Lattice Dynamics

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Phonon Heat Conduction

- Phonons are quantized lattice vibrations
- Govern thermal properties in electrical insulators and semiconductors
- Can be modeled to first order with spring-mass dynamics

Wave solutions
- Wave vector $K = \frac{2\pi}{\lambda}$
- Phonon energy $= \hbar \omega$
- Dispersion relations gives $\omega = f_n(K)$

![Diagram showing phonon wave vector](image)

- Optical branch
- Acoustic branch
- Sound speed (group velocity)
Heat Conduction Through Thin Films

- Experimental results for 3-micron silicon films
- Non-equilibrium scattering models work fairly well
- Crystalline structure often has larger impact than film thickness

Asheghi et al., 1999
Heat Conduction Through Multiple Thin Films

- Fine-pitch 5 nm superlattices
- Cross-thickness conductivity measurement
- Measured values are remarkably close to bulk alloy values (nearly within measurement error)
- Expected large reduction in conductivity not observed

Cahill et al., 2003
Lattice Vibrations

- Consider two neighboring atoms that share a chemical bond

\[ U(r) = k(r - r_0)^2 \]

- The bond is not rigid, but rather like a spring with an energy relationship such as…
Lattice Vibrations, cont’d

• Near the minimum, the energy is well approximated by a parabola
  \[ U = \frac{1}{2} gu^2 \]
  \( u = r - r_0 \) and \( g = \text{spring constant} \)

• Now consider a one-dimensional chain of molecules

\[ \text{spring constant} \quad g \quad \text{atom, mass} \quad m \]

\[ \text{a} \]
Lattice Energy and Motion

- **Harmonic** potential energy is the sum of potential energies over the lattice
  \[ U^{\text{harm}} = \frac{1}{2} g \sum_n \left\{ u[na] - u[(n+1)a] \right\}^2 \]

- Equation of motion of atom at location \( u(na) \)
  \[ F = m \frac{d^2 u(na)}{dt^2} = -\frac{\partial U^{\text{harm}}}{\partial u(na)} = -g \left\{ 2u(na) - u[(n-1)a] - u[(n+1)a] \right\} \]

- Simplified notation
  \[ m \frac{d^2 u_n}{dt^2} = -g \left\{ 2u_n - u_{n-1} - u_{n+1} \right\} \]
Lattice Motion, cont’d

• Seek solutions of the form

\[ u_n(t) \sim \exp\left\{ i \left( Kna - \omega t \right) \right\} \]

• Boundary conditions
  ♦ Born-von Karman: assume that the ends of the chain are connected
    • \( u_{N+1} = u_1 \)
    • \( u_0 = u_N \)
Lattice Motion, cont’d

• Then the boundary conditions become

\[ u_{N+1} \sim \exp\left\{ i \left[ K (N + 1) a - \omega t \right] \right\} \]

\[ u_1 \sim \exp\left\{ i \left[ Ka - \omega t \right] \right\} \]

\[ \rightarrow 1 = \exp\left\{ iKNa \right\} \rightarrow KNa = 2\pi n, \]

where \( n \) is an integer

• Let \( \lambda \) be the vibration wavelength, \( \lambda = aN/n \)

\[ K = \frac{2\pi n}{aN} = \frac{2\pi}{\lambda} \]

\( K = \) wave vector

• Minimum wavelength, \( \lambda_{\text{min}} = 2a = 2(\text{lattice spacing}) \)
Solution to the Equations of Motion

- Substitute exponential solution into equation of motion
  \[-m\omega^2 e^{i(Kn_a - \omega t)} = -g \left[ 2 - e^{-iKa} - e^{iKa} \right] e^{i(Kn_a - \omega t)}\]
  \[= -2g (1 - \cos Ka) e^{i(Kn_a - \omega t)}\]
- Solve for \(\omega\)
  \[\omega(K) = \sqrt{\frac{2g(1 - \cos Ka)}{m}} = 2\sqrt{\frac{g}{m}} \left| \sin(\frac{1}{2} Ka) \right|\]
- This is the dispersion relation for acoustic phonons
  - relates phonon frequency (energy) to wave vector (wavelength)
Dispersion Curve

- Changing $K$ by $2\pi/a$ leaves $u$ unaffected
  - Only $N$ values of $K$ are unique
  - We take them to lie in $-\pi/a < K < \pi/a$

\[
\omega(K) = 2(g/m)^{1/2}
\]
Wave Velocities

- Phase velocity: \( c = \frac{\omega}{K} \)
- Group velocity: \( v_g = \frac{\partial \omega}{\partial K} = a\left(\frac{g}{m}\right)^{1/2}\cos\left(\frac{Ka}{2}\right) \)
- For small \( K \):
  \[
  \lim_{K \to 0} \omega = a \sqrt{\frac{g}{m}} |K|
  \]
  \[
  \Rightarrow \lim_{K \to 0} v_g = a \sqrt{\frac{g}{m}} = \left| \frac{\omega}{K} \right| = c
  \]
- Thus, for small \( K \) (large \( \lambda \)), group velocity equals phase velocity (and speed of sound)
- We call these acoustic vibration modes
Notes on Lattice Vibrations

