NOTES 6: LATTICE WAVES

We derived the elastic wave equations in Notes#4 under the condition that the strain was non-uniform but much less than unity, corresponding to only slight distortion of the solid from its relaxed position. Such a description is independent of the atomic nature of the solid and, in fact, preceded the atomistic picture of matter in history by many decades in the 19th century. But an implicit assumption in the elastic theory is that the wavelength of any propagating wave is much longer than the interatomic separation.

Here we analyze the general case where the long-wavelength condition is not necessarily met and the wavelength of the elastic wave can be comparable to the interatomic separation. We are then forced to solve for the wave dynamics at the atomic level and the concept of an elastic wave gets extended to a "lattice wave." Our analysis starts with a simple model in which each atom is represented by a mass of integer index s, and each bond between atoms is represented by a mechanical spring of spring constant c. The model accounts for bonds along only one direction of the solid. Although simplified, such a model does provide a useful description of real three-dimensional lattice waves because as shown at the end of Notes#5, *every* atom in a crystalline solid can be associated with one of a large set of parallel lattice planes. Each set of lattice planes is associated with a reciprocal lattice vector **K** having magnitude $2\pi/d$, where d is the separation between planes. A subtle aspect of the analysis, which we will return to later, is that the spring constants are special *interplanar* spring constants that are not directly related to the elastic constants (e.g., stiffness coefficients). However, the two can be related knowing d and the density of atoms per plane.

We know from the treatment of cohesive energy in Notes#3 that the force between atoms is a rather complicated function of their separation, but the spring should follow the linear "Hooke's" law (force proportional to displacement) if we are interested only in a small displacement of each atom from its equilibrium position. Hence, the dynamical analysis can proceed by subtracting out the equilibrium force F_0 and position R_o of each atom in a differential manner, and the excess force on the sth atom becomes

$$\Delta F_S = F_S - F_{0,S} = \frac{dF_S}{dx}\Delta r_s$$

where Δr_s is the non-equilibrium displacement defined by (with s an integer)

$$\Delta r_s = R_S - R_{0,S}$$

The net non-equilibrium force on the sth atom can then be written as a sum over neighboring springs,

$$\Delta F_S = C_1\left(\Delta r_{s+1} - \Delta r_s\right) - C_1\left(\Delta r_s - \Delta r_{s-1}\right) + C_2\left(\Delta r_{s+2} - \Delta r_s\right) - C_2\left(\Delta r_s - \Delta r_{s-2}\right) + \ldots$$

From Newton's second law, this net force must equal the mass of the sth atom times its acceleration. So we end up with the equation of motion

$$F_S = m\frac{d^2\Delta r_S}{dt^2} = C_1\left(\Delta r_{s+1} - \Delta r_s\right) - C_1\left(\Delta r_s - \Delta r_{s-1}\right) + C_2\left(\Delta r_{s+2} - \Delta r_s\right) - C_2\left(\Delta r_s - \Delta r_{s-2}\right)\ldots$$

(1)

This expression is a linear second-order *difference* equation with constant coefficients. It is not generally soluble unless we state some further relationship between Δr_s and $\Delta r_{s+/-p}$. where p is an integer. Fortunately, in crystalline solids there is an ordering in the positions of atoms such that with respect to some arbitrary origin, r = sd where d is distance between lattice planes. It is then reasonable to seek solutions to (1) of the form

$$\Delta r_s = Ae^{j(kr-\omega t)} = Ae^{jksd}e^{-j\omega t} \tag{2}$$

Aside on Waves in Crystals: Minimum value of k.

To carry the solution further it important to understand some aspects of waves in general, which is the dependence of the solutions on the conditions at the solid boundaries and on the periodicity of the lattice. Both can be accounted for in terms of the general condition on the physical quantity f associated with any wave propagating along direction **r.** Namely, that at any point in time, the wave is always present everywhere in space but at different "phases". Hence we have the following *translational* expression

$$f(r+r') = f(r)e^{jkr'} \tag{3}$$

where $e^{jkr'}$ is the phase factor and $k = 2\pi/\lambda$ is the (circular) wavevector. The dependence on boundary conditions is similar to our solution of the Schrodinger's equation for free electrons in a "box", used to derive the Fermi free electron model for metals in Notes#2. In that case we assumed that the wavefunctions for the electrons must vanish at the boundaries and beyond, consistent with the common-sense observation that electrons are never observed outside of the box.

