ASESMA 2010
Added Material on Phonons

Background on phonons with simple models for the potential energy and forces

Part II

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These two lectures are taken from class notes for a solid state physics course at the University of Illinois

Phonons I - Crystal Vibrations Continued
(Kittel Ch. 4)

View of triple axis neutron scattering facility at National Research Council of Canada

http://neutron.nrc.ca/welcome.htm

Outline

- Finish examples in higher dimeansions

- Two atoms per cell: Acoustic and Optic Modes

- Quantization and Phonons

- Experimental observation by inelastic scattering

- (Read Kittel Ch 4)

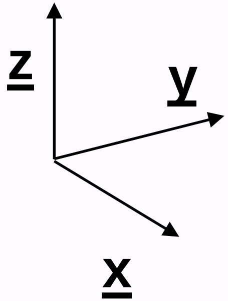
Repeat from Last Lecture

Displacements of Atoms

- Positions of atoms in their perfect lattice positions are given by:

$$\underline{\mathbf{R}}^0(n_1, n_2, n_3) = n_1\,\underline{\mathbf{a}}_1 + n_2\,\underline{\mathbf{a}}_2 + n_3\,\underline{\mathbf{a}}_3$$

For simplicity here we consider only one atom per cell and assume an orthogonal coordinate system



For convenience let $\underline{\mathbf{n}}_i = (n_{i1}, n_{i2}, n_{i3})$ denote atom i which has position $\underline{\mathbf{R}}^0_i$

- The displacement of atom i can be written

$$\Delta\underline{\mathbf{R}}_i = u_i\,\underline{\mathbf{x}} + v_i\,\underline{\mathbf{y}} + w_i\,\underline{\mathbf{z}}$$

Aside on Vector derivatives

- The definition of the derivative

  $$\underline{\mathbf{F}}_s = - dE/d\,\underline{\mathbf{R}}_s$$

  is that each component is defined by

  $F_{s,x} = - dE/d\,X_s$, $F_{s,y} = - dE/d\,Y_s$, $F_{s,z} = - dE/d\,Z_s$

- In the harmonic approximation the potential energy due to displacements of atoms $\Delta\underline{\mathbf{R}}_j$ is given by:

  $$E = E_0 + (1/2) \Sigma_{i\,j}\ \Delta\underline{\mathbf{R}}_i \cdot \mathbf{D}_{i\,j} \cdot \Delta\underline{\mathbf{R}}_j + \ldots.$$

  and the force on atom s is:

  $$\underline{\mathbf{F}}_s = - dE/d\,\underline{\mathbf{R}}_s = - \Sigma_j\ \mathbf{D}_{s\,j} \cdot \Delta\underline{\mathbf{R}}_j$$

  where the D's are force constants

- For central forces see following slides

Central Forces

- For Central Forces:

$$E = E_0 + (1/4N)\ \Sigma_{s\ i}\ \phi_i''\ (\Delta|\underline{\mathbf{R}}_s - \underline{\mathbf{R}}_{s+i}|)^2 + \ldots.$$

By the chain rule

$$F_{s,x} = -\ dE/d\ X_s = -\ \Sigma_i\ \phi_i''\ \Delta|\underline{\mathbf{R}}_s - \underline{\mathbf{R}}_{s+i}|)\ d\ |\underline{\mathbf{R}}_s - \underline{\mathbf{R}}_{s+i}|)\ /d\ X_s$$

Using the relation $|\underline{\mathbf{R}}_s - \underline{\mathbf{R}}_{s+i}| = \sqrt{(X_s - X_{s+i})^2 + \ldots}$

$$d\ |\underline{\mathbf{R}}_s - \underline{\mathbf{R}}_{s+i}|)\ /d\ X_s = \underbrace{(X^0_s - X^0_{s+i})/\ |\underline{\mathbf{R}}^0_s - \underline{\mathbf{R}}^0_{s+i}|}$$

evaluated at ideal positions $\underline{\mathbf{R}}^0$

x component of

unit vector $\underline{\mathbf{R}}^0_s - \underline{\mathbf{R}}^0_{s+i}$

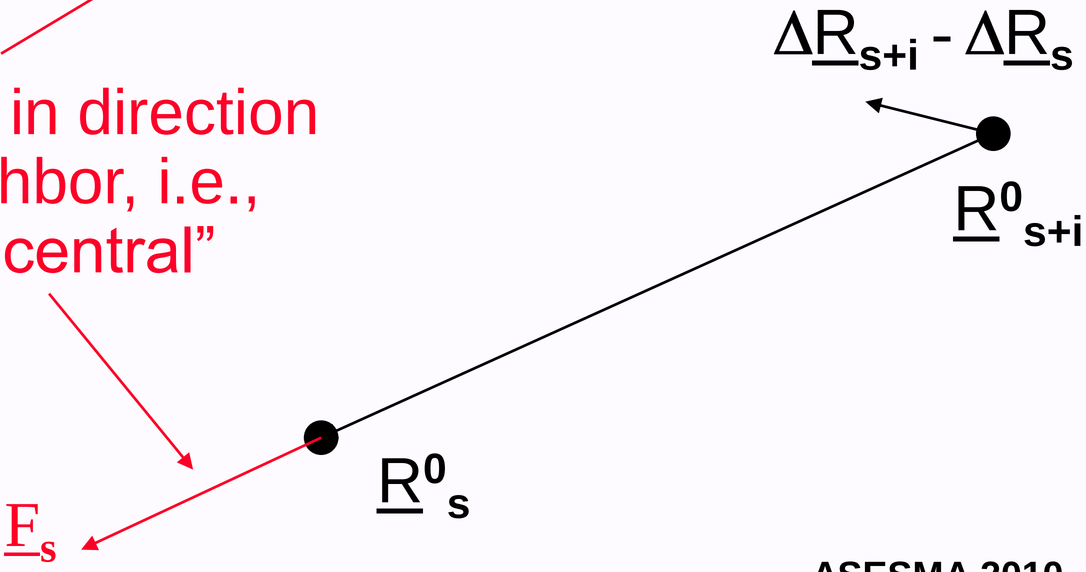
Similarly for $F_{s,y}$, $F_{s,x}$

- This leads to central forces as shown on the next slide

Geometric factors for Central Forces

- $\underline{\mathbf{F}}_s = - dE/d\,\underline{\mathbf{R}}_s = - \Sigma_i\, \phi_i''\, \underbrace{\Delta|\underline{\mathbf{R}}_s - \underline{\mathbf{R}}_{s+i}|}_{\text{To linear order}}\ \overbrace{\underline{\mathbf{R}}^0{}_s - \underline{\mathbf{R}}^0{}_{s+i}}^{\text{Unit vector}}$

