Molecular-Level Modeling of Phonon Transport: Formulation, Implementation, and Applications

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Outline

Session I (1:30 PM – 3:15 PM)
1. **Introduction** (McGaughey)
2. Harmonic lattice dynamics, MD simulation (Ruan)
3. Green Kubo, direct method, spectral methods (Ruan)

Session II (3:45 PM – 5:30 PM)
4. Anharmonic lattice dynamics, first principles (McGaughey)
5. Phonon-boundary and phonon-defect scattering (McGaughey)
6. Phonon-electron coupling and non-equilibrium (Ruan)
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Some History … 2002
Outline

1. Thermal Conductivity
2. Phonons
3. Computational Tools
4. Impactful Research
Stuff Gets Hot
The graph illustrates the trend in CPU power density with minimum IC feature size over time. The x-axis represents the minimum IC feature size in microns, while the y-axis shows the CPU power density in W/cm². The data points are color-coded to represent different manufacturers: AMD, Intel, and Power PC. Key milestones include the Pentium 4 (2005), Core 2 Duo (2006), and Atom (2008).
What is Thermal Conductivity?

Fourier law: \( q = -k \nabla T \)

heat flux = – thermal conductivity \( \times \) temperature gradient

- Defined empirically
- Any thermal conductivity prediction invokes the Fourier law
- Combines the effects of all energy carriers
- Provides no information about carrier-level transport
Experimental Thermal Conductivity Data

Atomic-Level Consideration

Size-Dependence in Nanostructures

Silicon thin films, in-plane direction

Jain et al., *PRB* 87 (2013) 195301.

Superlattices, carbon nanotubes, graphene, nanostructured alloys, ...

Silicon nanowires

Li et al., *APL* 83 (2003) 2934.
Why Simulations and Calculations?

- Nanoscale phenomena are challenging to observe experimentally
  - Small length (10^{-10} m) and time (10^{-15} s) scales

Why does one material have higher thermal conductivity than another? Can thermal conductivity be tailored by nanostructuring?
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2. Phonons
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What is a Phonon?

• Quantized lattice vibration with energy $\hbar \omega$

• Wave number (=2π/wavelength), frequency, polarization
  → Define the phonon mode

• Primary carriers of thermal energy in semiconductors and dielectrics (Si, GaN, graphene, quartz).

• There is a spectrum of phonons in every material.
Phonon Formula for Thermal Conductivity

Boltzmann transport equation + Fourier law

\[ \sum_{i} c_{v,i} v_{g,i,n} \tau_i = \sum_{i} c_{v,i} v_{g,i,n} \frac{\Lambda_i}{v_{g,i}} \]

- \( i \): indexes over all phonon modes
- \( c_{v,i} \): heat capacity
- \( v_{g,i} \): group velocity
- \( \tau_i \): lifetime
- \( \Lambda_i \): mean free path
Phonons scatter with:

- other phonons
- grain boundaries, interfaces, surfaces
- electrons
- defects (isotopes, vacancies, …)

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How are atomic trajectories & dynamics related to thermal conductivity?

Atom $\rightarrow$ Real
Molecular Dynamics

Phonon $\rightarrow$ Abstraction
Lattice Dynamics
What is Molecular Dynamics Simulations?

- Real-space technique (atoms, positions, velocities)
- Generate trajectories: Newton’s laws of motion & empirical potential

Water flow in a CNT
A. McGaughey, CMU

Bending a carbon CNT
J. Li, MIT

Assembling tethered nanoparticles
S. Glotzer, UMichigan
MD is Like a Mass-Spring System

1. Equations of Motion:

\[
\begin{bmatrix}
\ddot{x}_1 \\
\ddot{x}_2
\end{bmatrix} =
\begin{bmatrix}
-2 & 1 \\
1 & -1
\end{bmatrix}
\begin{bmatrix}
x_1 \\
x_2
\end{bmatrix}
\]

2. Convert to 1\textsuperscript{st} order equations:

\[
\begin{bmatrix}
\dot{x}_1 \\
\dot{x}_2 \\
\dot{x}_3 \\
\dot{x}_4
\end{bmatrix} =
\begin{bmatrix}
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1 \\
-2 & 1 & 0 & 0 \\
1 & -1 & 0 & 0
\end{bmatrix}
\begin{bmatrix}
x_1 \\
x_2 \\
x_3 \\
x_4
\end{bmatrix}
\]

3. Solve numerically
What is Lattice Dynamics Calculations?

• Reciprocal-space technique (vibrational modes)
• Taylor series of potential energy around equilibrium
  – Empirical potential or density functional theory
Harmonic Formulation

Find the natural frequencies and mode shapes for \( m = k = 1 \).

1. Equations of motion:
\[
\begin{bmatrix}
\ddot{x}_1 \\
\ddot{x}_2
\end{bmatrix} = \begin{bmatrix}
-2 & 1 \\
1 & -1
\end{bmatrix} \begin{bmatrix}
x_1 \\
x_2
\end{bmatrix}
\]

2. Assume harmonic solution:
\[
\omega^2 \begin{bmatrix}
e_1 \\
e_2
\end{bmatrix} = \begin{bmatrix}
2 & -1 \\
-1 & 1
\end{bmatrix} \begin{bmatrix}
e_1 \\
e_2
\end{bmatrix}
\]

3. Eigenvalue problem:
Solution 1: \( \omega = 0.618, e_2 = 1.618 e_1 \)
Solution 2: \( \omega = 1.618, e_2 = -0.618 e_1 \)
Normal Modes

Solution of form: \( \mathbf{x} = A \mathbf{e}_I + B \mathbf{e}_{II} \)
(coordinate transform)

Total potential energy = \( x_1^2 + (x_2 - x_1)^2 = 0.383 A^2 + 2.62 B^2 \)

Coordinates decoupled!
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2. Phonons

3. Molecular Dynamics and Lattice Dynamics

4. Impactful Research
Superlattices

Divergent Thermal Conductivity of Polyethylene

Henry and Chen,

Complex Hybrid Materials

Amorphous Materials


Thermal Conductivity from First Principles

Ultra-High Thermal Conductivity of BAs

Lindsay et al., *PRL* 111 (2013) 025901.
Nanostructure Thermal Conductivity

New Descriptions of Thermal Transport


Thermal Transport in Metals


A Huge Field! We Can’t Cover Everything.

- Focus on bulk crystals using MD and LD

- Disordered materials (alloys and amorphous)

- Solving the Boltzmann transport equation (key for interpreting experiments)
  - Theoretical and numerical approaches
  - Austin Minnich, Gang Chen, Sandip Mazumder

- Interfaces (will discuss a bit)

- Fluids (no phonons, but can apply MD)