For the case of elastic waves in a solid bounded by air, we have "free" boundary conditions meaning that the amplitude at the boundary is maximal and the phase is arbitrary. We are then "free" to choose the boundary condition, and a logical choice is the periodic condition $\Delta r(r = 0) = \Delta r(r = L)$, such $e^{jkL} = 1$. This implies $k = 2\pi m/L$ where m is any integer positive or negative. The two different signs of m is expected on physical grounds, corresponding to the usual two possible waves propagating in opposite directions. In essence, the boundary conditions have determined the discretization of k. .

To find $\Delta r_{s\pm p}$ at other sites relative to the reference s atom, we apply (3) again,

$$\text{so } \Delta r_{s\pm p} = \Delta r_s e^{jk(\pm p)d} = Ae^{jk(s\pm p)d}e^{-j\omega t}$$

Substituting this back into the difference wave equation (1) yields,

$$-m\omega^2 Ae^{jksd} =$$
$$= C_1 A\left\{e^{jk(s+1)d} + e^{jk(s-1)d} - 2e^{jksd}\right\} + C_2 A\left\{e^{jk(s+2)d} + e^{jk(s-2)d} - 2e^{jksd}\right\} + C_3 A\left\{e^{jk(s+3)d} + e^{jk(s-3)d} - 2e^{jksd}\right\} + \cdots$$

dividing by Ae^{jksd}, and re-arranging, we get

$$-m\omega^2 = C_1\left(2\cos kd - 2\right) + C_2\left(2\cos 2kd - 2\right) + C_3\left(2\cos 3kd - 2\right) + \cdots$$

$$\text{or } -m\omega^2 = \sum_{p=1}^{N} C_p\left[2\cos\left(pkd\right) - 2\right]$$

$$\text{or } \omega^2 = \frac{2}{m}\sum_{p=1}^{N} C_p\left[1 - \cos pkd\right]$$

In special case of nearest-neighbor interactions only, p = 1 only and the sum reduces to

$$\omega^2 = \frac{2}{m}C_1\left(1 - \cos kd\right) \text{ or } \omega = \sqrt{\frac{2C_1}{m_1}\left(1 - \cos kd\right)} \qquad (4)$$

The resulting expression is called a *dispersion* relation. Another way of writing it is

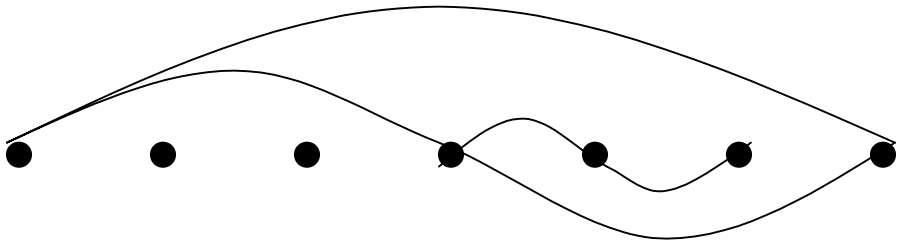
$$\omega = \sqrt{\frac{4C_1}{m_1}}\left|\sin\frac{1}{2}kd\right|$$

$$\rightarrow \omega = \sqrt{\frac{C_1}{m}}kd = \sqrt{\frac{C_1 d}{\rho}}\cdot k \text{ , } \rho \equiv \frac{\mathrm{m}}{\mathrm{d}}$$

So for small kd, $\frac{\omega}{k} = v_p = \sqrt{\frac{C_1 d}{\rho}}$, similar to the phase velocity of sound in the continuum limit of slowly varying strain:

But for larger kd, $\omega = \sqrt{\frac{2C_1}{m}\left(1 - \cos kd\right)}$ rolls over

This leads to the very interesting question of what is the largest significant value of k ?

