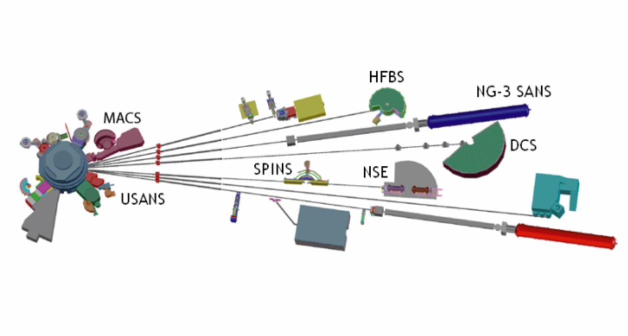

2012 NCNR Summer School on Fundamentals of Neutron Scattering

Basic Elements of Neutron Inelastic Scattering

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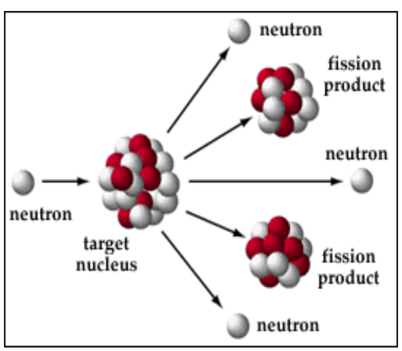
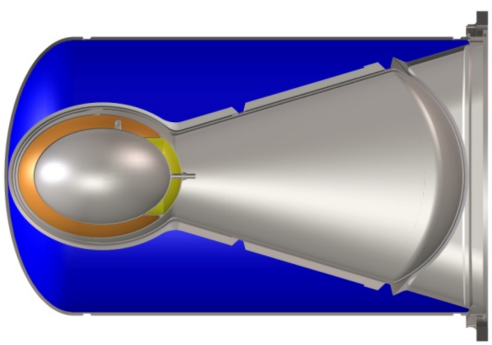
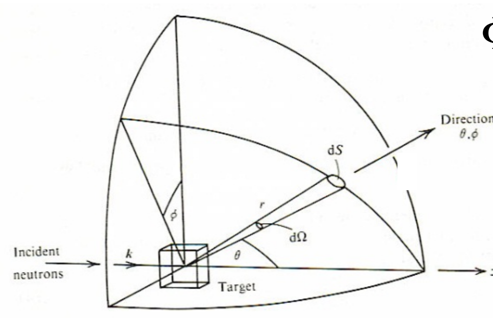
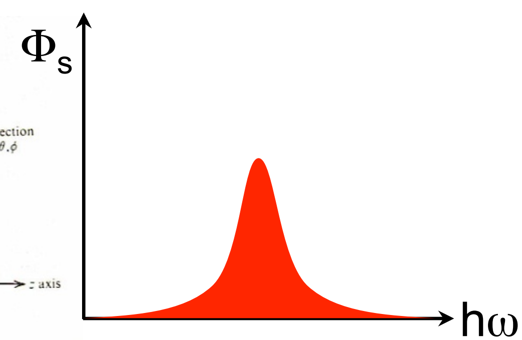
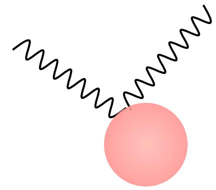




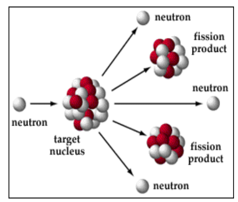




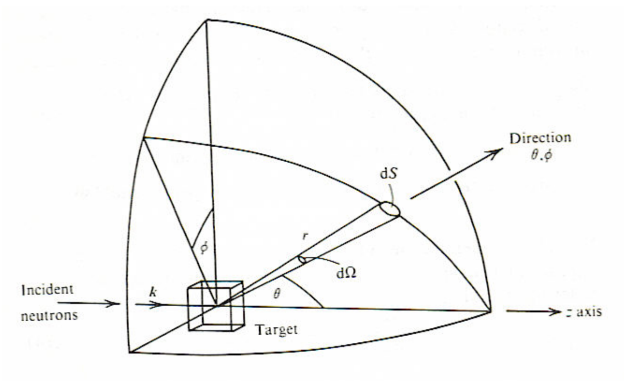
Outline



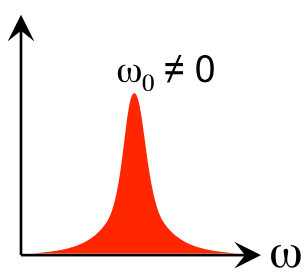
1. Introduction
   - Motivation
   - Types of Scattering

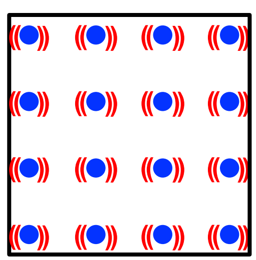


2. The Neutron
   - Production and Moderation
   - Wave/Particle Duality



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



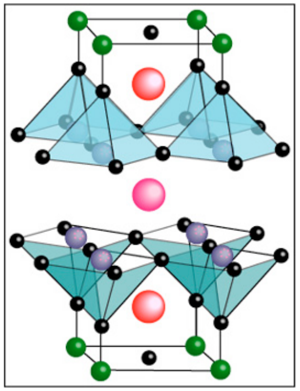


4. Summary of Scattering Cross Sections
   - Elastic (Bragg versus Diffuse)
   - Quasielastic (Diffusion)
   - Inelastic (Phonons)

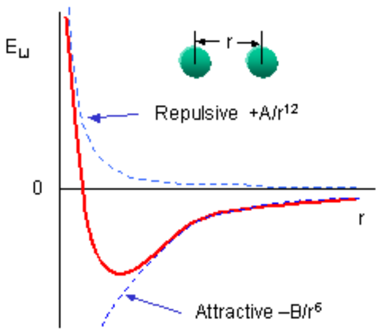
Motivation

Structure and Dynamics

The most important property of any material is its underlying atomic / molecular structure (structure dictates function).



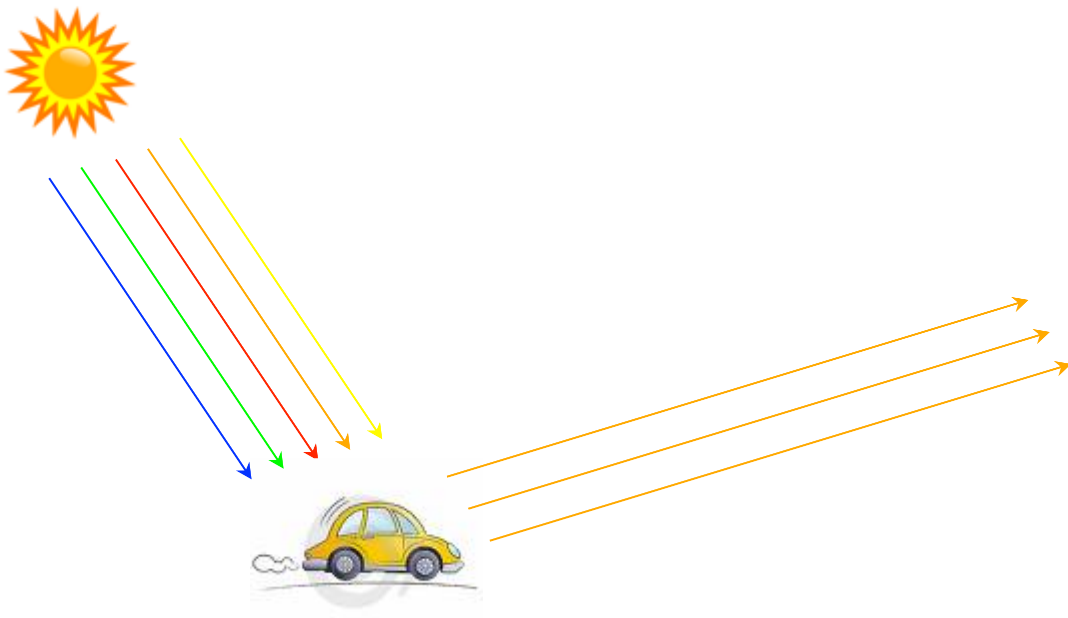
$Bi_2Sr_2CaCu_2O_{8+\delta}$



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.

Types of Scattering



How do we “see”?

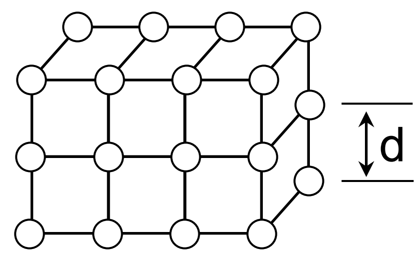
- We see something when light scatters from it. → Thus scattering conveys information!
- Light is composed of electromagnetic waves. → $\lambda \sim 4000$ A – 7000 A
- However, the details of what we see are ultimately limited by the wavelength.

Types of Scattering

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 1.6×10^{-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.



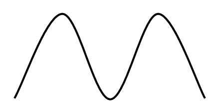
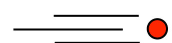
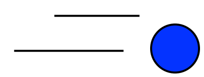
Problem: Distances between atoms in materials are of order Angstroms → light is inadequate. Moreover, most materials are opaque to light.

$\lambda_{Light} >> d \sim 4$ Å

Types of Scattering

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

Types of Scattering

Which one should we choose?

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

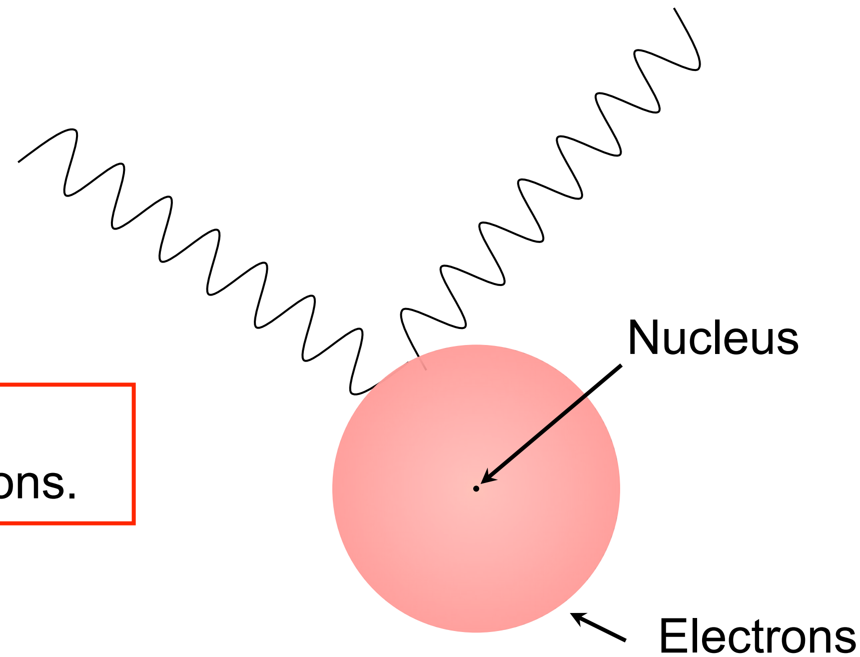
1. Relatively cheap

2. Sources are ubiquitous → easy access

3. Fluxes are extremely high → can study small samples

However …

Types of Scattering

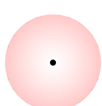


X-rays are electromagnetic radiation.
Thus they scatter from the atomic electrons.

Consequences:

Low-Z elements are hard to see.

Hydrogen



(Z = 1)

Elements with similar atomic numbers have very little contrast.

Cobalt   Nickel

(Z = 27)   (Z = 28)

X-rays are strongly attenuated as they pass through the walls of furnaces, cryostats, etc.

Types of Scattering

What about electrons?

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

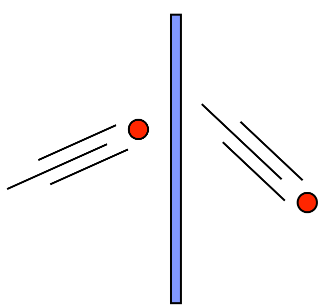
1. Relatively cheap
2. Sources are not uncommon → good access
3. Fluxes are extremely high → can study tiny crystals
4. Very small wavelengths → more information

However …

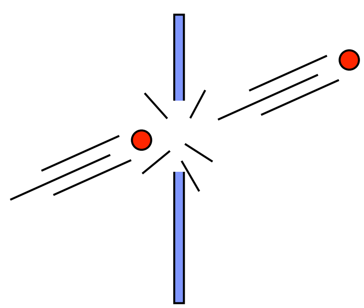
Types of Scattering

Electrons have some deficiencies too ...

