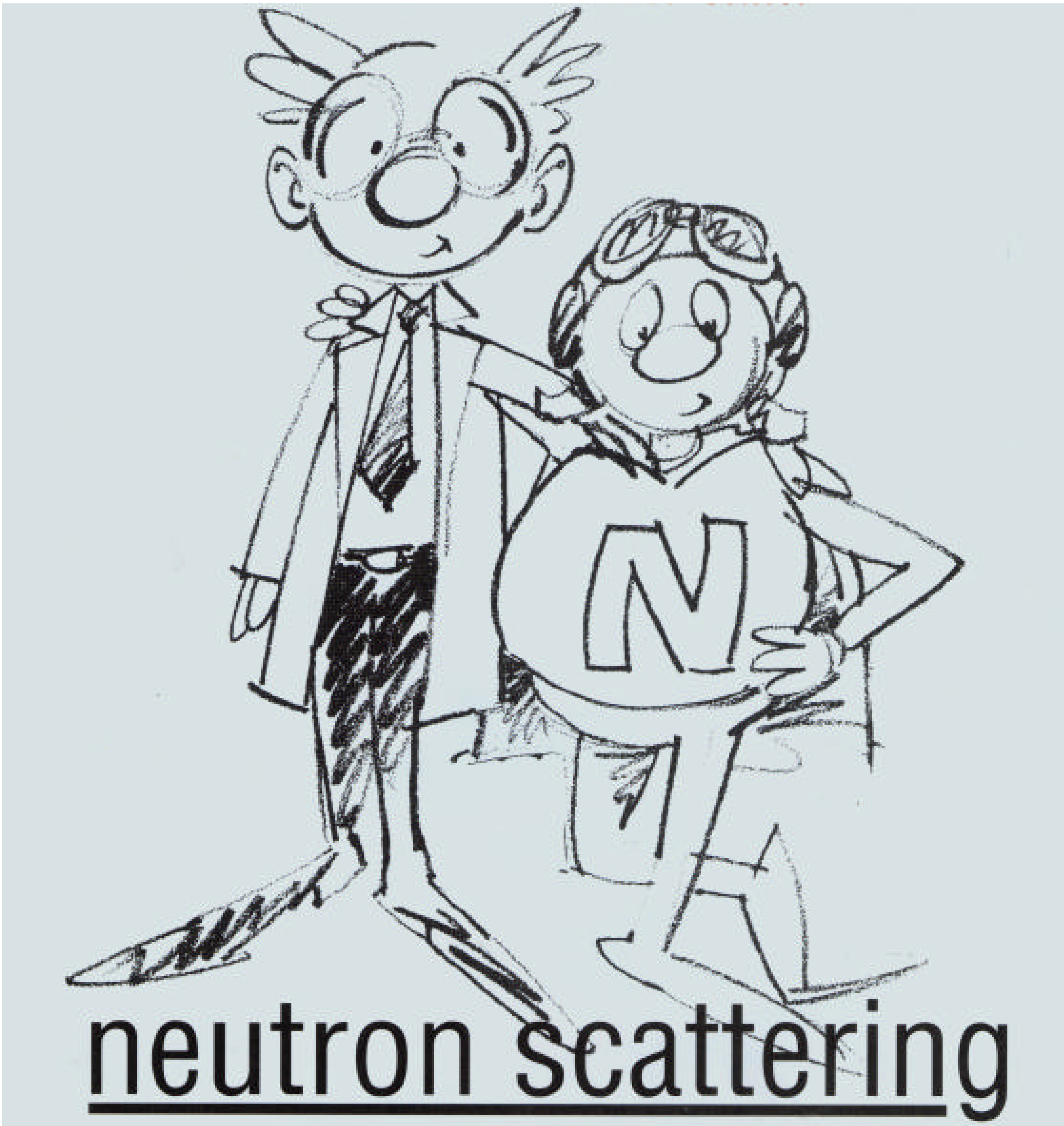


by

Roger Pynn

Los Alamos
National Laboratory

LECTURE 1: Introduction & Neutron Scattering “Theory”

Overview

1. Introduction and theory of neutron scattering
    1. Advantages/disadvantages of neutrons
    2. Comparison with other structural probes
    3. Elastic scattering and definition of the structure factor, S(Q)
    4. Coherent & incoherent scattering
    5. Inelastic scattering
    6. Magnetic scattering
    7. Overview of science studied by neutron scattering
    8. References
2. Neutron scattering facilities and instrumentation
3. Diffraction
4. Reflectometry
5. Small angle neutron scattering
6. Inelastic scattering

Why do Neutron Scattering?