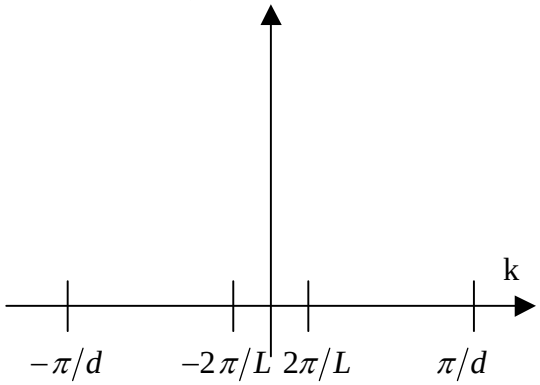


Aside on Nyquist sampling theorem in Lattices

The atoms of the crystal lattice can be thought of as discrete spatial sampling points as

shown in the Figure above. As such, there is a minimum wavelength (i.e., maximum spatial frequency) above which the wave can not be resolved without ambiguity. This is the subject of the Nyquist sampling theorem, which is fundamental to discrete sampling theory in the time domain. In the present context, the maximum spatial frequency is the one that the atoms sample twice per period, i.e., $|k_{max}| = 2\pi/\lambda_{max} = 2\pi/(2d) = \pi/d$.

Sampling waves of higher spatial frequency is ambiguous with waves at a lower spatial frequency, so the entire analysis can be reduced to the region $|k| < \pi/d$, or $-\pi/d < k < \pi/d$. In the language of digital sampling, π/d takes on the name Nyquist *spatial* frequency. In the language of solid-state (and for historic reasons), the region $-\pi/d < k < \pi/d$ π/d has the name 1st Brillouin zone. The 1st Brillouin zone for our one-dimensional analysis below including the discretization based on the solid boundary conditions.



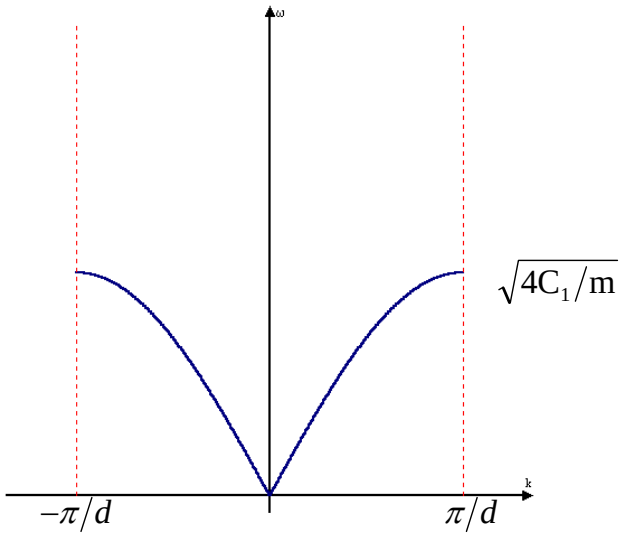
Given this clarification of the k values, the maximum significant evaluation of (4) occurs at $ka = \pi$ (Brillouin zone boundary), for which $\cos ka = -1$, $\omega = \sqrt{\frac{4C_1}{m}}$ and

$v_p = \frac{\omega}{k} = \sqrt{\frac{4C_1}{m}}\frac{d}{\pi}$ - a lower velocity than the speed of sound ! Note: v_p is the "phase" velocity. From wave propagation theory, energy propagates at the group velocity:

$$v_g \equiv \frac{d\omega}{dk} = \frac{\frac{C_1 d}{m}\sin kd}{\sqrt{\frac{2C_1}{m}\left(1-\cos kd\right)}}$$

and at $kd = \pi$, we get $v_g = 0$. This means that no energy is propagating.

- But we also know from wave phenomena that there are waves that don't transmit energy called *standing waves*.
- Standing waves are superposition of equal-amplitude positive- and negative-going traveling waves

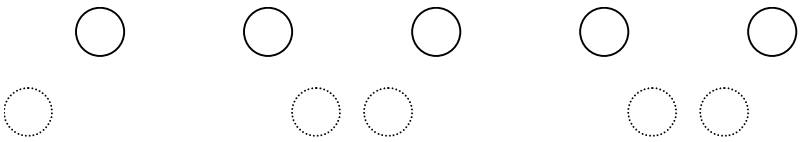


So at k = π/d, the assumed solution $\Delta r_s^+ = Ae^{j(ksd-\omega t)}$ must be accompanied by a negative going wave $\Delta r_s^- = Ae^{j(ksd+\omega t)}$ of equal amplitude. In other words, the chain of atoms reflects Δr_s^+ entirely. This very important effect is called Bragg Scattering

To understand it better, we solve for Δr_{s+p} for $p = 1$ using (3) at kd = 0 and kd = π
We find:

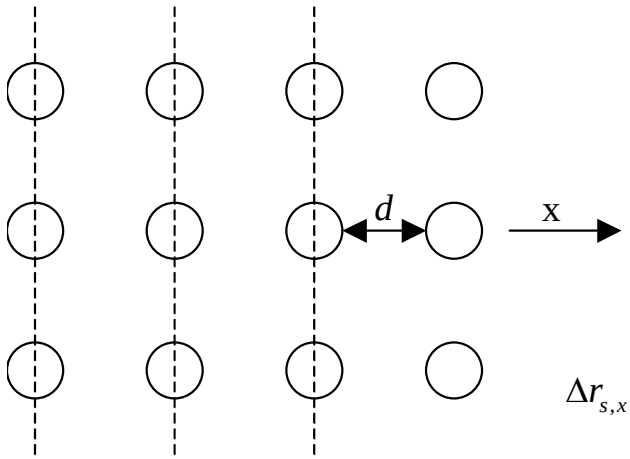
$$\frac{\Delta r_{s\pm1}}{\Delta r_s} = e^{\pm jkd} \qquad \begin{cases} 1 \text{ at } k = 0 \text{ (in phase)} \\ -1 \text{ at } k = \dfrac{\pi}{d} \text{ (out of phase)} \end{cases}$$

The latter case describes the standing wave (and Bragg Scattering)



Comments:

1) Although the derivation has been done for a linear chain, it can be thought as describing the deformation of all atoms in one of a "family" of parallel planes perpendicular to the propagation direction. This assumes the plane-wave form is a valid solution to the Newton's wave equation, which is certainly valid in crystals along high symmetry directions.



2) Guided by presence of shear waves in solid-state elasticity theory, we suspect that transverse waves will also be present

$$\Delta r_{s,y} = Be^{j(ksd-\omega t)}$$

Force equation on s^{th} atom (in s^{th} plane) for nearest neighbor interaction only will be:

$$F_s = C_T\left(\Delta r_{s+1} - \Delta r_s\right) - C_T\left(\Delta r_s - \Delta r_{s-1}\right)$$

The use of (3) with this force equation and Newton's law yields

$$-m\omega^2 Be^{jksd} = C_T B\left\{e^{jk(s+1)d} + e^{jk(s-1)d} - 2e^{jksd}\right\}$$

dividing by $Be^{jk_x sd}$ yields:

$$-m\omega^2 = C_T\left(e^{jkd} + e^{-jkd} - 2\right) = 2C_T\left(\cos kd - 1\right)$$

or

$$\omega = \sqrt{\frac{2C_T}{m}\left(1 - \cos kd\right)}$$

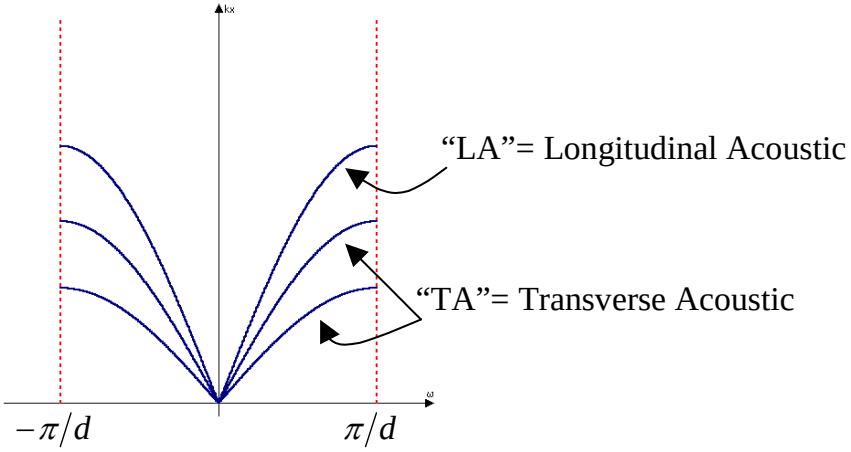
Same as longitudinal except for different C_T

3) As in elasticity theory, there are two possible shear waves which should have the same C value along directions of high symmetry, but in general will have different C values, C_{T1} and C_{T2}, along arbitrary directions. And generally the C_T are smaller than C_L (longitudinal spring constant).