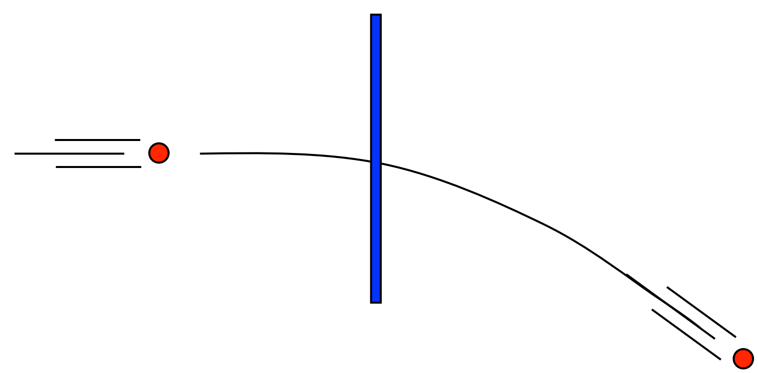
Requires very thin samples.



Radiation damage is a concern.



Magnetic structures are hard to determine because electrons are deflected by the internal magnetic fields.



Types of Scattering

What about neutrons?

Advantages

Wavelengths easily varied to match atomic spacings

Zero charge → not strongly attenuated by furnaces, etc.

Magnetic dipole moment → can study magnetic structures

Nuclear interaction → can see low-Z elements easily like H → good for the study of biomolecules and polymers.

Nuclear interaction is simple → scattering is easy to model

Low energies → Non-destructive probe

Disadvantages

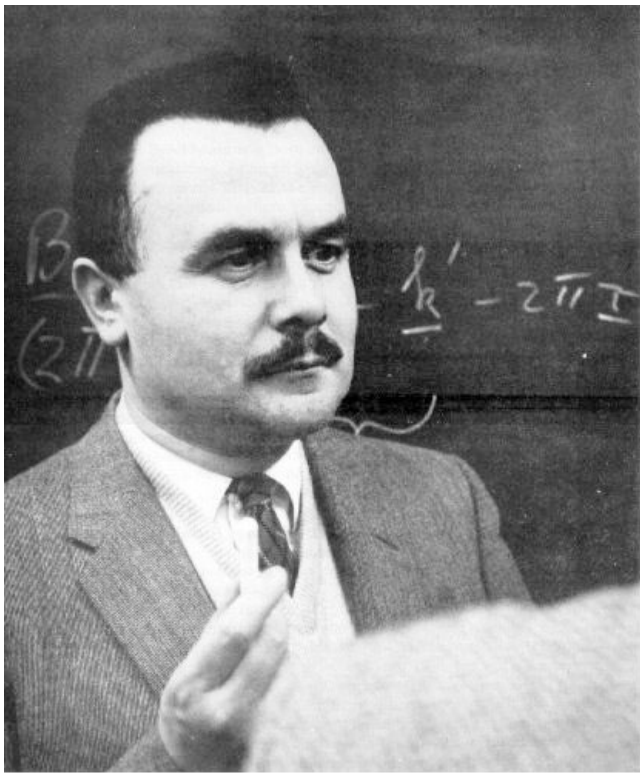
Neutrons are expensive to produce → access is not as easy

Interact weakly with matter → often require large samples

Available fluxes are low compared to those for x-rays

Let's consider neutrons …

The Neutron



"If the neutron did not exist, it would need to be invented."

Bertram Brockhouse
1994 Nobel Laureate in Physics

The Neutron

$m_n = 1.675 \times 10^{-27}$ kg
$Q = 0$
$S = ½\, h$
$\mu_n = -1.913\ \mu_N$

1926: de Broglie Relation

$$\lambda = h/p = h/m_n v$$

$\lambda = 1$ Å
$v = 4000$ m/s
$E = 82$ meV

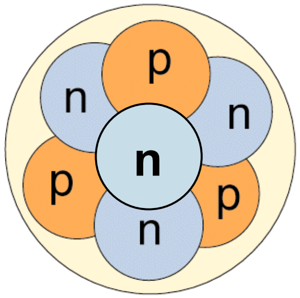
$\lambda = 9$ Å
$v = 440$ m/s
$E = 1$ meV

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).

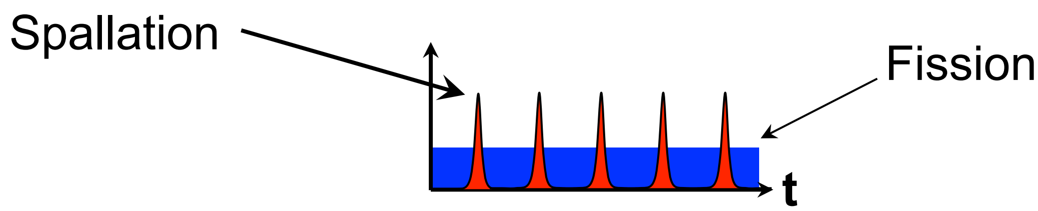
$$n \rightarrow p + e^- + \nu_e$$

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.



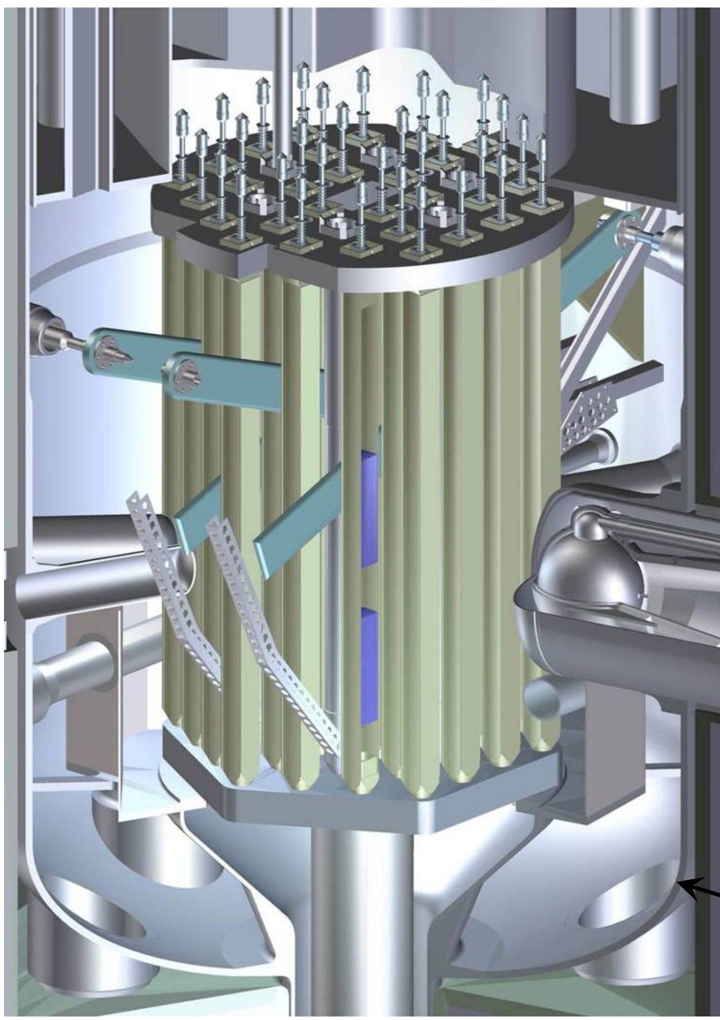
There are two such processes, spallation and fission …

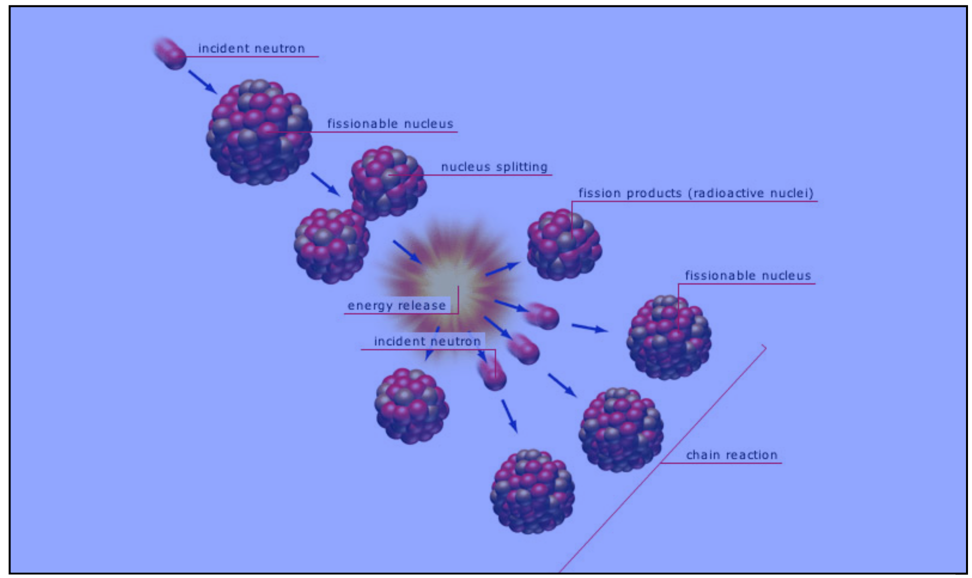
Neutron Production by Fission



Nuclear fission is used in power and research reactors.

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





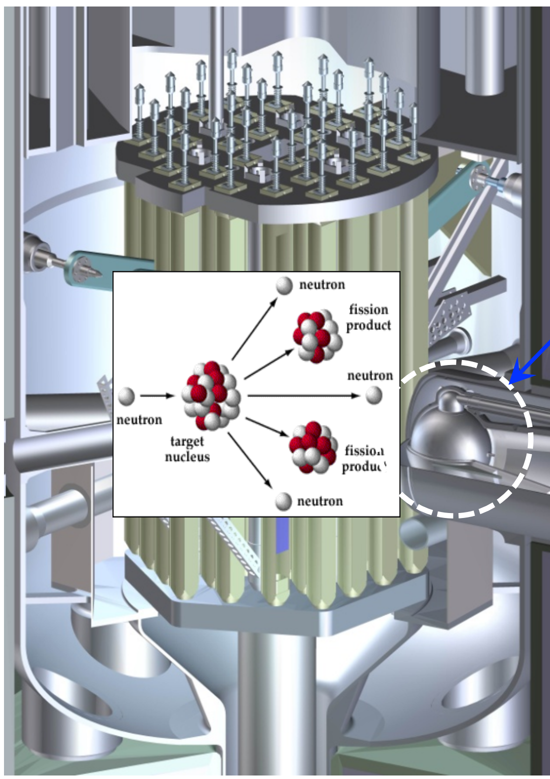
A liquid medium (D_2O, or heavy water) is used to moderate the fast fission neutrons to room temperature (2 MeV → 50 meV).

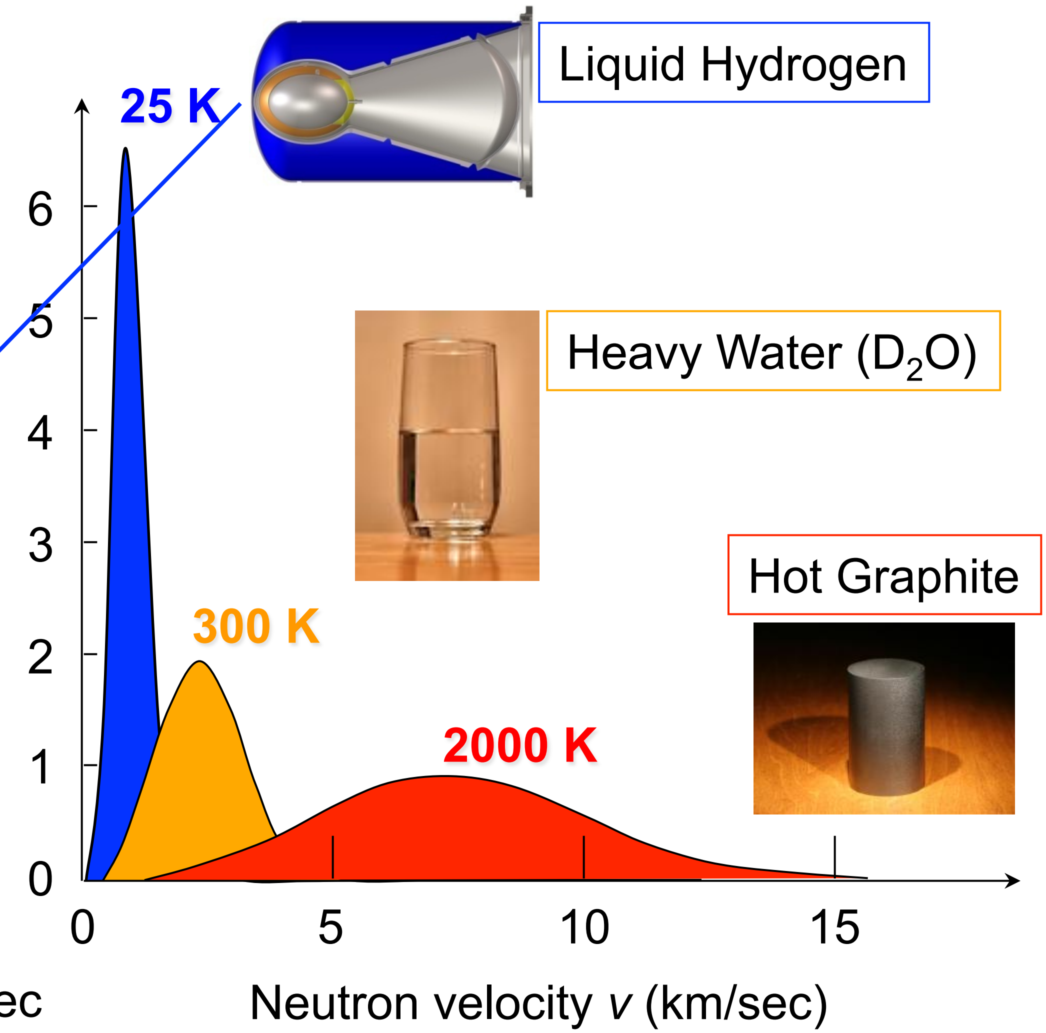
The fission process and moderator are confined by a large containment vessel.

