Outline

- Motivation – Inelastic Scattering
- Elementary excitations
  - Phonons
  - Magnons
- Practical aspects
- Scattering cross-section essentials
- One phonon cross-section
  - Example of Cu
- Magnons
- Damped harmonic oscillators
Inelastic Neutron Scattering

1994 Nobel prize poster (C. G. Shull and B. N. Brockhouse)
Excitations in solids

- Physical properties are determined by excitations, such as phonons and electronic excitations
Phonon

- Propagating lattice vibration
- Carries most of entropy in a material
- Responsible for many thermodynamic phenomena (Specific heat, thermal conduction, etc.)

http://spark.ucar.edu/molecular-vibration-modes
Phonon propagation

<table>
<thead>
<tr>
<th>Molecules/gas</th>
<th>Liquid/glass</th>
<th>Solid (crystal)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vibrational and rotational modes</td>
<td>Propagating vibrational mode – both longitudinal and transverse mode e.g. earthquakes</td>
<td></td>
</tr>
<tr>
<td>Do not propagate – no momentum dependence</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$c = \frac{\partial \omega}{\partial k}$

Longitudinal

Transverse

http://people.web.psi.ch/delley/nacl.html

http://www.tf.uni-kiel.de/matwis/amat/mw2_ge/kap_2/advanced/t2_1_1.html
Magnon (spin wave)

- Magnetic order (spin solid)
  - Ferromagnetic or
  - Antiferromagnetic
- Magnetic moment fluctuations can propagate in the presence of order
  - Spin wave or magnon
  - Only transverse!
- In general, material’s magnon dispersion is easier to calculate than its phonon counterpart. Why?
How to measure these in practice?

- Neutron scattering data $\xrightarrow{\text{Histogram of events}}$
- Inelastic scattering events different from diffraction
- Poisson distribution
  - $\sigma$ is given by $\sqrt{\# \text{ of events}}$
Scattering triangle

**Elastic:** \( \mathbf{k}_i = \mathbf{k}_f \)

**Neutron Energy loss:** \( \mathbf{k}_f < \mathbf{k}_i \)

**Neutron Energy gain:** \( \mathbf{k}_f > \mathbf{k}_i \)

Kinematic range that can be covered in a scattering event:

\[
\frac{\hbar^2}{2m} Q^2 = E_i + E_f - 2 \sqrt{E_i E_f} \cos 2\theta
\]

\[
Q = \mathbf{k}_i - \mathbf{k}_f = G_{hkl} + \mathbf{q}
\]

\[
Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta
\]

\[
E = E_i - E_f \rightarrow \frac{\hbar^2}{2m} Q^2 = 2E_f + E - 2 \sqrt{E_f (E_f + E)} \cos 2\theta
\]
Inelastic neutron scattering measures dynamic structure factor $S(Q, \omega)$:

$$S(Q, \omega) = \frac{1}{2\pi \hbar N} \sum_{j, j'} \int_{-\infty}^{\infty} dt \left\langle e^{iQ \cdot r_j(t)} e^{-iQ \cdot r_{j'}(0)} \right\rangle e^{-i\omega t}$$

Dynamic structure factor $S(Q, \omega)$ is the Fourier Transformation of space-time correlation function.

Dynamic structure factor $S(Q, \omega)$ is related to the imaginary part of the dynamic susceptibility $\chi(Q, \omega)$ through the fluctuation-dissipation theorem:

$$S(Q, \omega) = \frac{\chi''(Q, \omega)}{1 - e^{-\hbar \omega / k_B T}}$$
Coherent one phonon scattering:

\[
S(Q, \omega) \propto \frac{(Q \cdot e_s)^2}{\omega_{qs}} \left[ \langle n_s + 1 \rangle \delta(\omega - \omega_{qs}) + \langle n_s \rangle \delta(\omega + \omega_{qs}) \right]
\]

Detailed balance: Probability of a transition depends on statistical weight factor of the initial state:

\[
\langle n_s \rangle = \frac{1}{e^{\hbar \omega_{qs}/k_BT} - 1}
\]

\[
S(-Q, -\omega) = e^{-\hbar \omega/k_BT} S(Q, \omega)
\]
Derivations

- Can be found in references:
  - Squires, *Introduction to the theory of thermal neutron scattering*
  - Shirane, Shapiro, and Tranquada, *Neutron scattering with a triple-axis spectrometer*
  - Marder, *Condensed matter physics*
  - Dove, *Structure and Dynamics*
  - Chaikin and Lubensky, *Principles of condensed matter physics*
- Do it yourself!
- I will talk about rough steps for one phonon cross-section
\[ S(Q, \omega) = \frac{1}{2\pi\hbar N} \sum_{j,j'} \int_{-\infty}^{\infty} dt \exp \left( iQr_j(t) - iQr_j'(0) \right) e^{-i\omega t} \]

\[ r_j(t) = R_j + u_j(t) \]

\[ u_j = \frac{1}{\sqrt{N}} \sum_{k,v} \left[ \hat{u}_{kv} e^{ik \cdot R_j} + \hat{u}_{kv}^* e^{-ik \cdot R_j} \right] \hat{u}_{kv} = \sqrt{\frac{\hbar}{2M\omega_{kv}}} e_{kv} \hat{a}_{kv} \]