$$= - \Sigma_i\, \phi_i''\, \overbrace{\underline{\mathbf{R}}^0{}_s - \underline{\mathbf{R}}^0{}_{s+i}}\ \overbrace{\left(\overbrace{\underline{\mathbf{R}}^0{}_s - \underline{\mathbf{R}}^0{}_{s+i}} \right) \cdot \left(\Delta\, \underline{\mathbf{R}}_s - \Delta\, \underline{\mathbf{R}}_{s+i} \right)}^{\text{To linear order}}$$



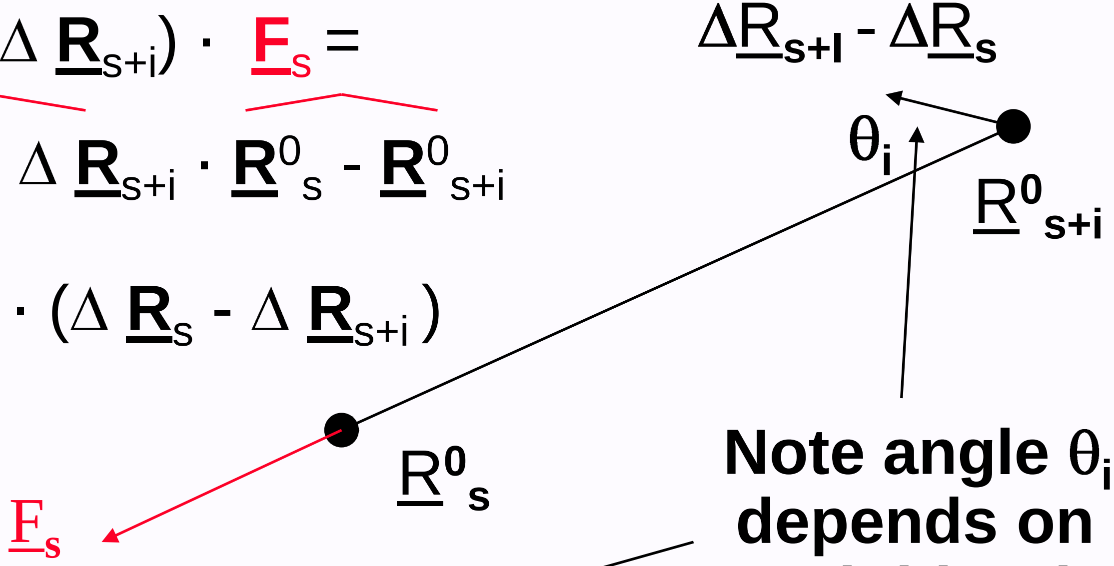
Force is in direction
of neighbor, i.e.,
it is “central”

Geometric factors for Central Forces

- For waves with each atom displaced in the same direction, we always need (see following slides) the force in the direction of the motion $F_{s||}$
- $F_{s||} = (\widehat{\Delta \underline{\mathbf{R}}_s - \Delta \underline{\mathbf{R}}_{s+i}}) \cdot \underline{\mathbf{F}}_s =$

$$- \Sigma_i \, \phi_i'' \; \widehat{\Delta \underline{\mathbf{R}}_s - \Delta \underline{\mathbf{R}}_{s+i}} \cdot \widehat{\underline{\mathbf{R}}^0_s - \underline{\mathbf{R}}^0_{s+i}}$$

$$(\widehat{\underline{\mathbf{R}}^0_s - \underline{\mathbf{R}}^0_{s+i}}) \cdot (\Delta \underline{\mathbf{R}}_s - \Delta \underline{\mathbf{R}}_{s+i})$$



Note angle θ_i depends on neighbor i

or

$$F_{s||} = - \Sigma_i \, \phi_i'' \, [\cos(\theta_i)]^2 \, |\Delta \underline{\mathbf{R}}_s - \Delta \underline{\mathbf{R}}_{s+i}|$$

Oscillations waves in 2 or 3 dimensions

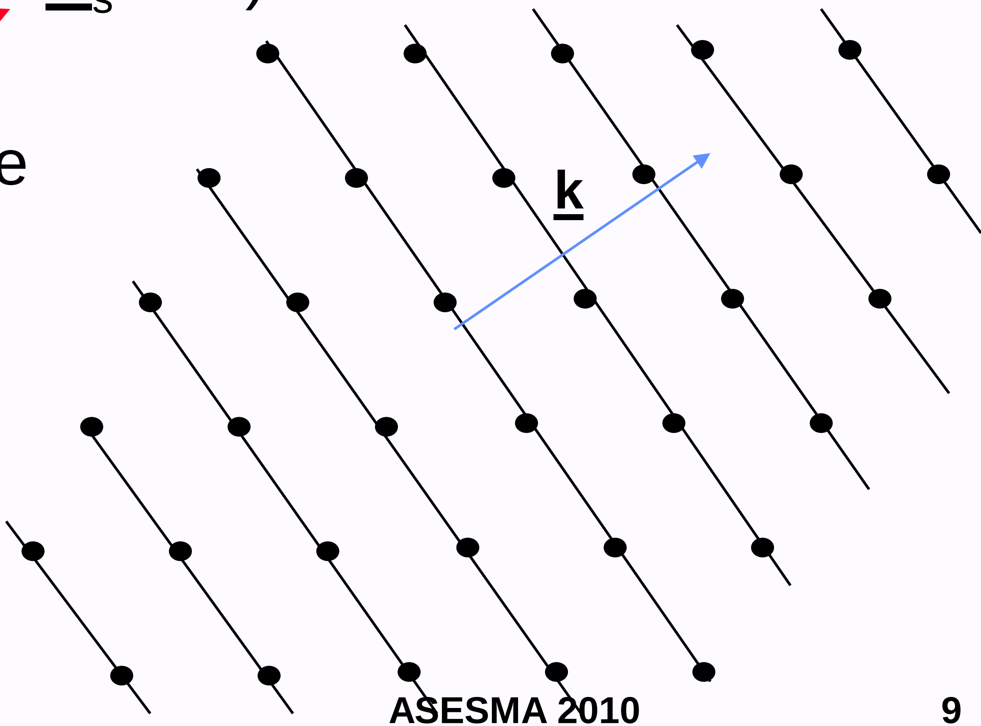
- Newton's Law:

$$M\, d^2\, \Delta\underline{\mathbf{R}}_s \,/\, dt^2 = \underline{\mathbf{F}}_s = -\, dE/d\,\underline{\mathbf{R}}_s \;\mathbf{=}\; -\, \Sigma_i\; \mathbf{D}_{s\;s+i} \cdot \Delta\underline{\mathbf{R}}_{s+i}$$

- General Solution:

$$\Delta\underline{\mathbf{R}}_s(t) = \Delta\underline{\mathbf{R}}\, \exp(i\underline{\mathbf{k}} \cdot \underline{\mathbf{R}}_s - i\omega t)$$

Vector dot product - same for all atoms in plane perpendicular to **k**



Oscillations waves in 2 or 3 dimensions

- Solution

$$M\,\omega^2\,\Delta\underline{\mathbf{R}} = -\sum_j \mathbf{D}_{s\,s+j}\cdot\Delta\underline{\mathbf{R}}\ \exp(i\underline{\mathbf{k}}\cdot\underline{\mathbf{R}}_s)$$

- For central forces

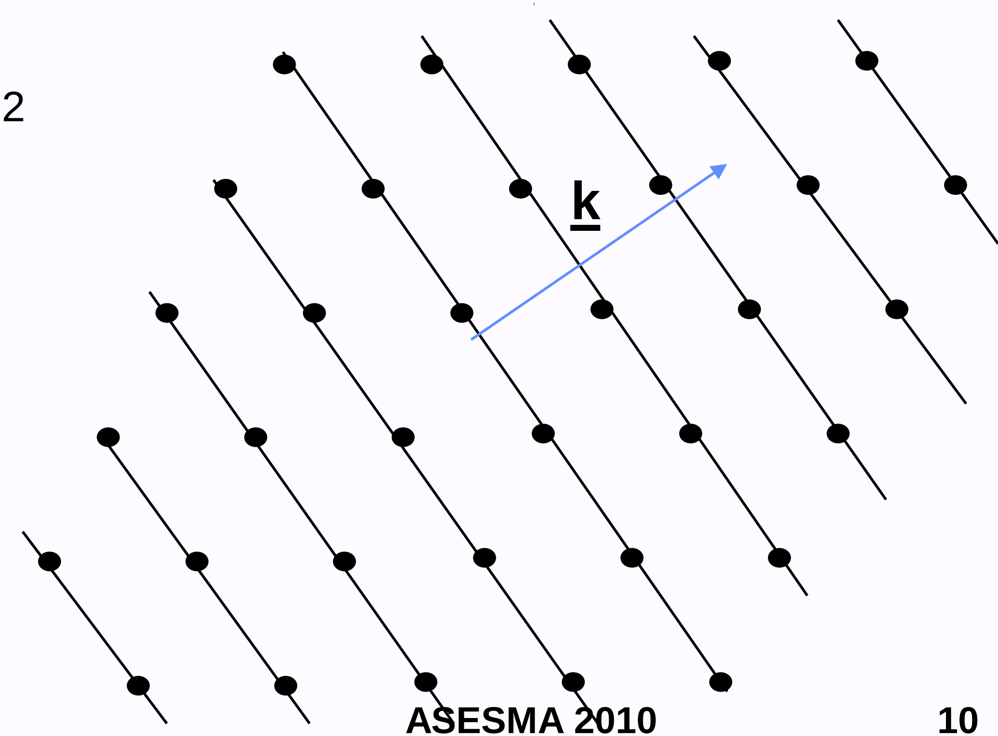
$$M\,\omega^2\,\Delta\underline{\mathbf{R}} =$$

$$-\sum_i \phi_i''\,\Delta\underline{\mathbf{R}}\ \widehat{[\cos}(\theta_i)\ ]^2$$