Neutron Moderation

Maxwellian Distribution

$$\Phi \sim v^3 e^{(-mv^2/2k_BT)}$$

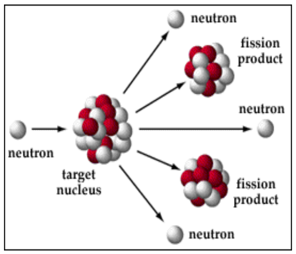


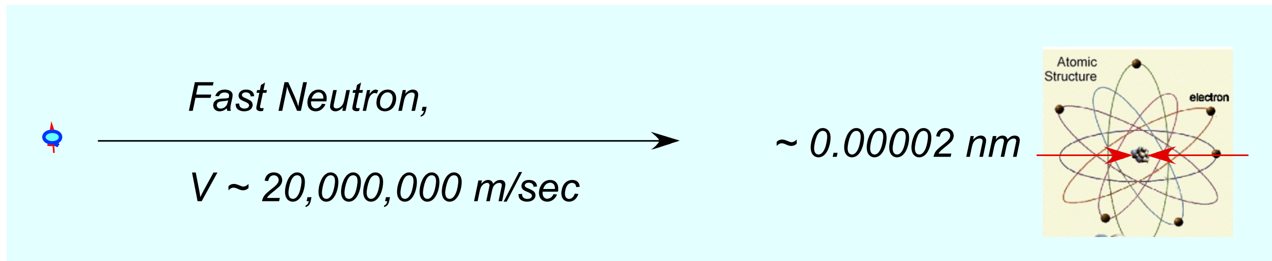


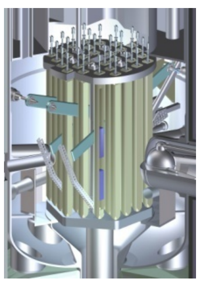
"Fast" neutrons: v = 20,000 km/sec

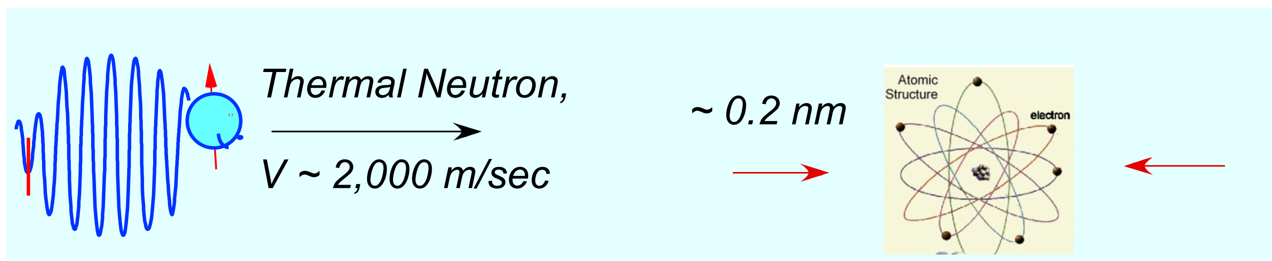
Wave - Particle Duality

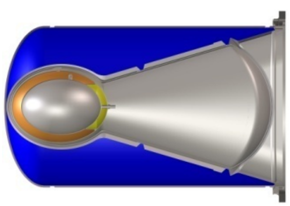
de Broglie Relation $\lambda = h/m_n v$

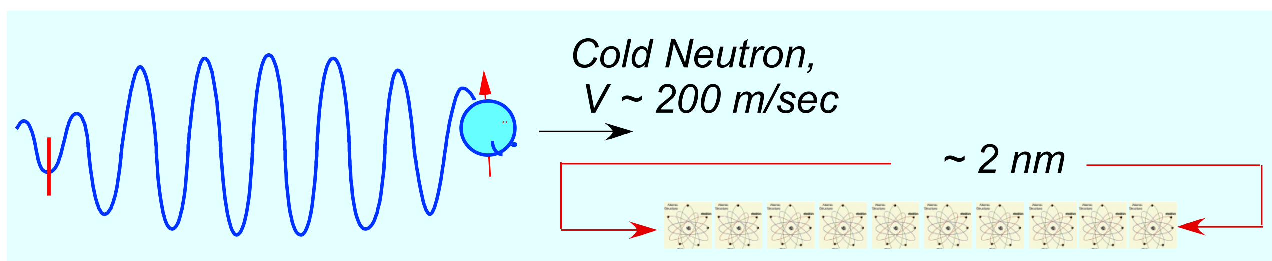










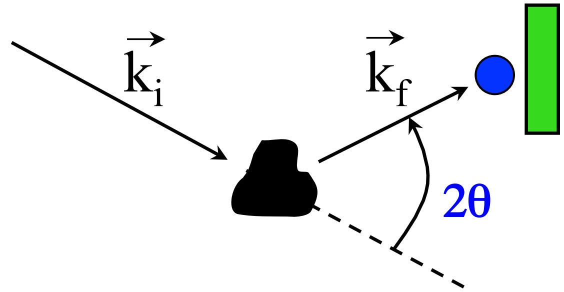


Basics of Neutron Scattering

Neutron scattering experiments measure the flux Φ_s of neutrons scattered by a sample into a detector as a function of the change in neutron wave vector ($\vec{Q}$) and energy ($\hbar\omega$).

$\Phi_s(Q,\hbar\omega) = \dfrac{\text{neutrons}}{\text{sec–cm}^2}$

<u>Momentum</u>	<u>Energy</u>
$\hbar k = \hbar(2\pi/\lambda)$	$\hbar\omega_n = \hbar^2 k_n{}^2/2m$
$\hbar\vec{Q} = \hbar\vec{k}_i - \hbar\vec{k}_f$	$\hbar\omega = \hbar\omega_i - \hbar\omega_f$



The expressions for the scattered neutron flux Φ_s involve the positions and motions of atomic nuclei or unpaired electron spins.

$\boxed{\Phi_s = f\{\vec{r}_i(t), \vec{r}_j(t), \vec{S}_i(t), \vec{S}_j(t)\}}$ ⇒ Φ_s provides information about <u>all</u> of these quantities!

Neutron Scattering Cross Sections

These "cross sections" are what we measure experimentally.

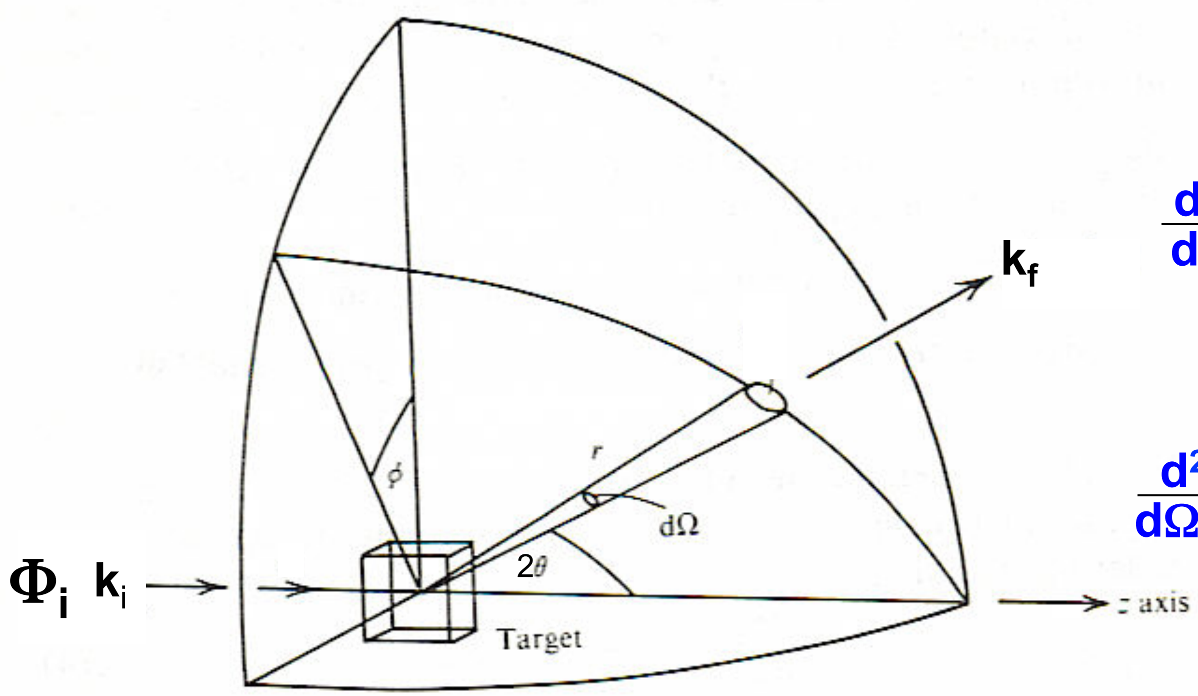
Consider an incident neutron beam with flux Φ_i (neutrons/sec/cm^2) and wave vector $\mathbf{k}_i$ on a non-absorbing sample.

We define three cross sections:

σ = 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?

σ Total # of neutrons scattered per second / Φ_i.

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

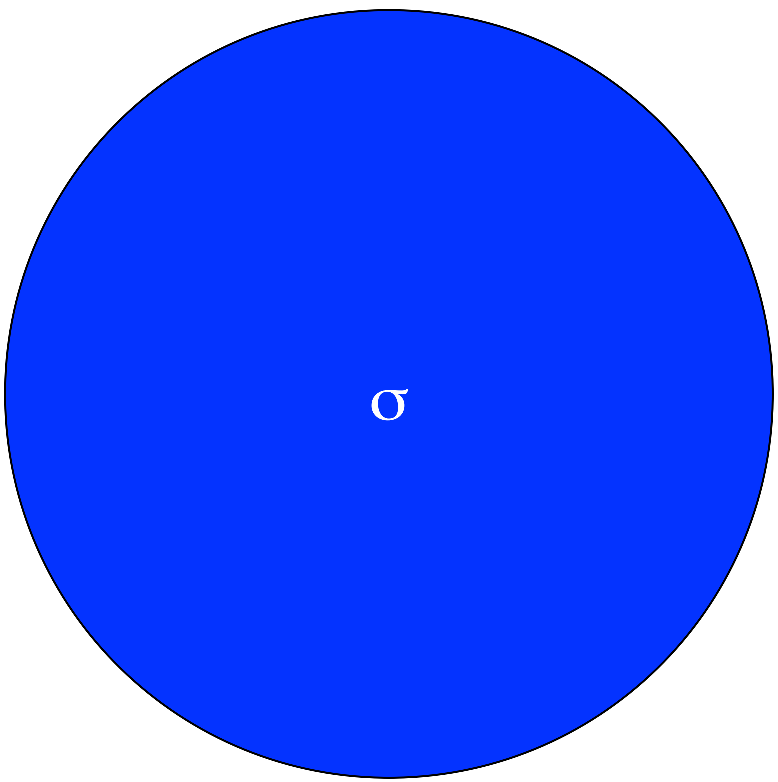
$\frac{d^2\sigma}{d\Omega dE_f}$ Total # of neutrons scattered per second into $d\Omega$ with a final energy between E_f and dE_f / $d\Omega\ dE_f\ \Phi_i$.
(Inelastic scattering → dynamics.