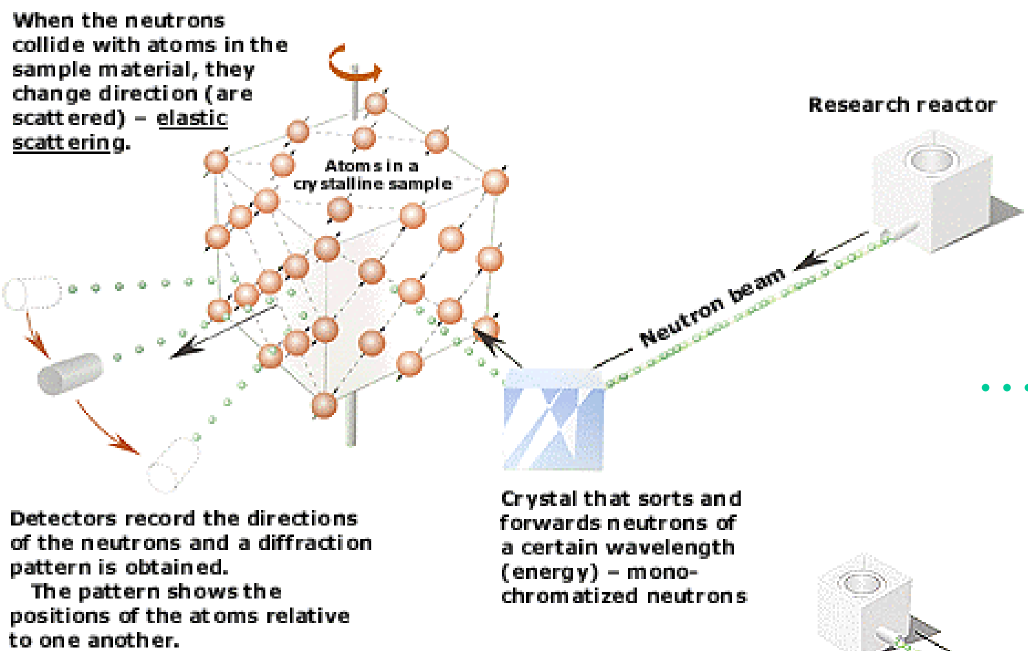
- To determine the positions and motions of atoms in condensed matter
  - 1994 Nobel Prize to Shull and Brockhouse cited these areas
    (see http://www.nobel.se/physics/educational/poster/1994/neutrons.html)

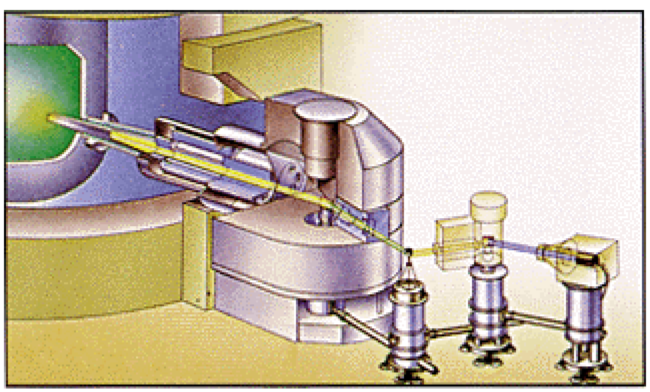
- Neutron advantages:
  - Wavelength comparable with interatomic spacings
  - Kinetic energy comparable with that of atoms in a solid
  - Penetrating => bulk properties are measured & sample can be contained
  - Weak interaction with matter aids interpretation of scattering data
  - Isotopic sensitivity allows contrast variation
  - Neutron magnetic moment couples to ***B*** **=>** neutron "sees" unpaired electron spins

- Neutron Disadvantages
  - Neutron sources are weak => low signals, need for large samples etc
  - Some elements (e.g. Cd, B, Gd) absorb strongly
  - Kinematic restrictions (can't access all energy & momentum transfers)

