Thermal boundary resistance predictions with non-equilibrium Green’s function and molecular dynamics simulations

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The non-equilibrium Green’s function (NEGF) method with Büttiker probe scattering self-energies is assessed by comparing its predictions for the thermal boundary resistance with molecular dynamics (MD) simulations. For simplicity, the interface of Si/heavy-Si is considered, where heavy-Si differs from Si only in the mass value. With Büttiker probe scattering parameters tuned against MD in homogeneous Si, the NEGF-predicted thermal boundary resistance quantitatively agrees with MD for wide mass ratios. Artificial resistances that the unaltered Landauer approach yield at virtual interfaces in homogeneous systems are absent in the present NEGF approach. Spectral information result from NEGF in its natural representation without further transformations. The spectral results show that the scattering between different phonon modes plays a crucial role in thermal transport across interfaces. Büttiker probes provide an efficient and reliable way to include anharmonicity in phonon related NEGF. NEGF including the Büttiker probes can reliably predict phonon transport across interfaces and at finite temperatures.

Semiconductor nanodevices such as quantum cascade lasers, LEDs and thermoelectric devices are typically composed of several semiconductor materials. Scattering of thermal energy carriers at the interface between two materials results in thermal boundary resistance. The size of the thermal boundary resistance was previously reported to be comparable to that of pure materials with lengths of a few to tens of nanometers. Predicting the thermal boundary resistance gives important insight into the device physics and enables design improvements. Often, molecular dynamics (MD) is used to model the thermal boundary resistance and reproduce experimental data. Inelastic phonon scattering is included in MD simulations through the anharmonicity of the interatomic potential. The non-equilibrium Green’s function (NEGF) method is widely accepted as one of the most consistent methods for electronic quantum transport in nanodevices. In particular for predicting stationary device physics, NEGF is potentially more attractive than MD given that it is a spectral approach when setup in energy space, though modal methods in non-equilibrium MD have just begun to be developed. When electrons and phonons are both solved in the NEGF framework, interparticle interactions and energy and momentum transfer in e.g. self-heating or thermoelectric situations can be described on equal footing with the predictions of the respective particles’ propagation. For phonon transport, however, the NEGF method has been used predominantly in the coherent (harmonic) regime due to the fact that the inclusion of incoherent scattering such as phonon-phonon decay usually requires solving polarization graphs in the self-consistent Born approximation which entails a large numerical load. It has been shown that the lack of anharmonicity in NEGF simulation gives incorrect thermal boundary resistance predictions at high temperatures.

In this work, a numerically efficient method to solve phonon transport in the NEGF framework including phenomenological phonon scattering with Büttiker probes is presented and benchmarked against MD. The artificial resistance at the virtual interface in a homogeneous structure that plagues the equilibrium Landauer approach is absent in the present NEGF approach. When solved for homogeneous systems, this NEGF method yields vanishing interface resistance. The thermal boundary resistance calculated with this NEGF method shows quantitative agreement with MD simulations. The extracted spectral transport information from NEGF shows that the different phonon modal contributions play an important role in thermal transport across the interface.

Fig. (a) shows the simulation domain considered in both MD and NEGF. The system consists of Si to the left and heavy-Si to the right of an interface at position 0. The heavy-Si differs from Si only in its atomic mass ratio vs. Si. For all simulations in this work, the lattice constant is fixed at 28.085 Å and a range of 1 to 10 is considered for . Transport is solved within a range L/2 to the left and right of the interface. The lattice temperature for regions further to the left and right of the interface is assumed constant and to equal 320 K and 280 K, respectively. Phonon transport occurs along the x direction and the system is considered to be periodic along y and z directions. The harmonic phonon bandstructure is described with a Tersoff potential and the lattice constant is set to 5.431 Å. Details of the thermal boundary resistance extraction are illustrated in Fig. (b). Firstly, linear fits are performed on the local temperature profiles to the left and to the right of the interface, respectively. The local temperature is obtained by minimizing the difference between the lo-
For MD calculations, the discretized device cross-section is in the energy domain, which gives spectral data without additional transformations. To calculate the harmonic interatomic force constants (IFCs), a $3 \times 3 \times 3$ unit cell bulk Si structure is relaxed in LAMMPS using the Tersoff potential. The derivatives of the forces between atoms with respect to the atom position variations give the harmonic IFCs. These values are then loaded into NEMO5 to construct the dynamical matrix. Only the transport direction ($x$) is discretized in real space. The periodic directions ($y$ and $z$) are represented with a single conventional unit cell. Longer-ranged periodicity is represented with the phonon momenta in reciprocal space. Anharmonic phonon decay is included via Büttiker probes.

