Semiclassical Phonon Transport in the Presence of Rough Boundaries

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Phonons in Nanostructures

- Phonons scatter with boundaries (mesoscale), imperfections (nanoscale) and isotope/dopant atoms (atomic scale).

- Strong interactions with rough boundaries or interfaces between materials

- Need to understand thermal transport in the presence of both roughness and internal scattering

Biswa et al., Nature 489, 414 (2012)

Lessons from electronic transport

Semiclassical (electron BTE and derivatives)
Drift-diffusion equations
Hydrodynamic equations
BTE in the relaxation-time approximation (RTA)
Ensemble Monte Carlo

Quantum corrections
Wigner transport equation (Wigner Monte Carlo)
Quantum hydrodynamic equations
Single-particle density matrix formalism

Quantum
Nonequilibrium Green’s functions
(NEGF+atomistic tight binding, NEGF+DFT for small systems)

More Accurate
More Complicated
Phonon transport

**Semiclassical (phonons BTE and derivatives))**
- Heat diffusion equation
- BTE in the RTA
- Ensemble Monte Carlo
- Discrete ordinate method
- Direct solutions to BTE

**Classical (atoms)**
- Molecular dynamics

**Quantum (phonons)**
- Nonequilibrium Green’s functions

**Great recent work to extract accurate rates from DFPT to use with Boltzmann transport kernels**

Currently not possible to address phonon-phonon scattering

**Great recent work on applying ab initio to extract accurate potentials for use in MD**
Why semiclassical phonon transport?

**Pros:**

✔ Computationally tractable for experimentally relevant sizes
✔ A variety of techniques adaptable to geometry
✔ All kinds of scattering mechanisms can be incorporated
✔ Intuitive (humans like the distribution function)

**Cons:**

↓ Cannot address coherent transport features
↓ Does not account for states being affected by interactions
↓ Must a priori decide which mechanisms to account for
↓ Only as good as the rates used, rates obtained from elsewhere
Semiclassical transport

From intro-level quantum-mechanics textbooks: The semiclassical (or WKB) approximation holds when the potential vary slowly on the scale of the particle wavelength. The concept of classical trajectories is viable.

When is the semiclassical approximation applicable to transport?

When **relevant length/time scales** on which the single-particle Green’s function varies are **much longer than coherence lengths and times**

\[
\left( \omega - \frac{p^2}{2m} - U(R,T) - \text{Re} \sum \left( p, \omega ; R, T \right) \right)^{-1}
\]

\[
+ \left[ \text{Re} g(p, \omega ; R, T), \sum^{<} (p, \omega ; R, T) \right]
\]

\[
= - \sum^{>} (p, \omega ; R, T) g^{<} (p, \omega ; R, T)
\]

\[
+ \sum^{<} (p, \omega ; R, T) g^{>} (p, \omega ; R, T)
\]

Kadanoff and Baym, Quantum Statistical Mechanics, W.A. Benjamin, New York, 1962
Example from the electronic world

What scattering does to coherences in a double-barrier tunneling structure

Scattering causes transition from resonant to sequential tunneling

Scattering kills off spatial coherences (off-diagonal terms)

Wigner Monte Carlo simulation of phonon-induced electron decoherence in semiconductor nanodevices

Damien Querlioz, Jérôme Saint-Martin, Arnaud Bournel, and Philippe Dollfus
Institut d’Electronique Fondamentale, CNRS, University Paris-Sud, 91405 Orsay, France
Semiclassical transport

Semiclassical approximation boils down to transport being diffusive:
Length scales longer than the mean free path, timescales longer than relaxation time

Technically, to obtain the Boltzmann equation, two more approximations are needed:
• The Born approximation (scattering event completed before the next one)
• Scattering does little to alter the density of states

\[
\int \frac{d\omega}{2\pi} \frac{g^>(p,\omega;R,T) + g^<(p,\omega;R,T)}{z - \omega} = g(p,z;R,T)
\]

Example of semiclassical phonon transport

A long smooth nanowire, room temperature

The same nanowire, much lower temperature

Length > mean free path

Length < mean free path
Phonons and boundary roughness

Depending on wavelength, they are treated as waves or classical objects when interacting with a rough boundary or rms roughness $\Delta$ and correlation length $L$.