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



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

Typically, $\frac{d\sigma}{d\Omega} \sim 10^6 \times \frac{d^2\sigma}{d\Omega dE_f}$

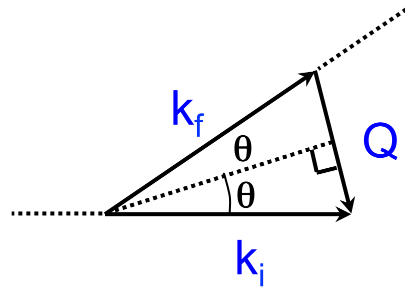
Elastic vs Inelastic Scattering

Note that both of these cases are described by … $\dfrac{d^2\sigma}{d\Omega dE_f}$

Elastic Scattering:

- Change in neutron energy = 0
- Probes changes in momentum only

Elastic ($k_i = k_f$) $\neq \dfrac{d\sigma}{d\Omega}$



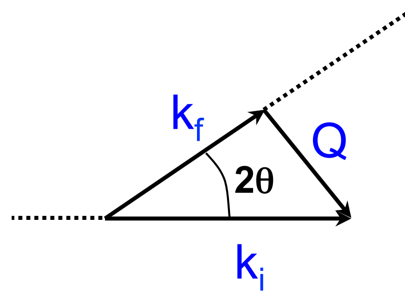
$\sin\theta = (Q/2)/k$

$Q = 2k\sin\theta = 4\pi\sin\theta/\lambda$

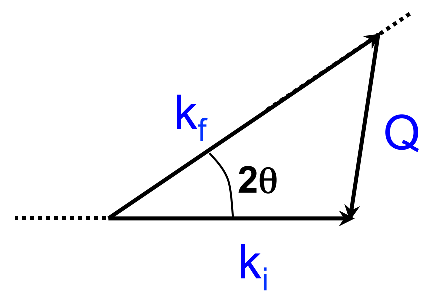
Inelastic Scattering:

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

Inelastic ($k_i \neq k_f$)



Energy loss ($h\omega > 0$)



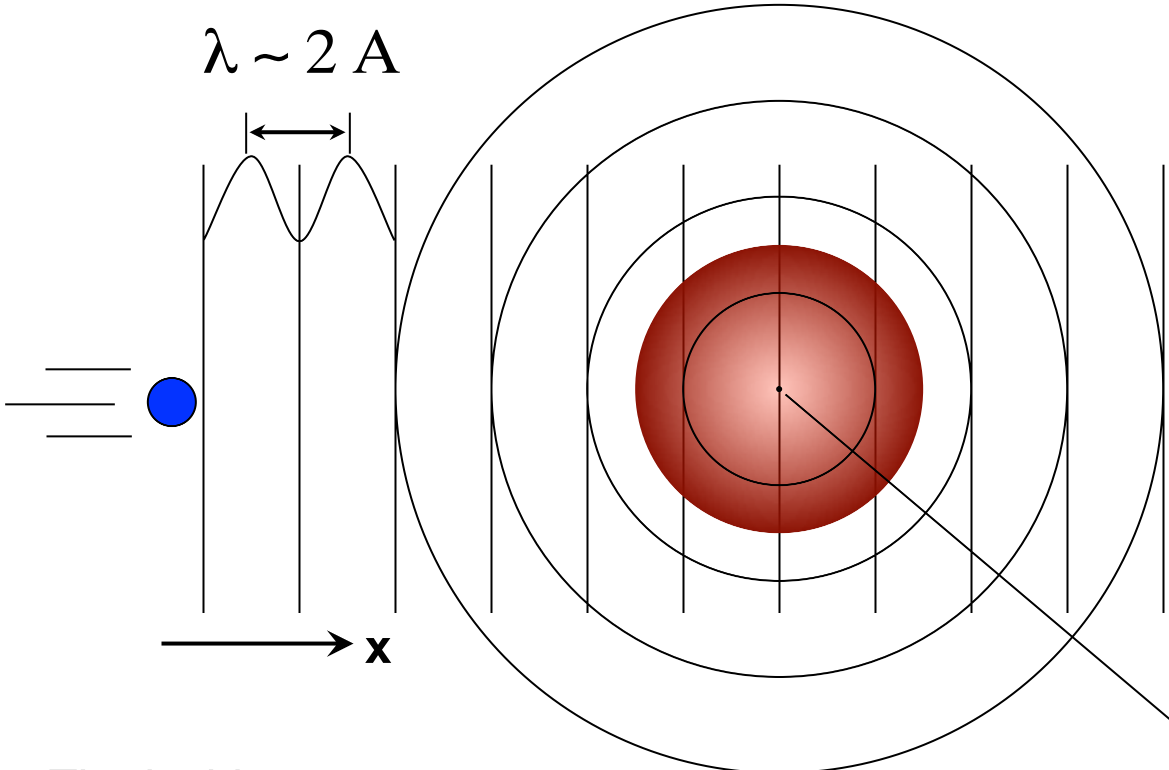
Energy gain ($h\omega < 0$)

Scattering From a Single Nucleus

Consider the simplest case:
A fixed, isolated nucleus.

The scattered (final) neutron Ψ is a <u>spherical</u> wave:

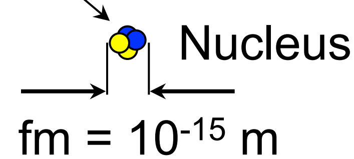
$$\boxed{\Psi_f(\mathbf{r}) \sim (-b/r)e^{ikr}}$$



QUESTIONS:

1. The scattering is <u>elastic</u> ($k_i = k_f = k$). Why?

2. The scattering is <u>isotropic</u>. Why?



The incident neutron Ψ_i is a <u>plane</u> wave:

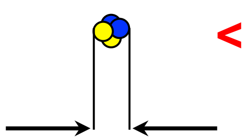
$$\boxed{\Psi_i(\mathbf{r}) \sim e^{ikx}} \quad (k = 2\pi/\lambda)$$

Scattering From a Single Nucleus

ANSWERS:

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 << λ, then the scattered waves are spherically symmetric. (This is also known as s-wave scattering.)



<< λ

~ 10^{-15} m << 10^{-10} m

$$V(r) = \left(\frac{2\pi h^2}{m_n}\right) \sum_{j=1}^{N} b_j \, \delta(r - r_j)$$

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

Scattering From a Single Nucleus

The Neutron Scattering Length - b

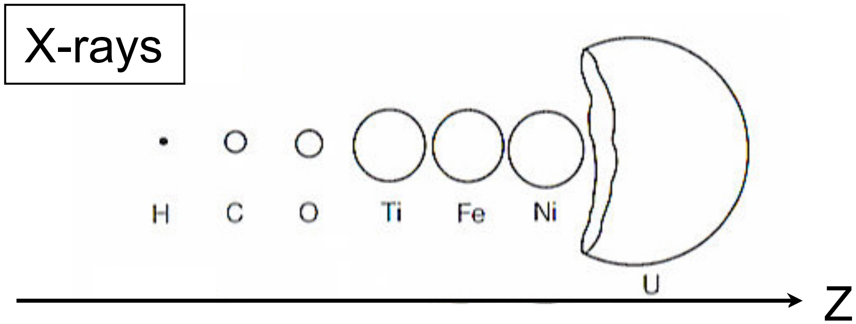
The parameter b is known as the neutron “scattering length” and has units of length ~ 10^{-12} cm.

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.

X-rays



b varies randomly with Z and isotope → allows access to atoms that are usually unseen by x-rays.

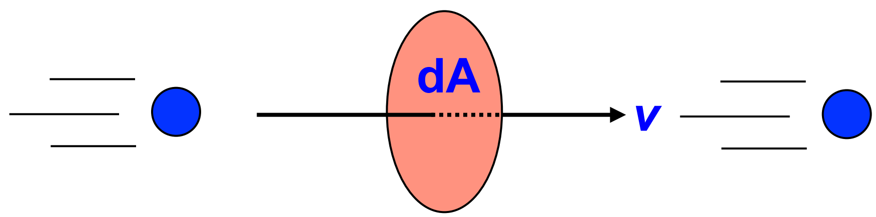
Scattering From a Single Nucleus

We can easily calculate σ for a single, fixed nucleus:

Def.

$\sigma \cdot \Phi_i$ = Total number of neutrons scattered per second by the nucleus.

v = Velocity of neutrons (elastic → same before and after scattering).



$v\, dA\, |\Psi_f|^2$ = Total number of neutrons scattered per second through **dA**.

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

Since $\Phi_i = v\, |\Psi_i|^2 = v \rightarrow$ $\boxed{\sigma = 4\pi b^2}$

Try calculating $\frac{d\sigma}{d\Omega}$

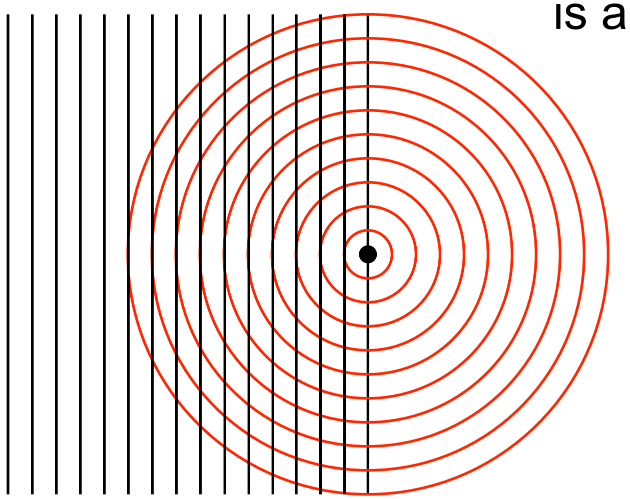
Scattering From Many Nuclei

What if many atoms are present?

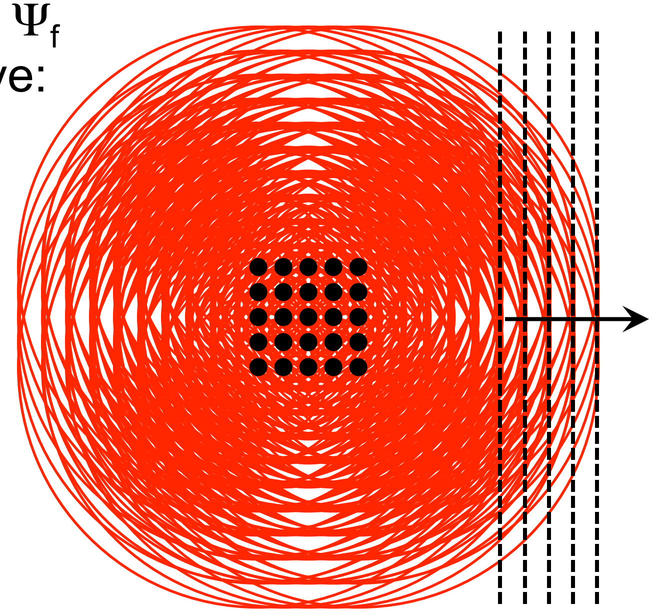
Scattering from one nucleus

Scattering from many nuclei

Scattered neutron Ψ_f is a <u>spherical</u> wave:



The incident neutron Ψ_i is a <u>plane</u> wave:

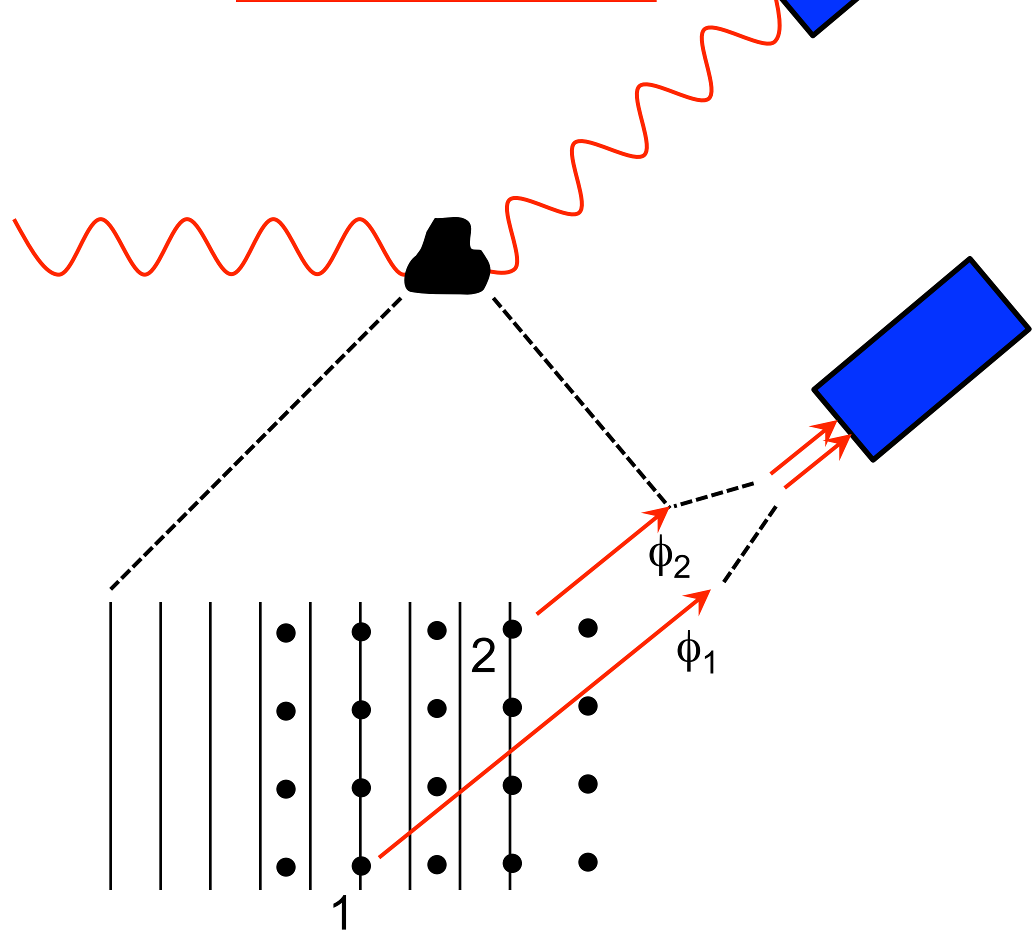


Constructive interference

Get strong scattering in some directions, but not in others. Angular dependence yields information about how the nuclei are arranged or <u>correlated</u>.