• For \( K = \pm \pi/a \), the group velocity is zero
  ∗ why? \[ \frac{u_{n+1}}{u_n} = \exp\{iKa\} = \exp\{i\pi\} = \cos \pi + i \sin \pi = -1 \]
  ∗ neighbors are 180 deg out of phase

• The region \(-\pi/a < K < \pi/a\) is the first Brillouin zone of the 1D lattice

• We must extrapolate these results to three dimensions for bulk crystals
Density of Phonon States

• Consider a 1D chain of total length $L$ carrying $M+1$ particles (atoms) at a separation $a$
  ♦ Fix the position of atoms 0 and $M$
  ♦ Each normal vibrational mode of polarization $p$ takes the form of a standing wave

$$u_n \sim \sin(nKa) \exp(-i\omega_{Kp} t)$$

♦ Only certain wavelengths (wavevectors) are allowed

$$\lambda_{\text{max}} = 2L \quad (K_{\text{min}} = \pi/L), \quad \lambda_{\text{min}} = 2a \quad (K_{\text{max}} = \pi/a = M\pi/L)$$

♦ In general, the allowed values of $K$ are

$$K = \frac{\pi}{L}, \frac{2\pi}{L}, \frac{3\pi}{L}, ..., \frac{(M-1)\pi}{L}$$

Note: $K = M\pi/L$ is not included because it implies no atomic motion, i.e.,
$$\sin(nM\pi a/L) = \sin(n\pi) = 0.$$
Density of States, cont’d

• Thus, we have M-1 allowed, independent values of \( K \)
  ♦ This is the same number of particles allowed to move
  ♦ In \( K \)-space, we thus have \( M-1 \) allowable wavevectors
  ♦ Each wavevector describes a single mode, and one mode exists in each distance \( \pi/L \) of \( K \)-space
  ♦ Thus, \( dK/dN = \pi/L \), where \( N \) is the number of modes

Discrete \( K \)-space representation

\[ \pi/a \]

\[ \pi/(M-1)a \]  \[ \pi/L \]  \[ \pi/a \]
Density of States, cont’d

- The phonon density of states gives the number of modes per unit frequency per unit volume of real space

\[
D(\omega) = \frac{1}{L^{\alpha=1}} \frac{dN}{d\omega} = \frac{1}{L} \frac{dN}{dK} \frac{dK}{d\omega} = \frac{1}{\pi} \frac{1}{d\omega / dK}
\]

- The last denominator is simply the group velocity, derived from the dispersion relation

\[
D(\omega) = \frac{1}{\pi v_g (\omega)} = \left[ \pi a \sqrt{\frac{g}{m}} \cos \left( \frac{1}{2} K(\omega)a \right) \right]^{-1}
\]

Note singularity for \( K = \pi / a \)
Periodic Boundary Conditions

• For more generality, apply periodic boundary conditions to the chain and find

\[ K = 0, \pm \frac{2\pi}{L}, \pm \frac{4\pi}{L}, ..., \frac{M\pi}{L} \]

♦ Still gives same number of modes (one per particle that is allowed to move) as previous case, but now the allowed wavevectors are separated by \( \Delta K = \frac{2\pi}{L} \)

♦ Useful in the study of higher-dimension systems (2D and 3D)
2D Density of States

- Each allowable wavevector (mode) occupies a region of area \((2\pi/L)^2\)
- Thus, within the circle of radius \(K\), approximately \(N=\pi K^2/(2\pi/L)^2\) allowed wavevectors exist
- Density of states

\[
D(\omega) = \frac{1}{L^\alpha=2} \frac{dN}{d\omega} = \frac{1}{V} \frac{dN}{dK} \frac{dK}{d\omega} = \frac{K(\omega)}{2\pi} \frac{1}{v_g(\omega)}
\]
3D Density of States

• Using periodic boundary conditions in 3D, there is one allowed value of $K$ per $(2\pi/L)^3$ volume of $K$-space.

