Lattice Dynamics

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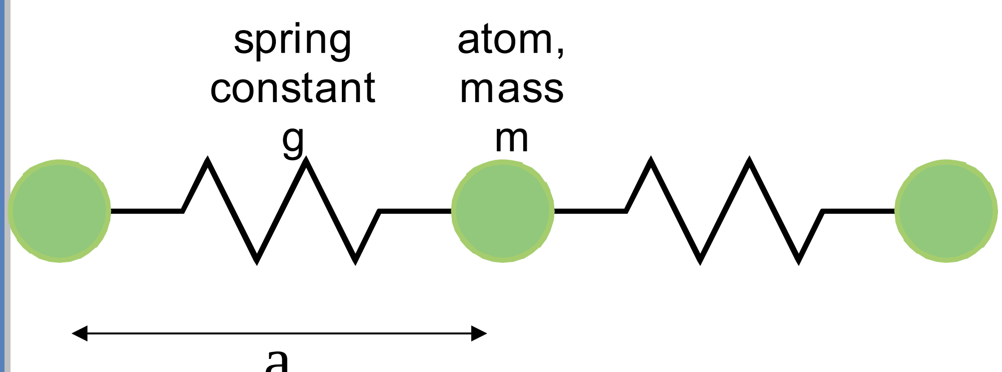
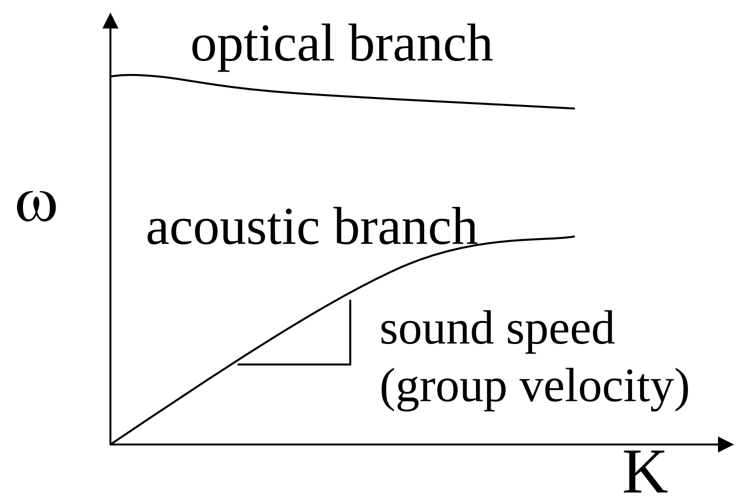
Phonon Heat Conduction

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

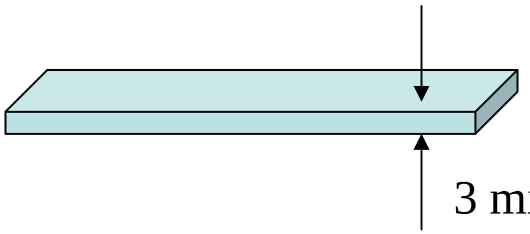


- Wave solutions
  - wave vector $K=2\pi/\lambda$
  - phonon energy=$\hbar\omega$
  - dispersion relations gives ω = fn(K)

