A Survey of Inelastic Neutron Scattering

- Properties of the neutron
- The neutron scattering cross section
- The triple axis spectrometer
- Phonons
- Time-of-flight spectrometry
- Experimental details
Neutrons:
- no charge
- spin = 1/2
- massive:
  - $mc^2 \approx 1\text{GeV}$

$$^{235}\text{U} + n \rightarrow \text{daughter nuclei} + 2\text{--}3\ n + \gamma s$$
The Neutron as a Wave

Energy, wave vector, wavelength, velocity:

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

\[ E = k_B T = 0.08617 \text{meV} \cdot K^{-1} \times T \]

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

Neutrons with \( \lambda \) typical of interatomic spacings (~ 2 Å) have energies typical of elementary excitations in solids (~ 20 meV)
What are we typically trying to understand?

• What is the atomic and magnetic structure of new materials?
• What are the dynamic properties of the atoms and the magnetic moments?
• How are structure and dynamics related to physical properties?

Bragg’s law: \( n\lambda = 2d \sin(\theta) \)
The Basic Neutron Scattering Experiment

- Incident Beam
  - Monochromatic
  - “White”
  - “Pink”
- Scattered Beam
  - Resolve its energy
  - Don’t resolve its energy
  - Filter its energy

\( \theta, \phi \)
Fermi’s Golden Rule within the 1st Born approximation

\[ W = \frac{2\pi}{\hbar} \left| \langle f | V | i \rangle \right|^2 \rho(E_f) \]

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

\[ \frac{\partial^2 \sigma}{\partial \Omega \partial E_f} = \frac{k_f}{k_i} \frac{\sigma_{\text{coherent}}}{4\pi} N S_{\text{coherent}}(\mathbf{Q}, \hbar \omega) \]
Correlation Functions

Pair correlation function

\[ G(\vec{r}, t) = \frac{1}{N} \int \sum_{j, j'} \delta(\vec{r}' - \vec{R}_j(0)) \delta(\vec{r}' + \vec{r} - \vec{R}_j(t)) \, d\vec{r}' \]

Intermediate function

\[ I(\vec{Q}, t) = \int G(\vec{r}, t) \, e^{i\vec{Q} \cdot \vec{r}} \, d\vec{r} = \frac{1}{N} \sum_{j, j'} e^{-i\vec{Q} \cdot \vec{R}_j(0)} \, e^{i\vec{Q} \cdot \vec{R}_j(t)} \]

Scattering function

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

Pair correlation function

\[ G(\mathbf{r}, t) = \frac{1}{N} \int \sum_{j, j'} \delta(\mathbf{r}' - R_j'(0)) \delta(\mathbf{r}' + \mathbf{r} - \mathbf{R}_j(t)) \, d\mathbf{r}' \]

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Scattering function

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

Neutrons “see” nuclei and magnetism

X-rays - electromagnetic radiation “see” electrons
Dipole moment of the neutron interacts with the magnetic field generated by the electron

\[ \mu_n = -\gamma \mu_N \sigma \]

\( \gamma = 1.913 \)  
\( \text{nuclear magneton} = e \frac{\hbar}{2m_n} \)  
Pauli spin operator

\[ V_M = -\mu_n \cdot B \]

Dipole field due to orbital currents  
Dipole field due to Spin of the electron(s)
Diffraction in Momentum (Q) space

In momentum space, our sample is represented by its reciprocal lattice.

\[ \frac{2\pi}{a} \]
Diffraction in Momentum (Q) space

Origin of reciprocal space

Remains fixed for all sample orientations

\[ \frac{2\pi}{a} \]
Diffractection in Momentum $\mathcal{Q}$ space

Bragg diffraction

constructive interference when

$$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f = \mathbf{\tau}$$

= a reciprocal lattice vector

$$\mathbf{Q} = \frac{2\pi}{a}$$
Diffraction in Momentum (Q) space

Bragg diffraction: Constructive interference when

\[ \vec{Q} = \vec{k}_i - \vec{k}_f = \vec{\tau} \]

= a reciprocal lattice vector

\( \frac{2\pi}{a} \)
Elementary Excitations

Momentum $Q = k_i - k_f$

Energy $\hbar \omega$

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

$Q = k_i - k_f$
Phonon Polarizations

Longitudinal Acoustic

Transverse Acoustic

Transverse Acoustic

Transverse Optic
Phonon eigenvectors and eigenvalues

\[ \hbar \omega \]

\[ \text{Momentum } Q = k_i - k_f \]
Phonons in 3D


FCC Brillouin zone
Phonons in more complicated 3D structures

**KBr** - two atoms/unit cell
- 3 acoustic phonon branches
- 3 optic phonon branches

**La$_2$CuO$_4$**
- many atoms/unit cell
- 3 acoustic phonon branches
- $3n-3 = $ many optic phonon branches


The High Flux Reactor at the ILL and its moderators and beam ports.
Betram N. Brockhouse, McMaster University, Hamilton, Ontario, Canada, receives one half of the 1994 Nobel Prize in Physics for the development of neutron spectroscopy.
Brockhouse’s Triple Axis Spectrometer

\[ n\lambda = 2d \sin \theta \]
Brockhouse’s Triple Axis Spectrometer

$$| \vec{k}_i | = \frac{2\pi}{\lambda_i}$$

$$| \vec{k}_f | = \frac{2\pi}{\lambda_f}$$
Brockhouse's Triple Axis Spectrometer

\[ \vec{Q} = \vec{k}_i - \vec{k}_f \]