**Wave limit**
- Phonons should be treated as waves.
- Interference effects important.
- Specularity parameter OK.

**“Ray phononics” (particle) limit**
- Semiclassical approximation valid.
- Treat phonons as little balls with classical trajectories.
The role of correlation length

$L=0$: Nearly isotropic diffuse portion of the scattered wave

$L>0$: With increasing correlation length, the diffusely scattered wave collimates along the direction obeying Snell’s law

$L \to \infty$: Even a rough surface appears completely smooth, as the diffuse and specular reflected waves align
Specularity parameter $p$

Intuitively plausible: 

- $p$ – probability of specular reflection
- $1-p$ – probability of having momentum randomized

Often assumed as a constant, but should be momentum-dependent

Technically valid for $L \to 0$

OK for phonons whose wavelengths are considerably larger than the correlation length

\[
p(\vec{q}) = \exp(-\langle \phi^2 \rangle)
\]

\[
\phi(\vec{q}, \vec{r}) = 2\vec{q} \cdot \hat{s}_z(\vec{r}) = 2qz(\vec{r})\cos \Theta_B
\]

\[
\langle \phi^2 \rangle = (2q \cos \Theta_B)^2 \langle z^2 \rangle = 4q^2 \Delta^2 \cos^2 \Theta_B
\]

\[
p(\vec{q}) = \exp(-4q^2 \Delta^2 \cos^2 \Theta_B)
\]
Examples of momentum-dependent specularity parameter $\rho(q)$ used in conjunction with the single-mode RTA and full dispersions on:

1) Thin silicon nanomembranes
2) Graphene nanoribbons
3) SiGe superlattices
Thin Si Nanomembranes

LA phonons scatter strongly from \{111\} and \{100\}

TA phonons scatter strongly from \{100\}
Effective specularity parameter is dependent on branch and temperature. At high roughnesses, here is still a considerable specular component.

Z. Aksamia* and I. Knezevic*

PHYSICAL REVIEW B 82, 045319 (2010)
Thin Si Nanomembranes

FIG. 3. (Color online) Lattice thermal conductivity of 100 nm (filled circles), 30 nm (open circles), and 20 nm (squares) thick SOI, according to the measurements of Refs. 46 and 47. The curves present our calculations with $\Delta=0.35$ nm for 100 nm SOI, $\Delta=0.4$ nm for 30, and $\Delta=0.45$ nm for 20 nm SOI, in very good agreement with experiment. Thermal conductivity is lower by almost an order of magnitude than the room-temperature bulk value of 148 W/mK, owing primarily to boundary scattering at the rough Si/SiO$_2$ interfaces.

Z. Aksamiia and I. Knezevic
PHYSICAL REVIEW B 82, 045319 (2010)
When is it OK to treat boundary scattering through an effective scattering rate?

\[ \tau_{j,B}(\vec{q}) = \left[ \frac{1 + p(\vec{q})}{1 - p(\vec{q})} \right] \frac{L}{v_{j,\perp}(\vec{q})} \]

When the presence of boundaries doesn’t affect the solution to the BTE, i.e. when boundaries are farther than the mean free path due to internal scattering, so phonons can thermalize between successive hits.

Casimir rate gives an estimate for how important boundary scattering is

Diffuse (Casimir) limit

\[ \lim_{p \to 0} \tau_{j,B} = \frac{L}{v_{j,\perp}} \]

Specular limit

\[ \lim_{p \to 1} \tau_{j,B} = \infty \]
Examples: Partially diffuse interfaces

Examples of momentum-dependent specularity parameter $\rho(q)$ used in conjunction with the single-mode RTA and full dispersions on:

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2) Graphene nanoribbons

3) SiGe superlattices
Boundary vs. internal scattering

- Model using the steady-state phonon Boltzmann transport equation

\[
\vec{v}_q \cdot \nabla r T \frac{\partial N^0_q(T)}{\partial T} + \nu_\perp(\vec{q}) \frac{\partial n_q(y)}{\partial y} = \frac{n_q(y)}{\tau_{\text{int}}(\vec{q})}
\]

- In the absence of boundaries, RTA solution:

\[
R_q = \tau_{\text{int}}(\vec{q}) \vec{v}_q \cdot \nabla r T \frac{\partial N^0_q(T)}{\partial T}
\]

- Each boundary contributes one term:

\[
p(\vec{q}) \exp \left[-W/\Lambda_{\text{int}}(\vec{q})\right]
\]

- Sum the infinite series:

\[
n_q^\pm(y) = R_q \left\{ 1 - \frac{\left[1 - p(\vec{q})\right] \exp \left[-y/\Lambda_{\text{int}}(\vec{q})\right]}{1 - p(\vec{q}) \exp \left[-W/\Lambda_{\text{int}}(\vec{q})\right]} \right\}
\]

- Heat flux is weaker near boundaries/interfaces due to roughness scattering
Suspended graphene nanoribbons (GNRs)

Thermal conductivity very hard to measure - useful to have theory!