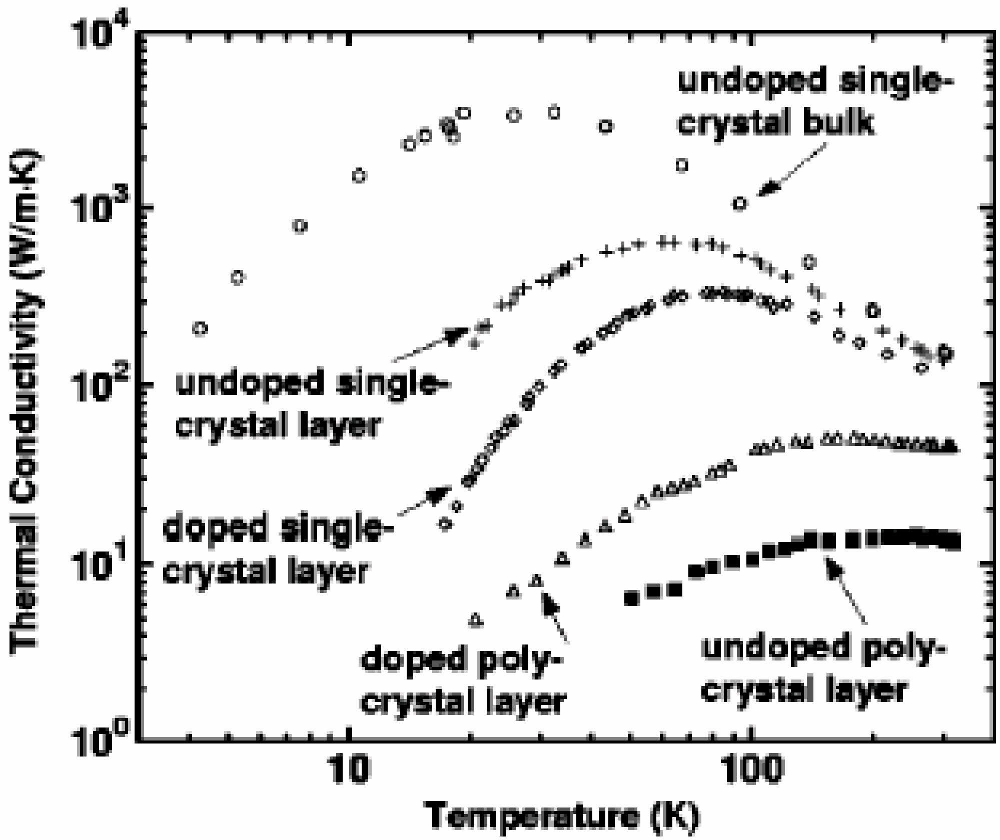


Heat Conduction Through Thin Films

- Experimental results for 3-micron silicon films



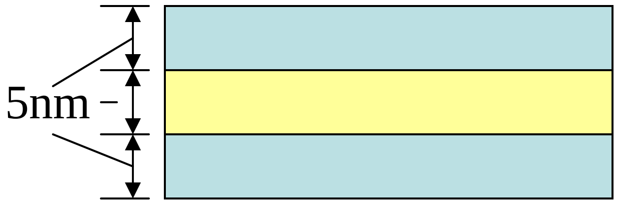
- Non-equilibrium scattering models work fairly well
- Crystalline structure often has larger impact than film thickness