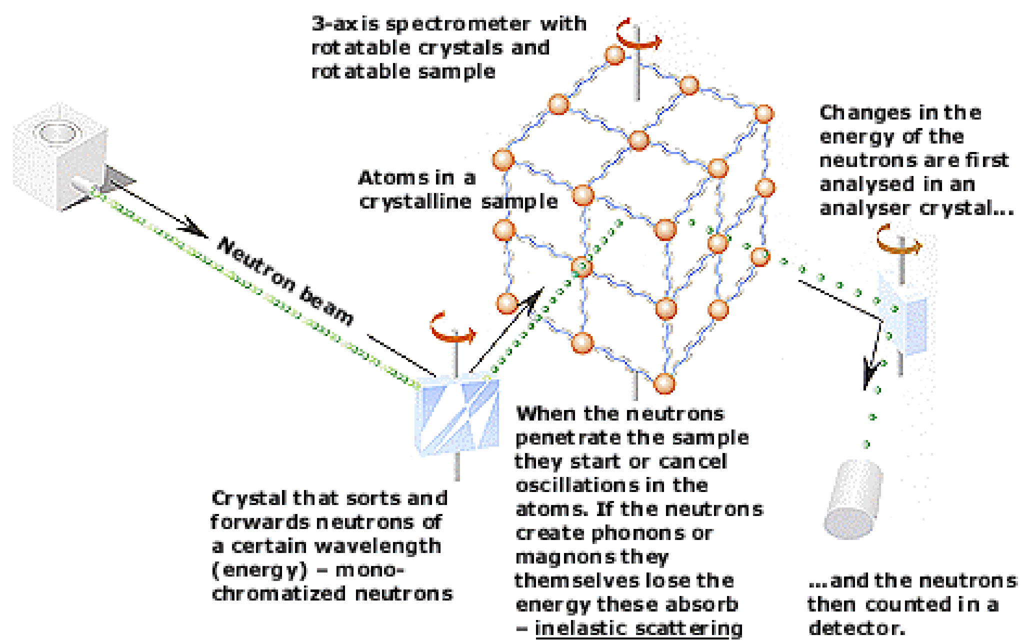
The 1994 Nobel Prize in Physics – Shull & Brockhouse

Neutrons show where the atoms are….




3-axis spectrometer

…and what the atoms do.



The Neutron has Both Particle-Like and Wave-Like Properties

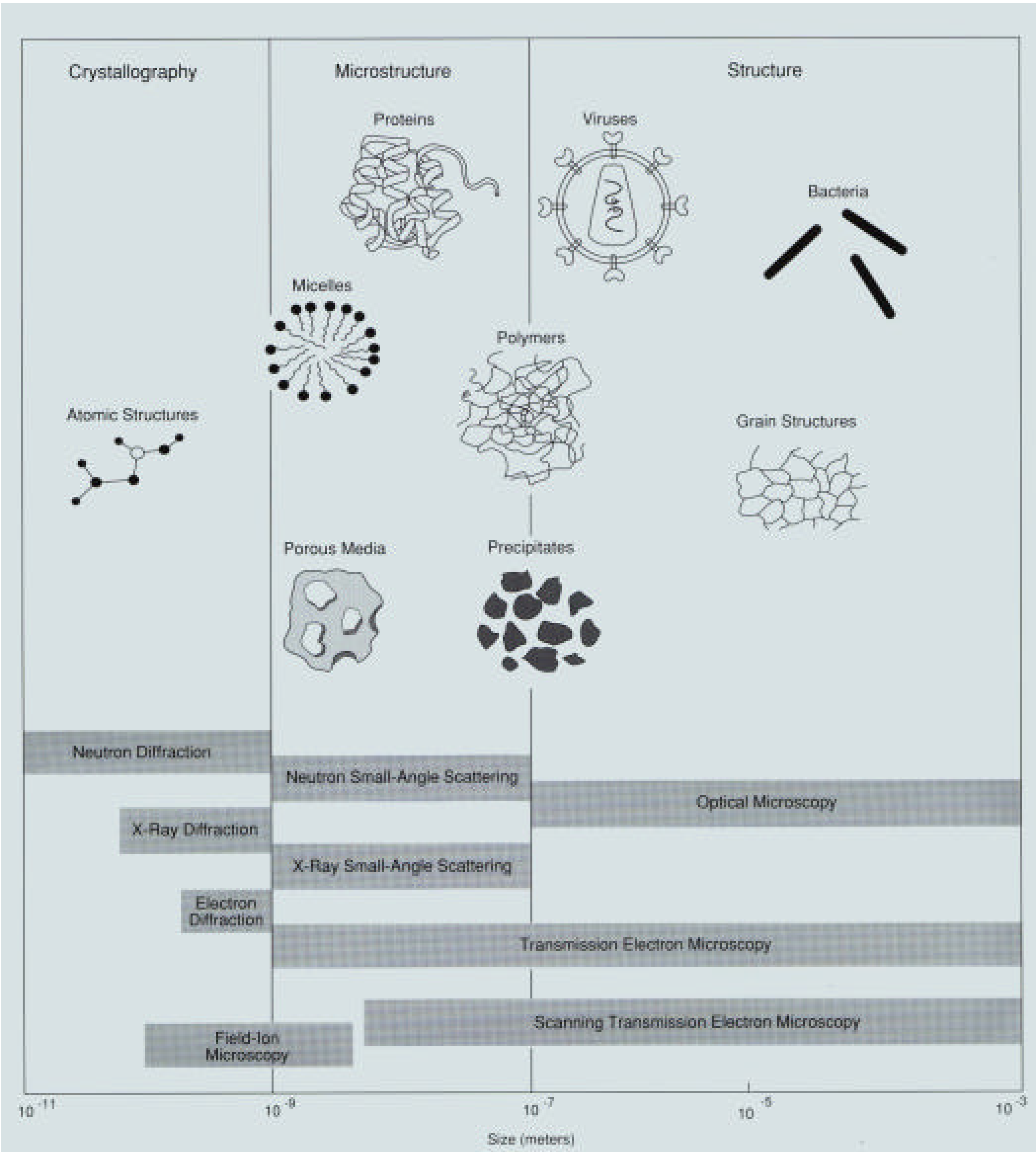
- Mass: $m_n = 1.675 \times 10^{-27}$ kg
- Charge = 0; Spin = ½
- Magnetic dipole moment: $\mu_n = - 1.913\ \mu_N$
- Nuclear magneton: $\mu_N = eh/4\pi m_p = 5.051 \times 10^{-27}\ J\ T^{-1}$
- Velocity (v), kinetic energy (E), wavevector (k), wavelength (λ), temperature (T).
- $E = m_n v^2/2 = k_B T = (hk/2\pi)^2/2m_n$; $k = 2\ \pi/\lambda = m_n v/(h/2\pi)$