The Büttiker probe acts as a 2D array with phonon momenta in reciprocal space. The local phonon density of states is then switched to a microcanonical ensemble (NVE) and a stationary Green’s functions are solved with the recursive Green’s function (RGF) method. The phonon density of states is then loaded into NEMO5 to construct the dynamical matrix. The spatial distribution of local phonon density of states (LDOS) can illustrate the contributions of different phonon-phonon Umklapp processes.

Anharmonic phonon decay is included via Büttiker probes. To ensure energy conservation, the Büttiker probe temperature is solved iteratively by Newton’s method until the integrated energy current vanishes for each Büttiker probe.

The LAMMPS package is used for all MD simulations in this work. The lengths of both the heat source and the heat sink are $L/10$ and the simulation timestep is $0.4$ fs. Periodic boundary conditions are applied in $y$ and $z$ directions while the fixed boundary condition is applied in the $x$ direction. For MD calculations, the discretized device cross-section is of $8 \times 8$ conventional unit cells, with each conventional cell containing 8 atoms. First, a canonical ensemble (NVT) is considered and run for $1.2$ ns to relax the structure, allowing the system to reach thermal equilibrium at $300$ K. The system is then switched to a microcanonical ensemble (NVE) and a constant heat flux is added to the heat source and extracted from the heat sink for $12$ ns. After the system reaches steady state, the local temperature of each cell is obtained by averaging over ten million timesteps in the last $4$ ns. $L$ of $92$, $130$ and $184$ unit cells is used for MD based simulation of each $MR$ value.

For all NEGF simulations, the nanodevice simulation tool NEM05 is used. Stationary Green’s functions are solved in the energy domain, which gives spectral data without additional transformations. To calculate the harmonic interatomic local phonon energy density and the product of a local Bose-Einstein distribution and the local phonon density of states.

For a given atom, the local temperature $T$ solves the equation

$$
\sum_{q_{\parallel}} \int_{0}^{\infty} \omega \rho(\omega) d\omega = \sum_{q_{\parallel}} \int_{0}^{\infty} \omega D(\omega) f_{BE}(\omega, T) d\omega,
$$

where $\omega$ is the phonon frequency and $q_{\parallel}$ is the transverse phonon wave vector. $\rho$ is the local phonon number density and $D$ is the local phonon density of states. The temperatures $T_1$ and $T_2$ of the Si and heavy Si in the vicinity of the interface (see Fig. 1(b)) are determined with the fitted temperature profiles. Finally, the thermal boundary resistance is calculated as $R = (T_1 - T_2)/q$, where $q$ is the simulated heat flux.

The Büttiker probe self-energies at atom $i$ can be solved with the recursive Green’s function (RGF) method, the device is partitioned into slabs perpendicular to transport direction. This limits the peak memory usage during computation, but requires the Büttiker probe self-energies to be equal throughout each slab.

The spatial distribution of local phonon density of states (LDOS) can illustrate the contributions of different phonon-phonon Umklapp processes. Anharmonic phonon decay is included via Büttiker probes. To ensure energy conservation, the Büttiker probe temperature is solved iteratively by Newton’s method until the integrated energy current vanishes for each Büttiker probe.