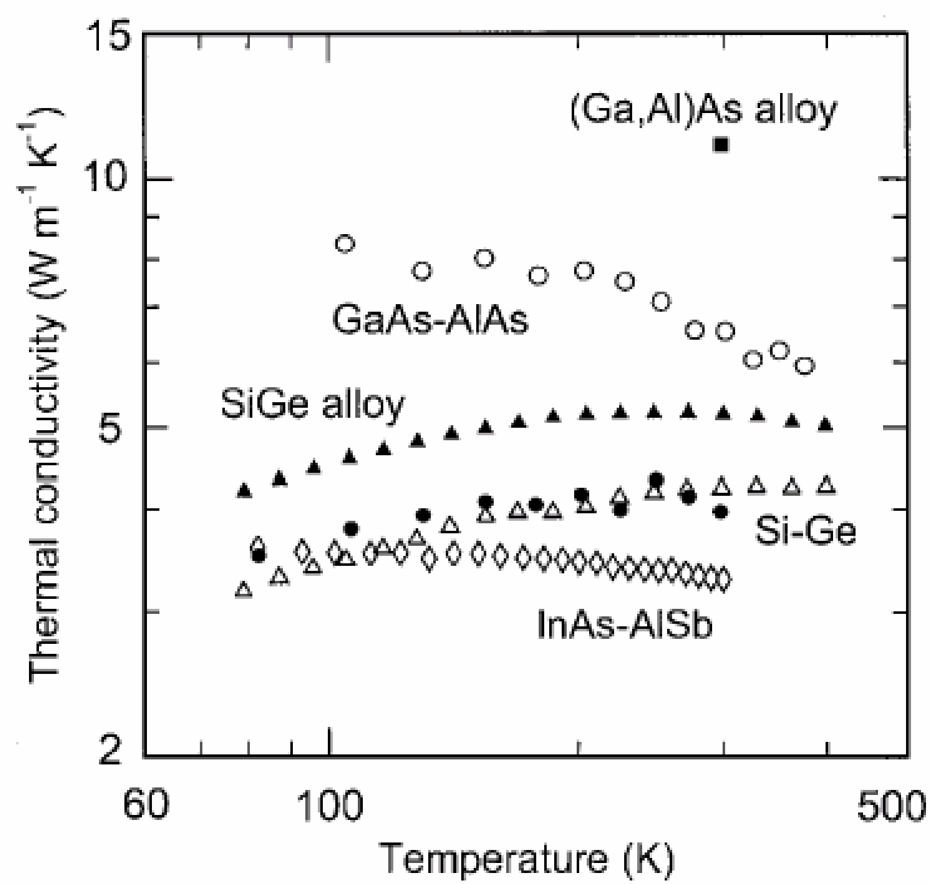
Asheghi et al., 1999

Heat Conduction Through Multiple Thin Films

- Fine-pitch 5 nm superlattices



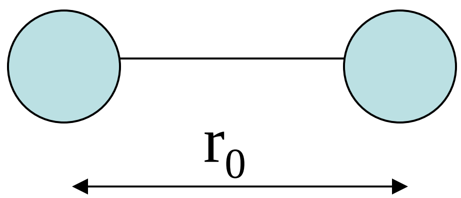
- Cross-thickness conductivity measurement
- Measured values are remarkably close to bulk alloy values (nearly within measurement error)
- Expected large reduction in conductivity not observed



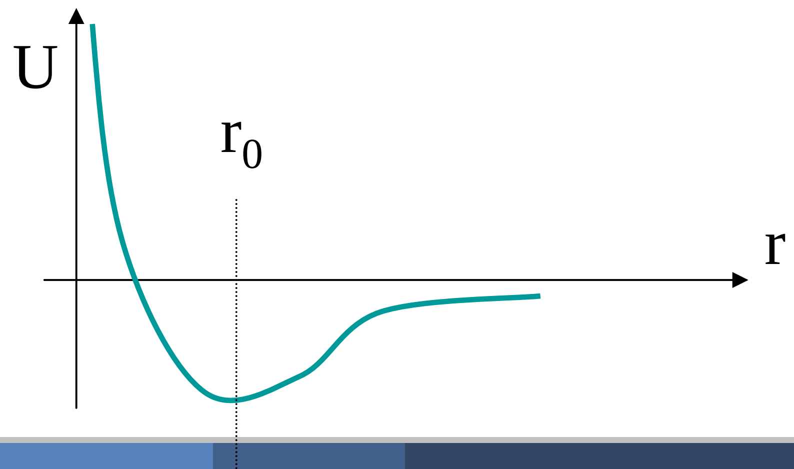
Cahill et al., 2003

Lattice Vibrations

- Consider two neighboring atoms that share a chemical bond



- The bond is not rigid, but rather like a spring with an energy relationship such as…