So we have:

$$\omega_L^2 = \frac{2C_L}{m}\left(1-\cos kd\right)$$

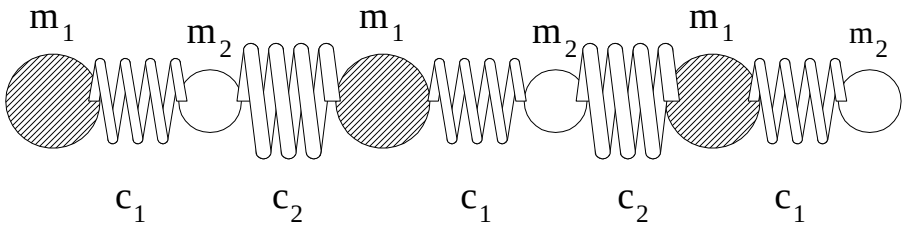
$$\omega_{T1}^2 = \frac{2C_{T1}}{m}\left(1-\cos kd\right);\ \omega_{T2}^2 = \frac{2C_{T2}}{m}\left(1-\cos kd\right)$$



These are all "acoustic" waves (or modes) because in the limit that kd ≈ 0, $\omega \propto$ k as in elasticity theory

$$\omega_L \approx \sqrt{\frac{C_L d}{\rho}}\cdot k;\ \omega_{T1} \approx \sqrt{\frac{C_{T1} d}{\rho}}\cdot k;\ \omega_{T2} \approx \sqrt{\frac{C_{T2} d}{\rho}}\cdot k$$

General Solution for linear chain (Two masses Two spring constants):
A good exercise in Coupled Resonator Theory



The preceding analysis is valid for crystals in the form of a pure Bravais lattice. But as we have said, most naturally occurring and many artificial (i.e., human made) crystals have the form of a Bravais lattice plus a basis. Our analysis can be generalized to this case by the addition of a second atom, its mass, and its displacement from equilibrium $\Delta r_{2,S}$ as a second mechanical degree-of-freedom. Note: the integer index S now refers to a unit cell, not a specific atom !

Equation for Δr_1 (Note: general enough to be $\Delta r_1^L, \Delta r_1^{T1}, or\ \Delta r_1^{T2}$):

$$m_1 \frac{d^2 \Delta r_{(1,s)}}{dt^2} = F_s = C_1\left(\Delta r_{(2,s)} - \Delta r_{(1,s)}\right) - C_2\left(\Delta r_{(1,s)} - \Delta r_{(2,s-1)}\right) \qquad \text{(i)}$$

Equation for Δr_2 (could also be $\Delta r_2^L, \Delta r_2^{T1}, or\ \Delta r_2^{T2}$ with change of spring constants)

$$m_2 \frac{d^2 \Delta r_{(2,s)}}{dt^2} = F_s = C_2\left(-\Delta r_{(2,s)} + \Delta r_{(1,s+1)}\right) - C_1\left(\Delta r_{(2,s)} - \Delta r_{(1,s)}\right)$$

(ii)

As in single atom case, assume plane-wave solutions:

$$\Delta r_{(1,s)} = A_1 e^{j(ksd-\omega t)},\ \Delta r_{(2,s)} = A_2 e^{j(ksd-\omega t)}$$

Utilize wave translation property: $\Delta r_{(s\pm1)} = \Delta r_s e^{j(\pm kd)}$

Get for (i) and (ii)

$$-m_1\omega^2 A_1 e^{jksd} = \left(-C_1 - C_2\right) A_1 e^{jksd} + \left(C_1 e^{jksd} + C_2 e^{jk(s-1)d}\right) A_2$$

$$-m_2\omega^2 A_2 e^{jksd} = \left(-C_1 - C_2\right) A_2 e^{jksd} + \left(C_1 e^{jksd} + C_2 e^{jk(s+1)d}\right) A_1$$

Divide both sides by e^{jksd} to get:

$$-m_1\omega^2 A_1 + \left(C_1 + C_2\right) A_1 - \left(C_1 + C_2 e^{-jkd}\right) A_2 = 0$$

$$-m_2\omega^2 A_2 + \left(C_1 + C_2\right) A_2 - \left(C_1 + C_2 e^{jkd}\right) A_1 = 0$$

or in matrix form

$$\begin{pmatrix} m_1\omega^2 - C_1 - C_2 & C_1 + C_2 e^{-jkd} \\ C_1 + C_2 e^{jkd} & m_2\omega^2 - C_1 - C_2 \end{pmatrix} \begin{pmatrix} A_1 \\ A_2 \end{pmatrix} = 0$$

Aside: Reminder of Important (and often forgotten) Theorems from Linear Algebra

Suppose we have a system of linear equations represented by M·V = 0 where M is a matrix and V is a column vector. Now suppose that M and V represent a real physical system for which we know a real, unique, non-zero solution exists. Then the trival solution, V = 0 can not exist. So the fact that there is a unique solution forces us to discard the trivial solution as a possibility.