The local phonon density of states is

$$
\sum_{\omega}^{RBP_{i,m}}(\omega) = -i \frac{2 \omega \hbar^2}{\tau_{i,m}^{-1}(\omega)}. \tag{2}
$$

$T_i$ is the phonon Büttiker probe temperature of the atom $i$. It represents the local temperature for the limit of complete phonon thermalization at each atom. The actual local temperature differs from the Büttiker probe one depending on the scattering strength.

The spatial distribution of local phonon density of states (LDOS) can illustrate the contributions of different phonon modes to the heat flux in various regions. For this purpose, four regions of interest, consisting of three atomic layers each are defined in Fig. 1(a). The LDOS $\phi_{i}(\omega)$ of any region $i$ of these regions is averaged over all of its atoms. For a given phonon frequency, the L1-norm of the LDOS summed over all regions is defined as

$$
\|\phi_{\text{tot}}(\omega)\| = \sum_{i=1}^{4} \phi_{i}(\omega). \tag{4}
$$

Each discretized atom in the system has a Büttiker probe applied to it. Following Ref. [21] we approximate the phonon frequency $\omega$ dependent lifetime $\tau$ as isotropic and assume it represents only the phonon-phonon Umklapp process.

$$
\tau_{-1}(\omega) = \tau_{i}^{-1}(\omega) = BT_{i} \omega^{2} e^{-C/T_{i}}. \tag{3}
$$
The relative contribution of LDOS of region $i$ is then defined as

$$R_i(\omega) = \frac{\phi_i(\omega)}{||\phi_{tot}(\omega)||}.$$  

Fig. 2(b) shows the thermal boundary resistance as a function of $MR$ calculated with NEGF agrees quantitatively with the MD predictions. For both methods, the thermal boundary resistance increases exponentially with $MR$ and vanishes when $MR$ tends to unity (i.e. for a homogeneous system). Small remaining difference between the MD and NEGF results are systemic to the different treatment of phonon modes perpendicular to the transport direction: In NEGF, the phonon momentum perpendicular to transport is explicitly resolved as a parameter in the Dyson and Keldysh equations. In contrast, MD calculations require as large as possible unit cells perpendicular to transport to cover as many phonon modes with long wave lengths in these directions as feasible. Another source of differences can be the open boundary condition in transport direction: Figs. 3(a) and (b) benchmark the open boundary condition treatment in MD and NEGF, since they illustrate finite-size effects with the thermal boundary resistance $R$ as a function of the inverse system length $1/L$. For MD, the slope of $R$ vs $1/L$ increases with the mass ratio $MR$, i.e. with reducing average sound velocity in the device. This agrees with similar findings discussed in detail in Ref. [7]. Note that the mean free path of Si at room temperature is about 300 nm. Therefore, it is practically impossible to completely eliminate the size effect along the transport direction in MD due to computational cost. In contrast to the finite sized boundary reservoirs of MD, contact self-energies in NEGF incorporate semi-infinite leads as phonon reservoirs. Accordingly, we observe the slope of $R$ vs. $1/L$ in NEGF predictions is much smaller than that of MD. Also its increase with $MR$ is comparably negligible. This different boundary treatment is another source of some differences between MD and NEGF seen in Fig. 2(b).

FIG. 2. (a) Thermal conductivity of bulk Si calculated with MD and NEGF. The Büttiker probe parameters $B$ and $C$ are fitted such that the transport results of NEGF agree with MD. (b) Thermal boundary resistance as a function of the mass ratio ($MR$) calculated by NEGF with the fitted parameters $B$ and $C$ of (a) and MD. The error bar of MD simulation is calculated but not plotted since the standard deviation for independent simulations is only 1.6% to 4.6% of the average resistance at the Si/heavy-Si interface for different mass ratios. The dotted line is the relative difference of the two methods and is defined as $(\text{NEGF} - \text{MD})/\text{MD} \times 100\%$. The green curve shows the results calculated by NEGF without Büttiker probe self-energies. With increasing masses, the maximum phonon frequency and (with the frequency dependence of the Büttiker probes, Eq. 2) the average scattering strength reduces. Consequently, the impact of scattering on $R_{\text{interface}}$ declines with increasing mass of heavy-Si.