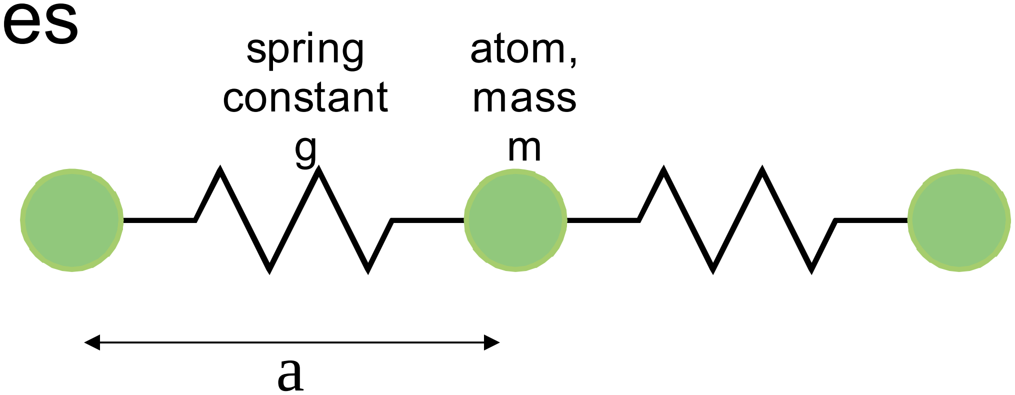
Lattice Vibrations, cont'd

- Near the minimum, the energy is well approximated by a parabola

$$U = \frac{1}{2} g u^2$$

  - $u = r - r_0$ and g = spring constant

- Now consider a one-dimensional chain of molecules



Lattice Energy and Motion

- Harmonic potential energy is the sum of potential energies over the lattice

$$U^{harm} = \frac{1}{2} g \sum_n \{u[na] - u[(n+1)a]\}^2$$

- Equation of motion of atom at location *u*(*na*)

$$F = m\frac{d^2u(na)}{dt^2} = -\frac{\partial U^{harm}}{\partial u(na)} = -g\{2u(na) - u[(n-1)a] - u[(n+1)a]\}$$

- Simplified notation

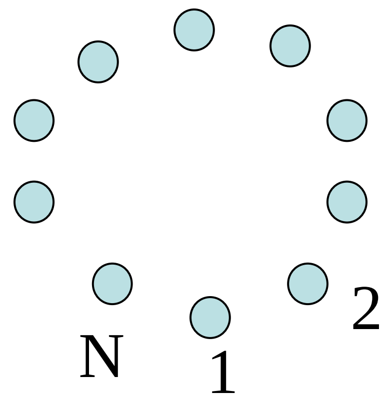
$$m\frac{d^2u_n}{dt^2} = -g\{2u_n - u_{n-1} - u_{n+1}\}$$

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

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

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

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

where n is an integer

- Let λ be the vibration wavelength, λ = *aN*/*n*

$$K = \frac{2\pi n}{aN} = \frac{2\pi}{\lambda}$$

K = wave vector

- Minimum wavelength, λ_{min} = 2*a* = 2(lattice spacing)

Solution to the Equations of Motion

- Substitute exponential solution into equation of motion

$$-m\omega^2 e^{i(Kna-\omega t)} = -g\left[2-e^{-iKa}-e^{iKa}\right]e^{i(Kna-\omega t)}$$

