Introduction to Inelastic Neutron Scattering

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- Neutrons: Properties and Cross Sections
- Excitations in solids
- Triple Axis and Chopper Techniques
- Practical concerns
\[ ^{235}\text{U} + n \rightarrow \text{daughter nuclei} + 2-3 n + \text{gammas} \]

neutrons:

no charge

s=1/2

massive: \( mc^2 \approx 1 \text{ GeV} \)
How do we produce neutrons

Fission
- chain reaction
- continuous flow
- 1 neutron/fission

Spallation
- no chain reaction
- pulsed operation
- 30 neutrons/proton
Neutron interactions with matter

- **Properties of the neutron**
  - Mass $m_n = 1.675 \times 10^{-27}$ kg
  - Charge $0$
  - Spin-1/2, magnetic moment $\mu_n = -1.913 \mu_N$

- **Neutrons interact with...**
  - Nucleus
  - Crystal structure/excitations (e.g., Phonons)
  - Unpaired electrons via dipole scattering
  - Magnetic structure and excitations

Nuclear scattering

Magnetic dipole scattering
Wavelength-energy relations

- Neutron as a wave ...
  - Energy ($E$), velocity ($v$), wavenumber ($k$), wavelength ($\lambda$)

\[
k = \frac{m_n v}{h} = \frac{2\pi}{\lambda}
\]

\[
E = \frac{\hbar^2 k^2}{2m_n} = \frac{\hbar^2}{2m_n} \left( \frac{2\pi}{\lambda} \right)^2 = \frac{81.81\text{meV} \cdot \text{Å}^2}{\lambda^2}
\]

\[
E = k_B T = \left( 0.08617\text{meV} \cdot \text{K}^{-1} \right)T
\]

$\lambda \sim$ interatomic spacing $\rightarrow$ $E \sim$ excitations in condensed matter

<table>
<thead>
<tr>
<th></th>
<th>Energy (meV)</th>
<th>Temperature (K)</th>
<th>Wavelength (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cold</td>
<td>0.1 – 10</td>
<td>1 – 120</td>
<td>4 – 30</td>
</tr>
<tr>
<td>Thermal</td>
<td>5 – 100</td>
<td>60 – 1000</td>
<td>1 – 4</td>
</tr>
<tr>
<td>Hot</td>
<td>100 – 500</td>
<td>1000 – 6000</td>
<td>0.4 – 1</td>
</tr>
</tbody>
</table>
The Basic Experiment:

Incident Beam:
- monochromatic
- “white”
- “pink”

Scattered Beam:
- Resolve its energy
- Don’t resolve its energy
- Filter its energy
Fermi’s Golden Rule within the 1st Born Approximation

\[ W = \frac{2\pi}{\hbar} |<f|V|i>|^2 \rho(E_f) \]

\[ \delta\sigma = \frac{W}{\Phi} = \left(\frac{m}{2\pi\hbar^2}\right)^2 \frac{k_f}{k_i} |<f|V|i>|^2 \delta\Omega \]

\[ \frac{\delta^2\sigma}{\delta\Omega\delta E_f} = \frac{k_f}{k_i} \sigma_{\text{coh}}/4\pi N S_{\text{coh}}(Q, \omega) \]

\[ + \frac{k_f}{k_i} \sigma_{\text{incoh}}/4\pi N S_{\text{incoh}}(Q, \omega) \]
Nuclear correlation functions

Pair correlation function

\[ G(r, t) = \frac{1}{N} \int \sum_{jj'} \delta(r' - R_{j'}(0)) \delta(r' + r - R_j(t)) dr' \]

Intermediate function

\[ I(Q, t) = \int G(r, t)e^{iQ \cdot r} dr = \frac{1}{N} \sum_{jj'} \exp(-iQ \cdot R_{j'}(0)) \exp(iQ \cdot R_j(t)) \]

Scattering function

\[ S(Q, \omega) = \frac{1}{2\pi\hbar} \int I(Q, t)e^{-i\omega t} dt \]

Differential scattering cross-section

\[ \frac{d^2\sigma}{d\Omega dE_f} = \frac{\sigma_{\text{scat}}}{4\pi} \frac{k_f}{k_i} NS(Q, \omega) \]
Nuclear (lattice) excitations

Neutron scattering measures simultaneously the wavevector and energy of collective excitations $\rightarrow$ dispersion relation, $\omega(q)$

In addition, local excitations can of course be observed

- Commonly studied excitations
  - Phonons
  - Librations and vibrations in molecules
  - Diffusion
  - Collective modes in glasses and liquids

- Excitations can tell us about
  - Interatomic potentials & bonding
  - Phase transitions & critical phenomena (soft modes)
  - Fluid dynamics
  - Momentum distributions & superfluids (eg. He)
  - Interactions (eg. electron-phonon coupling)
Atomic diffusion

For long times compared to the collision time, atom diffuses

\[ \langle r^2(t) \rangle \approx 6Dt \]