Scattering From Many Nuclei

Two-particle or pair correlations



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

(2) Superposition:
Amplitudes of scattered neutrons ϕ_n add linearly.

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

$$\text{Intensity} = |\Phi|^2 = |\phi_1 + \phi_2 + \ldots|^2 = |\phi_1|^2 + |\phi_2|^2 + \ldots + \underbrace{\phi_1{}^*\phi_2{}^* + \phi_2{}^*\phi_1}_{\text{Depends on relative positions of 1 and 2} \rightarrow \text{pair correlations!}} + \ldots$$

After Andrew Boothroyd
PSI Summer School 2007

Pair Correlation Functions

From Van Hove (1954) …

The measured quantity Φ_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

$$\frac{d\sigma}{d\Omega} = \sum_{i,j} b_i b_j \, e^{-i(\mathbf{k}_i-\mathbf{k}_j)\cdot(\mathbf{r}_i-\mathbf{r}_j)}$$

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

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

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

Neutron Structure Factor

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

Fourier Transform

Pair Correlation Function

$$G(\mathbf{r},t) = (\frac{1}{2\pi})^3 \frac{1}{N}\int \sum_{jj'} e^{i\mathbf{Q}\cdot\mathbf{r}} < e^{-i\mathbf{Q}\cdot\mathbf{r}_{j'}(0)} e^{i\mathbf{Q}\cdot\mathbf{r}_j(t)} > d\mathbf{Q}$$

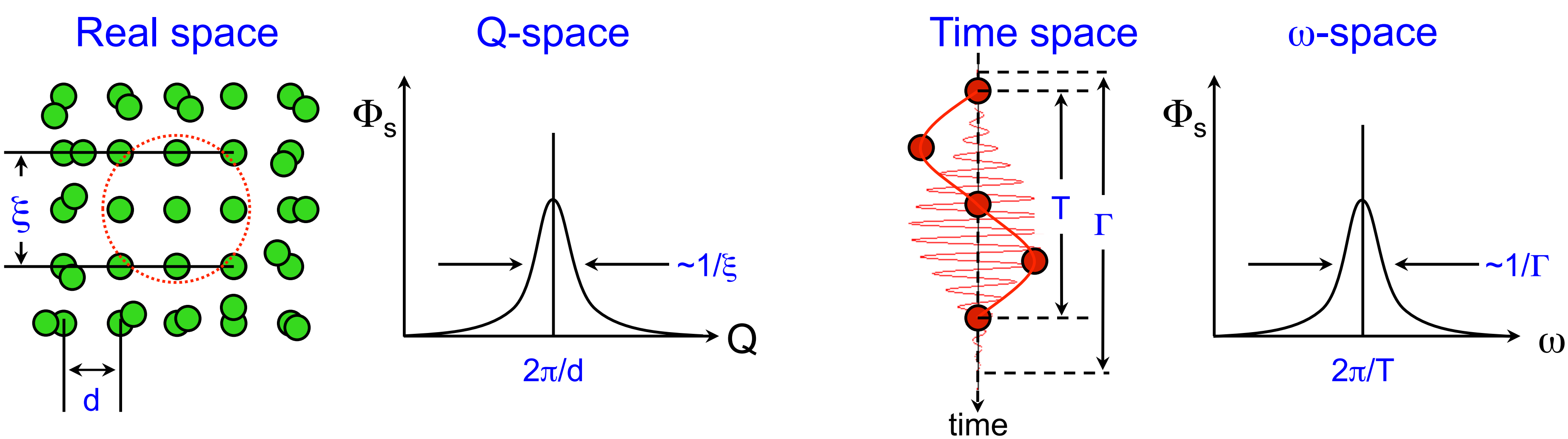
Squires, Introduction to the theory of thermal neutron scattering (1996)

Pair Correlation Functions

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

The scattered neutron flux $\Phi_s(\vec{Q},\hbar\omega)$ 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.

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

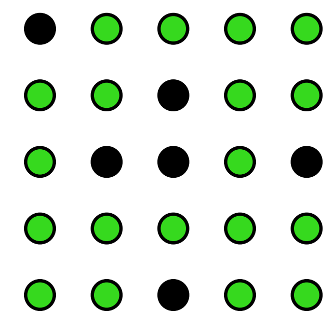


Neutron Coherent and Incoherent Scattering

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 <u>nuclear</u>.



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

Then the above equation must be generalized:

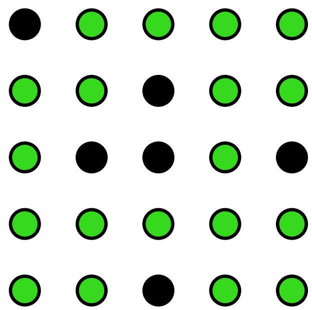
$$\frac{d^2\sigma}{d\Omega dE_f} = \sum_{i,j} \overline{b_i b_j}\, S_{ij}(Q,\omega)$$

$$\overline{b_i b_j} = (\overline{b})^2, \text{ for } i=j$$

$$\overline{b_i b_j} = \overline{b^2}, \text{ for } i \neq j$$

—— = average

Neutron Coherent and Incoherent Scattering



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

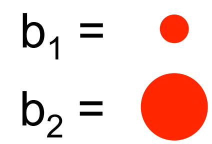
$$\boxed{\frac{d^2\sigma}{d\Omega dE_f} = \sigma_c S_c(Q,\omega) + \sigma_i S_i(Q,\omega)}$$, where

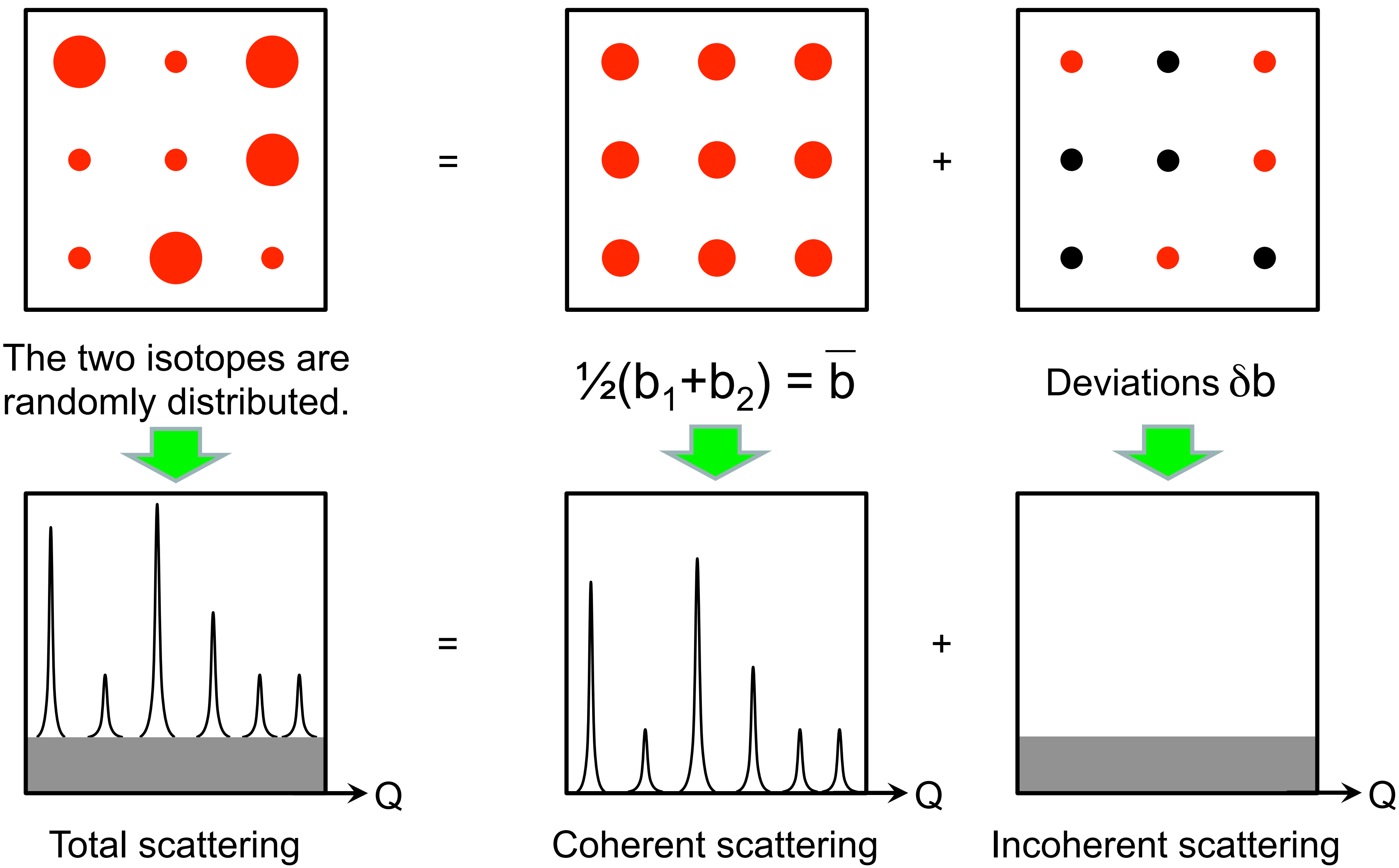
$$\sigma = \sigma_c + \sigma_i, \qquad \sigma_c = 4\pi(\overline{b})^2 \qquad \text{c = coherent}$$

$$\sigma_i = 4\pi\{\overline{b^2} - (\overline{b})^2\} \qquad \text{i = incoherent}$$

Neutron Coherent and Incoherent Scattering

Consider a system composed of two different scattering lengths, b_1 and b_2.





Neutron Coherent and Incoherent Scattering

What do these expressions mean physically?