	Energy (meV)	**Temp (K)**	**Wavelength (nm)**
Cold	**0.1 – 10**	**1 – 120**	**0.4 – 3**
Thermal	**5 – 100**	**60 – 1000**	**0.1 – 0.4**
Hot	**100 – 500**	**1000 – 6000**	**0.04 – 0.1**

l (nm) = 395.6 / v (m/s)

E (meV) = $0.02072\ k^2$ (k in nm^{-1})

Comparison of Structural Probes



Note that scattering methods provide statistically averaged information on structure rather than real-space pictures of particular instances

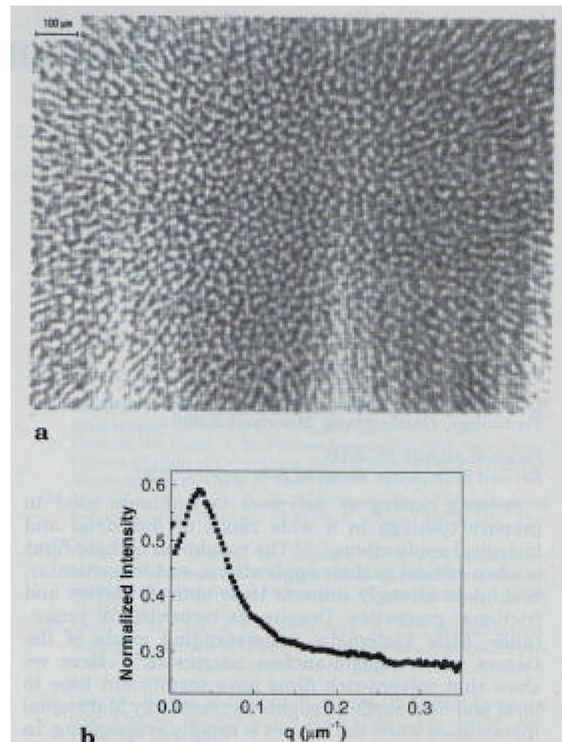
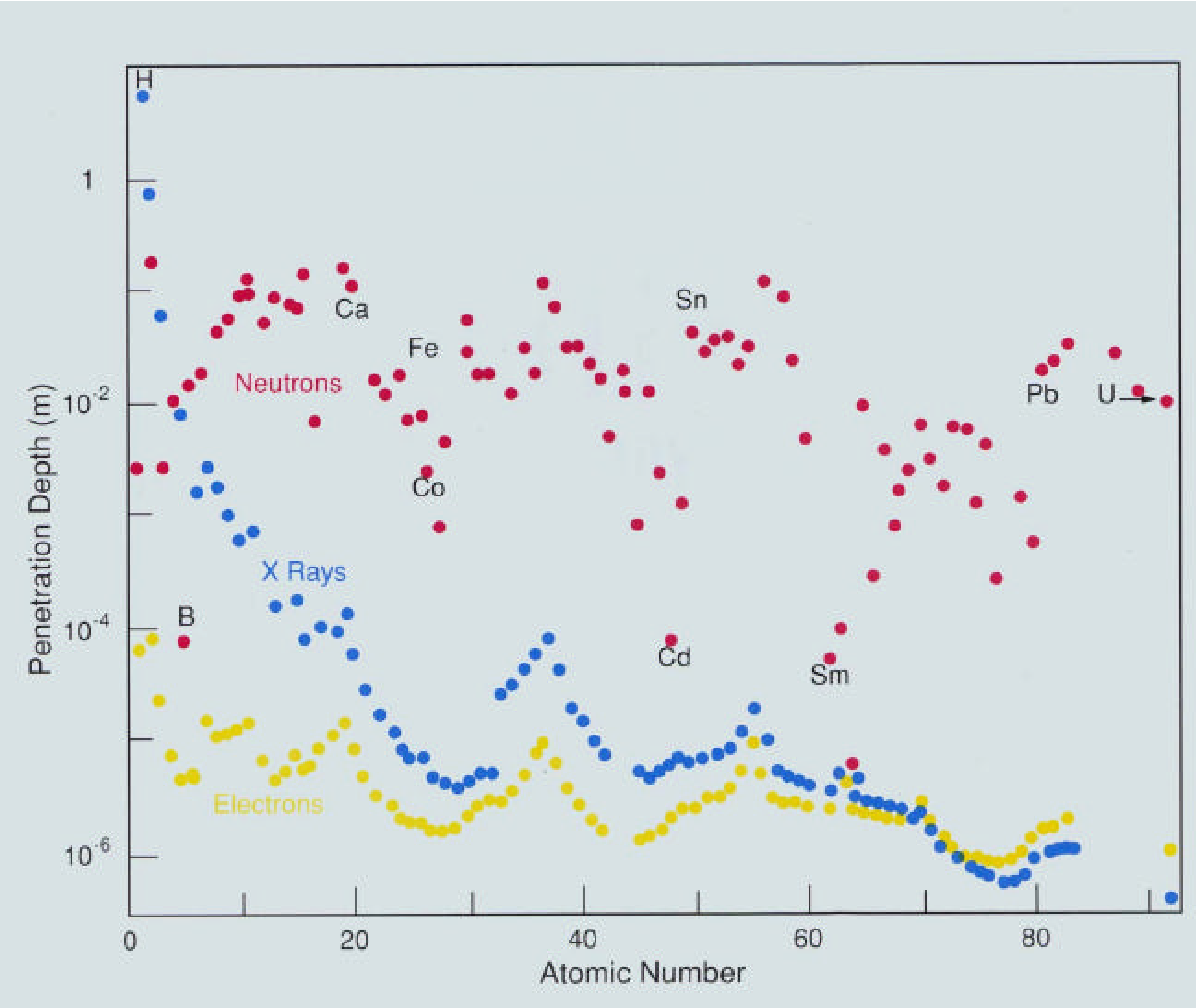


Figure 2. (a) Optical micrograph of PS-200 spin cast at 7500 rpm using THF (P_{vp} = 0.215 bar). (b) One-dimensional Fourier transform of central (isotropic) region of optical micrograph.

Macromolecules, 34, 4669 (2001)