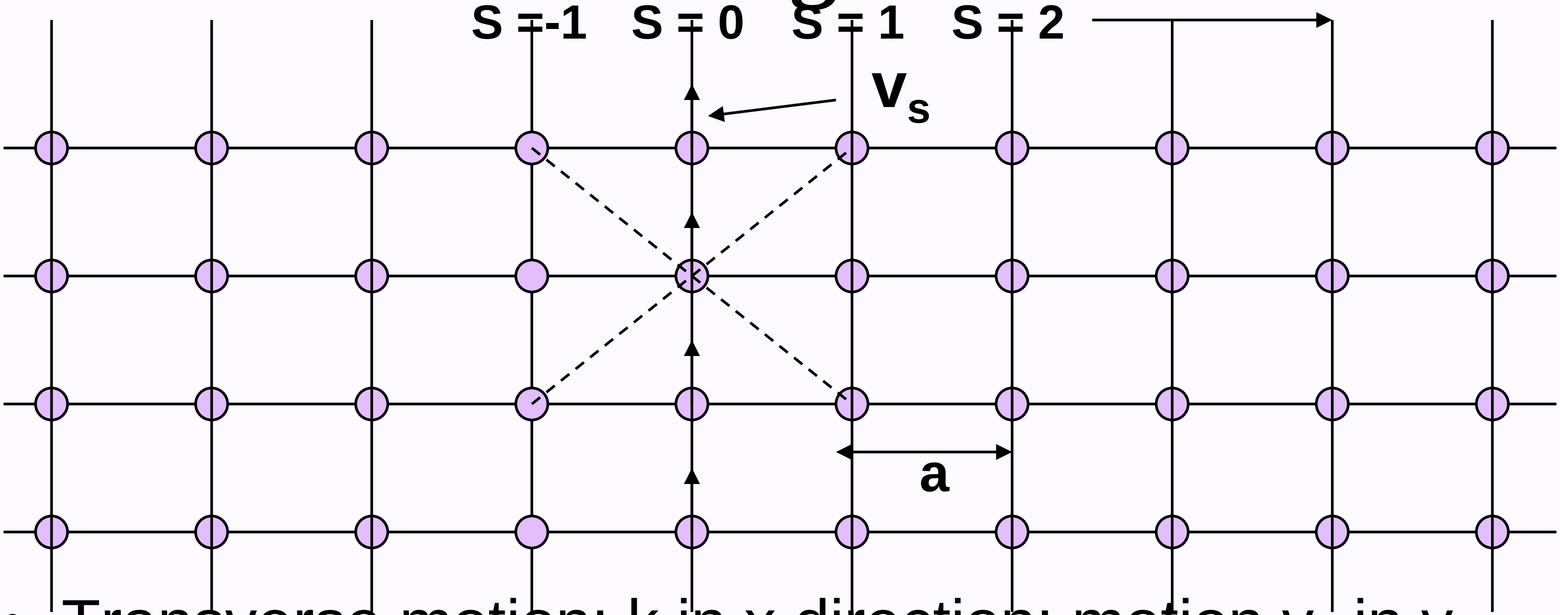
or

$$M\,\omega^2 =$$

$$-\sum_i \phi_i''\,[\cos(\theta_i)\ ]^2$$



Oscillations in higher dimensions



- Transverse motion: k in x direction; motion v_s in y direction; s labels plane perpendicular to x

$$v_s = v_s = v \exp(ik\,(s\,a) - i\omega\,t)$$

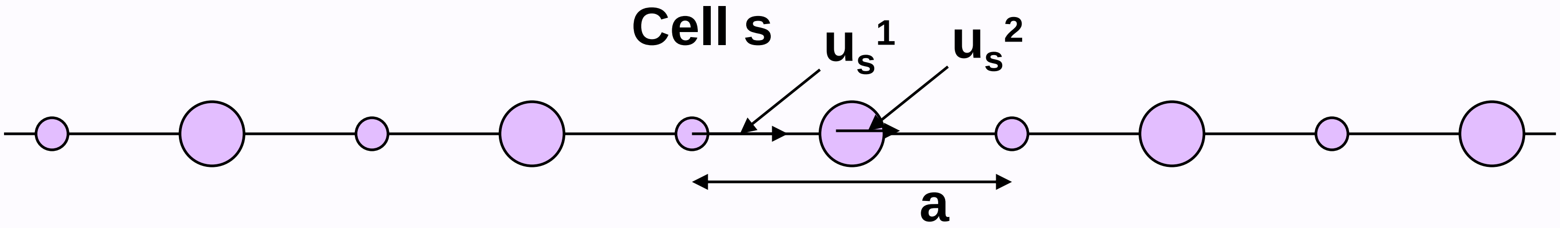
- Second neighbor forces

4 neighbors

$$\omega^2 = \underbrace{(1/2)}_{\text{Geometric factor} = \cos^2(\pi/4)}(\,\phi_2'' / M\,)\,[4\cos(ka) - 4]$$

Two atoms per cell - Linear chain

- To illustrate the effect of having two different atoms per cell, consider the simplest case atoms in a line with nearest neighbor forces only



- Now we must calculate force and acceleration of each of the atoms in the cell

$$F_s^1 = \phi'' \, [\, u_{s-1}^2 + u_s^2 - 2\, u_s^1] \; = M_1 \, d^2 \, u_s^1 \, / \, dt^2$$

and

$$F_s^2 = \phi'' \, [\, u_{s+1}^1 + u_s^1 - 2\, u_s^2] \; = M_2 \, d^2 \, u_s^2 \, / \, dt^2$$

Note subscripts

Oscillations with two atoms per cell

- Since the equation is the same for each cell s, the solution must have the same form at each s differing only by a phase factor. This is most easily written

$$u_s^1 = u^1 \exp(ik\,(s\,a) - i\omega\, t\,)$$
$$u_s^2 = u^2 \exp(ik\,(s\,a) - i\omega\, t\,)$$