\[ \sum_{j,j'} e^{iQ(R_j - R_{j'})} \int_{-\infty}^{\infty} dt \exp \left( iQ(u_j(t) - u_{j'}(0)) \right) e^{-i\omega t} \]

\[ \langle e^{(A+B)} \rangle = e^{\frac{(A+B)^2}{2}} \]

\[ \langle \cdots \rangle = e^{-\frac{[Q \cdot u_j(t)]^2}{2}} + \frac{[Q \cdot u_j(t)] [Q \cdot u_{j'}(0)]}{2} - \frac{[Q \cdot u_{j'}(0)]^2}{2} \]

\[ \text{mean square ionic displacement (time indep.)} \]
\[ e^{-2W} = e^{-\langle (Q \cdot u_j)^2 \rangle} \quad \text{: Debye – Waller factor} \]

\[ e^{\langle [Q \cdot u_j(t)] [Q \cdot u_j'(0)] \rangle} = 1 + \langle [Q \cdot u_j(t)] [Q \cdot u_j'(0)] \rangle + \cdots \]

\[ u_j = \frac{1}{\sqrt{N}} \sum_{kv} \left[ \hat{u}_{kv} e^{ik \cdot R_j} + \hat{u}_{kv}^+ e^{-ik \cdot R_j} \right] \quad \hat{u}_{kv} \equiv \sqrt{\frac{\hbar}{2M\omega_{kv}}} \mathbf{e}_{kv} \hat{a}_{kv} \]

\[ \text{time dependence } \hat{u}_j(t) = \hat{u}_j e^{-i\omega_{kv}t} \quad \text{(Heisenberg)} \]

\[ \langle [Q \cdot u_j(t)] [Q \cdot u_j'(0)] \rangle = \sum_{kv} \frac{1}{N} \frac{\hbar^2 (\mathbf{e}_{kv} \cdot Q)^2}{2M\hbar \omega_{kv}} \times \left\langle \hat{a}_{kv}^+ \hat{a}_{kv} e^{-i\omega_{kv}t} + \hat{a}_{kv} \hat{a}_{kv}^+ e^{i\omega_{kv}t} \right\rangle e^{-ik \cdot (R_j - R_j)} \]

\[ \langle n \rangle \quad \delta(\omega + \omega_{kv}) \quad \langle n + 1 \rangle \quad \delta(\omega - \omega_{kv}) \]
One phonon x-section revisited

\[ S(Q, \omega) \propto \frac{(Q \cdot e_s)^2}{\omega_{qs}} \left[ \langle n_s + 1 \rangle \delta(\omega - \omega_{qs}) + \langle n_s \rangle \delta(\omega + \omega_{qs}) \right] \]

Creation and annihilation

Delta function part

![Graph showing constant-q and constant-E](image)

Neutron energy gain
Excited state
\[ \hbar \omega \]

Neutron energy loss
Ground state
\[ \hbar \omega \]

Phonon annihilation
No transition at \( T = 0 \)

Phonon creation
Transition at all \( T \)

\[ T = 0 \]

\[ -\omega_0 \quad \omega_0 \]

\[ k_B T \sim \hbar \omega \]

\[ -\omega_0 \quad \omega_0 \]
Example of Cu phonon

\[ S(Q, \omega) \propto \frac{(Q \cdot e_s)^2}{\omega_{qs}} \left[ \langle n_s + 1 \rangle \delta(\omega - \omega_{qs}) + \langle n_s \rangle \delta(\omega + \omega_{qs}) \right] \]

Chesser and Axe 1973
Svensson, Brockhouse, and Rowe 1967
Magnon scattering

\[ S(Q, \omega) \propto \sum_{\alpha, \beta} \left( \delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta \right) \frac{1}{\omega_{qs}} \left[ \langle n_s + 1 \rangle \delta(\omega - \omega_{qs}) + \langle n_s \rangle \delta(\omega + \omega_{qs}) \right] \]

- Similar to phonon scattering, except for the polarization factor ➔ More by Prof. Broholm
- Magnons ➔ Transverse mode

Headings et al. PRL 105, 247001 (2010)
Damped harmonic oscillator

- In real compounds, delta function broadened due to phonon-phonon and electron-phonon interaction
- Damped harmonic oscillator is used instead

\[
\frac{1}{\omega_{qs}} \delta(\omega \pm \omega_{qs}) \rightarrow \frac{1}{\pi \omega'_{qs}} \frac{\Gamma_{qs}}{(\omega \pm \omega'_{qs})^2 + \Gamma_{qs}^2}
\]

\[
\omega'_{qs}^2 = \omega_{qs}^2 - \Gamma_{qs}^2
\]

- What about overdamping?
  - Diffusion \(\Rightarrow\) Quasielastic scattering
  - Critical scattering
Conclusions

- Studying excitations are essential for understanding materials properties
- INS can tell us about
  - Phonons
  - Magnons
  - Diffusive excitations
  - Molecular level excitations
- INS from phonons
  - Energy loss vs. energy gain
  - Use of Q
    - phonon polarization vector
    - High Q Brillouin zone