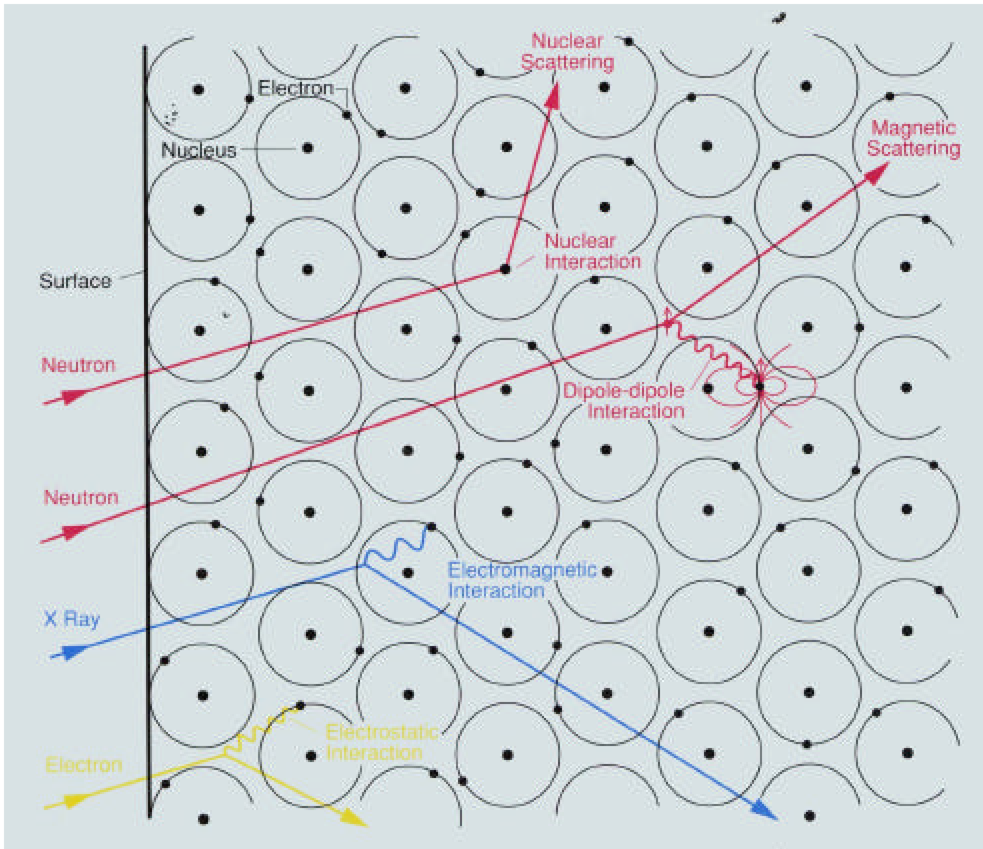
Thermal Neutrons, 8 keV X-Rays & Low Energy Electrons:- Absorption by Matter



Note for neutrons:

- H/D difference
- Cd, B, Sm
- no systematic A dependence

Interaction Mechanisms

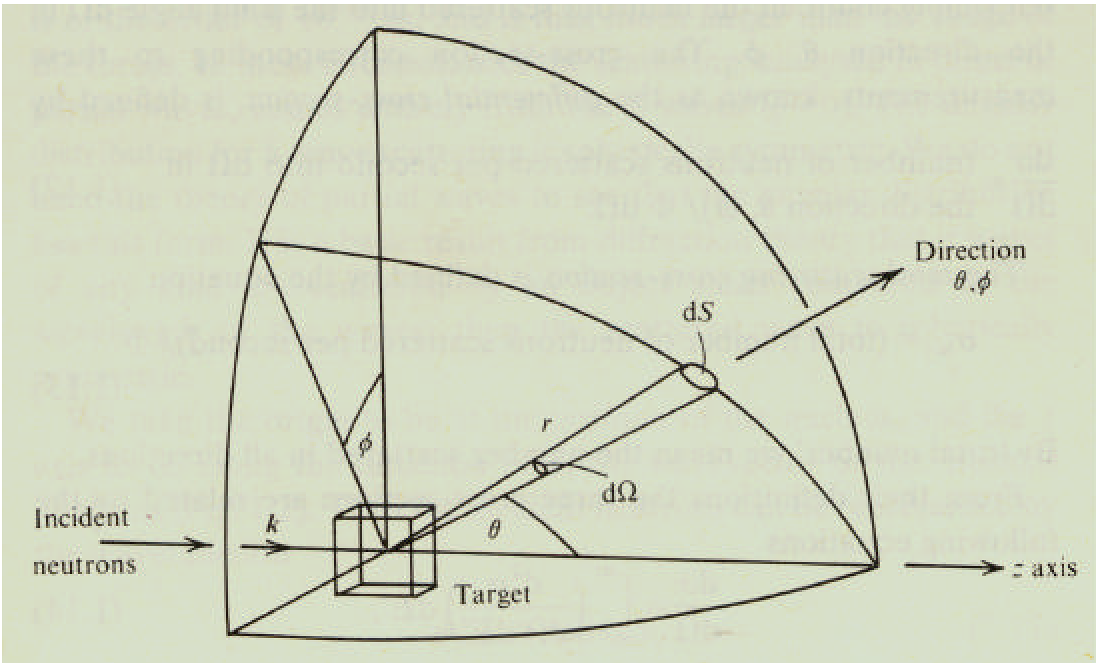


• Neutrons interact with atomic nuclei via very short range (~fm) forces.
• Neutrons also interact with unpaired electrons via a magnetic dipole interaction.