Coherent Scattering

Measures the Fourier transform of the *pair* correlation function G(r,t) → <u>interference effects.</u>

This cross section reflects <u>collective</u> phenomena such as:

Bragg Peaks

Phonons

Spin Waves

Incoherent Scattering

Measures the Fourier transform of the *self* correlation function $G_s(r,t)$ → <u>no interference effects.</u>

This cross section reflects <u>single-particle</u> scattering:

Atomic Diffusion

Vibrational Density of States

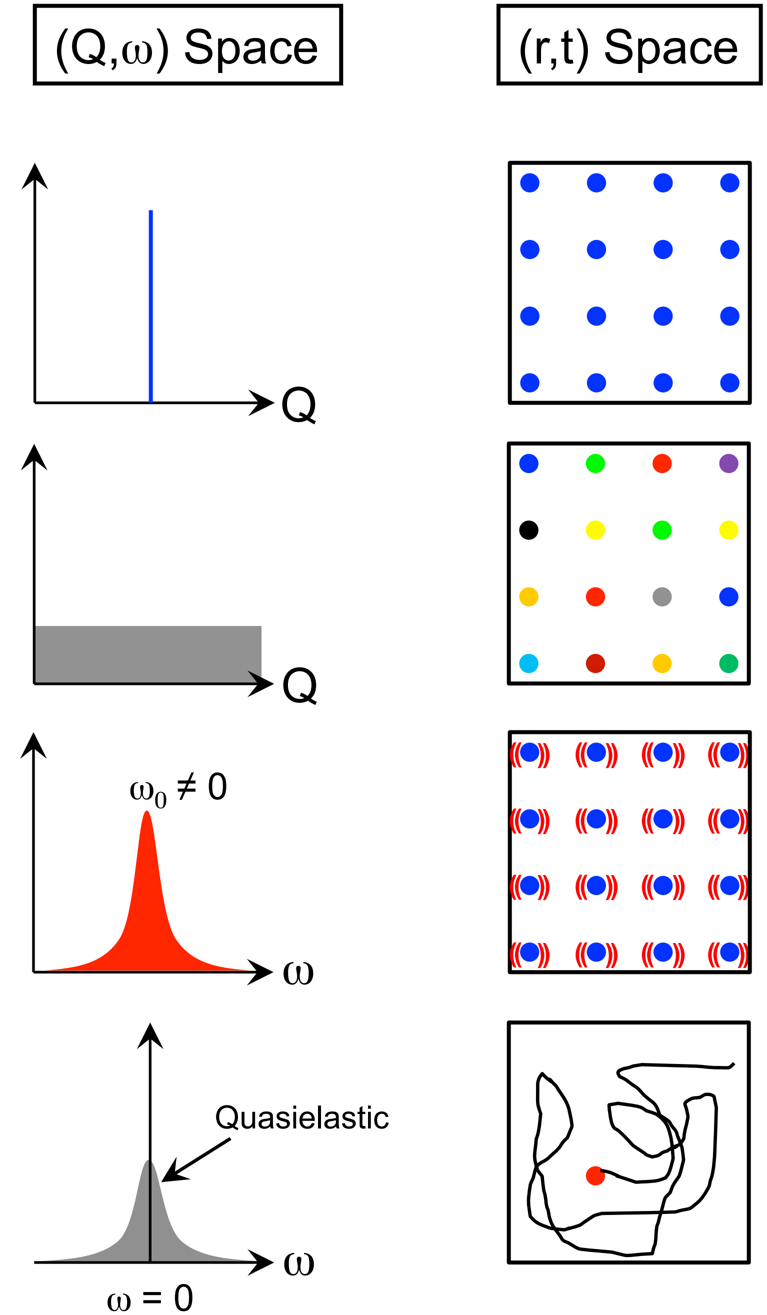
Summary of Cross Sections

$$\left.\frac{d\sigma}{d\Omega}\right|_{coh} = \frac{\sigma_{coh}}{4\pi} S(Q)$$

$$\left.\frac{d\sigma}{d\Omega}\right|_{inc} = \frac{\sigma_{inc}}{4\pi}$$

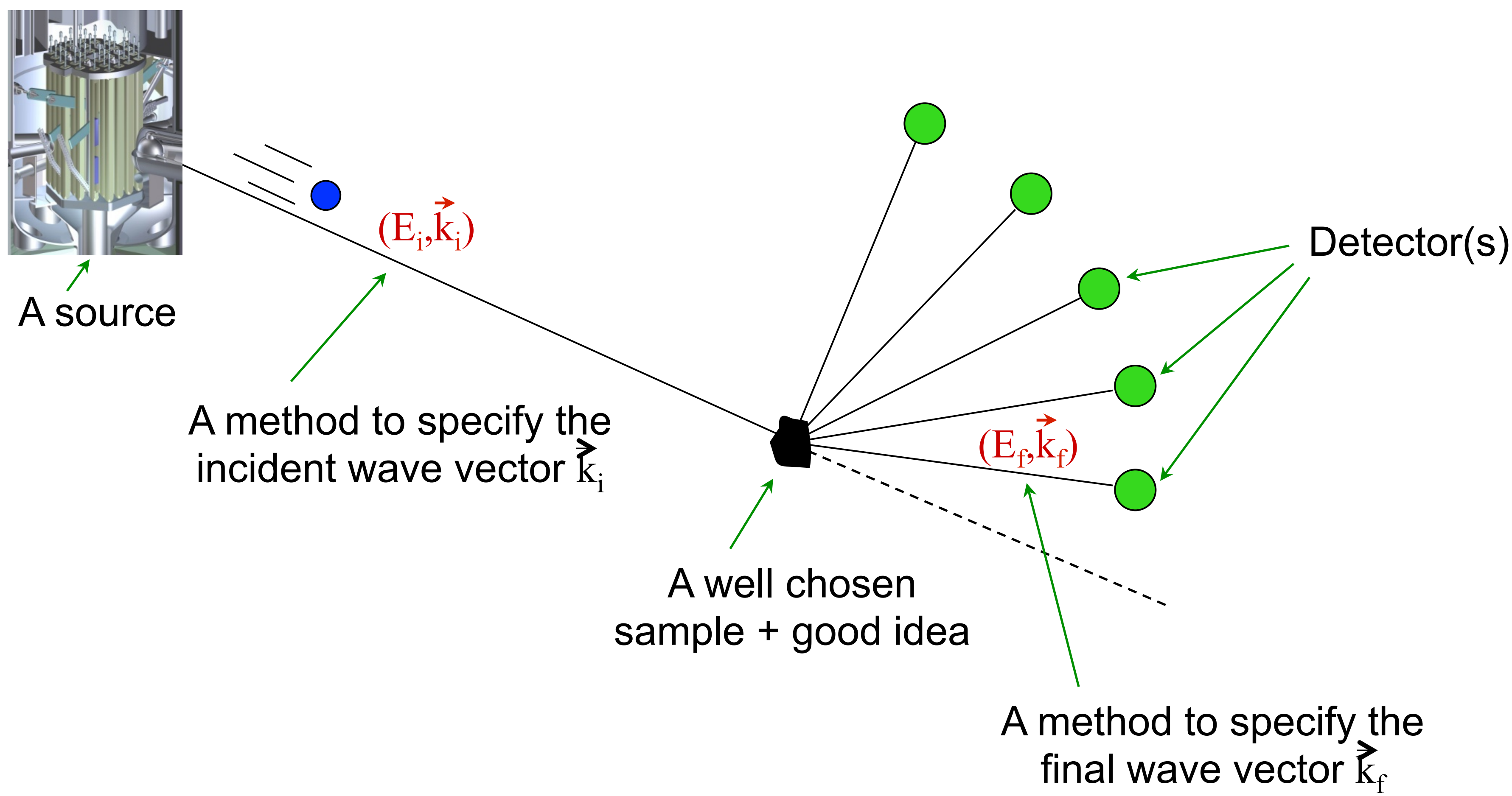
$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right|_{coh} = \frac{k_f}{k_i}\frac{\sigma_{coh}}{4\pi} S_{coh}(Q,\omega)$$

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right|_{inc} = \frac{k_f}{k_i}\frac{\sigma_{inc}}{4\pi} S_{inc}(Q,\omega)$$



Basics of Neutron Scattering Methods

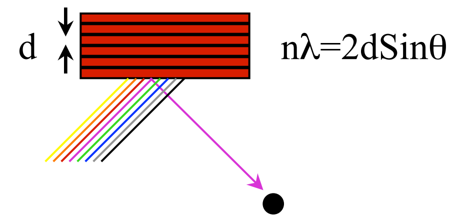
Elements of all scattering experiments



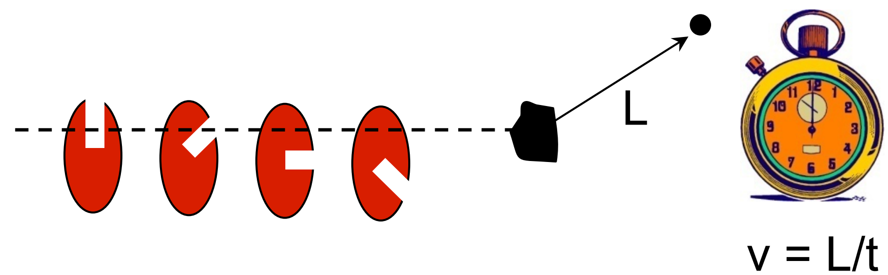
Basics of Neutron Scattering Methods

Methods of specifying and measuring $\vec{k}_i$ and $\vec{k}_f$

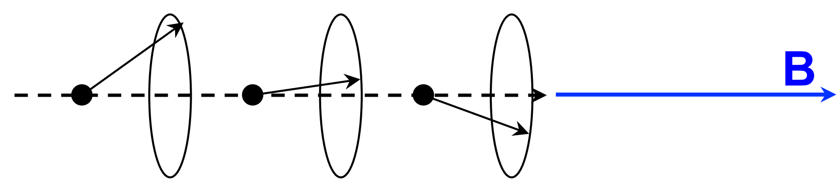
1. Bragg Diffraction



2. Time-of-Flight (TOF)

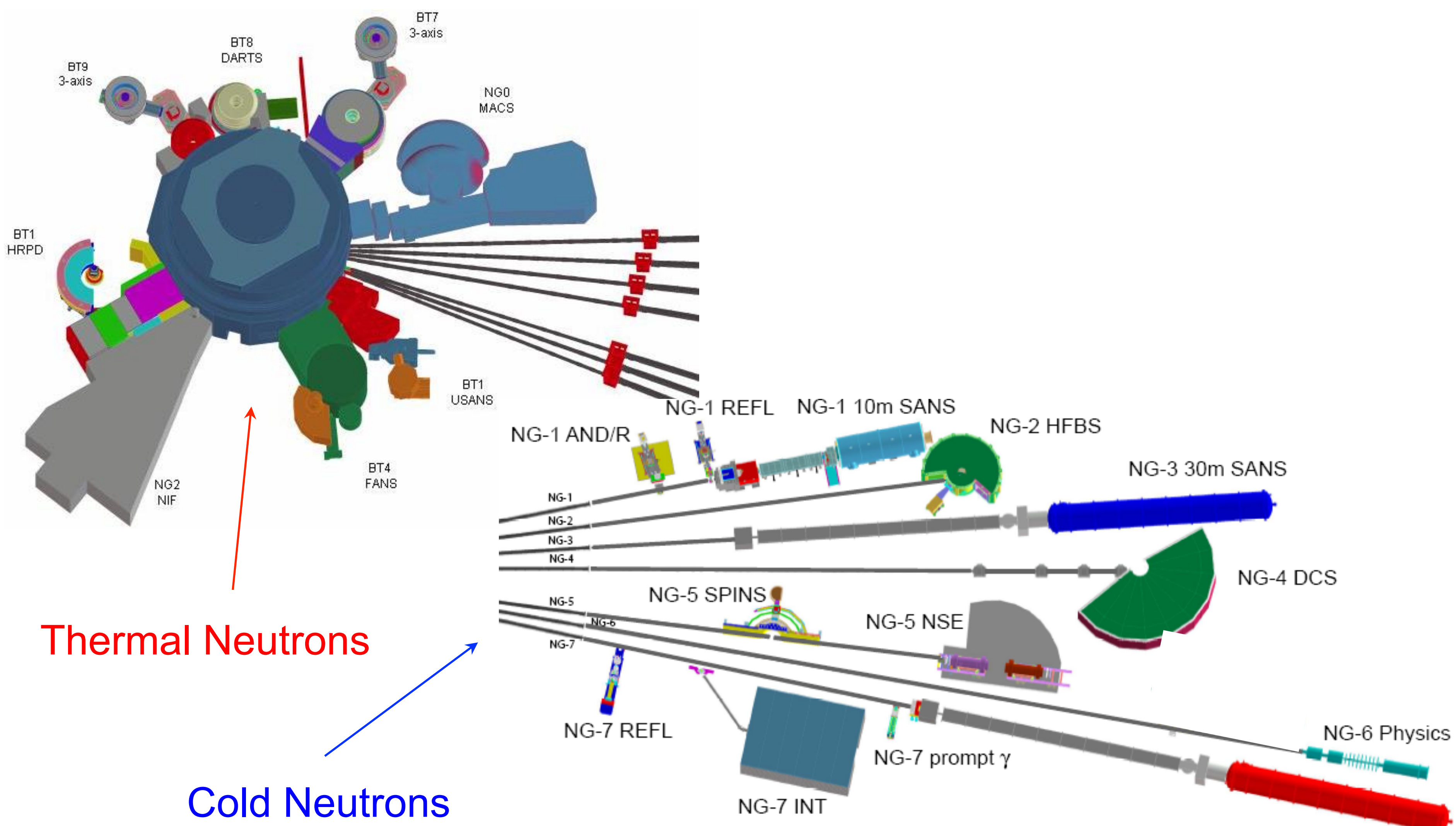


3. Larmor Precession



Different Types of Neutron Instruments

You will see many of these on the tour …

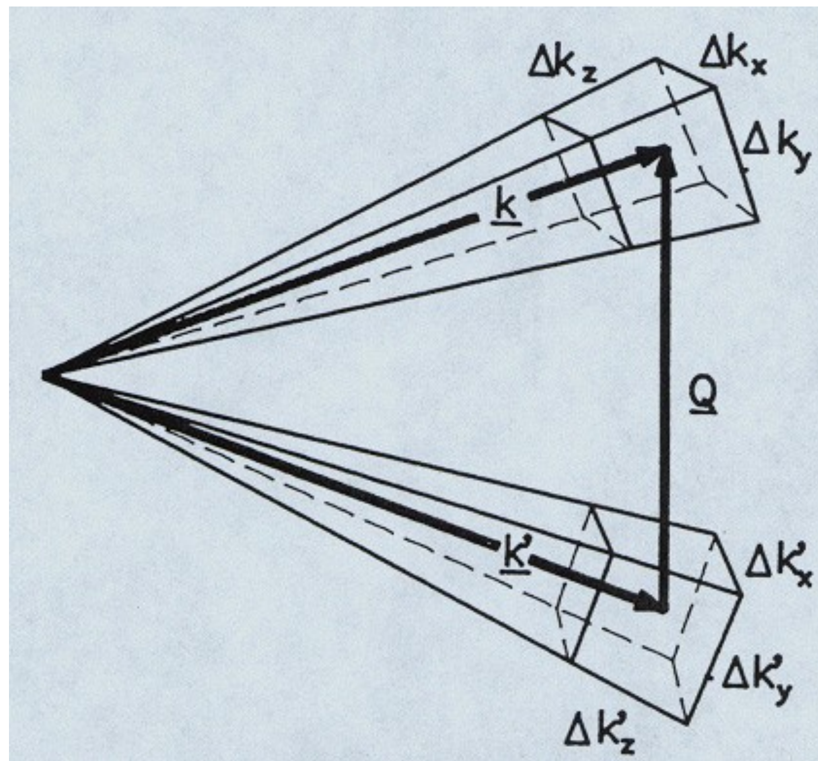


Different Types of Neutron Instruments

Why so many different kinds?

