Lecture #12 PHYS0551

Title: Quantization of the Elastic Energy and Interaction of Incident Waves with a Vibrating Crystal

Quantization of the Elastic Energy (PHONONS):

Up until this point, we have been discussing elastic waves in a crystal. There has been no explicit way to introduce the quantization of these waves. (Except to state that $E = \hbar\omega$.) This is not entirely correct, ω really represents a particular mode of the lattice. If the total energy of a particular mode is to be calculated, then $E_\omega = (n + \frac{1}{2})\hbar\omega$ where n is the sum number of phonons, $\hbar\omega/2$ is the quantum mechanical zero point energy and $\hbar\omega$ is the unit energy per mode of vibration (i.e., per phonon).

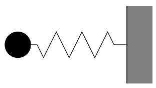
In theory, there is no limit to the number phonons of any particular mode.

To justify this we note that at any instant in time an atom s will have kinetic and potential energy for a particular vibrational mode. Thus the total energy is (monoatomic 1-D chain)

$$H = \sum_{s=1}^{N}[\frac{1}{2m}p_s^2 + \frac{1}{2}c(q_{s+1} - q_s)^2]$$

assuming the atoms form a very large ring. (Atom N is next to atom 1.) Notice the analogy to the harmonic oscillator

$$H = \frac{1}{2m}p^2 + \frac{1}{2}cx^2$$



If $H\psi = E\psi$ is solved, then

$$\epsilon_n = (n + \frac{1}{2})\hbar\omega \qquad \omega = \sqrt{c/m}$$

$$\psi_n(x) = \left(\frac{\alpha}{\sqrt{\pi}2^n n!}\right)^{1/2} \exp(-\alpha^2 x^2/2)\ H_n(\alpha x) \quad \text{where} \quad \alpha = (mc/\hbar^2)^{1/2}$$

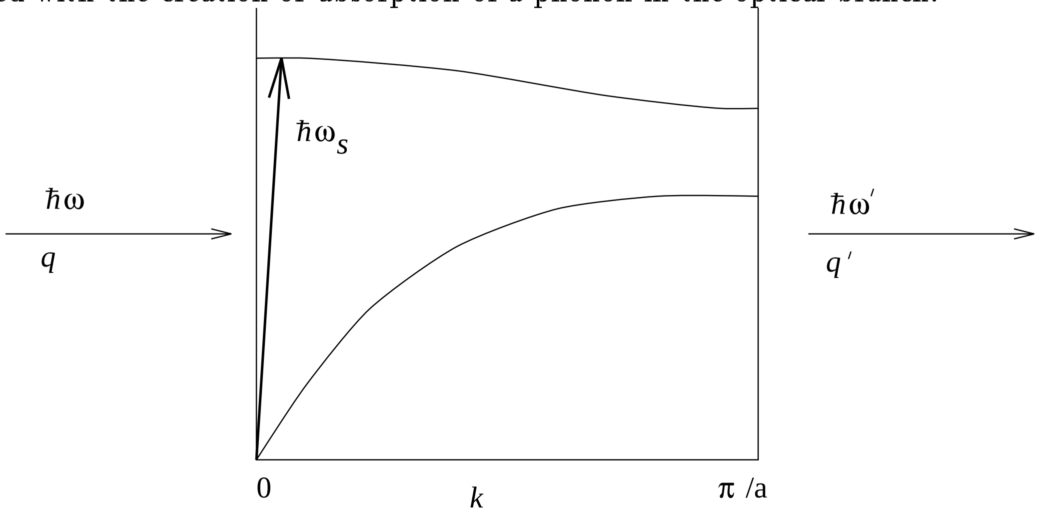
Generalizing this argument for each mode of ω,
then $\epsilon_{n,\omega} = (n + 1/2)\hbar\omega$ and $\omega = \sqrt{4c/m}\ |\sin(ka/2)|$.
Thus at absolute zero, the elastic energy stored in a crystal is

$$E(T = 0) = \sum_{\omega' s}^{all} \frac{\hbar\omega}{2}$$

and we can add and subtract energy to the crystal in increments of $\hbar\omega$!

Interaction of incident waves with a vibrating crystal:

Perhaps the simplest interaction is Raman scattering. Here a photon of visible light is scattered with the creation or absorption of a phonon in the optical branch.



