Basic Elements of Neutron Inelastic Scattering

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Outline

1. Introduction
   - Motivation
   - Types of Scattering

2. The Neutron
   - Production and Moderation
   - Wave/Particle Duality

3. Basic Elements of Neutron Scattering
   - The Scattering Length, \( b \)
   - Scattering Cross Sections
   - Pair Correlation Functions
   - Coherent and Incoherent Scattering
   - Neutron Scattering Methods

4. Summary of Scattering Cross Sections
   - Elastic (Bragg versus Diffuse)
   - Quasielastic (Diffusion)
   - Inelastic (Phonons)
The most important property of any material is its underlying atomic / molecular structure (structure dictates function).

In addition the motions of the constituent atoms (dynamics) are extremely important because they provide information about the interatomic potentials.

An ideal method of characterization would be one that can provide detailed information about both structure and dynamics.
We see something when light scatters from it. Thus scattering conveys information!

Light is composed of electromagnetic waves. \( \lambda \sim 4000 \text{ A} - 7000 \text{ A} \)

However, the details of what we see are ultimately limited by the wavelength.
The tracks of a compact disk act as a diffraction grating, producing a separation of the colors of white light.

From this one can determine the nominal distance between tracks on a CD, which is $1.6 \times 10^{-6}$ meters = 16,000 Angstroms.

To characterize materials we must determine the underlying structure. We do this by using the material as a diffraction grating.

**Problem**: Distances between atoms in materials are of order Angstroms $\rightarrow$ light is inadequate. Moreover, most materials are opaque to light.
To “see” atomic structure, we require a probe with a wavelength $\lambda \sim$ length scale of interest.

Some candidates …

- X-rays (EM - wave)
- Electrons (Charged particle)
- Neutrons (Neutral particle)
Which one should we choose?

If we wish only to determine relative atomic positions, then we should choose x-rays almost every time.

1. Relatively cheap

2. Sources are ubiquitous → easy access

3. Fluxes are extremely high → can study small samples

However …
X-rays are electromagnetic radiation. Thus they scatter from the atomic electrons.

**Consequences:**

- **Low-Z elements are hard to see.**
- **Elements with similar atomic numbers have very little contrast.**
- **X-rays are strongly attenuated as they pass through the walls of furnaces, cryostats, etc.**

### Types of Scattering

- **Hydrogen** (Z = 1)
- **Cobalt** (Z = 27)
- **Nickel** (Z = 28)
What about electrons?

Electrons are charged particles → they see both the atomic electrons and nuclear protons at the same time.

1. Relatively cheap

2. Sources are not uncommon → good access

3. Fluxes are extremely high → can study tiny crystals

4. Very small wavelengths → more information

However …
Requires very thin samples. Radiation damage is a concern. Magnetic structures are hard to determine because electrons are deflected by the internal magnetic fields.
What about neutrons?

<table>
<thead>
<tr>
<th>Advantages</th>
<th>Disadvantages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelengths easily varied to match atomic spacings</td>
<td>Neutrons are expensive to produce → access is not as easy</td>
</tr>
<tr>
<td>Zero charge → not strongly attenuated by furnaces, etc.</td>
<td>Interact weakly with matter → often require large samples</td>
</tr>
<tr>
<td>Magnetic dipole moment → can study magnetic structures</td>
<td>Available fluxes are low compared to those for x-rays</td>
</tr>
<tr>
<td>Nuclear interaction → can see low-Z elements easily like H → good for the</td>
<td></td>
</tr>
<tr>
<td>study of biomolecules and polymers.</td>
<td></td>
</tr>
<tr>
<td>Nuclear interaction is simple → scattering is easy to model</td>
<td>Low energies → Non-destructive probe</td>
</tr>
</tbody>
</table>
“If the neutron did not exist, it would need to be invented.”

Bertram Brockhouse
1994 Nobel Laureate in Physics
1926: de Broglie Relation

\[ \lambda = \frac{h}{p} = \frac{h}{m_n \nu} \]

<table>
<thead>
<tr>
<th>( \lambda )</th>
<th>( \nu )</th>
<th>( E )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Å</td>
<td>4000 m/s</td>
<td>82 meV</td>
</tr>
<tr>
<td>9 Å</td>
<td>440 m/s</td>
<td>1 meV</td>
</tr>
</tbody>
</table>

\( m_n = 1.675 \times 10^{-27} \text{ kg} \)
\( Q = 0 \)
\( S = \frac{1}{2} h \)
\( \mu_n = -1.913 \mu_N \)
Neutron Production

Neutrons not bound to a nucleus decay via the weak force at a rate characterized by a lifetime of ~ 888 seconds (15 minutes).

\[ n \rightarrow p + e^- + \nu_e \]

A useful source of neutrons requires a nuclear process by which bound neutrons can be freed from the nuclei of atoms and that is easily sustainable.