Anisotropy in terms of ribbon orientation

\[
\tau_{LER}(\vec{q}) = \frac{v_{\perp}}{W} F_P(\vec{q}) \left[ 1 - \frac{\Lambda_{int.}(\vec{q})}{W} F_P(\vec{q}) \right]
\]

Thermal transport in supported graphene


Examples of momentum-dependent specularity parameter $\rho(q)$ used in conjunction with the single-mode RTA and full dispersions on:

1) Thin silicon nanomembranes

2) Graphene nanoribbons

3) SiGe superlattices
SiGe alloy superlattices

Lattice with reasonably thick layers and partially diffuse interfaces

**Roughness + mismatch?**

Experimentally observed:
- cross-plane/in-plane anisotropy of $\kappa$
- increasing $\kappa$ with increasing period

Separate roughness from mismatch: Calculate $\kappa$ for sublattices with rough interfaces; mismatch figures in $\kappa$, not the BTE itself
Solving the BTE for superlattices

A single sublattice with partially diffuse interfaces

\[ n_b(\vec{q}, y) = R_b(\vec{q}) \left\{ 1 - \left[ \frac{1 - p(\vec{q})}{1 - p(\vec{q}) \exp[-L/\tau_{b, \text{Internal}}(\vec{q}) \nu_{b, \perp}(\vec{q})]} \right] \exp[-y/\tau_{b, \text{Internal}}(\vec{q}) \nu_{b, \perp}(\vec{q})] \right\}. \]

Effective interface scattering rate

\[ \tau_{\text{Interface}}(\vec{q}) = \frac{\nu_{b, \perp}(\vec{q})}{L} \frac{F_p(\vec{q}, L)}{1 - \frac{\tau_{b, \text{Internal}}(\vec{q}) \nu_{b, \perp}(\vec{q})}{L} F_p(\vec{q}, L)} \]

\[ F_p(\vec{q}, L) = \frac{[1 - p(\vec{q})] \{1 - \exp[-L/\tau_{b, \text{Internal}}(\vec{q}) \nu_{b, \perp}(\vec{q})] \}}{1 - p(\vec{q}) \exp[-L/\tau_{b, \text{Internal}}(\vec{q}) \nu_{b, \perp}(\vec{q})]} \]

Si/Ge and SiGe alloy superlattices

Si/Ge: in-plane vs cross-plane anisotropy

Si/Ge: $\kappa$ increases w/increasing period

Alloy superlattices $\rightarrow$
- $\kappa$ increases w/increasing period
- the role of alloying


When is $p$ not enough?

Wavelength-independent:

- Diffuse scattering angle from $\cos \theta \, d\Omega$
- Probability distribution
- 100% diffuse scattering called the Casimir Limit
- Thermal conductivities below this limit have been measured in Si nanowires

Wavelength-dependent:

- $p(\theta_i) > p$

- For zero correlation length, and large wavelength:
  $$p(\theta_i) = e^{-(2q\sigma \cos \theta_i)^2}$$
- Diffuse scattering angle from
  $$\left[1 - e^{-(2q\sigma \cos \theta)^2}\right] \cos \theta \, d\Omega$$
- Same limit of 100% diffuse scattering as previous model
When things get really really rough....

- Rough Si nanowires show greatly reduced $\kappa$ vs bulk
- Potential thermoelectric applications
- Lower than the Casimir limit, showing limitation of the specularity model
- Depends on $\sigma$ and $L$

Phonon EMC

Heat flux ($\phi$)

$\phi = -\kappa \frac{dT}{dx}$

Real-space rough surfaces

- Generates a random rough surface from an autocorrelation function
- Captures important features, like multiple scattering
- Allows lower thermal conductivities

We can make a surface to match any ACF.

\[
ACF(\mathbf{r}) = \sigma^2 e^{-|\mathbf{r}|/L}
\]

C. Buran, M. G. Pala, M. Bescond, M. Dubois, and M. Mouis,
Models at room temperature

- Constant specularity
- Wavelength-dependent specularity

Generated Surfaces
- 2.5 nm $L$

Graph showing probability of specular scattering vs. RMS roughness (nm) for 10x10x1000nm Si wires.
Temperature dependence

Wavelength-dependent specularity ($\sigma = 0.6$ nm)

Casimir Limit

Generated surface ($\sigma = 0.3$ nm, $L = 2.5$ nm)

$k$ (W/m·K)

Temperature (K)

10x10x2000 nm Si wires
Dependence on rms roughness

L.N. Maurer and I. Knezevic,
Dependence on correlation length

Exponential autocorrelation

Gaussian autocorrelation

Phonon trapping

70 nm thick, **3 nm rms roughness**, 10 nm correlation length

70 nm thick, **6 nm rms roughness**, 10 nm correlation length

Conclusions

• Semiclassical transport simulations of great practical utility
• Specularity parameter limited to moderately rough interfaces with very low correlation
• Very rough or correlated surfaces require a different approach
• High roughness – phonon trapping
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