Auto-correlation function

\[ G_s(r, t) = \left\{ 6\pi \langle r^2(t) \rangle \right\}^{3/2} \exp\left( -\frac{r^2}{6\langle r^2(t) \rangle} \right) \]

\[ S(Q, \omega) = \frac{1}{\pi\hbar} \exp\left( \frac{\hbar\omega}{2k_B T} \right) \frac{DQ^2}{\omega^2 + (DQ^2)^2} \]

Molecular vibrations

- Large molecule, many normal modes
- Harmonic vibrations can determine interatomic potentials

Origin of reciprocal space;
Remains fixed for any sample rotation

Mapping Momentum – Energy (Q-E) space
Bragg diffraction:

Constructive Interference

\[ Q = \text{Reciprocal Lattice Vector} \]

Elastic scattering: \[ | k_i | = | k_f | \]
Bragg diffraction:

Constructive Interference

$Q = \text{Reciprocal Lattice Vector}$

Elastic scattering: $|k_i| = |k_f|$
Elementary Excitations in Solids

- Lattice Vibrations (Phonons)
- Spin Fluctuations (Magnons)

Energy vs Momentum

- Forces which bind atoms together in solids
Phonons

- Normal modes in periodic crystal $\rightarrow$ wavevector
  
  $$ \mathbf{u}(l,t) = \frac{1}{\sqrt{NM}} \sum_{j} \epsilon_j(q) \exp(iq \cdot l) \hat{B}(q_j, t) $$

- Energy of phonon depends on $q$ and polarization

FCC structure

Phonon intensities

\[ S_{1+}(Q, \omega) = \frac{1}{2NM} e^{-\frac{Q^2 \langle u^2 \rangle}{2}} \sum_{jq} \frac{Q \cdot \varepsilon_j(q)}{\omega_j(q)} \left( 1 + n(\omega) \right) \delta(Q - q - \tau) \delta(\omega - \omega_j(q)) \]

Structure (polarization) factor

More complicated structures


Acoustic phonon
Optical phonon

La$_2$CuO$_4$
Spin excitations

- **Spin excitations**
  - Spin waves in ordered magnets
  - Paramagnetic & quantum spin fluctuations
  - Crystal-field & spin-orbit excitations

- **Magnetic inelastic scattering can tell us about**
  - Exchange interactions
  - Single-ion and exchange anisotropy (determine Hamiltonian)
  - Phase transitions & critical phenomena
  - Quantum critical scaling of magnetic fluctuations
  - Other electronic energy scales (eg. CF & SO)
  - Interactions (eg. spin-phonon coupling)
Spin waves


Ferromagnetic

Antiferromagnetic


Scattering experiments

Instrument and sample (powder or single-crystal) determine how \((\mathbf{Q}, \omega)\) space is sampled

\[ \hbar \omega = \frac{\hbar^2}{2m} (k_i^2 - k_f^2) \]

\[ \mathbf{Q} = k_i - k_f \]
Bragg's Law: \( n\lambda = 2d \sin(\theta) \)
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Brockhouse’s Triple Axis Spectrometer

\[ |k_i| = 2 \frac{\pi}{\lambda_i} \]

\[ |k_f| = 2 \frac{\pi}{\lambda_f} \]
Momentum Transfer:

\[ Q = k_i - k_f \]

Energy Transfer:

\[ \delta E = \frac{h^2}{2m} (k_i^2 - k_f^2) \]
Two Axis Spectrometer:

- 3-axis with analyser removed
- Powder diffractometer
- Small angle diffractometer
- Reflectometers

Diffractometers often employ working assumption that all scattering is elastic.
Soller Slits: Collimators

Define beam direction to +/- 0.5, 0.75 etc. degrees
Filters:
Remove $\lambda/n$ from incident or scattered beam, or both
Single crystal monochromators:

Bragg reflection and harmonic contamination

\[ n\lambda = 2d \sin(\theta) \]

Get: \( \lambda, \lambda/2, \lambda/3, \text{etc.} \)
Pyrolitic graphite filter:

- $E = 14.7$ meV
- $\lambda = 2.37$ Å
- $v = 1.6$ km/s
- $2xv = 3.2$ km/s
- $3xv = 4.8$ km/s
Two different ways of performing constant-Q scans

Constant $k_f$  

Constant $k_i$
Origin of reciprocal space;
Remains fixed for any sample rotation

Mapping Momentum – Energy (Q-E) space
Elementary Excitations in Solids

- Lattice Vibrations (Phonons)
- Spin Fluctuations (Magnons)

Energy vs Momentum

- Forces which bind atoms together in solids
Constant $Q$, Constant $E$
3-axis technique allow us to
Put $Q$-Energy space on a grid,
And scan through as we wish

Map out elementary excitations
In $Q$-energy space (dispersion Surface)
Samples

- **Samples need to be BIG**
  - ~ gram or cc
  - Counting times are long (mins/pt)