There are two such processes, spallation and fission …
Nuclear fission is used in power and research reactors.

The fission process and moderator are confined by a large containment vessel.

A liquid medium (D\textsubscript{2}O, or heavy water) is used to moderate the fast fission neutrons to room temperature (2 MeV $\rightarrow$ 50 meV).

\[ ^{235}_{92}U_{143} + n \rightarrow \left[ ^{236}_{92}U_{144} \right]^* \rightarrow X + Y + 2.44n \]
Maxwellian Distribution

\[ \Phi \sim v^3 e^{-mv^2/2k_B T} \]

“Fast” neutrons: \( v = 20,000 \text{ km/sec} \)

Neutron velocity \( v \) (km/sec)
Wave - Particle Duality

de Broglie Relation $\lambda = \frac{h}{m_n \nu}$

Fast Neutron,
$V \sim 20,000,000 \text{ m/sec}$
$\sim 0.00002 \text{ nm}$

Thermal Neutron,
$V \sim 2,000 \text{ m/sec}$
$\sim 0.2 \text{ nm}$

Cold Neutron,
$V \sim 200 \text{ m/sec}$
$\sim 2 \text{ nm}$
Neutron scattering experiments measure the flux \( \Phi_s \) of neutrons scattered by a sample into a detector as a function of the change in neutron wave vector (\( Q \)) and energy (\( \hbar \omega \)).

**Momentum**

\[
\hbar k = \hbar \left(2\pi/\lambda\right)
\]

\[
hQ = \hbar k_i - \hbar k_f
\]

**Energy**

\[
\hbar \omega_n = \hbar^2 k_n^2/2m
\]

\[
h\omega = h\omega_i - h\omega_f
\]

The expressions for the scattered neutron flux \( \Phi_s \) involve the positions and motions of atomic nuclei or unpaired electron spins.

\[
\Phi_s = f\{\vec{r}_i(t), \vec{r}_j(t), \vec{S}_i(t), \vec{S}_j(t)\}
\]

\( \Phi_s \) provides information about all of these quantities!
These “cross sections” are what we measure experimentally.

Consider an incident neutron beam with flux $\Phi_i$ (neutrons/sec/cm$^2$) and wave vector $k_i$ on a non-absorbing sample.

We define three cross sections:

- $\sigma$ = Total cross section
- $\frac{d\sigma}{d\Omega}$ = Differential cross section
- $\frac{d^2\sigma}{d\Omega dE_f}$ = Partial differential cross section
Neutron Scattering Cross Sections

What are the physical meanings of these three cross sections?

- $\sigma$: Total # of neutrons scattered per second / $\Phi_i$.

- $\frac{d\sigma}{d\Omega}$: Total # of neutrons scattered per second into $d\Omega$ / $d\Omega \Phi_i$. (Diffraction $\rightarrow$ structure.)

- $\frac{d^2\sigma}{d\Omega dE_f}$: Total # of neutrons scattered per second into $d\Omega$ with a final energy between $E_f$ and $dE_f$ / $d\Omega dE_f \Phi_i$. (Inelastic scattering $\rightarrow$ dynamics.)
Neutron Scattering Cross Sections

What are the relative sizes of the cross sections?

Clearly:

$$\sigma = \int \frac{d\sigma}{d\Omega} d\Omega = \int \frac{d^2\sigma}{d\Omega dE_f} d\Omega dE_f$$

Thus:

$$\sigma >> \frac{d\sigma}{d\Omega} >> \frac{d^2\sigma}{d\Omega dE_f}$$

Typically,

$$\frac{d\sigma}{d\Omega} \sim 10^6 \times \frac{d^2\sigma}{d\Omega dE_f}$$
Elastic Scattering:
- Change in neutron energy = 0
- Probes changes in momentum only

Inelastic Scattering:
- Change in neutron energy ≠ 0
- Probes changes in both momentum and energy

Note that both of these cases are described by...

\[
\frac{d^2\sigma}{d\Omega dE_f}
\]

Elastic (\(k_i=k_f\))

\[\sin\theta = (Q/2)/k\]
\[Q = 2k\sin\theta = 4\pi\sin\theta/\lambda\]

Inelastic (\(k_i\neq k_f\))

Energy loss (\(h\omega>0\))

Energy gain (\(h\omega<0\))
The incident neutron $\Psi_i$ is a plane wave:

$$\Psi_i(r) \sim e^{ikx} \quad (k = 2\pi/\lambda)$$

The scattered (final) neutron $\Psi$ is a spherical wave:

$$\Psi_f(r) \sim (-b/r)e^{ikr}$$

Consider the simplest case: A fixed, isolated nucleus.