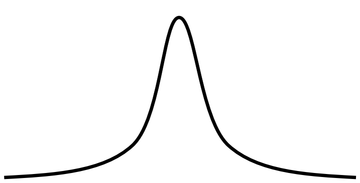
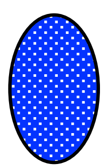
Because neutron scattering is an intensity-limited technique. Thus detector coverage and resolution MUST be tailored to the science.

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



Courtesy of R. Pynn

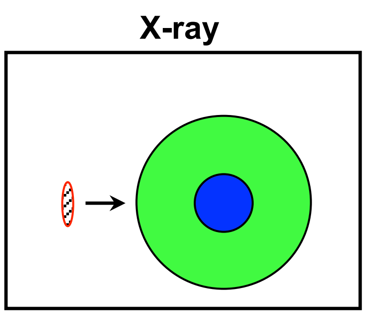
The total signal in a scattering experiment is proportional to the resolution volume → better resolution leads to lower count rates! *Choose carefully* …

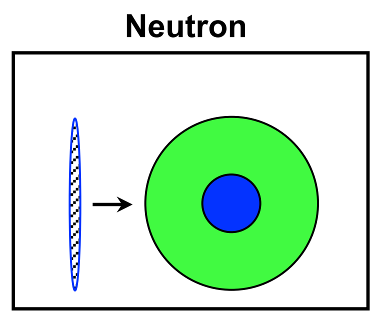
 $\propto$ 

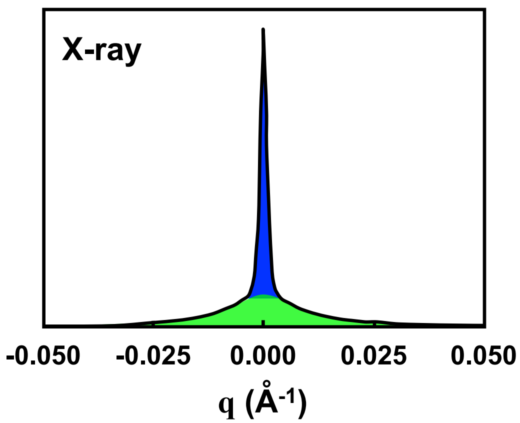
Instrumental Q-Resolution

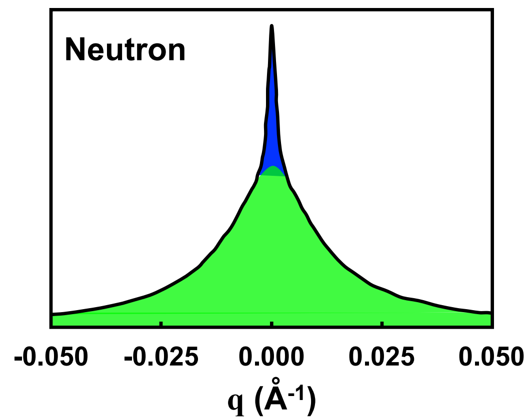
Q-Resolution Matters!

The "right" resolution depends on what you want to study.







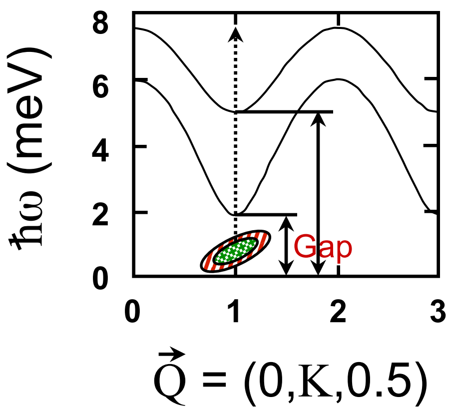


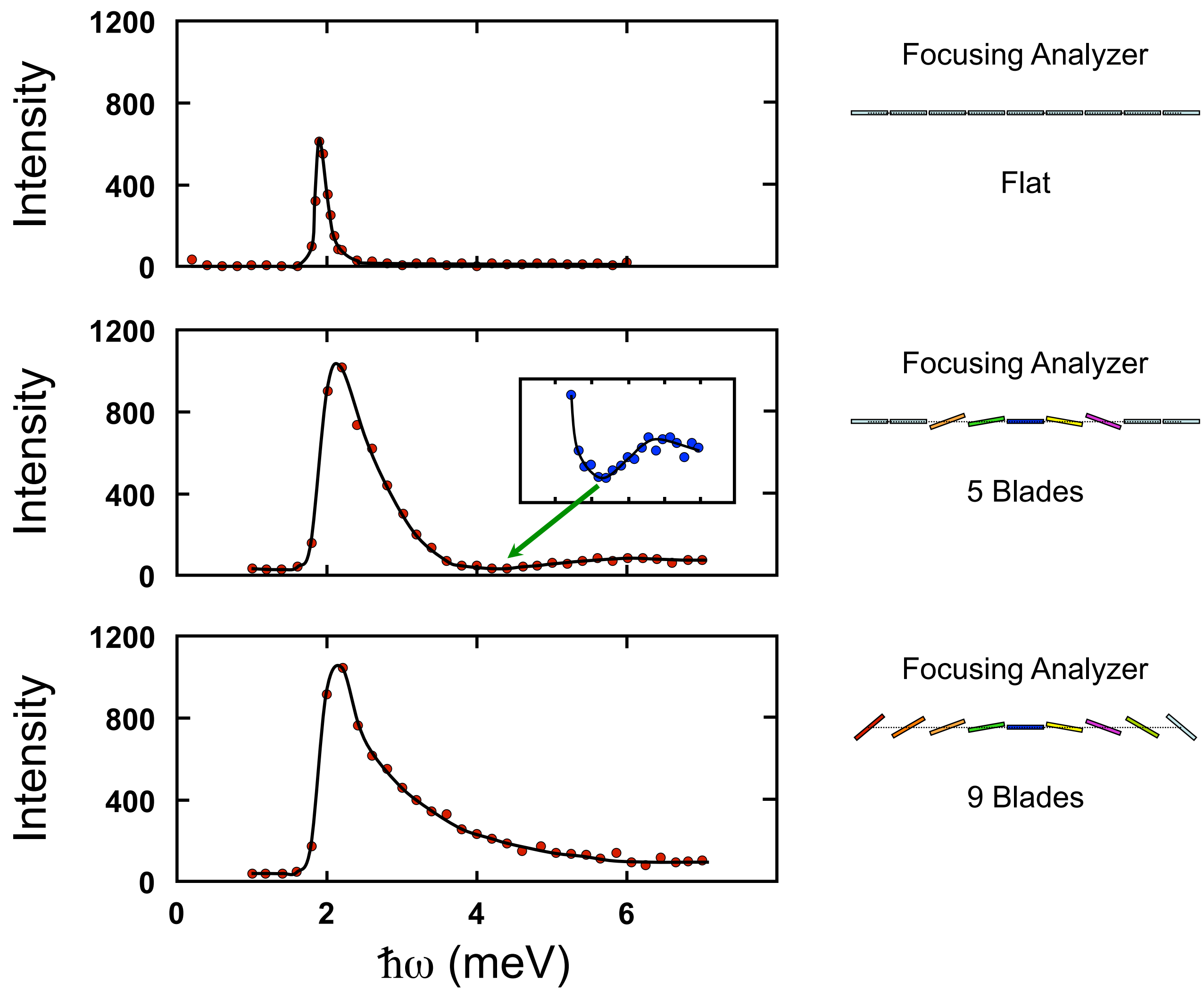
Instrumental ħω-Resolution

ħω-Resolution Matters!

Example – $CuGeO_3$

SPINS





A Quick Review …

Please try to remember these things …

1. Neutrons scattering probes <u>two-particle</u> 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 $G(r,t)$ → <u>interference effects.</u>

   This cross section reflects <u>collective</u> phenomena.

4. Incoherent Scattering

   Measures the Fourier transform of the self correlation function $G_s(r,t)$→ <u>no interference effects.</u>

   This cross section reflects <u>single-particle</u> scattering.