Now suppose that the matrix is invertible, i.e., M^{-1} exists such that $M^{-1} \cdot M = I$, where I is the identity matrix (one in diagonal, zeroes everywhere else). If this is true, then we could invert M·V = 0 by multiplying both sides by M^{-1} to get V = 0 ! To prevent this possibility, we must require that M not be invertible.

But from yet another theorem of linear algebra, the condition that M not be invertible also means that the determinant of M vanish, i.e., det{M} = 0.

So the coupled pair of equations has a unique, non-trivial solution only if the determinant of 2x2 matrix vanishes

$$\rightarrow \left[m_1\omega^2 - \left(C_1 + C_2\right)\right]\left[m_2\omega^2 - \left(C_1 + C_2\right)\right] - \left[C_1^2 + C_2^2 + 2C_1C_2 \cos kd \right] = 0$$

or

$$m_1m_2\omega^4 + \left(C_1 + C_2\right)^2 - \left(m_1 + m_2\right)\left(C_1 + C_2\right)\omega^2 - \left[C_1^2 + C_2^2 + 2C_1C_2 \cos kd \right] = 0$$

$$m_1m_2\omega^4 + \left(C_1 + C_2\right)^2 - \left(m_1 + m_2\right)\left(C_1 + C_2\right)\omega^2 - \left[C_1^2 + C_2^2 + 2C_1C_2 \cos kd \right] = 0$$

Define: $\dfrac{m_1 + m_2}{m_1m_2} = \dfrac{1}{\mu}$ $\mu \equiv$ reduced mass

get $\omega^4 - \dfrac{\left(C_1 + C_2\right)}{\mu}\omega^2 + \dfrac{2C_1C_2}{m_1m_2}\left(1 - \cos kd\right) = 0$

let $v = \omega^2$, solve by quadratic formula

$\omega = \sqrt{v}$ positive root only

and we get $\omega^2 = \dfrac{C_1 + C_2}{2\mu} \pm \dfrac{1}{2}\sqrt{\left(\dfrac{C_1 + C_2}{\mu}\right)^2 - \dfrac{8C_1C_2}{m_1m_2}\left(1 - \cos kd\right)}$

Interesting solutions again occur at kd = 0 and kd = π

(1) kd = 0 $\Rightarrow$ cos kd = 1

$$\rightarrow \omega^2 = \frac{C_1 + C_2}{2\mu} \pm \frac{C_1 + C_2}{2\mu} = \begin{cases} \dfrac{C_1 + C_2}{\mu} + branch \\ 0 \ - branch \end{cases}$$

Note + branch reduces to $\omega^2 = \dfrac{2C\left(m_1 + m_2\right)}{m_1 m_2} = 2C\left(\dfrac{1}{m_1} + \dfrac{1}{m_2}\right)$

when $C_1 = C_2 \equiv C$ (as in Kittel)

Also note + branch reduces to $\omega^2 = \dfrac{2\left(C_1 + C_2\right)}{m}$ when $m_1 = m_2 \equiv m$

But in general $m_1 \neq m_2$, $C_1 \neq C_2$

(2) kd = π $\Rightarrow$ cos kd = -1

$$\rightarrow \omega^2 = \frac{C_1 + C_2}{2\mu} \pm \frac{1}{2}\sqrt{\left(\frac{\left(C_1 + C_2\right)^2}{\mu}\right) - \frac{16 C_1 C_2}{m_1 m_2}}$$

$$= \frac{C_1 + C_2}{2\mu}\left\{1 \pm \sqrt{1 - \frac{16\mu^2 C_1 C_2}{m_1 m_2 \left(C_1 + C_2\right)^2}}\right\}$$

Can go back and solve for A_1/A_2:

$$\frac{A_1}{A_2} = \frac{C_1 + C_2 e^{-jkd}}{C_1 + C_2 - m_1\omega^2}$$

$$\frac{A_1}{A_2} = \frac{C_1 + C_2 e^{-jkd}}{C_1 + C_2 - m_1\left[\dfrac{C_1 + C_2}{2\mu} \pm \dfrac{1}{2}\sqrt{\left(\dfrac{C_1 + C_2}{\mu}\right)^2 - \dfrac{8 C_1 C_2}{m_1 m_2}\left(1 - \cos kd\right)}\right]}$$