- Inserting in Newton’s equations gives the coupled equations

$$-M_1\, \omega^2\, u^1 = \phi_1'' \,[(\exp(-ik\,a) + 1)\, u^2 - 2\, u^1]$$

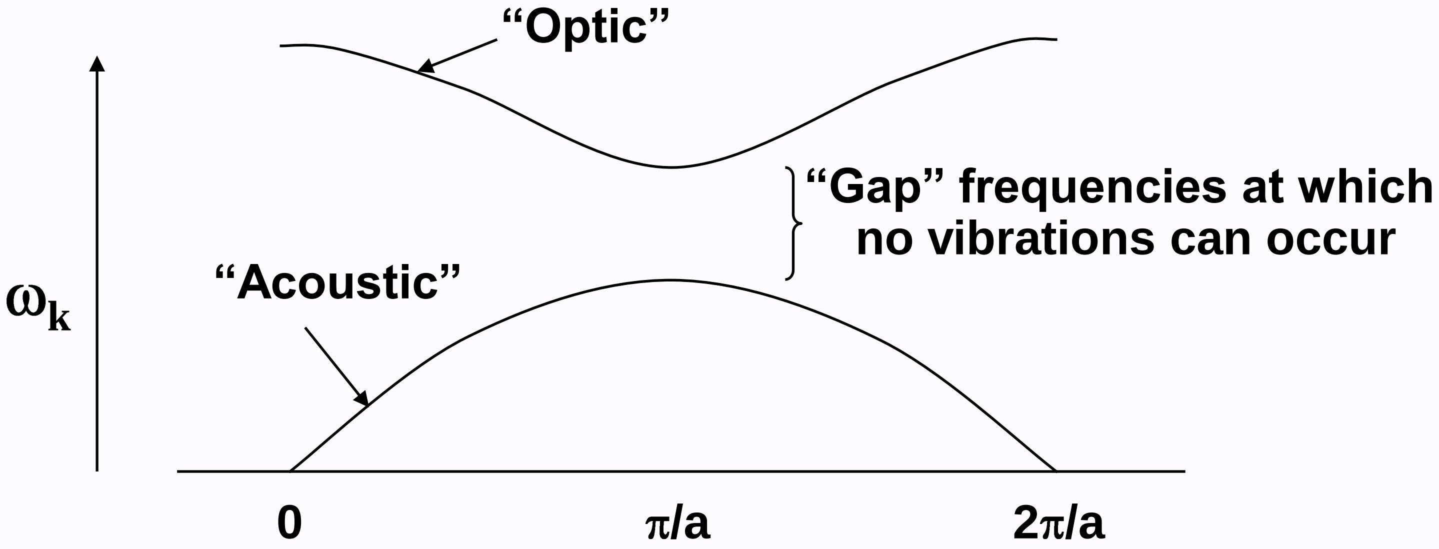
and

$$-M_2\, \omega^2\, u^2 = \phi_1'' \,[(\exp(\,ik\,a) + 1)\, u^1 - 2\, u^2]$$

$$\begin{vmatrix} 2\,\phi'' - M_1\,\omega^2 & -\,\phi''\,(\exp(-ik\,a) + 1) \\ -\,\phi''\,(\exp(\,ik\,a) + 1) & 2\,\phi'' - M_2\,\omega^2 \end{vmatrix} \mathbf{= 0}$$

Oscillations with two atoms per cell

- Solution



“Gap” frequencies at which no vibrations can occur

Oscillations with two atoms per cell

Acoustic -
Total Mass

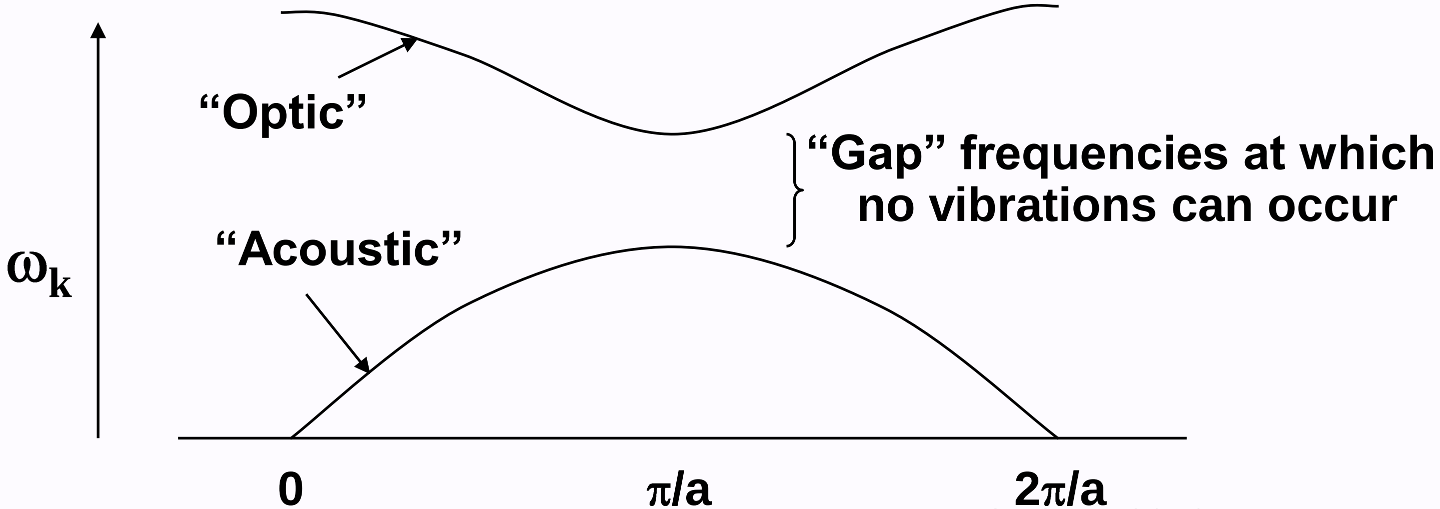
Optic -
Reduced Mass

- Limits:
- k ~ 0

  Acoustic: $\omega^2 = (1/2) (\phi_1'' / (M_1 + M_2)) k^2 a^2$

  Optic: $\omega^2 = 2 \phi_1'' [(1 / M_1) + (1/M_2)] = 2 \phi_1'' / \mu$
- $k = \pi/a$

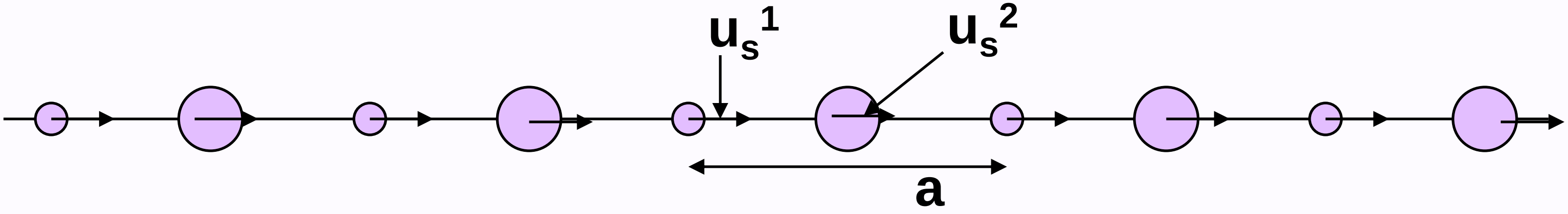
  Acoustic: $\omega^2 = 2 \phi_1''/ M_{large}$ Optic: $\omega^2 = 2 \phi_1''/ M_{smaLL}$



