Electron-phonon scattering from green’s function transport combined with molecular dynamics: Applications to mobility predictions

Daniele Stradi

www.quantumwise.com
daniele.stradi@quantumwise.com
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What if ...

- ... the system is large?
- ... we want to study an interface?
- ... the material is not crystalline?
- ... we want to include anharmonic effects?
Objective

Obtain mobilities and resistivities without the explicit evaluation of the electron-phonon coupling matrix.

Two general methods with increasing efficiency:

- **MD-Landauer method [1]**: Finite-temperature effects are accounted for by using molecular dynamics (MD).
- **STD-Landauer method**: Finite-temperature effects are accounted for by a special thermal displacement (STD).

Computational methods

- Electronic structure
  - ATK-DFT or ATK-DFTB
  - LCAO basis set: DZP or SG15-M
  - LDA or GGA-PBE xc-functionals

- Molecular dynamics and phonons
  - ATK-ForceField
  - EAM or Tersoff potentials
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In the MD region with length $L$ the atomic positions are evolved using MD simulations at finite temperature.

A number (10-50) of MD trajectories is equilibrated at a target temperature using a random Maxwell-Boltzmann distribution of initial velocities to create an ensemble of snapshots of the structure at that temperature.

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The electronic transmission is calculated for all the snapshots using the NEGF approach and the resulting transmission functions are averaged to obtain the finite-temperature transmission $\langle T \rangle$.

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The finite-temperature resistance is calculated from the finite-temperature conductance $\langle G \rangle$ obtained using the Landauer formula:

$$\langle R^{-1} \rangle = \langle G \rangle = \frac{2e^2}{h} \int \langle T \rangle \left( -\frac{\partial f}{\partial E} \right) dE$$
The finite-temperature resistance is calculated for increasingly longer lengths $L$ of the MD region.
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The 1D resistivity $\rho_{1D}$ is obtained by a linear-regression of the $R$ vs. $L$ data using:

$$R(L) = R_c + \rho_{1D} \cdot L$$

MD-Landauer method [1]: Finite-temperature bulk resistivity

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- The bulk resistivity $\rho_{\text{Bulk}}$ is obtained by multiplying $\rho_{1D}$ for the cross-sectional area of the system $A$:

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- The electron mobility can be calculated from $\rho_{\text{Bulk}}$ as:

$$\mu = \frac{1}{q n \rho_{\text{Bulk}}} = \frac{A}{q \tilde{n} \rho_{1D}}$$

$$\tilde{n} = \frac{A}{n} = \int_{E_g}^{\infty} f(E_g, E, T) D(E) dE$$

For bulk Si, the mobility calculated with the MD-Landauer method agrees with that obtained using the BTE and with that measured experimentally [2].

The MD-Landauer methods captures the reduced mobility in the 1D nanowire compared to the bulk.

For a number of systems, the mobilities calculated using the 1D method agree with those calculated using the BTE within a factor \(~2\).

The method is applicable to:
- Metallic systems
- Semiconducting systems
- 1D, 2D and 3D systems

MD-Landauer method [1]: Pros and cons

😊 **Pros:**
- Conceptually simple
- Mobilities and resistivities agree semi-quantitatively with BTE and experiments and trends are correctly reproduced
- Can be applied to complex geometries
- Includes anharmonic effects

👎 **Cons:**
- Requires extensive sample averaging
- Too time consuming for large device calculations

STD-Landauer method: Device set-up

Central region

Electrode

MD region

Electrode

$L$
Key idea: all temperature effects are accounted for by a single distorted atomic configuration based on a canonical average over all phonon modes [1].

In the STD (Special Thermal Displacement) region each $\kappa$-th atom is displaced by $\Delta r_{\kappa\alpha}$ along the Cartesian direction $\alpha$ according to:

$$
\Delta r_{\kappa\alpha} = (M_p/M_\kappa)^{1/2} \sum_{\nu} (-1)^{\nu-1} e^\nu r_{\kappa\alpha,\nu} \sigma^\nu_{\nu,T}
$$

$$
\sigma^\nu_{\nu,T} = (2n_{\nu,T} + 1) \ell^2_{\nu}
$$

For bulk Au, the mobility calculated at 300 K using the **STD-Landauer** method matches that calculated using the **MD-Landauer** method.
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Using the STD-Landauer method, fewer transmission calculations are performed, and the time-to-result (~time of Landauer transmission calculations) is reduced by one order of magnitude.

### Comparison of STD-Landauer and MD-Landauer

<table>
<thead>
<tr>
<th></th>
<th>MD-Landauer</th>
<th>STD-Landauer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nr. of calculations</td>
<td>31</td>
<td>4</td>
</tr>
<tr>
<td>Time-to-result (h)</td>
<td>18.5</td>
<td>2.0</td>
</tr>
</tbody>
</table>

\[ \rho_{\text{bulk, MD-Landauer}} = 3.28 \times 10^{12} \Omega \cdot m \]

\[ \rho_{\text{bulk, STD-Landauer}} = 3.23 \times 10^{12} \Omega \cdot m \]
The $\Sigma 3$ twin boundary is related to the pristine $\langle 111 \rangle$-oriented system by a $60^\circ$ rotation around the C vector.
The Σ3 twin boundary is related to the pristine (111)-oriented system by a 60° rotation around the C vector.

The specific resistivity $\gamma_R$ of the twin grain boundary can be calculated from the resistance of the grain boundary $R_{\Sigma3}$ and that of the pristine system $R_{Pristine}$ [1]:

$$\gamma_R = \frac{(R_{\Sigma3} - R_{Pristine}) \cdot A}{N_{\Sigma3}}$$

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Including finite-temperature effects brings $\gamma_R$ above the experimental value.

Conclusions

- The MD-Landauer and STD-Landauer methods allow for the calculation of mobilities and resistivities including electron-phonon coupling effects without explicitly evaluating the electron-phonon scattering matrix.
- The MD-Landauer approach is validated against mobility calculations using the Botzmann transport equations and describes well the temperature dependence of the mobility for a wide range of materials.
- The STD-Landauer approach gives results in close agreement with the MD-Landauer method at a fraction of the computational cost, and can be applied routinely to investigate the temperature dependence of the resistivity in complex systems.
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Troels Markussen,1 Mattias Palsgaard,1,2 Daniele Stradi,1 Tue Gunst,2 Mads Brandbyge,2 and Kurt Stokbro3

1QuantumWise A/S, Prøfbjerget 3, Postbox 4, DK-2180 Copenhagen, Denmark
2Department of Micro- and Nanotechnology (DTU Nanotech), Center for Nanostructured Graphene (CNG), Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark
3Department of Physics, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

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Thank You!

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