1. The scattering is elastic ($k_i = k_f = k$). Why?

2. The scattering is isotropic. Why?

Scattering From a Single Nucleus

$\lambda \sim 2$ A

QUESTIONS:

1. The scattering is elastic ($k_i = k_f = k$). Why?

2. The scattering is isotropic. Why?

Nucleus

1 fm = $10^{-15}$ m
1. The scattering is elastic because the nucleus is fixed, so no energy can be transferred to it from the neutron (ignoring any excitations of the nucleus itself).

2. A basic result of diffraction theory states: if waves of any kind scatter from an object of a size $\ll \lambda$, then the scattered waves are spherically symmetric. (This is also known as s-wave scattering.)

\[ V(r) = \left( \frac{2\pi \hbar^2}{m_n} \right) \sum_{j=1}^{N} b_j \delta(r-r_j) \]

The details of $V(r)$ are unimportant – $V(r)$ can be described by a scalar parameter $b$ that depends only the choice of nucleus and isotope!

Fermi Pseudopotential
The parameter $b$ is known as the neutron “scattering length” and has units of length $\sim 10^{-12}$ cm.

It is independent of neutron energy.

It is (usually) a real number; imaginary for absorption ...

It corresponds to the “form factor” for x-ray scattering.

$b$ varies randomly with $Z$ and isotope $\rightarrow$ allows access to atoms that are usually unseen by x-rays.
We can easily calculate \( \sigma \) for a single, fixed nucleus:

**Def.** \[ \sigma \cdot \Phi_i = \text{Total number of neutrons scattered per second by the nucleus.} \]

\[ \nu = \text{Velocity of neutrons (elastic } \rightarrow \text{ same before and after scattering).} \]

\[ \nu \, dA \, |\Psi_f|^2 = \text{Total number of neutrons scattered per second through } dA. \]

\[ \int \nu \, dA \, |\Psi_f|^2 = \nu \int (r^2 d\Omega)(b/r)^2 = \nu \int b^2 d\Omega = \nu \, 4\pi b^2 = \sigma \cdot \Phi_i \]

Since \( \Phi_i = \nu \, |\Psi_i|^2 = \nu \rightarrow \sigma = 4\pi b^2 \)
Scattering From Many Nuclei

What if many atoms are present?

Scattering from one nucleus

Scattered neutron $\Psi_f$ is a spherical wave:

The incident neutron $\Psi_i$ is a plane wave:

Get strong scattering in some directions, but not in others. Angular dependence yields information about how the nuclei are arranged or correlated.
Scattering From Many Nuclei

(1) Born Approximation:
Assumes neutrons scatter only once (single scattering event).

(2) Superposition:
Amplitudes of scattered neutrons $\phi_n$ add linearly.

$$\Phi_s = \phi_1 + \phi_2 + \ldots$$

Intensity = $|\Phi|^2 = |\phi_1 + \phi_2 + \ldots|^2 = |\phi_1|^2 + |\phi_2|^2 + \ldots + \phi_1^*\phi_2^* + \phi_2^*\phi_1 + \ldots$

Depends on relative positions of 1 and 2 $\rightarrow$ pair correlations!

After Andrew Boothroyd
PSI Summer School 2007
The measured quantity $\Phi_s$ depends only on time-dependent correlations between the positions of pairs of atoms.

This is true because neutrons interact only weakly with matter. Thus only the lowest order term in the perturbation expansion contributes.