$$= -2g\left(1-\cos Ka\right)e^{i(Kna-\omega t)}$$

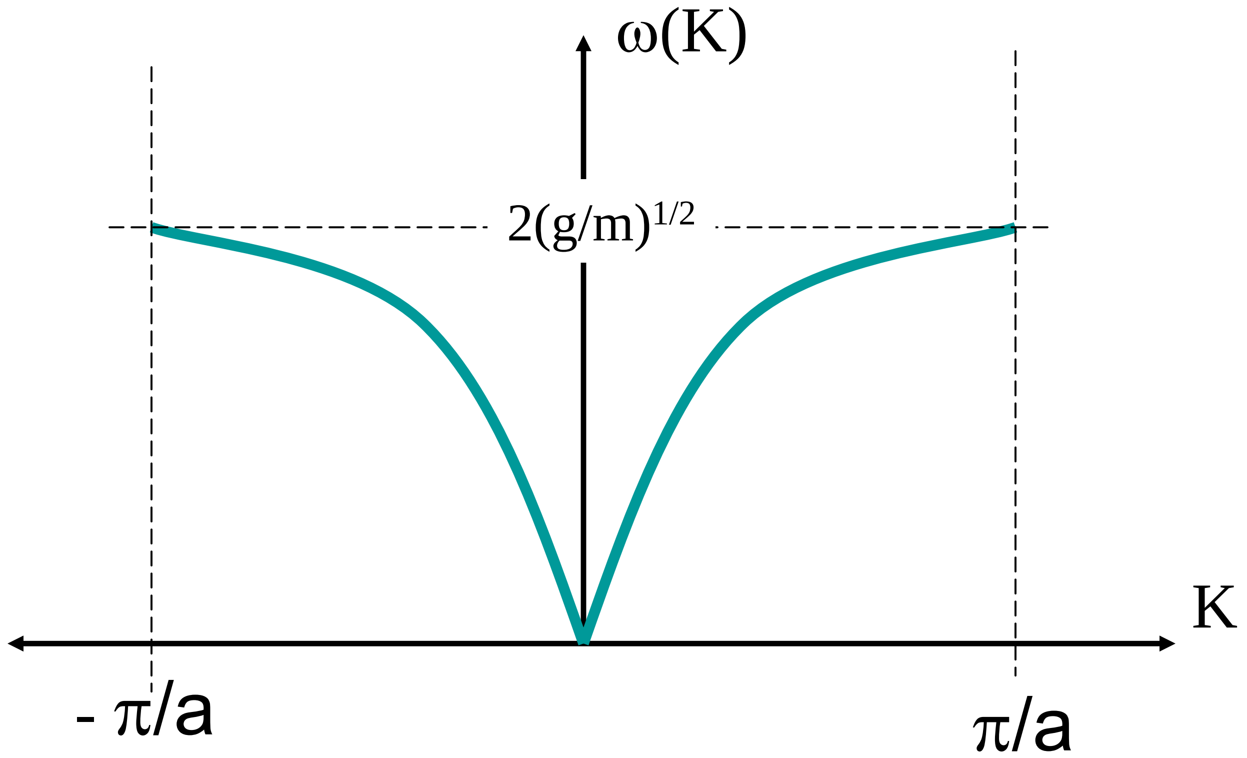
- Solve for ω

$$\omega(K) = \sqrt{\frac{2g(1-\cos Ka)}{m}} = 2\sqrt{\frac{g}{m}}\left|\sin\left(\tfrac{1}{2}Ka\right)\right|$$

- 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 $-\pi/a < K < \pi/a$



Wave Velocities

- Phase velocity: $c = \omega/K$
- Group velocity: $v_g = \partial\omega/\partial K = a(g/m)^{1/2}\cos(Ka/2)$
- For small K:

$$\lim_{K\to 0} \omega = a\sqrt{\frac{g}{m}}\,|K|$$

$$\to \lim_{K\to 0} v_g = a\sqrt{\frac{g}{m}} = \left|\frac{\omega}{K}\right| = c$$

- Thus, for small K (large λ), 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

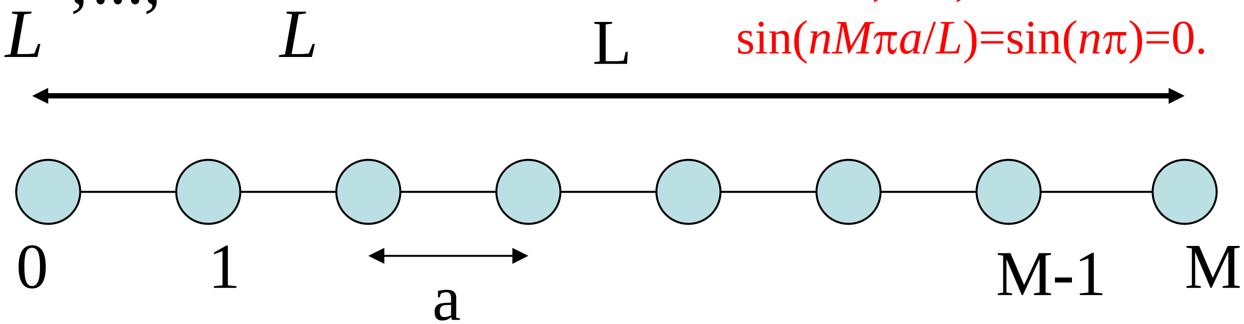
- Consider a 1D chain of total length *L* carrying *M*+1 particles (atoms) at a separation *a*
  - Fix the position of atoms 0 and *M*
  - Each normal vibrational mode of polarization p takes the form of a standing wave