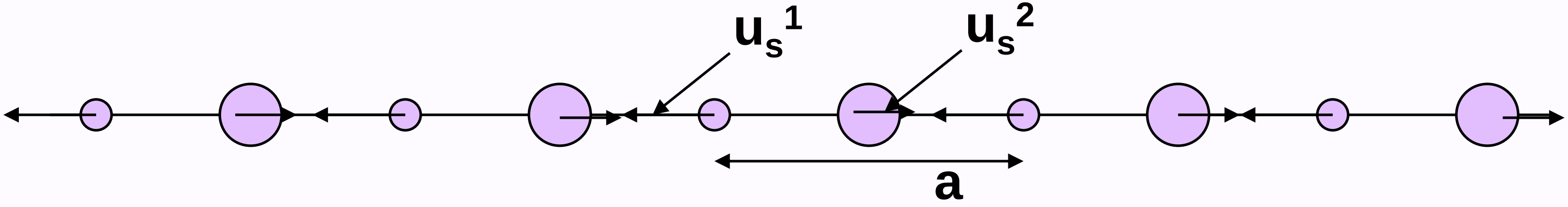
"Gap" frequencies at which no vibrations can occur

Modes for k near 0

- Acoustic at k near 0 - motion of cell as a whole

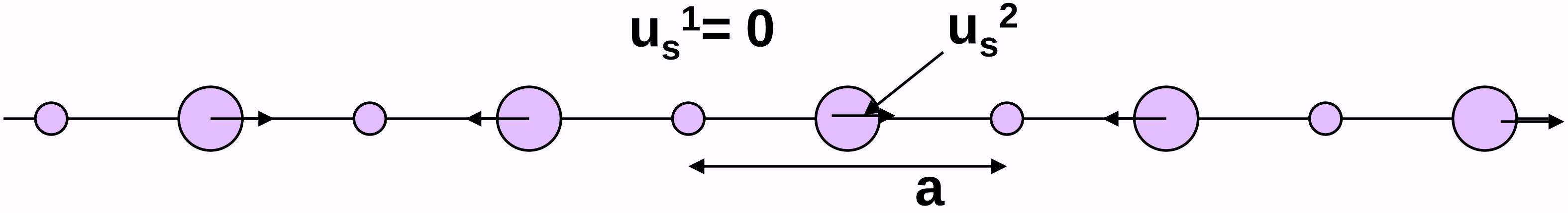


- Optic at k = 0 - opposed motion - larger displacement of smaller mass

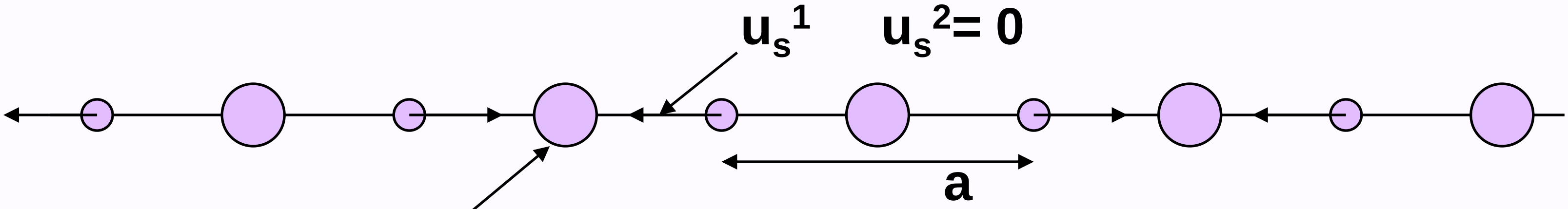


Modes for k at BZ boundary

- Each type of atom moves in opposite directions in adjacent cells
- Leads to two modes, each with only one type of atoms moving
- Acoustic at k = π/a - motion of larger mass



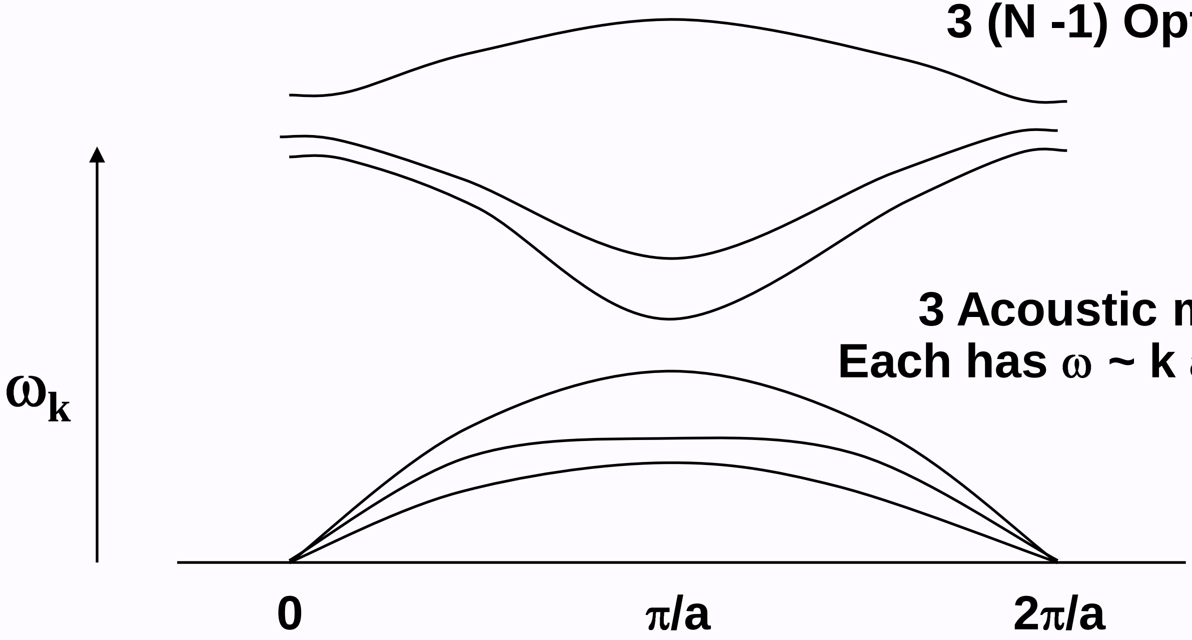
- Optic at k = π/a - motion of smaller mass



Atom 2 does not move because there are no forces on it!