Differential Cross-Section

$$\frac{d\sigma}{d\Omega} = \sum_{i,j} b_i b_j e^{-i(k_i - k_j) \cdot (r_i - r_j)}$$

Depends only on: where the atoms are and what the atoms are.
**Pair Correlation Functions**

These formulae are stated without proof …

**Partial Differential Cross-Section**

\[
\left( \frac{d^2\sigma}{d\Omega dE} \right)_{k_0 \to k_1} = \frac{1}{N} \frac{k_f}{k_i} \left( \frac{m}{2\pi \hbar^2} \right)^2 \sum p_i p_f \sum |<k_f|V|k_i>|^2 \delta(E+E_i-E_f)
\]

\[
\left( \frac{d^2\sigma}{d\Omega dE} \right)_{k_0 \to k_1} = N \frac{k_f}{k_i} b^2 S(Q, \omega)
\]

**Neutron Structure Factor**

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

**Pair Correlation Function**

\[
G(r, t) = \left( \frac{1}{2\pi} \right)^3 \frac{1}{N} \int \sum_{jj'} e^{iQ \cdot r} <e^{-iQ \cdot r_j(0)} e^{iQ \cdot r_j(t)}> dQ
\]

Squires, Introduction to the theory of thermal neutron scattering (1996)
The scattered neutron flux $\Phi_s(Q,\hbar\omega)$ is proportional to the space ($\vec{r}$) and time ($t$) Fourier transform of the probability $G(\vec{r},t)$ of finding an atom at $(\vec{r},t)$ given that there is another atom at $r = 0$ at time $t = 0$.

$$\Phi_s \propto \frac{\partial^2 \sigma}{\partial \Omega \partial \omega} \propto \int \int e^{i(Q\cdot\vec{r} - \omega t)} G(\vec{r},t) d^3\vec{r} dt$$

**KEY IDEA** – Neutron interactions are weak → Scattering only probes two-particle correlations in space and time, but does so simultaneously!
Neutron Coherent and Incoherent Scattering

What happens when there are 2 or more different nuclei in the sample, each having a different value of \( b \), and these nuclei are randomly distributed?

This situation could arise for two reasons.
1. Isotopic incoherence
2. Nuclear spin incoherence

Both reasons can occur because the scattering interaction is nuclear.

Recall that

\[
\left( \frac{d^2 \sigma}{d\Omega dE} \right)_{k_0 \rightarrow k_1} = N \frac{k_f}{k_i} b^2 S(Q, \omega)
\]

Then the above equation must be generalized:

\[
\frac{d^2 \sigma}{d\Omega dE} = \sum_{i,j} b_i b_j S_{ij}(Q, \omega)
\]

\( b_i b_j = \langle b \rangle^2 \), for \( i=j \)

\( b_i b_j = b^2 \), for \( i \neq j \)

= average
Our partial differential cross section can then be recast into the form:

\[
\frac{d^2\sigma}{d\Omega dE_f} = \sigma_c S_c(Q, \omega) + \sigma_i S_i(Q, \omega),
\]

where

\[\sigma = \sigma_c + \sigma_i,\]

\[\sigma_c = 4\pi (b)^2\quad \text{c = coherent}\]

\[\sigma_i = 4\pi \{b^2 - (b)^2\}\quad \text{i = incoherent}\]
Consider a system composed of two different scattering lengths, $b_1$ and $b_2$.

The two isotopes are randomly distributed.

$$\frac{1}{2}(b_1 + b_2) = \bar{b}$$

Deviations $\delta b$
<table>
<thead>
<tr>
<th>Neutron Coherent and Incoherent Scattering</th>
</tr>
</thead>
<tbody>
<tr>
<td>What do these expressions mean physically?</td>
</tr>
<tr>
<td><strong>Coherent Scattering</strong></td>
</tr>
<tr>
<td>Measures the Fourier transform of the <em>pair</em> correlation function $G(r,t) \rightarrow$ interference effects.</td>
</tr>
<tr>
<td>This cross section reflects collective phenomena such as:</td>
</tr>
<tr>
<td>Bragg Peaks</td>
</tr>
<tr>
<td>Phonons</td>
</tr>
<tr>
<td>Spin Waves</td>
</tr>
<tr>
<td><strong>Incoherent Scattering</strong></td>
</tr>
<tr>
<td>Measures the Fourier transform of the <em>self</em> correlation function $G_s(r,t) \rightarrow$ no interference effects.</td>
</tr>
<tr>
<td>This cross section reflects single-particle scattering:</td>
</tr>
<tr>
<td>Atomic Diffusion</td>
</tr>
<tr>
<td>Vibrational Density of States</td>
</tr>
</tbody>
</table>
Summary of Cross Sections

\[
\frac{d\sigma}{d\Omega} \bigg|_{\text{coh}} = \frac{\sigma_{\text{coh}}}{4\pi} S(Q)
\]

\[
\frac{d\sigma}{d\Omega} \bigg|_{\text{inc}} = \frac{\sigma_{\text{inc}}}{4\pi}
\]

\[
\frac{d^2\sigma}{d\Omega dE_f} \bigg|_{\text{coh}} = \frac{k_f}{k_i} \frac{\sigma_{\text{coh}}}{4\pi} S_{\text{coh}}(Q,\omega)
\]

\[
\frac{d^2\sigma}{d\Omega dE_f} \bigg|_{\text{inc}} = \frac{k_f}{k_i} \frac{\sigma_{\text{inc}}}{4\pi} S_{\text{inc}}(Q,\omega)
\]
Elements of all scattering experiments