FIG. 3. Linear extrapolation of (a) NEGF and (b) MD for three $MR$ values. MD results show a stronger dependence on the length of the device. Open boundary conditions included in NEGF with contact self-energies give almost device size independent results.

Fig. 4(a) shows the phonon density of states (DOS) in homogeneous Si and heavy-Si solved with NEGF. The DOS of heavy-Si is limited to energies at or below 33.3 meV, two times less than in native Si in agreement with the applied $MR = 4$. Incoherent phonon scattering allows phonons with energies above this heavy-Si cutoff energy to propagate. This is illustrated in Figs. 4(b), (c) and (d) with the energy resolved current densities of the heat source and heat sink of the device shown in Fig. 1(a) when solved in NEGF with different scattering strengths. While the energy distribution changes with scattering, the total energy current (on the order of $10^3 W/m^2$) is conserved, and the difference of the total energy current between the source and the sink is on the order of 1 W/m². In Fig. 4(b), normal scattering strength (fitted to reproduce the MD-calculated bulk thermal conductivity) is applied. The source current with energies above the energy cutoff is finite since its corresponding phonons can relax to lower energies at the heavy-Si side via inelastic scattering. In Fig. 4(c), an artificially weak scattering strength is used ($1/20 \times$ normal scattering strength). Accordingly, the profiles of current in both the heat source and sink follow the profile of the heavy-Si DOS. In contrast, Fig. 4(d) shows the NEGF results when artificially strong scattering is used ($20 \times$ normal scattering strength). The current profiles in the heat source and heat sink follow the profiles of the DOS in the Si and the heavy-Si leads, respectively. The results show that stronger inelastic scattering brings the system closer to local thermodynamic equilibrium.

In Fig. 4(b), four current peaks in the heavy-Si heat sink are located at 12 meV, 22 meV, 27 meV and 32 meV, respectively. They correspond to the four peaks of the heavy-Si DOS shown in Fig. 4(a). The relative magnitudes of the four current peaks do not follow the relative magnitudes of the four DOS peaks (i.e., the list of current peaks arranged in decreasing order of magnitude is 22 meV > 12 meV > 27 meV > 32 meV, whereas the same list according to the DOS magnitude is
32 meV > 12 meV > 22 meV > 27 meV). Without interfaces involved, the current is expected to be proportional to the product of the DOS and the group velocity. The results of Figs. (a) can be understood in Fig. since it illustrates the different relative contributions of phonon modes in different device areas. Phonon modes around 12 meV reside nearly exclusively in the heavy-Si. Consequently, they contribute less to the overall heat current. In contrast, phonon modes around 22 meV are present in all 4 device regions considered in Fig. Therefore, these modes can maintain a higher contribution to the total heat current.

In conclusion, NEGF with Büttiker probe scattering is applied to the thermal boundary resistance of the Si/heavy-Si interface. The empirical NEGF scattering parameters are tuned to reproduce the thermal conductivity of homogeneous Si predicted by MD. The scattering parameters proved to be transferable, since the NEGF results of the thermal boundary resistance quantitatively agrees with MD results for mass ratios ranging from 1 to 10. The artificial resistance at the virtual interface in a homogeneous structure that plagues the equilibrium Landauer approach is absent in the presented NEGF approach. Besides, the present NEGF approach is found to be numerically more efficient than MD. Thanks to the open boundary conditions, NEGF shows virtually no finite-size effects compared to MD. The analysis of the NEGF spectral information shows that the scattering between different phonon modes determines the phonon energy current flow across interfaces. Future improvements of our approach can include a more accurate description of inelastic scattering at the interface and enable computationally efficient calculation of spectral current adjacent to the interface.

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