- Sample rotation
- Sample tilt

Co-aligned CaFe$_2$As$_2$ crystals
Monochromators

- Selects the incident wavevector

\[ Q(hkl) = \frac{2\pi}{d(hkl)} = 2k_i \sin \theta \]

- Reflectivity
- focusing
- high-order contamination eg. \( \lambda/2 \) PG(004)

<table>
<thead>
<tr>
<th>Mono</th>
<th>d(hkl)</th>
<th>uses</th>
</tr>
</thead>
<tbody>
<tr>
<td>PG(002)</td>
<td>3.353</td>
<td>General</td>
</tr>
<tr>
<td>Be(002)</td>
<td>1.790</td>
<td>High ( k_i )</td>
</tr>
<tr>
<td>Si(111)</td>
<td>3.135</td>
<td>No ( \lambda/2 )</td>
</tr>
</tbody>
</table>
Detectors

- **Gas Detectors**
- \( n + ^3\text{He} \rightarrow ^3\text{H} + p + 0.764 \text{ MeV} \)
- Ionization of gas
- \( e^- \) drift to high voltage anode
- High efficiency

- **Beam monitors**
- Low efficiency detectors for measuring beam flux
Resolution

- **Resolution ellipsoid**
  - Beam divergences
  - Collimations/distances
  - Crystal mosaics/sizes/angles

- **Resolution convolutions**

\[ I(Q_0, \omega_0) = \int S(Q_0, \omega_0) R(Q - Q_0, \omega - \omega_0) \, dQ \, d\omega \]
Resolution focusing

- Optimizing peak intensity
- Match slope of resolution to dispersion
Neutrons have mass so higher energy means faster – lower energy means slower.

\[ v \text{ (km/sec)} = \frac{3.96}{\lambda \text{ (A)}} \]

- 4 A neutrons move at ~ 1 km/sec
- DCS: 4 m from sample to detector
- It takes 4 msec for elastically scattered 4 A neutrons to travel 4 m
- msec timing of neutrons is easy
- \( \delta E / E \sim 1-3\% \) - very good!

We can measure a neutron’s energy, wavelength by measuring its speed.
Time-of-flight methods

- Effectively utilizes time structure of pulsed neutron groups

\[ t = \frac{d}{v} = \left( \frac{m}{\hbar} \right) \lambda \]

Spallation neutron source

Pharos – Lujan Center

velocity selector

detector banks

sample

Scattered neutrons

NXS School

Detector

Sample

Fermi chopper

Distance

Time
A single (disk) chopper pulses the neutron beam.

A second chopper selects neutrons within a narrow range of speeds.

Counter-rotating choppers (close together), with speed $\omega$, behave like single choppers with speed $2\omega$. They can also permit a choice of pulse widths.

Additional choppers remove "contaminant" wavelengths and reduce the pulse frequency at the sample position.
The DCS has seven choppers, 4 of which have 3 "slots"
Fermi Choppers

- Body radius ~ 5 cm
- Curved absorbing slats
  - B or Gd coated
  - ~mm slit size
- $f = 600$ Hz (max)
- Acts like shutter, $\Delta t \sim \mu s$

Figure 1. ISIS MAPS chopper and slit package assembly – exploded view
T-zero chopper

- Background suppression
- Blocks fast neutron flash
Position sensitive detectors

- $^3$He tubes (usu. 1 meter)
- Charge division
- Position resolution ~ cm
- Time resolution ~ 10 ns

MAPS detector bank
Sample environment

- Temperature, field, pressure
- Heavy duty for large sample environment
  - CCR
  - He cryostats
  - SC magnets
  - ...
- Can be machined from Al
  ~ neutron transparent
  relatively easy to work with
Guides

- Transport beam over long distances
- Background reduction
- Total external reflection
  - Ni coated glass
  - Ni/Ti multilayers (supermirror)
Size matters

- **Length = resolution**
  - Instruments ~ 20 – 40 m long
  - E-resolution ~ 2-4% $E_i$

- **More detectors**
  - SEQUOIA – 1600 tubes, 144000 pixels
  - Solid angle coverage 1.6 steradians

- **Huge data sets**
  - 0.1 – 1 GB
Kinematic limitations

• Many combinations of $k_i, k_f$ for same $Q, \omega$
  – Only certain configurations are used (e.g. $E_f$-fixed)

• Cannot “close triangle” for certain $Q, \omega$
  due to kinematics

Minimum accessible $Q$
Data visualization

- Large, complex data from spallation sources
- Measure $S(Q, \omega) – 4D$ function


La$_{1-x}$Ca$_x$MnO$_3$
Field-induced order in the Pyrochlore Yb$_2$Ti$_2$O$_7$:

Weak magnetic field // [110] induces LRO

**appearance of long-lived spin waves at low $T$ and moderate $H$**
References

**General neutron scattering**

**Polarized neutron scattering**

**Triple-axis techniques**

**Time-of-flight techniques**