Brightness & Fluxes for Neutron & X-Ray Sources

	Brightness *(s^{-1} m^{-2} $ster^{-1}$)*	*dE/E* *(%)*	*Divergence* *($mrad^2$)*	*Flux* *(s^{-1} m^{-2})*
Neutrons	10^{15}	2	10 x 10	10^{11}
Rotating Anode	10^{16}	3	0.5 x 10	5×10^{10}
Bending Magnet	10^{24}	0.01	0.1 x 5	5×10^{17}
Wiggler	10^{26}	0.01	0.1 x 1	10^{19}
Undulator (APS)	10^{33}	0.01	0.01 x 0.1	10^{24}

Cross Sections

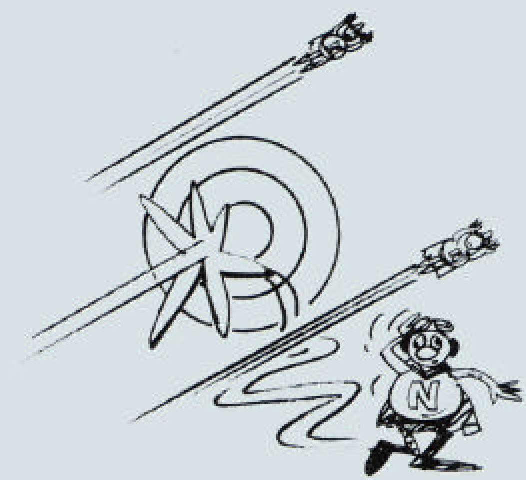


Φ = number of incident neutrons per cm^2 per second

s = total number of neutrons scattered per second / Φ

$$\frac{ds}{d\Omega} = \frac{\text{number of neutrons scattered per second into d}\Omega}{\Phi\ d\Omega}$$

$$\frac{d^2 s}{d\Omega dE} = \frac{\text{number of neutrons scattered per second into d}\Omega\ \&\ \text{dE}}{\Phi\ d\Omega\ dE}$$



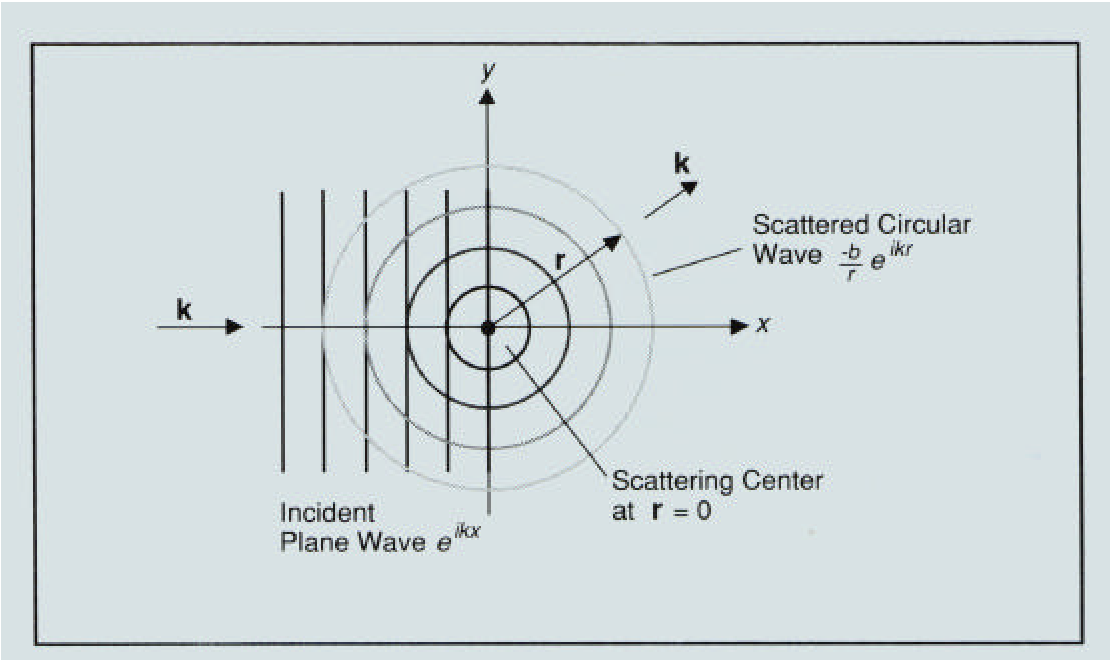
cross section

The effective area presented by a nucleus to an incident neutron. One unit for cross section is the barn, as in "can't hit the side of a barn!"