• The total number of modes with wavevectors of magnitude less than a given $K$ is thus

$$N = \left(\frac{L}{2\pi}\right)^3 \left(\frac{4}{3} \pi K^3\right) = \frac{VK^3}{6\pi^2}$$

• The 3D density of states becomes

$$D(\omega) = \frac{1}{L^{\alpha=3}} \frac{dN}{d\omega} = \frac{1}{V} \frac{dN}{dK} \frac{dK}{d\omega} = \frac{K(\omega)^2}{2\pi^2} \frac{1}{v_g(\omega)}$$
Glossary for Lattice Descriptions and Lattice Dynamics

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Lattice Structure

• **a = lattice constant**

• **Common crystal structures**
  - Body centered (bcc)
  - Face centered (fcc)
  - Diamond (dia)

• **Bravais lattice**: an infinite array of discrete points whose position vectors can be expressed as:

\[ \mathbf{R} = n_1 \mathbf{a}_1 + n_2 \mathbf{a}_2 + n_3 \mathbf{a}_3 \]

where \( \mathbf{a}_i \) are PRIMITIVE VECTORS
and \( n_i \) are integers
Primitive Unit Cells

- A primitive unit cell is a volume of real space that, when translated through all \( \mathbf{R} \), just fills all space without overlaps or voids and contains one lattice point.
- 2D examples

Jarrell (2) Fig2
Lattice with a Basis

- Often, we need to describe a crystalline material’s structure by placing a primary atom at each lattice point and one or more basis atoms relative to it.
  - For compound materials (e.g., CuO₂), this is an obvious requirement.
  - Also applies to some monoatomic crystals (e.g., Si).

Jarrell (2) Fig3
Reciprocal Lattice

• More convenient to express spatial dependencies in terms of wave vectors, instead of wavelengths

• Reciprocal lattice (RL) is like the inverse of a Bravais lattice

• \( \mathbf{G} \) is the vector that satisfies

\[
\mathbf{G} \cdot \mathbf{R} = 2\pi \times \text{integer} \quad \Rightarrow \quad \mathbf{G} = m_1 \mathbf{b}_1 + m_2 \mathbf{b}_2 + m_3 \mathbf{b}_3
\]

\( m_i \) are integers, and

\[
\mathbf{b}_1 = 2\pi \frac{\mathbf{a}_2 \times \mathbf{a}_3}{\mathbf{a}_1 \cdot (\mathbf{a}_2 \times \mathbf{a}_3)}; \quad \mathbf{b}_2 = 2\pi \frac{\mathbf{a}_3 \times \mathbf{a}_1}{\mathbf{a}_1 \cdot (\mathbf{a}_2 \times \mathbf{a}_3)}; \quad \mathbf{b}_3 = 2\pi \frac{\mathbf{a}_1 \times \mathbf{a}_2}{\mathbf{a}_1 \cdot (\mathbf{a}_2 \times \mathbf{a}_3)}
\]
Primitive Cells & Miller Indices

- **Primitive Cell**: A region of space that is closer to one point than any others
- **1st Brillouin Zone**: The primitive cell of the reciprocal lattice
- For a given lattice plane, *Miller indices* are coordinates of the shortest reciprocal lattice vector normal to the plane
  - A plane with Miller indices \((hkl)\) is perpendicular to the vector
    \[
    \vec{G} = h\vec{b}_1 + k\vec{b}_2 + l\vec{b}_3
    \]
Dispersion Curves

- **Phase velocity:** \( c = \frac{\omega}{q} \)
- **Group velocity:** \( v_g = \frac{\partial \omega}{\partial q} \)
- **Acoustic phonons:** determine the speed of sound in a solid and are characterized by \( \omega \sim q \) for \( q \to 0 \)
- **Optical phonons:** occur for lattices with more than one atom per unit cell and are characterized by flat dispersion curves with relatively high frequencies
- **Branch:** acoustic or optical
- **Polarization:** defines the direction of oscillation of neighboring atoms of a given dispersion curve
  - **Longitudinal:** atomic displacements aligned with wave direction
  - **Transverse:** atomic displacements perpendicular to wave direction

Animated chains from http://www.chembio.uoguelph.ca/educmat/chm729/Phonons/movies.htm

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Phonons

- **Phonon**: a quantized lattice vibration (i.e., one that can take on only a discrete energy, $\hbar \omega$)

- **Normal mode**: a lattice wave that is characterized by a branch, polarization, wave vector, and frequency

- **Occupation number (or excitation number) $n_{Kp}$**: the number of phonons of a given wave vector ($K$) and branch/polarization $p$
  - Note that $n_{Kp}$ depends on frequency, which in turn depends on wave vector and branch/polarization as defined by the dispersion curve
  - Note also that, in this context, the term $p$ implies both branch and polarization
Overview of Phonon Simulation Tools

• **Boltzmann Transport Equation (BTE)**
  - Requires boundary scattering models
  - Requires detailed understanding of phonon scattering and dispersion for rigorous inclusion of phonon physics

• **Molecular Dynamics (MD)**
  - Computationally expensive
  - Not strictly applicable at low temperatures
  - Handling of boundaries requires great care for links to larger scales and simulation of functional transport processes

• **Atomistic Green’s Function (AGF)**
  - Efficient handling of boundary and interface scattering
  - Straightforward links to larger scales
  - Inclusion of anharmonic effects is difficult