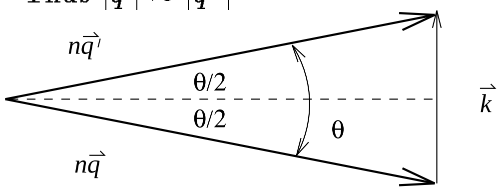
Note that the photon wave vectors are changed inside the crystal due the index of refraction.

Conservation of energy $\quad \hbar\omega' = \hbar\omega \pm \hbar\omega_s(k)$

Conservation of momentum $\quad \hbar n\vec{q}\,' = \hbar n\vec{q} \pm \hbar\vec{k} + \hbar\vec{G}$

Since $|\vec{q}\,'|$ and $|\vec{q}|$ are exceedingly small, $\vec{G} = 0$ and $\hbar\omega \sim 2eV \;\; \hbar\omega_s \sim 10^{-2}$ eV.

Thus $|\vec{q}| \approx |\vec{q}\,'|$



θ is the scattering angle

$|k| \approx 2nq \sin\frac{\theta}{2} = 2(\omega n/c)\sin\frac{\theta}{2}$

$k^2 = \frac{4\omega^2 n^2}{c^2}\sin\frac{\theta}{2}$

$k_{max} = 2\omega n/c \sim 10^{-2}\text{Å}^{-1} \approx 0$ (compared to the B.Z. boundary).

So from the previous lecture (#11)

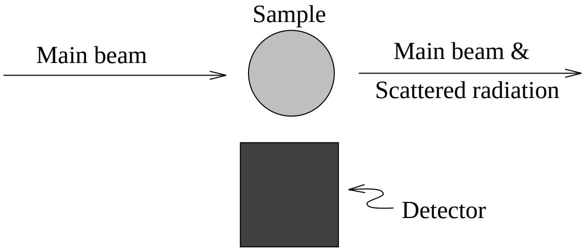
$$\omega_s^2 \approx 2c_s(\frac{1}{M} + \frac{1}{m}) + \frac{c_s}{M+m}\frac{k^2 a^2}{2}$$

(and the 2^{nd} is *small* compared to first term) giving

$$\omega_s^2 = 2c_s(\frac{1}{M} + \frac{1}{m}) + \frac{c_s}{M+m}\frac{a^2}{2}\frac{4\omega^2 n^2}{c^2}\sin\frac{\theta}{2}$$

with $\qquad \omega = \omega' + \omega_s$

Only a small angular dependence of the energies.



(These modes must also satisfy symmetry considerations.)

Scattering of Neutrons (Coherent Inelastic Diffraction):

In the previous case we discussed the scattering in the context of particles interacting. However, we could have just as easily focused on the wave nature of matter.

In view of the fact that our crystal is not static, we will examine this system when a monochromatic beam of neutrons (or He atoms for that matter) is incident on this crystal.

(1) We see the scattering due to the equilibrium position of the atoms (i.e., simple Bragg scattering).

(2) We observe the absorption and creation of phonons.

Mathematically:

For each atom $\vec{r}_j = \vec{r}_{0_j} + \vec{u}_j$ where $\vec{r}_{0_j} \equiv$ equilibrium position
$\vec{u}_j \equiv$ displacement from equilibrium

Note: $\vec{r}_{0_j} = n_1\vec{a}_1 + n_2\vec{a}_2 + n_3\vec{a}_3$

$\vec{u}_j$ is the composite sum due to *all* phonons.

$$\vec{r}_j = \vec{r}_{0_j} + \sum_{q>0}[u_q\ e^{i(\vec{q}\cdot\vec{r}_{0_j} - \omega t)} + u_q^*\ e^{i(-\vec{q}\cdot\vec{r}_{0_j} - \omega t)}]\ ,\ \omega = f(\vec{q})$$

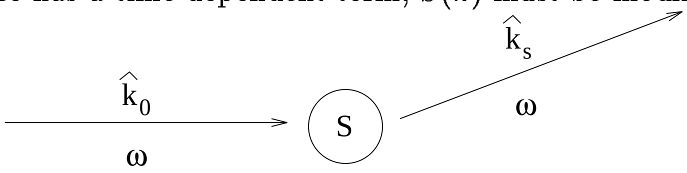
Notice $u_q^* \equiv u_{-q}$ by symmetry arguments.