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

  - Only certain wavelengths (wavevectors) are allowed
    $\lambda_{max}=2L$ ($K_{min}=\pi/L$), $\lambda_{min}=2a$ ($K_{max}=\pi/a=M\pi/L$)
  - In general, the allowed values of K are

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

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

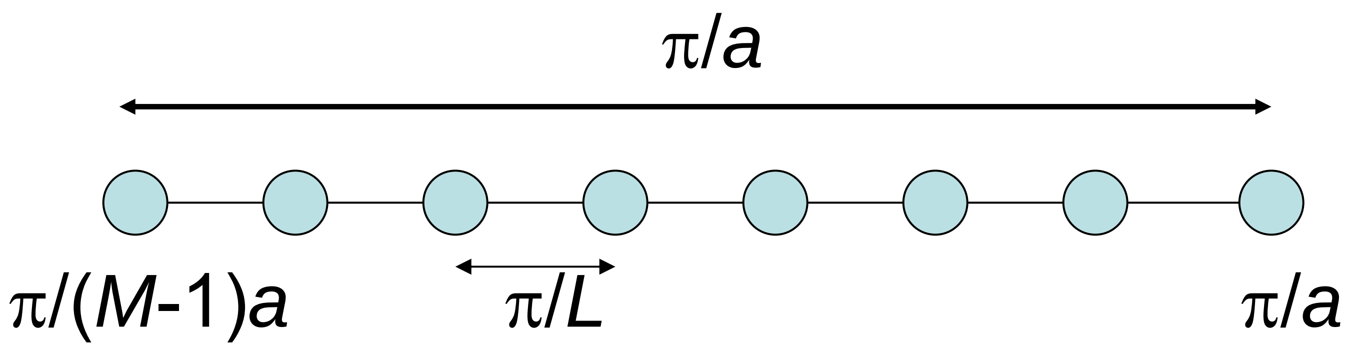


See Kittel, Ch5, Intro to Solid-State Physics, Wiley 1996

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 π/L of K-space
  - Thus, $dK/dN = \pi/L$, where N is the number of modes

Discrete K-space representation



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

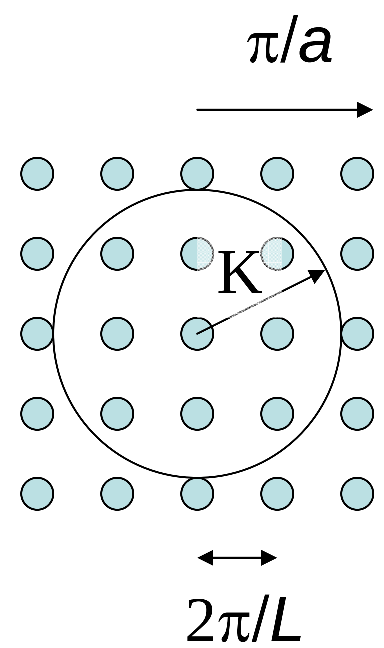
$$K = 0, \pm\frac{2\pi}{L}, \pm\frac{4\pi}{L}, \ldots, \frac{M\pi}{L}$$

  - ♦ 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 $\Delta K = 2\pi/L$
  - ♦ 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

K-space



$$D(\omega)=\frac{1}{L^{\alpha=2}}\frac{dN}{d\omega}=\frac{1}{V}\frac{dN}{dK}\frac{dK}{d\omega}=\frac{K(\omega)}{2\pi}\frac{1}{v_g(\omega)}$$