(1) at kd = 0, this goes to

$$\frac{A_1}{A_2} = \frac{C_1 + C_2}{C_1 + C_2 - m_1\left[\frac{C_1 + C_2}{2\mu} \pm \frac{C_1 + C_2}{2\mu}\right]}$$

$$\rightarrow \frac{C_1 + C_2}{C_1 + C_2 - \frac{m_1}{\mu}(C_1 + C_2)} = \frac{1}{1 - \frac{m_1}{\mu}} \quad \text{for + branch}$$

always negative

$\rightarrow 1$ for – branch

- So that for + branch, main atom A_1 and its satellite A_2 are always out of phase by 180˚ no matter what m_1, m_2, C_1 or C_2. This leads us to call the + branch the optical branch.
- And for - branch, main atom and its satellite are always in phase. This leads us to call the - branch the acoustical branch.

(2) now at kd = π , cos kd = -1, sin kd = 0, and we get

$$\frac{A_1}{A_2} = \frac{C_1 - C_2}{C_1 + C_2 - m_1\left[\frac{C_1 + C_2}{2\mu} \pm \frac{1}{2}\sqrt{\left(\frac{C_1 + C_2}{\mu}\right)^2 - \frac{16C_1C_2}{m_1m_2}}\right]}$$

and there is, in general, no special relationship between A_1 and A_2 for either branch

<u>However</u>, if $C_1 = C_2$, $\frac{A_1}{A_2} = 0$ which implies $A_1 = 0$ for arbitrary A_2.

A similar analysis of 2 x 2 matrix for $\frac{A_1}{A_2}$ produces an equally good expression

$$\frac{A_1}{A_2} = \frac{C_1 + C_2 - m_2\omega^2}{C_1 + C_2e^{ika}} \rightarrow \frac{C_1 + C_2 - m_2\omega^2}{C_1 - C_2} \quad \text{at kd} = \pi$$

so now $C_1 = C_2 \rightarrow \frac{A_1}{A_2} \rightarrow \infty$ or $A_2 = 0$ for arbitrary A_1. Hence for $C_1 = C_2$, m_1 and m_2 are "decoupled"

Summary and Important Remarks

We now see some very interesting and important contrasts between the elastic theory of solids and the lattice-wave, or discrete-mass theory:

(1) Both predict the existence of compressional and shear waves

(2) But the lattice wave theory predicts something new – the *optical* wave – when the crystal is in the form of a lattice plus a basis. The optical wave arises from the new degree-of-freedom allowed by the relative motion of the primary atom and each of the "satellite" atoms of the primitive cell.

(3) With elasticity theory the dispersion relation between ω and k for both the compressional and shear waves is linear to arbitrarily high k, meaning that both waves have constant phase and group velocities (i.e., they are dispersionless).

(4) But lattice-wave theory predicts that all wave types, compressional and shear, acoustic and optical, approach zero group velocity at the special value of $k = \pi/d$, where d is the distance between adjacent parallel (Bravais) lattice planes perpendicular to the direction of propagation. This is important result is caused by Bragg scattering of the lattice wave from all the atoms acting collectively.

(5) Convenient nomenclature: the compression and shear waves of lattice-wave theory that map into the compressional and shear waves of elasticity theory are called longitudinal acoustic (LA) and transverse acoustic (TA), respectively. The new compressional and shear waves of lattice-wave theory (lattice plus basis) are called longitudinal optic (LO) and transverse optic (TO), respectively.

(6) We have seen that a monatomic Bravais lattice has three acoustical and zero optical lattice waves. A Bravais lattice-plus-basis with two atoms per primitive cell (one primary, one sattelite) has three acoustic and three optical lattice waves. By deduction, we can say that a Bravais lattice plus basis having P atoms per primitive cell will have 3 acoustical waves and 3P – 3 optical waves. Of the 3 acoustic waves, one will always be the LA, and two will be TA. Of the 3P-3 optical waves, P-1 of them will be LO waves, and 3P – 3 – (P-1) , or 2P – 2 will be TO waves.

The above statements are very useful to remember for examination and research purposes !