Notice also the factor of $e^{i\omega t}$; this term *cannot* be ignored. As a result, the "static" distortions are seen in the laboratory.

Recall the structure factor S($\vec{k}$),

$$S(\vec{k}) = \frac{1}{N} f(\vec{k}) \sum_{\vec{r}}^{\substack{\text{All}\\ \text{space}}} e^{-i\vec{k}\cdot\vec{r}_j}$$

Since the lattice has a time dependent term, $S(\vec{k})$ must be modified

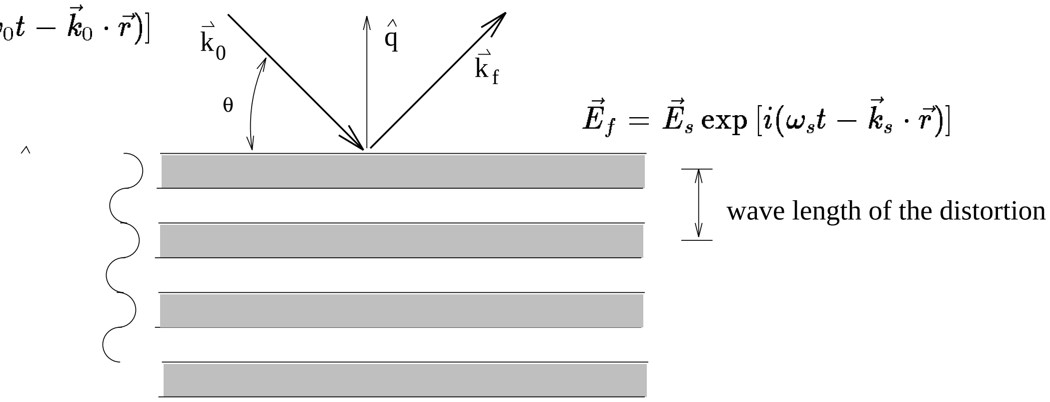


A digression:

If we move at a v_{phase} along the $\vec{q}$ direction, then those phonons whose speed we match will appear stationary. Thus, this distortion can now act as a diffraction grating.

$\vec{E}_i = \vec{E}_0 \exp\left[i(\omega_0 t - \vec{k}_0 \cdot \vec{r})\right]$

$\vec{E}_f = \vec{E}_s \exp\left[i(\omega_s t - \vec{k}_s \cdot \vec{r})\right]$



Thus $n\lambda = 2d\sin\theta$. In the laboratory frame, there will be Doppler shifting of the neutron frequencies.

$$
\begin{aligned}
d &= \frac{2\pi}{q} \\
n\lambda &= \frac{4\pi}{q}\sin\theta \qquad \lambda = \frac{2\pi}{|k|} \\
n/|k| &= \frac{2}{q}\sin\theta \\
nq &= 2|k|\sin\theta \qquad -\vec{k}_0\cdot\hat{q} = |k_0|\sin\theta \qquad \vec{k}_f\cdot\hat{q} = |k_f|\sin\theta \\
nq &= (\vec{k}_f - \vec{k}_0)\cdot\hat{q} \\
& \quad \text{Now } \vec{k}_f - \vec{k}_0 \parallel \hat{q} \\
n\vec{q} &= \vec{k}_f - \vec{k}_0 \qquad v_p = \omega/q \qquad \vec{v}_p = \frac{\omega}{q}\hat{q}
\end{aligned}
$$

$$
\text{Moving frame}\left\{\begin{array}{rcl}
\omega' & = & \omega - \vec{q}\cdot\vec{v} \\
\frac{\overline{E}_0}{\hbar} & = & \frac{E_0}{\hbar} - \vec{k}_0\cdot\vec{v} \\
\frac{\overline{E}_f}{\hbar} & = & \frac{E_f}{\hbar} - \vec{k}_f\cdot\vec{v}
\end{array}\right\}\text{Laboratory Frame}
$$

$$
\overline{E}_0 = \overline{E}_f\ ,\ \frac{E_f}{\hbar} = \frac{E_0}{\hbar} + (\vec{k}_f - \vec{k}_0)\cdot\vec{v}
$$

Substituting $\quad n\vec{q} = \vec{k}_f - \vec{k}_0$

$$E_f = E_0 + n\vec{q}\cdot\vec{v}$$

$$E_f = E_0 + n\hbar\omega$$

Thus the "process" of Bragg reflection corresponds to simultaneous creation or absorption of a phonon and scattering of the resultant neutron. For values of $n > 1$ this corresponds to a multiphonon process.

