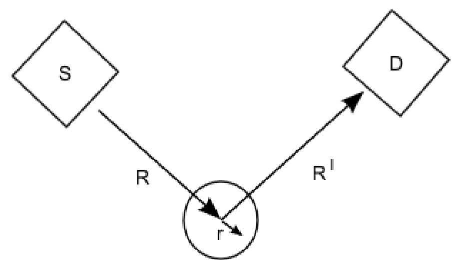
Topic 7-1: Inelastic Scattering Theory
Ibach and Luth Pages: 91-93

**Summary:** We begin by ascertaining an expression for the amplitude at the detector with inelastic scattering of vibrating atoms based off of the expression developed for elastic scattering. We then explore neutron scattering as well as Raman scattering to give physical examples of inelastic scattering.

- <u>Elastic scattering</u>: found constructive interference at the detector when $\Delta\vec{k} = \vec{G}$ for a periodic sample




- Amplitude of elastic scattering was given by $F \propto \int_V n(\vec{r}) e^{-i(\Delta\vec{k}\cdot\vec{r}+\omega_o t)} d\vec{r}$
- Now we consider *vibrating* atoms; the scattering density $n(\vec{r})$ is now a function of time as well as position
- To make this manageable, we're going to make as many approximations as possible. Treat the atoms as *point* scattering sources with one atom at each lattice point (trivial basis)
    - $n(\vec{r},t) \propto \sum_{\vec{T}} \delta(\vec{r} - r_{\vec{T}}(t))$ where $r_{\vec{T}}(t)$ is the position of the atom based at $\vec{T}$ as a function of time, there will be motion of the atom over time off of $\vec{T}$.
    - At 0 K, $r_{\vec{T}}(t) = \vec{T}$ for all time.
- Putting $n(\vec{r},t)$ into the integral as our new scattering density, the delta functions in $n(\vec{r},t)$ will destroy the integral and leave $F_D \propto \sum_{\vec{T}} e^{-i(\Delta\vec{k}\cdot\vec{r}_{\vec{T}}(t)+\omega_o t)}$
- Let $r_{\vec{T}}(t) = r_{\vec{T}} + \vec{u}_{\vec{T}}(t)$ which is the original placement of the atom at $\vec{T}$ plus the displacement off of $\vec{T}$
- Now $F_D \propto e^{-i\Delta\vec{k}\cdot\vec{r}_{\vec{T}}} e^{-i\Delta\vec{k}\cdot\vec{u}_{\vec{T}}(t)} e^{-i\omega_o t}$
- Now we can expand $\vec{u}_{\vec{T}}(t)$ for small displacements as $e^{-i\Delta\vec{k}\cdot\vec{u}_{\vec{T}}(t)} \rightarrow (1 - i\Delta\vec{k}\cdot\vec{u}_{\vec{T}}(t))$

- Also recall displacement $\vec{u}_{\vec{T}} = \hat{u}e^{\pm i(\vec{q}\cdot r_{\vec{T}} - \omega(\vec{q})t)}$ where $\vec{q}$ is the same wave vector from phonons and has an associated frequency, just a normal mode in the lattice
- **Conclusion:** Now putting it all together we get the following expression for the amplitude at the detector

$$F_D \propto \sum_{\vec{T}} e^{-i(\Delta\vec{k}\cdot\vec{r}_{\vec{T}} - \omega_o t)} - \sum_{\vec{T}} i\Delta\vec{k}\cdot\hat{u}e^{-i\Delta\vec{k}\cdot r_{\vec{T}}}e^{\pm i(\vec{q}\cdot r_{\vec{T}})}e^{\mp i\omega(q)t}\, e^{-i\omega_o t} \quad [1]$$

  - First sum is the elastic scattering term, second sum is the inelastic scattering term $F_{inelastic} = \sum_{\vec{T}} e^{-i(\Delta\vec{k}\mp\vec{q})\cdot r_{\vec{T}}} i\Delta\vec{k}\cdot\hat{u}\, e^{-i(\omega_{o\pm}\omega(q)t)}$
- Condition for constructive interference from inelastic component is $\Delta\vec{k} \pm \vec{q} = \vec{G}$ or $\Delta\vec{k} = \vec{G} \pm \vec{q}$
- ω at the detector will be $\omega = \omega_o \pm \omega(q)$

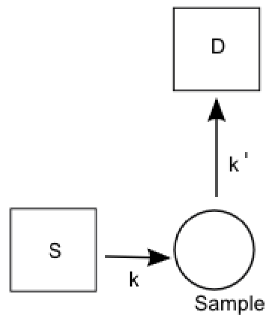
**Physical example: Neutron scattering**