Oscillations in 3 dimension with N atoms per cell

- Result



3 (N -1) Optic Modes

3 Acoustic modes
Each has $\omega \sim k$ at small k

Quantization of Vibration waves

- Each independent harmonic oscillator has quantized energies:

  $$e_n = (n + 1/2)\, h\nu = (n + 1/2)\, \hbar\omega$$

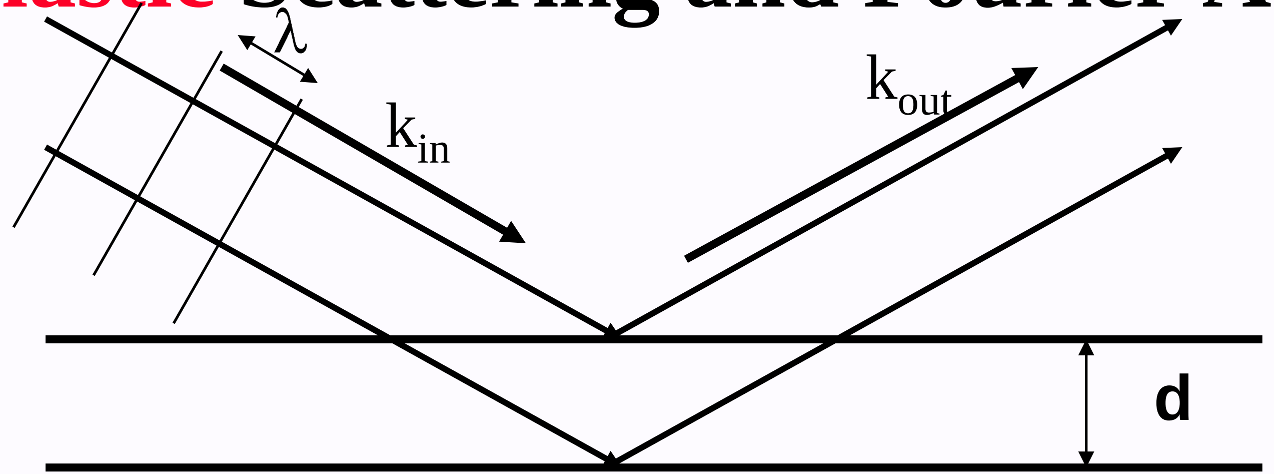
- We can use this her because we have shown that vibrations in a crystal are independent waves, each labeled by **k** (and index for the type of mode - 3N indices in a 3 dimen. crystal with N atoms per cell)

- Since the energy of an oscillator is 1/2 kinetic and 1/2 potential, the mean square displacement is given by $(1/2)\, M\, \omega^2\, u^2 = (1/2)\, (n + 1/2)\, h\omega$
  where M and u are appropriate to the particular mode (e.g. total mass for acoustic modes, reduced mass for optic modes , ….)

Quantization of Vibration waves

- Quanta are called phonons
- Each phonon carries energy $\hbar\omega$
- For each independent oscillator (i.e., for each independent wave in a crystal), there can be any integer number of phonons
- These can be viewed as particles
- They can be detected experimentally as creation or destrucion of quantized particles
- Later we will see they can transport energy just like a gas of ordinary particles (like molecules in a gas).

Inelastic Scattering and Fourier Analysis



- The in and out waves have the form:

  $\exp(i\, \underline{k}_{in} \cdot r - i\, \omega_{in} t)$ and $\exp(i\, \underline{\mathbf{k}}_{out} \cdot r - i\, \omega_{out} t)$
- For elastic scattering we found that diffraction occurs only for $\underline{\mathbf{k}}_{in} - \underline{\mathbf{k}}_{out} = \underline{\mathbf{G}}$
- For inelastic scattering the lattice planes are vibrating and the phonon supplies wavevector $\underline{\mathbf{k}}_{phonon}$ and frequency ω_{phonon}

Inelastic Scattering and Fourier Analysis

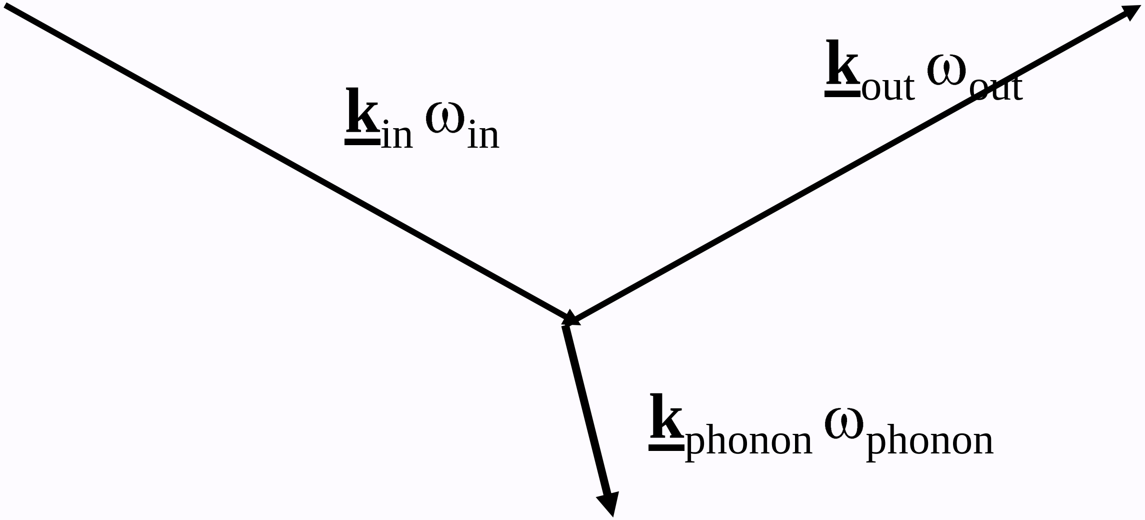
- Result:
- Inelastic diffraction occurs for

$$\underline{\mathbf{k}}_{in} - \underline{\mathbf{k}}_{out} = \underline{\mathbf{G}} \pm \underline{\mathbf{k}}_{phonon}$$