Back to $S(\vec{k}) \implies S(\vec{k},\omega)$

$$S(\vec{k},\omega) = \frac{1}{N}\int_{-\infty}^{\infty}\frac{dt}{2\pi}\sum_{\vec{r}}^{\substack{\text{All}\\ \text{space}}} e^{-i\vec{k}\cdot\vec{r}_j}\, e^{-i\frac{E_f}{\hbar}t}\, e^{+i\frac{E_0}{\hbar}t}$$

$$S(\vec{k},\omega) = \frac{1}{N}\sum_{\vec{r}}^{\substack{\text{All}\\ \text{space}}} \underbrace{e^{-i\vec{k}\cdot\vec{r}_j}}_{\text{No more }\underline{t}\text{ dependence}} \qquad \underbrace{\delta(E_0 - E_f \pm \sum_{\omega}\hbar\omega)}_{\text{Conservation of energy}}$$

If ω=0, elastic scattering

Now to examine $\sum_{\vec{r}}\exp\ i\vec{k}\cdot\vec{r}_j$ in greater detail

(1) $\sum_{\vec{r}} e^{i\vec{k}\cdot\vec{r}_j} = \sum_{\vec{r}}\exp[-i\vec{k}\cdot\{\vec{r}_0 + \sum_{q>0}(u_q\ e^{i\vec{q}\cdot\vec{r}_0} + u_q\ e^{-i\vec{q}\cdot\vec{r}_0})\}]$

(The j indexing is now implicit)

If u_q's are small (HARMONIC approximation)

(2) $\exp\{-i\vec{k}\cdot u_q\ e^{i\vec{q}\cdot\vec{r}_0}\} \approx 1 - i\vec{k}\cdot u_q\ e^{i\vec{q}\cdot\vec{r}_0} - \frac{1}{2}|\vec{k}\cdot\vec{u}_q|^2 + ...$

So by rearranging (1)

$$\sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}_j} = \sum_{\vec{r}}\exp(-i\vec{k}\cdot\vec{r}_0)\Pi_{q>0}\exp\{-i\vec{k}\cdot(u_q e^{i\vec{q}\cdot\vec{r}_0} + u_{-q}e^{-i\vec{q}\cdot\vec{r}_0})\}$$

Substituting (2)

$$\sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}} = \sum_{\vec{r}}\exp(-i\vec{k}\cdot\vec{r}_0)\Pi_{q>0}\{1 - i\vec{k}\cdot(u_q\ e^{i\vec{q}\cdot\vec{r}_0} + u_{-q}e^{-i\vec{q}\cdot\vec{r}_0}) - \frac{1}{2}|\vec{k}\cdot u_q|^2 + ...\}$$

(Assume $u_q \approx u_{-q}$)

The $\Pi_{q>0}$ implies a product $(1-\frac{q_1}{....})(1-\frac{q_2}{....})(1-\frac{q_3}{....})$ for all q values.

So we get

$$\Pi_{q>0}\{....\} = 1 + \sum_{q>0} -i\vec{k}\cdot(u_q\, e^{i\vec{q}\cdot\vec{r}_0} + u_{-q}\, e^{-i\vec{q}\cdot\vec{r}_0}) - \underbrace{2\sum_{q>0}|\vec{k}\cdot u_q|^2}_{\text{Net displacement } \equiv W \propto k^2} + ...$$

Let

$$W = 2\sum_{q>0}|\vec{k}\cdot\vec{u}_q|^2$$

Thus

$$\sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}} = \sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}_0}\{1 - W + \sum_{q>0} -i\vec{k}\cdot(u_q\, e^{i\vec{q}\cdot\vec{r}_0} + u_{-q}e^{-i\vec{q}\cdot\vec{r}_0})\}$$