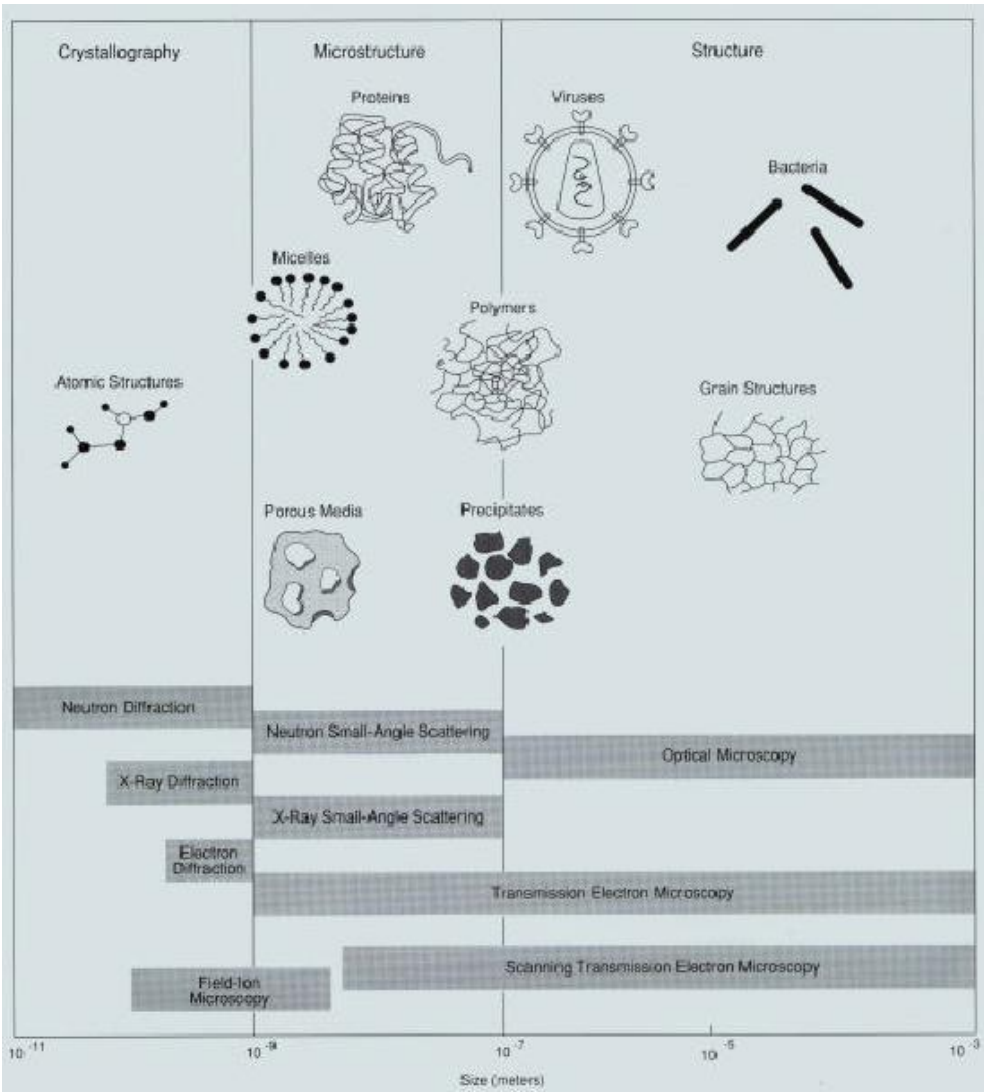
Some Examples

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

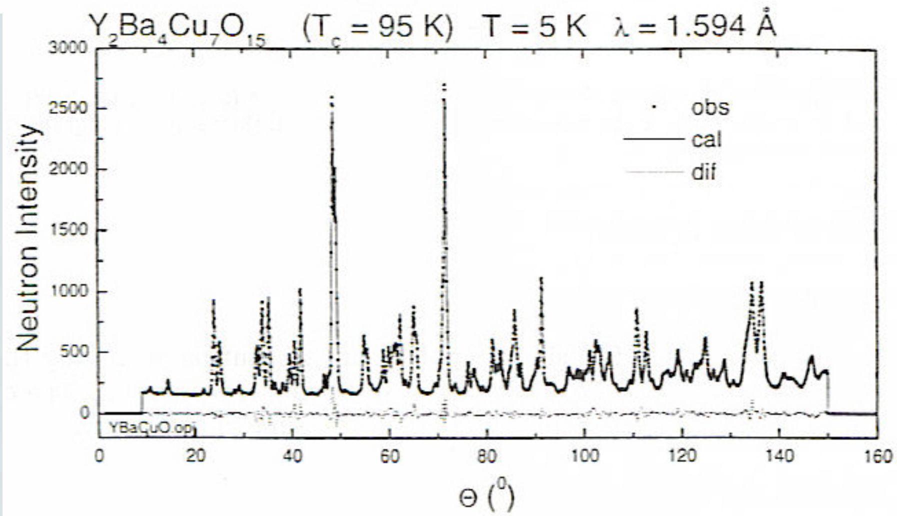
Let’s look at a few examples …

Neutron Elastic Scattering

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



Pynn, *Neutron Scattering: A Primer* (1989)



Mitchell et. al, *Vibrational Spectroscopy with Neutrons* (2005)

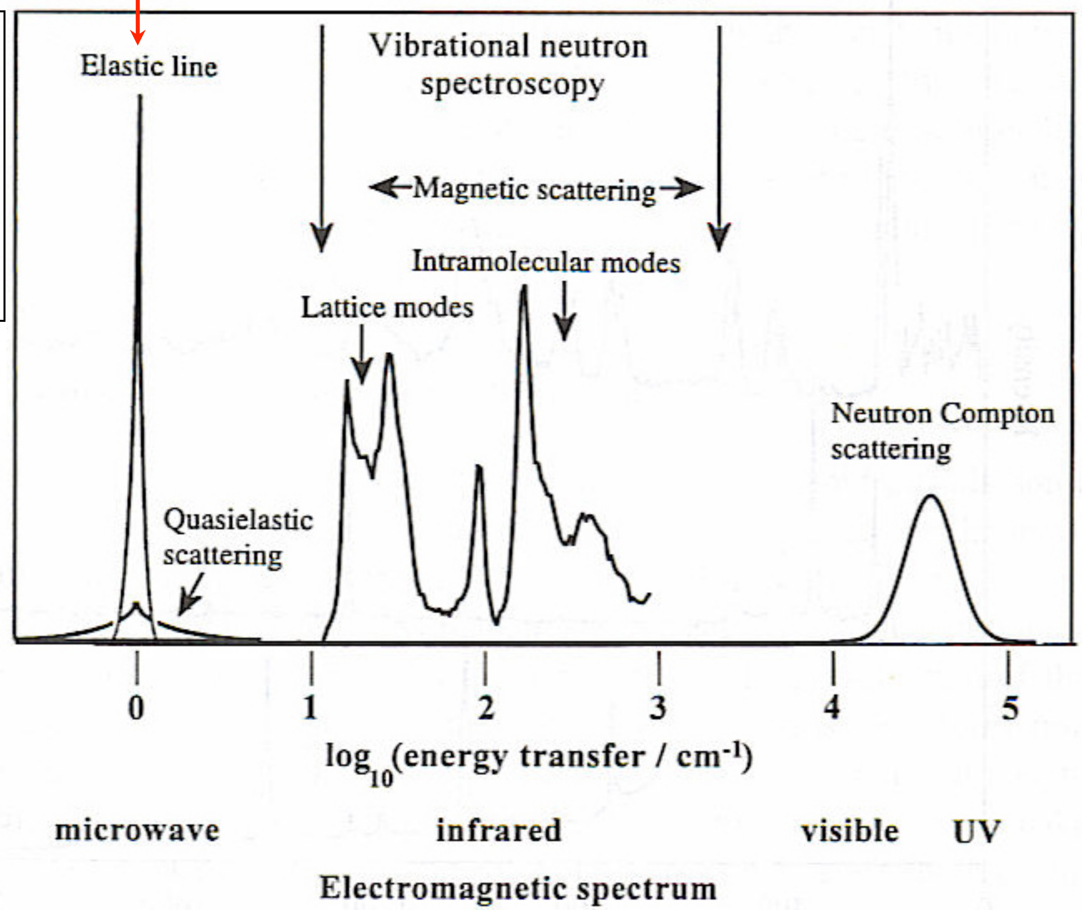
- Used to determine structure of 123 high-T_c cuprates as x-rays weren't sufficiently sensitive to the oxygen atoms.

Neutron Inelastic Scattering

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

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

$\hbar\omega = 0$

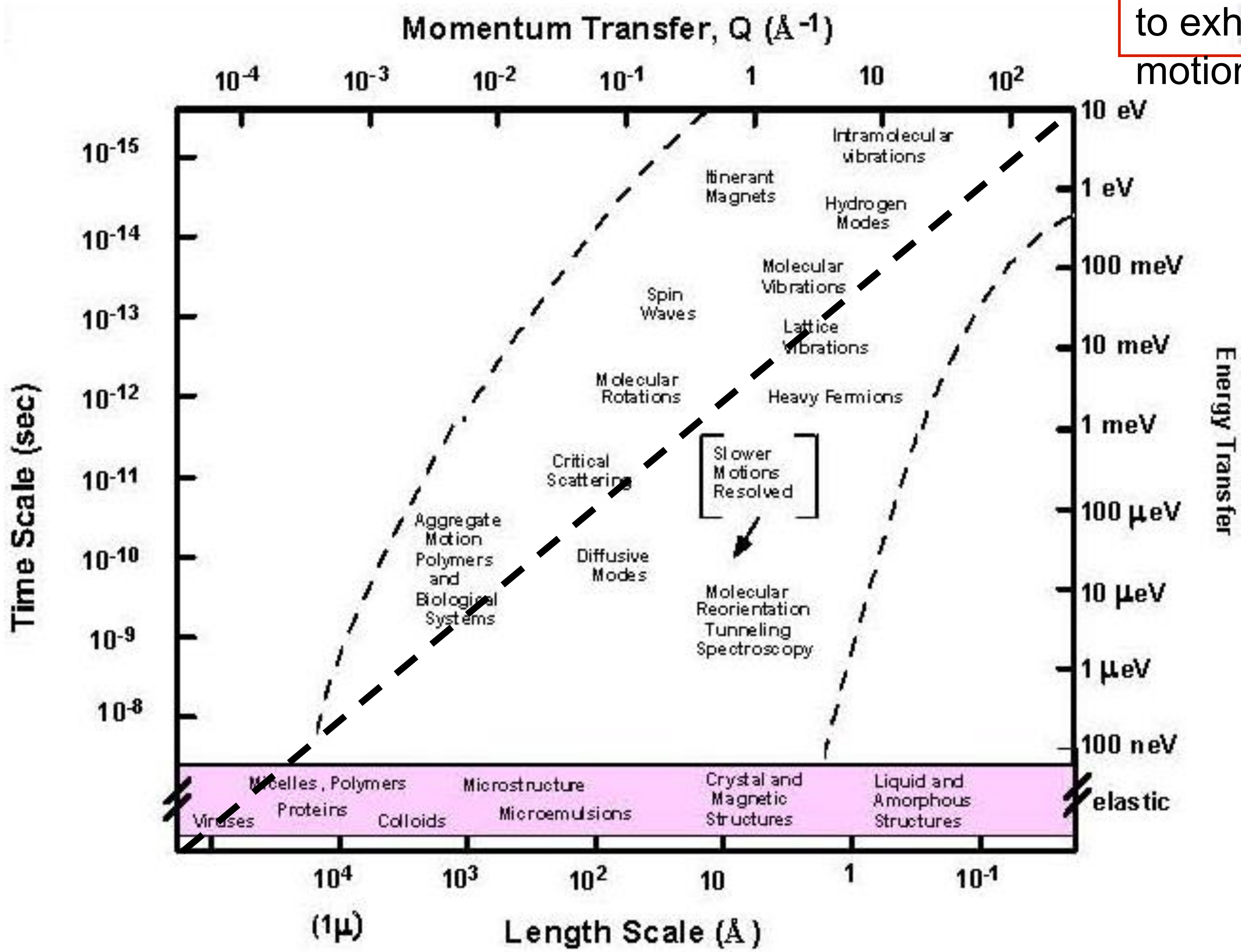


Mitchell et. al, *Vibrational Spectroscopy with Neutrons* (2005)

Neutron Length and Time Scales

Do you see a pattern here?

Larger "objects" tend to exhibit slower motions.



Neutron Elastic Scattering

Pop Quiz

Can one measure elastic scattering from a liquid?

If Yes, explain why?
If No, explain why not?

Hint: What is the correlation of one atom in a liquid with another after a time t?

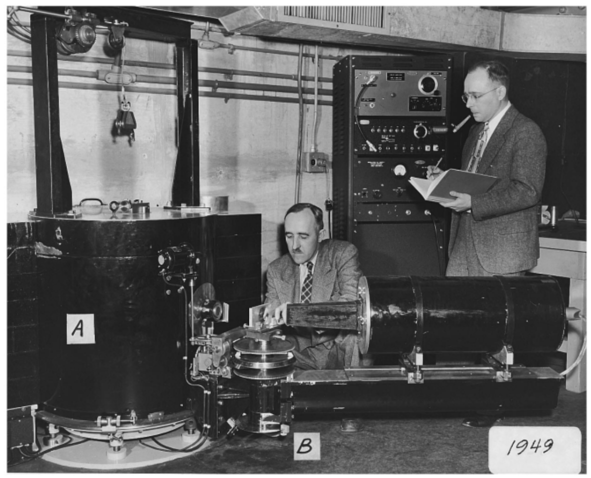
Nobel Prize in Physics 1994

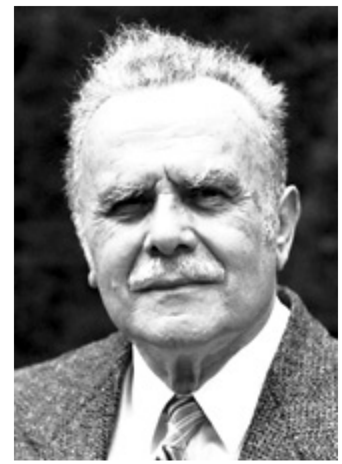
The Fathers of Neutron Scattering

"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 …

References for Further Reading

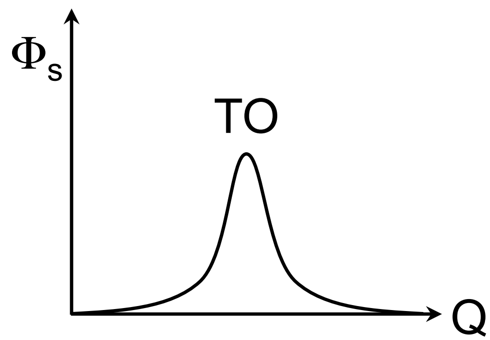
- http://www.mrl.ucsb.edu/~pynn/primer.pdf
- "Introduction to the Theory of Thermal Neutron Scattering"
  - G. L. Squires, Cambridge University Press
- "Theory of Neutron Scattering from Condensed Matter"
  - S. W. Lovesey, Oxford University Press
- "Neutron Diffraction" (Out of print)
  - G. E. Bacon, Clarendon Press, Oxford
- "Structure and Dynamics"
  - M. T. Dove, Oxford University Press
- "Elementary Scattering Theory"
  - D. S. Sivia, Oxford University Press

Neutron Inelastic Scattering

Neutron Measurements of Phonon Dispersions.

There are two primary methods of measuring the neutron scattering cross section S(Q,ω).

- Constant-E scans: vary Q at fixed hω.



- Constant-Q scans: vary hω at fixed Q.