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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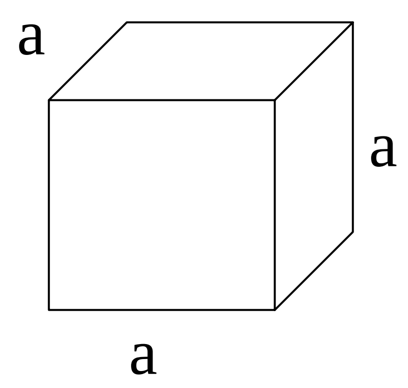
Lattice Structure



- 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:

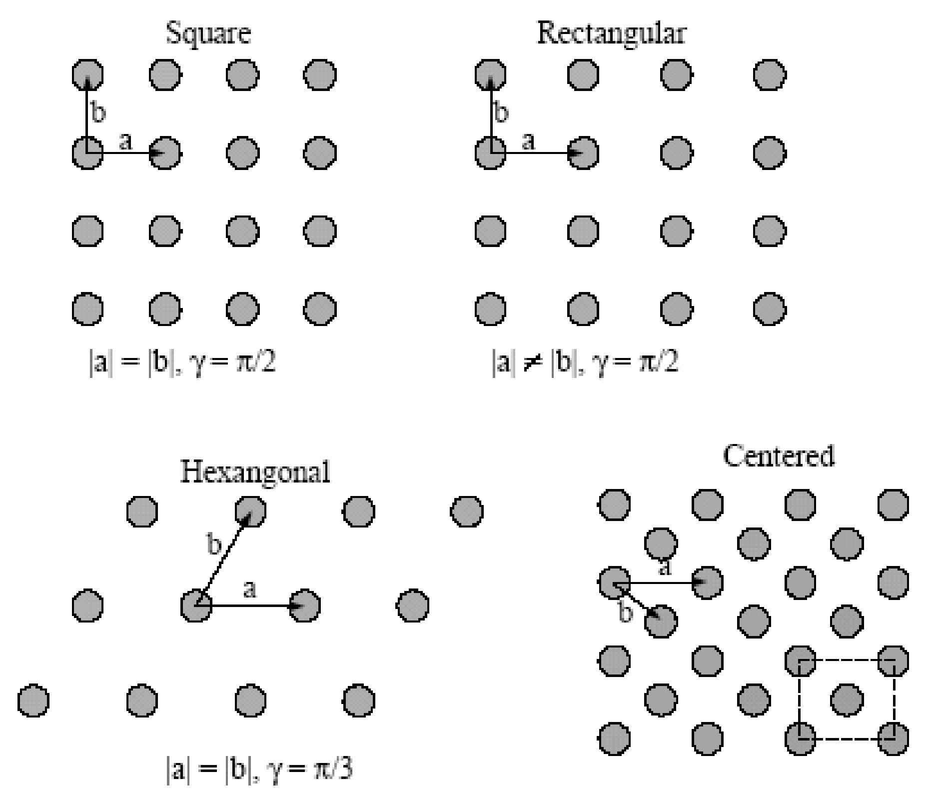
$$\vec{R} = n_1 \vec{a_1} + n_2 \vec{a_2} + n_3 \vec{a_3}$$

