ddots & \ddots & 0 & \cdots & \cdots \\
\vdots & 0 & \ddots & \ddots & \ddots & \ddots & \cdots \\
\vdots & 0 & \cdots & C_p & \cdots & \ddots & \ddots \\
\vdots & 0 & \cdots & \cdots & \cdots & B_{p-1} & A_p \\
\end{pmatrix}
\]

- White = 0
- Black ≠ 0

Elec-1 →
Elec-2 →
Elec-3 →
Elec-4 →
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\end{pmatrix}
\]

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\[\text{Elec-1} \rightarrow \text{Elec-2} \rightarrow \text{Elec-3} \rightarrow \text{Elec-4}\]
Memory and time for varying system sizes (3D bulk):

- **x-axis**: length
- **y-axis**: width

Brandbyge et.al.: 10.1103/PhysRevB.65.165401
Papior et.al.: 10.1016/j.cpc.2016.09.022
Speedup and scalability

Memory and time for varying system sizes (3D bulk):

- $x$-axis: length
- $y$-axis: width

Threading performance
Reduces memory consumption by $1/N_T$

Brandbyge et al.: 10.1103/PhysRevB.65.165401
Papior et al.: 10.1016/j.cpc.2016.09.022
Currently known investigated systems

- ~2000 atoms
- ~3000 atoms
- ~5000 atoms

Brandimarte et al.: 10.1063/1.4974895, Obesteiner et al.: 10.1021/acs.nanolett.7b03066
Brandimarte et al.: under review
Optimizing the integration

- Integration in the complex plane for the equilibrium contour $G - G^\dagger$

\[ \mathcal{R}^+ = \int G(z) \]

\[ \mathcal{R}^- = -\int G^\dagger(z) \]
Optimizing the integration

- Integration in the complex plane for the equilibrium contour $G - G^\dagger$
- Gaussian quadrature have more points close to the integration boundaries

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Optimizing the integration

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\[ R^+ = \int G(z) \]
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Eigenvalues
\[ \Re E - E_F \text{ [eV]} \]
\[ \Im E - E_F \text{ [eV]} \]

Total # of contour points
Abs. error
Cont. fraction
Circle contour
C+
C+ + C−
Simpson

Papior et al.: 10.1016/j.cpc.2016.09.022
Optimizing the integration

$k$-points on

- Bias window calculation $\text{GG}^\dagger$
- Only states close to the reference energy are important!
Optimizing the integration

- Bias window calculation $G \Gamma G^\dagger$
- Only states close to the reference energy are important!
DFT couple to tight-binding – graphene

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3. DFT coupled to tight-binding – graphene
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Current problems involve a model-tip coupling via $\Gamma$

For complex chemical defects and/or complex electrode attachments, tight-binding is not accurate enough.
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For complex chemical defects and/or complex electrode attachments, tight-binding is not accurate enough.

1. Calculate the self-energy from the tip-electrode region
2. Propagate the self-energy onto the red atoms
3. Cut the self-energies to the parameterized orbitals

Gaetano Calogero

sisl: 10.5281/zenodo.597181
DFT coupled to tight-binding – graphene

DFT → TB

**DFT**

Two DFT calculations are carried out:

1. Pristine graphene calculation from which TB parameters are extracted.
2. Calculation of a defected region surrounded by pristine (in every sense) graphene. The pristine graphene region must be big enough to screen the “defected” region.

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From pristine DFT calculation extract the non-orthogonal parameters up to a user-specified range. These parameters are used to construct a non-orthogonal tight-binding model of pristine graphene to a user-defined size. Next, any region of the system may be replaced by the self-energy describing local defects.
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**Tight-binding**

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- Selecting the parameters is done using the orbitals that have all the weight in the energy range specified.

- It is crude, but it works!
DFT coupled to tight-binding – graphene

Gold STM tip

Injection into infinite graphene using an Au-tip structure

\[ E = -0.8 \text{eV} \]

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DFT coupled to tight-binding – graphene

Gold STM tip

Injection into infinite graphene using an Au-tip structure

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More than one region of defects

Two-tip structure by inserting the same tip in two different places. Bond-currents from the left tip are seen.
Co-workers and collaborators

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