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

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

\[ \hbar \omega = \frac{\hbar^2 k_i^2}{2m} - \frac{\hbar^2 k_f^2}{2m} \]
Two different ways of performing constant-$Q$ scans

\[ Q = k_i - k_f \]

\[ Q = \text{Constant } k_f \quad \text{and} \quad Q = \text{Constant } k_i \]
Mapping Momentum ($Q$) and Energy ($\hbar \omega$) space

Origin of reciprocal space

Remains fixed for all sample orientations
Putting the Q-map of the scattering with the reciprocal lattice of the crystal
Putting the Q-map of the scattering with the reciprocal lattice of the crystal
Constant-Q triple axis data


Constant Q, constant E triple axis techniques allow us to put Q and E on a grid, and scan through as we choose.
Constant-Q, constant E triple axis techniques allow us to put Q and E on a grid, and scan through as we choose.
Elastic scattering with a Triple Axis Spectrometer

\[ |k_f| = |k_i| = \frac{2\pi}{\lambda_i} \]
The assumption is often made that the scattering is elastic - but, *this is an assumption*!

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

Two Axis “Spectrometer” integrates over \( k_f \) : diffraction.
\[
S(\vec{Q}, \hbar \omega) = \frac{1}{2NM} e^{-Q^2 \langle u^2 \rangle} \sum_{j, q} \left| \vec{Q} \cdot \vec{\varepsilon}_{j}(\vec{q}) \right|^2 \frac{1}{\omega_{j}(\vec{q})}
\]

The displacement (eigenvectors) of the atoms must be parallel to the momentum transfer.

\[
\times (1 + n(\hbar \omega)) \delta(\vec{Q} - \vec{q} - \vec{\tau}) \delta(\hbar \omega - \hbar \omega_{j}(\vec{q}))
\]

The neutron can always create a phonon, but it cannot destroy a phonon unless one is already present.

Momentum must be conserved
Energy must be conserved
The coherent neutron scattering cross section for phonons

\[
S(Q, \hbar \omega) = \frac{1}{2NM} e^{-Q^2 \langle u^2 \rangle} \sum_{j, q} |Q \cdot \varepsilon_j(q)|^2 \frac{1}{\omega_j(q)} \times (1 + n(\hbar \omega)) \delta(Q - q - \tau) \delta(\hbar \omega - \hbar \omega_j(q))
\]
The coherent neutron scattering cross section for phonons

\[ S(\vec{Q}, \hbar \omega) = \frac{1}{2NM} e^{-Q^2 \langle u^2 \rangle} \sum_{j, \vec{q}} |\vec{Q} \cdot \vec{\epsilon}_j(\vec{q})|^2 \frac{1}{\omega_j(\vec{q})} \]
Neutrons have mass so higher energy means faster – lower energy means slower

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

- 4 Å neutrons move at ~ 1 km/sec
- DCS: 4 m from sample to detector
- It takes 4 msec for elastically scattered 4 Å 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 Neutron Scattering

\[ t = \frac{d}{v} = \left( \frac{md}{h} \right) \lambda \]

Detector
Sample
Fermi chopper

velocity selector

detector banks

sample
Scattered neutrons
Time-of-flight Neutron Scattering

- Optical Filter
- Crystal Filter
- White Beam Monitor
- Shutter
- Monochromatic Beam Monitor
- Neutron Guide
- Sample Chamber
- Radial Collimator
- Sample
- Transmitted Beam Monitor
- Argon gas-filled Flight Chamber
- Frame Overlap Chopper
- Order Removal Choppers
- Pulsing Choppers
- Monochromating Choppers
- 913 $^3$He detectors, 400x31x11 mm$^3$
4D data sets for single crystals can be very large ~ 2 Tbyte
Time-of-flight Neutron Scattering: Disc Choppers

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”

Monochromating Choppers
Order Removal Choppers
Frame Removal Chopper
Pulsing Choppers

Disk 4B
Time-of-flight Neutron Scattering: Fermi Choppers
Resolution Considerations

Resolution “ellipse” is defined by:

- Beam divergences
- Collimation and distances
- Crystal mosaic, sizes
- Beam energy

\[
I(\vec{Q}_0, \hbar \omega_0) = \int S(\vec{Q}_0 - \vec{Q}, \hbar \omega_0 - \hbar \omega) R(\vec{Q}_0, \hbar \omega_0) \, d\vec{Q} \, d\hbar \omega
\]
Resolution focussing
Resolution focussing
Resolution focussing
Resolution focussing
Neutron Detectors

Gas Detectors

- $n + ^3\text{He} \rightarrow ^3\text{H} + p + 0.764 \text{ MeV}$
- Ionization of gas
- High efficiency

Beam monitors

- Low efficiency detectors for monitoring beam flux
Q or angular resolution improved by using collimation (Soller slits)

Soller slit collimators
neutron channels
with absorbing walls

Allows the angular resolution of $k_i$, $k_f$ to be selected
Harmonic contamination from crystal monochromators

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

\[ \lambda, \frac{\lambda}{2}, \text{and} \frac{\lambda}{3} \] appear at the same \( \theta \) with different \( n \)
Neutron filters remove $\lambda/n$ from incident or scattered beam, or both.

$n\lambda = 2d \sin \theta$
Harmonic contamination from crystal monochromators: Pyrolitic Graphite

\[ E = 14.7 \text{ meV} \]
\[ \lambda = 2.37 \text{ Å} \]
\[ v = 1.6 \text{ km/s} \]
\[ 2 \times v = 3.2 \text{ km/s} \]
\[ 3 \times v = 4.8 \text{ km/s} \]