Basics of Neutron Scattering Methods

A source

A method to specify the incident wave vector $\mathbf{k}_i$

A well chosen sample + good idea

Detector(s)

A method to specify the final wave vector $\mathbf{k}_f$
1. Bragg Diffraction

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

2. Time-of-Flight (TOF)

\[ v = \frac{L}{t} \]

3. Larmor Precession
Different Types of Neutron Instruments

You will see many of these on the tour…

Thermal Neutrons

Cold Neutrons
Because neutron scattering is an intensity-limited technique. Thus detector coverage and resolution MUST be tailored to the science.

Uncertainties in the neutron wavelength and direction imply $Q$ and $\hbar\omega$ can only be defined with a finite precision.

The total signal in a scattering experiment is proportional to the resolution volume → better resolution leads to lower count rates! Choose carefully …

Courtesy of R. Pynn
The “right” resolution depends on what you want to study.
Instrumental $h\omega$-Resolution

Example – CuGeO$_3$

$\vec{Q} = (0, K, 0.5)$

$Focusing Analyzer$

Flat

5 Blades

9 Blades
**A Quick Review ...**

1. Neutrons scattering probes **two-particle** correlations in both space and time (simultaneously!).

2. The neutron scattering length, \( b \), varies randomly with \( Z \) allows access to atoms that are usually unseen by x-rays.

3. **Coherent Scattering**
   
   Measures the Fourier transform of the pair correlation function \( G(r,t) \) → interference effects.
   
   This cross section reflects **collective** phenomena.

4. **Incoherent Scattering**
   
   Measures the Fourier transform of the self correlation function \( G_s(r,t) \) → **no interference effects**.
   
   This cross section reflects **single-particle** scattering.

Please try to remember these things ...
Some Examples

OK, after all of this, just exactly what can neutron scattering do for you?

Let’s look at a few examples …
Neutron Elastic Scattering

- Neutrons can probe length scales ranging from ~0.1 Å to ~1000 Å

- Used to determine structure of 123 high-T\textsubscript{c} cuprates as x-rays weren’t sufficiently sensitive to the oxygen atoms.


Neutron Inelastic Scattering

Probes the vibrational, magnetic, and lattice excitations (dynamics) of materials by measuring changes in the neutron momentum and energy simultaneously.

$h\omega = 0$

Neutrons can probe time scales ranging from $\sim 10^{-14}$ s to $\sim 10^{-8}$ s.

Do you see a pattern here?

Larger “objects” tend to exhibit slower motions.
Can one measure elastic scattering from a liquid?

Hint: What is the correlation of one atom in a liquid with another after a time $t$?
Nobel Prize in Physics 1994

The Fathers of Neutron Scattering

“For pioneering contributions to the development of neutron scattering techniques for studies of condensed matter”

“For the development of the neutron diffraction technique”

“For the development of neutron spectroscopy”

Clifford G Shull
MIT, USA
(1915 – 2001)

Ernest O Wollan
ORNL, USA
(1910 – 1984)

Bertram N Brockhouse
McMaster University, Canada
(1918 – 2003)

Showed us where the atoms are …

Did first neutron diffraction expts …

Showed us how the atoms move …
http://www.mrl.ucsb.edu/~pynn/primer.pdf

“Introduction to the Theory of Thermal Neutron Scattering”
- G. L. Squires, Cambridge University Press

“Theory of Neutron Scattering from Condensed Matter”
- S. W. Lovesey, Oxford University Press

“Neutron Diffraction” (Out of print)
- G. E. Bacon, Clarendon Press, Oxford

“Structure and Dynamics”
- M. T. Dove, Oxford University Press

“Elementary Scattering Theory”
- D. S. Sivia, Oxford University Press
There are two primary methods of measuring the neutron scattering cross section $S(Q, \omega)$.

- **Constant-E scans:** vary $Q$ at fixed $h\omega$.
- **Constant-Q scans:** vary $h\omega$ at fixed $Q$. 

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**Neutron Inelastic Scattering**

**Neutron Measurements of Phonon Dispersions.**

$Q = (0,0,\zeta)$ (r.l.u.)