Note: if $W << 1 \quad \exp(-W) \approx\ 1 - W$ (This is done for conventional reasons)

$$\begin{aligned}
\sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}} &= \sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}_0}\exp(-W) + \sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}}\sum_{q>0}\left\{-i\vec{k}\cdot(u_q\, e^{i\vec{q}\cdot\vec{r}_0} + u_{-q}\, e^{-i\vec{q}\cdot\vec{r}_0})\right\} \\
\Downarrow &= \sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}_0}\ \exp(-W) + \sum_{q>0}\sum_{\vec{r}}\left[\{e^{-i(\vec{k}-\vec{q})\cdot\vec{r}_0}\}(-i\vec{k}\cdot\vec{u}_q) + \{e^{-i(\vec{k}+\vec{q})\cdot\vec{r}_0}\}(-i\vec{k}\cdot\vec{u}_q)\right] \\
\text{or} & \\
\sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}} &= \sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}_0}\exp(-W) + \sum_{q>0}\delta(\vec{k}\pm q, \vec{G}_{hk\ell})(-i\vec{k}\cdot u_q) \\
\text{So } S(\vec{k},\omega) &= \frac{1}{N}f(|\vec{k}|)\sum_{\vec{r}} e^{-i\vec{k}\cdot\vec{r}_0} + \frac{1}{N}f(|\vec{k}|)\sum_{q>0}\delta(\vec{k}\pm q, \vec{G}_{hk\ell})(ik\cdot u_q) \\
& \qquad \times \text{ conservation of energy} \\
S(\vec{k},\omega) &= S_{\text{Elastic}}(\vec{G}_{hk\ell})\exp(-W)\delta(\vec{k},\vec{G})\delta(E_0 - E_f) \\
&+ f'(\vec{k})\sum_{q>0}\underbrace{\delta(\vec{k}\pm\vec{q}, \vec{G}_{nk\ell})}_{\text{conservation of “crystal” momentum}}(-i\vec{G}\cdot u_q)\ \underbrace{\delta(E_0 = E_f \pm \hbar\omega)}_{\text{conservation of Energy}}
\end{aligned}$$

Since neutrons are incident $\Longrightarrow E = \hbar^2 k^2/2M$ for a particle

$$\frac{\hbar^2}{2M_{\text{neutron}}}(k_i^2 - k_f^2) = \pm\hbar\omega_q$$

Recall $\exp(-2W) \equiv$ Debye-Waller factor and $\omega \propto k^2$

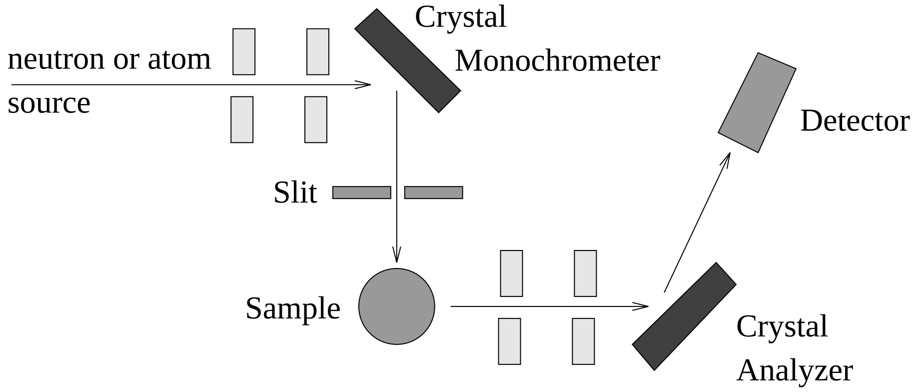
*Thus Bragg peaks have reduced intensity.

*2^{nd} term corresponds to the creation/absorption of a phonon.