where $\vec{a_i}$ are PRIMITIVE VECTORS

and $\mathrm{n_i}$ are integers

Primitive Unit Cells

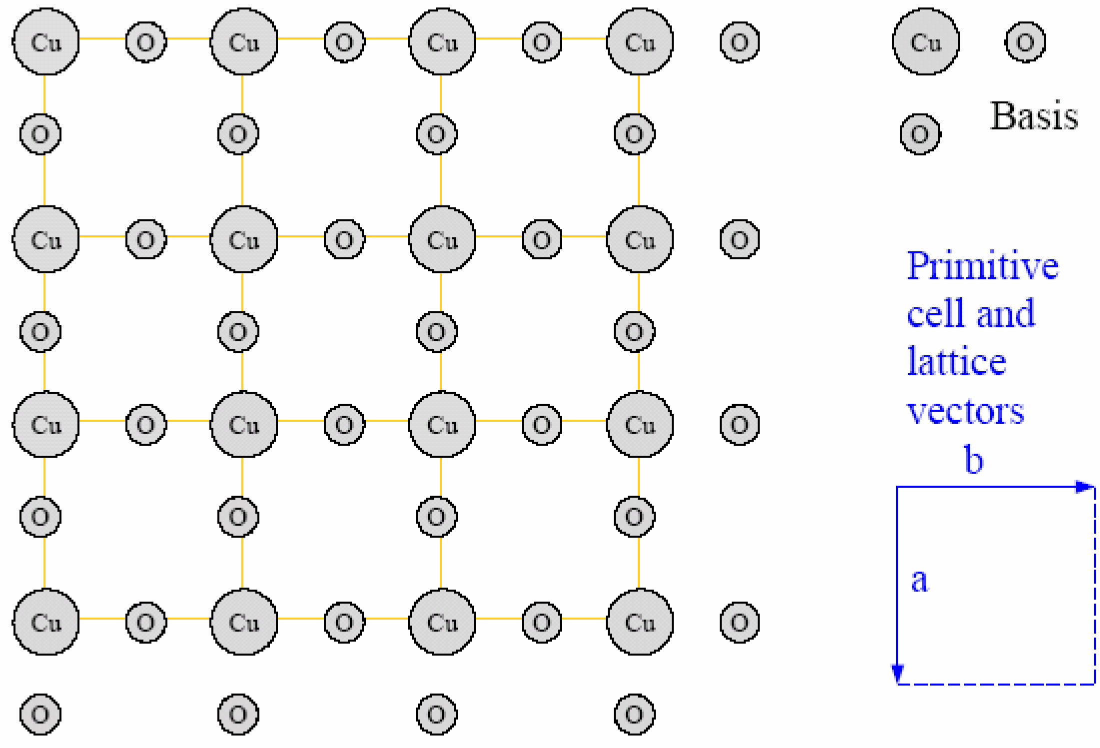
- A primitive unit cell is a volume of real space that , when translated through all **R**, just fills all space without overlaps or voids and contains one lattice point
- 2D examples



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 (eg CuO_2), this is an obvious requirement
  - Also applies to some monoatomic crystals (eg 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
- **G** is the vector that satisfies

$$\mathbf{G} \bullet \mathbf{R} = 2\pi \times \text{integer} \quad \Longrightarrow \quad \vec{G} = m_1 \vec{b}_1 + m_2 \vec{b}_2 + m_3 \vec{b}_3$$

  - m_i are integers, and

$$\vec{b}_1 = 2\pi \frac{\vec{a}_2 \times \vec{a}_3}{\vec{a}_1 \bullet \left(\vec{a}_2 \times \vec{a}_3\right)}; \quad \vec{b}_2 = 2\pi \frac{\vec{a}_3 \times \vec{a}_1}{\vec{a}_1 \bullet \left(\vec{a}_2 \times \vec{a}_3\right)}; \quad \vec{b}_3 = 2\pi \frac{\vec{a}_1 \times \vec{a}_2}{\vec{a}_1 \bullet \left(\vec{a}_2 \times \vec{a}_3\right)}$$

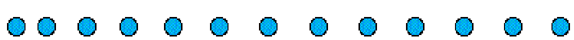
Primitive Cells & Miller Indices

- 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

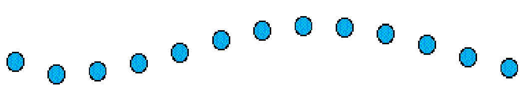
$$\vec{G} = h\vec{b}_1 + k\vec{b}_2 + l\vec{b}_3$$

Dispersion Curves

- Phase velocity: $c = \omega/q$
- Group velocity: $v_g = \partial\omega/\partial q$
- Acoustic phonons: determine the speed of sound in a solid and are characterized by $\omega \sim q$ for $q \rightarrow 0$
- 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

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