  - Neutron incident on sample then diffracted
  - There is a high probability that elastic diffraction happens and it's just the usual business.
  - Occasionally, neutron absorbs a phonon, gaining both its momentum and energy or the neutron will create a phonon in the sample as it passes through, losing both that momentum and energy.
  - If we can resolve position and energy at the detector, we can figure out what happened for every neutron.
- Vector picture for inelastic scattering is more complicated than the elastic case!
- In inelastic scattering $|k'| \neq |k|$
  - When a phonon absorbed by the neutron, the exit $|k'| > |k|$ (incident neutron wavevector) since the final wavelength is smaller because the exit energy is greater than the incident energy, this creates a Δk closer to k'
  - When a phonon is created, $|k'| < |k|$ which creates a Δk closer to k
- Use inelastic scattering because it gives a lot of information about the phonon population within the sample.

**Physical example: Raman scattering**

- Now consider the inelastic scattering of visible light, known as Raman scattering

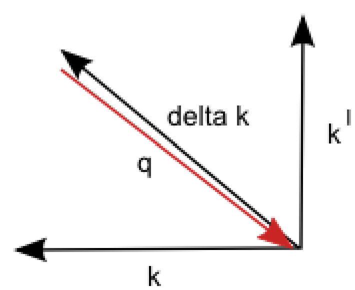
- New setup as below



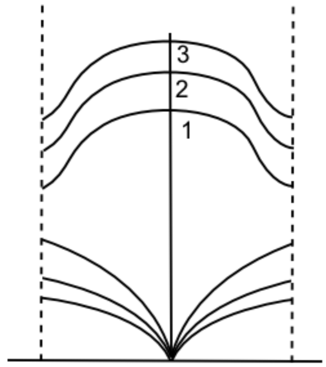

- Detector is now frequency dependent
  - Splitter separates out frequencies spatially
- Consider $|k'|\ vs\ |k|$
  - Elastic scattering and visible light were incompatible because Δk is too short to reach a nonzero G vector, this is why we used X-rays! (larger ks, could have a reasonably large Ewald sphere)
- Start with change in energy from creating or absorbing a phonon in a solid with visible light
- Visible light energy: $E = \frac{1240}{\lambda\, nm} = \frac{1240}{620\, nm} = 2eV$
  - 620 is the wavelength of red light
- $|k| = \frac{2\pi}{620}$
- If light has highest energy at 2 eV and the highest energy phonon is at about 0.05 eV our exit light energy will be between 1.95 and 2.05eV
  - This is an upper bound because we are invoking that the highest energy phonon is either being created or destroyed

| Scenario: | Phonon absorbed by incident red light | Phonon created by incident red light |
|---|---|---|
| New wavelength | 605 nm, orange | 636 nm, deep red |
| $\lvert k'\rvert$ | $\frac{2\pi}{605}$ | $\frac{2\pi}{636}$ |
| $\Delta\lvert k\rvert$ | $\frac{2\pi}{25000\ nm}$ | $\frac{2\pi}{24600\ nm}$ |

- $\Delta|k|$ is really small especially compared to the size of the reciprocal lattice
- **Intermediate conclusion:** For visible light undergoing inelastic scattering, $|k'| \approx |k|$ and $|k'|\ and\ |k| \ll |g_1|$
- Remember need $\Delta\vec{k} = \vec{G} \pm \vec{q}$ for constructive interference in inelastic diffraction
    - But from the phonon, $|q_{max}| \leq \frac{\pi}{a}$
- Thus, the only $\vec{G}$ that can satisfy $\Delta\vec{k} = \vec{G} \pm \vec{q}$ is $\vec{G} = 0$
    - $\Delta\vec{k} = \pm\vec{q}$ for constructive interference at the detector
- Elastic case: if $\Delta\vec{k} = \vec{G} = 0$ this was transmission
- Recall $\Delta|k| \ll |g_1|$
- Only phonons extremely close to gamma (with $\vec{q}$ = $\pm$ light's $\Delta\vec{k}$), at the top of the dispersion graph, will be involved in inelastic scattering of visible light as $\Delta\vec{k} = \pm\vec{q}$ must be satisfied.




- Above for phonon absorbed by red light. g1, g2 vectors are way larger than the graph region.
- $\omega = \omega_o + \omega(q)$ if phonon absorbed or $\omega = \omega_o - \omega(q)$ if phonon created by light
- Will have a 3D solid in a real experiment




- Detector measures frequency with a spatial dependence
- Will see one large intensity peak associated with specularly scattered light at $\omega_o$ then 3 little peaks associated the 3 optical modes
    - 3 to the right (higher energy) due to absorbed phonons and 3 to the left due to created phonons (lower energy)
- We now have a spectroscopic way to look at phonon modes at the gamma point using visible light!