To determine ω_q vs. q from neutron diffraction

$\vec{k}_f = \vec{k}_i \pm \vec{q} + \vec{G}$ Conservation of Crystal Momentum

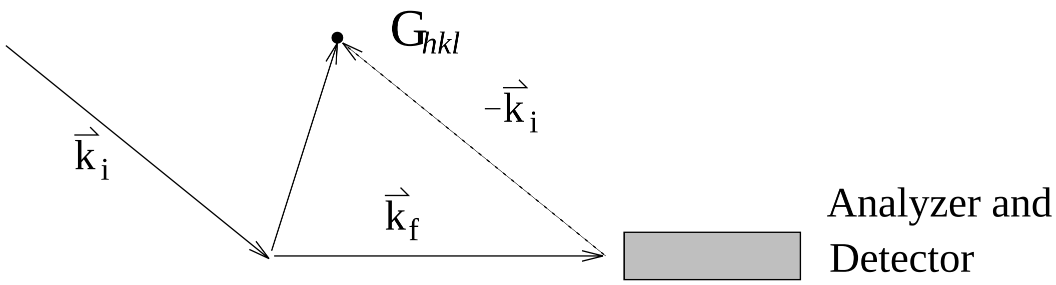
$\frac{\hbar^2}{2m_n}(k_f^2 - f_i^2) = \pm\hbar\omega_q$ Conservation of Energy



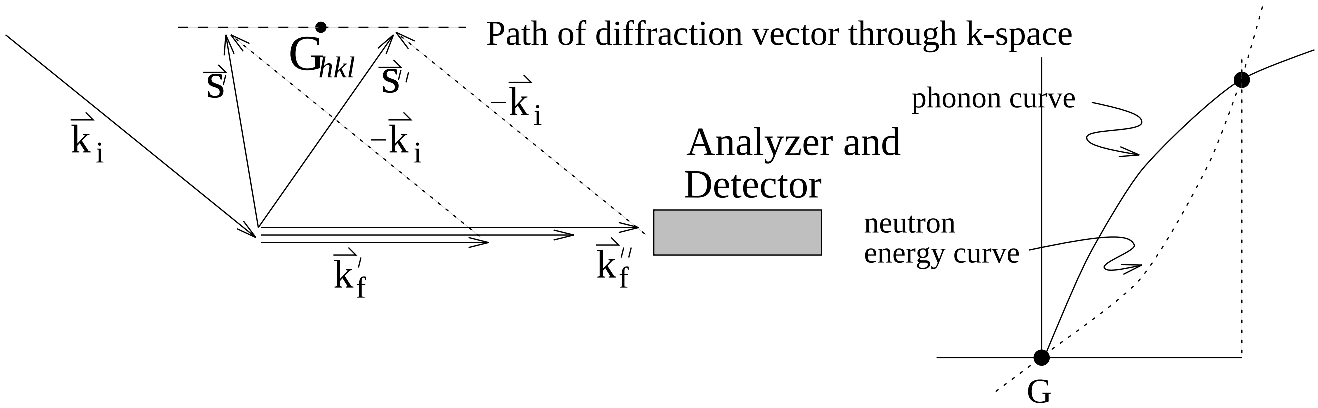
$E_n = \hbar^2 K_n^2 / 2m_n$

To do an actual experiment, the crystal orientation *must* be known

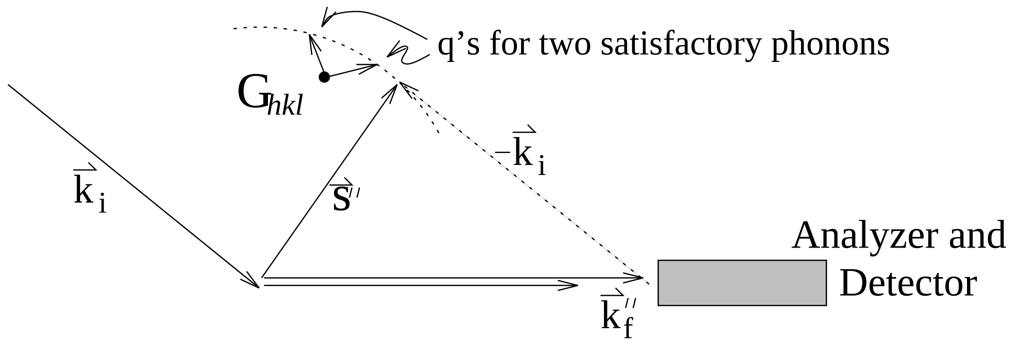
Let $|\vec{k}_i| = |\vec{k}_f|$ (elastic scattering)



Sweep Energy: Now adjust $E_n(f), |k_f| = \sqrt{\frac{2mE}{\hbar}}$ $\frac{dE}{dk} = \hbar k / m_n$



Fix Energy, Rotate crystal



Fix Energy, Rotate detector