$$\omega_{in} - \omega_{out} = \pm \omega_{phonon} \text{ or }$$

$$\boxed{E_n - E_{out} = \pm \hbar\omega_{phonon}}$$

Quantum Mecchanics



Experimental Measurements of Dispersion Curves

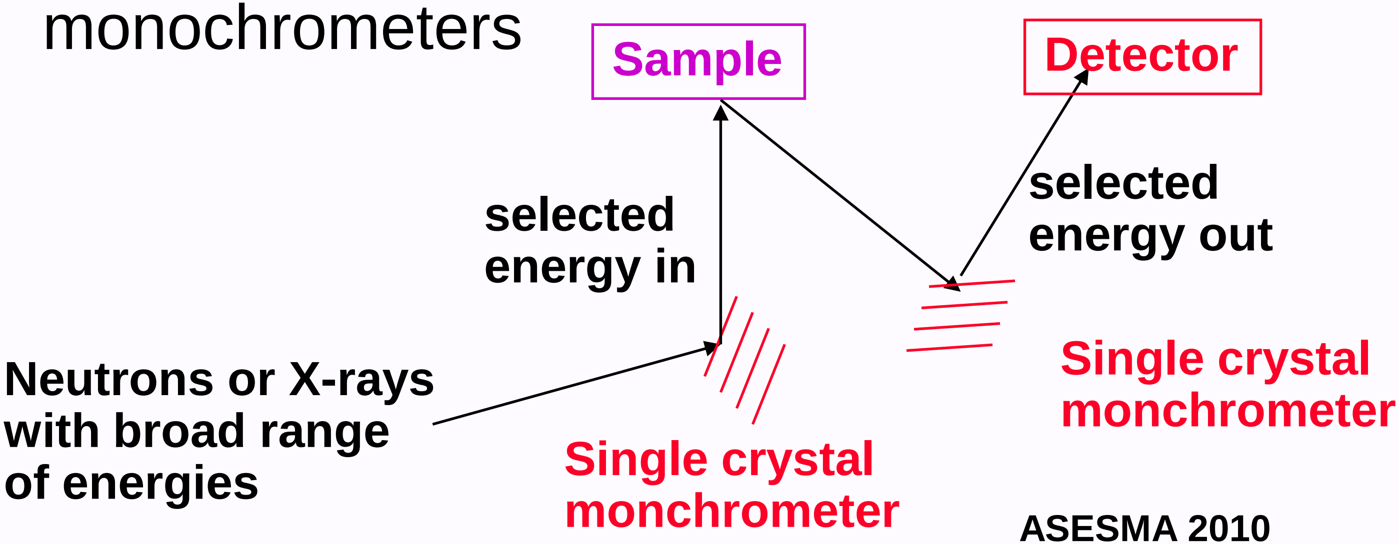
- Dispersion curves ω as a function of k are measured by inelastic diffraction

- If the atoms are vibrating then diffraction can occur with energy loss or gain by scattering particle

- In principle, can use any particle - neutrons from a reactor, X-rays from a synchrotron, He atoms which scatter from surfaces, …...

Experimental Measurements of Dispersion Curves

- Neutrons are most useful for vibrations
  For λ ~ atomic size, energies ~ vibration energies
  BUT requires very large crystals (weak scattering)
- X-ray - only recently has it been possible to have enough resolution (meV resolution with KeV X-rays!)
- “Triple Axis” - rotation of sample and two monochrometers



Sample

Detector

selected energy in

selected energy out

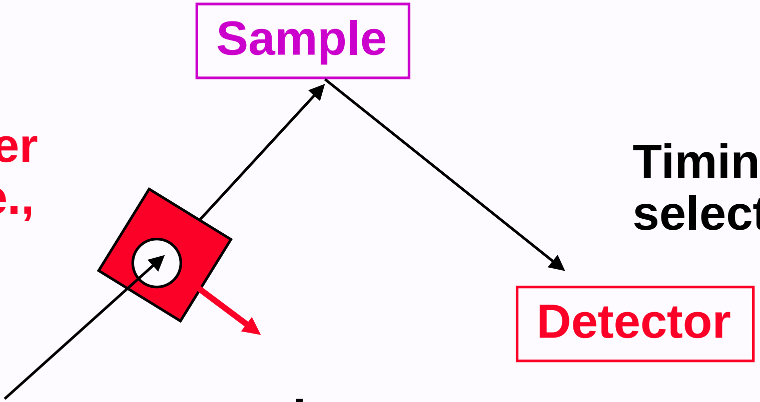
Neutrons or X-rays with broad range of energies

Single crystal monchrometer

Single crystal monchrometer

Experimental Measurements of Dispersion Curves

- Alternate approach for Neutrons
  Use neutrons from a sudden burst, e.g., at the new “spallation” source being built at Oak Ridge
- Measure in and out energies by “time of flight”



Sample

Mechanical chopper selects velocity, i.e., energy in

Timing at detector selects energy out

Detector

Busrt of neutrons at measured time (broad range of energies)

More on Phonons as Particles

- Quanta are called phonons, each with energy $\hbar\omega$
- $\underline{\mathbf{k}}$ can be interpreted as "momentum"
- What does this mean?
  NOT really momentum - a phonon does not change the total momentum of the crystal
  But $\underline{\mathbf{k}}$ is "conserved" almost like real momentum - when a phonon is scattered it transfers "$\underline{\mathbf{k}}$" plus any reciprocal lattice vector, i.e.,

$$\sum \underline{\mathbf{k}}_{\mathbf{before}} = \sum \underline{\mathbf{k}}_{\mathbf{after}} + \underline{\mathbf{G}}$$

- Example : scattering of particles

$$\underline{\mathbf{k}}_{\underline{\mathrm{in}}} = \mathbf{k}_{\mathrm{out}} + \underline{\mathbf{G}} \pm \underline{\mathbf{k}}_{\mathrm{phonon}}$$

where + means a phonon is created, - means a phonon is destroyed

Summary

- Normal modes of harmonic crystal:
  Independent oscillators labeled by wavevector k and having frequency ω_k
- The relation ω_k as a function of k is called a dispersion curve - 3N curves for N atoms/cell in 3 dimensions
- Quantized energies (n + 1/2) h ω_k
- Can be viewed as particles that can be created or destroyed - each carries energy and “momentum”
- “Momentum” conserved modulo any **G** vector
- Measured directly by inelastic diffraction - difference in in and out energies is the quantized phonon energy
- Neutrons, X-rays, …..