σ measured in barns:
1 barn = 10^{-24} cm^2

Attenuation = $\exp(-N\sigma t)$
N = # of atoms/unit volume
t = thickness

Scattering by a Single (fixed) Nucleus



- range of nuclear force (~ 1fm) is << neutron wavelength so scattering is "point-like"
- energy of neutron is too small to change energy of nucleus & neutron cannot transfer KE to a fixed nucleus => scattering is elastic
- we consider only scattering far from nuclear resonances where neutron absorption is negligible

If v is the velocity of the neutron (same before and after scattering), the number of neutrons passing through an area dS per second after scattering is :

$$v\, dS \left|\psi_{scat}\right|^2 = v\, dS\, b^2/r^2 = v\, b^2\, d\Omega$$

Since the number of incident neutrons passing through unit area is : $\Phi = v\left|\psi_{incident}\right|^2 = v$

$$\frac{d\sigma}{d\Omega} = \frac{v\, b^2\, d\Omega}{\Phi d\Omega} = b^2 \qquad \text{so } \sigma_{total} = 4\pi b^2$$

Adding up Neutrons Scattered by Many Nuclei

At a nucleus located at $\vec{R}_i$ the incident wave is $e^{i\vec{k}_0.\vec{R}_i}$

so the scattered wave is $\psi_{scat} = \sum e^{i\vec{k}_0.\vec{R}_i}\left[\frac{-b_i}{|\vec{r}-\vec{R}_i|}e^{i\vec{k}'.(\vec{r}-\vec{R}_i)}\right]$

$$\therefore \frac{d\sigma}{d\Omega} = \frac{v dS|\psi_{scat}|^2}{v d\Omega} = \frac{dS}{d\Omega}\left|b_i e^{i\vec{k}'.\vec{r}}\sum \frac{1}{|\vec{r}-\vec{R}_i|} e^{i(\vec{k}_0-\vec{k}').\vec{R}_i}\right|^2$$

If we measure far enough away so that $r >> R_i$ we can use $d\Omega = dS/r^2$ to get

$$\frac{d\sigma}{d\Omega} = \sum_{i,j} b_i b_j e^{i(\vec{k}_0-\vec{k}').(\vec{R}_i-\vec{R}_j)} = \sum_{i,j} b_i b_j e^{-i\vec{Q}.(\vec{R}_i-\vec{R}_j)}$$

where the wavevector transfer Q is defined by $\vec{Q} = \vec{k}' - \vec{k}_0$

Coherent and Incoherent Scattering

The scattering length, b_i , depends on the nuclear isotope, spin relative to the neutron & nuclear eigenstate. For a single nucleus:

$$b_i = \langle b \rangle + db_i \quad \text{where } d\text{b averages to zero}$$

$$b_i b_j = \langle b \rangle^2 + \langle b \rangle (db_i + db_j) + db_i db_j$$

$$\text{but } \langle db \rangle = 0 \text{ and } \langle db_i db_j \rangle \text{ vanishes unless i = j}$$

$$\langle db_i^2 \rangle = \langle b_i - \langle b \rangle \rangle^2 = \langle b^2 \rangle - \langle b \rangle^2$$

$$\therefore \frac{ds}{d\Omega} = \langle b \rangle^2 \sum_{i,j} e^{-i\vec{Q}.(\vec{R}_i - \vec{R}_j)} + (\langle b^2 \rangle - \langle b \rangle^2) N$$

Coherent Scattering
(scattering depends on the direction of **Q**)

Incoherent Scattering
(scattering is uniform in all directions)

Note: N = number of atoms in scattering system

Values of σ_{coh} and σ_{inc}

Nuclide	s_{coh}	s_{inc}	Nuclide	s_{coh}	s_{inc}
^{1}H	1.8	80.2	V	0.02	5.0
^{2}H	5.6	2.0	Fe	11.5	0.4
C	5.6	0.0	Co	1.0	5.2
O	4.2	0.0	Cu	7.5	0.5
Al	1.5	0.0	^{36}Ar	24.9	0.0

- Difference between H and D used in experiments with soft matter (contrast variation)
- Al used for windows
- V used for sample containers in diffraction experiments and as calibration for energy resolution
- Fe and Co have nuclear cross sections similar to the values of their magnetic cross sections
- Find scattering cross sections at the NIST web site at:

  http://webster.ncnr.nist.gov/resources/n-lengths/

Coherent Elastic Scattering measures the Structure Factor S(Q) I.e. correlations of atomic positions

$\frac{ds}{d\Omega} = \langle b \rangle^2 N.S(\vec{Q})$ for an assembly of similar atoms where $S(\vec{Q}) = \frac{1}{N}\left\langle \sum_{i,j} e^{-i\vec{Q}.(\vec{R}_i - \vec{R}_j)} \right\rangle_{\text{ensemble}}$

Now $\sum_i e^{-i\vec{Q}.\vec{R}_i} = \int d\vec{r}.e^{-i\vec{Q}.\vec{r}} \sum_i d(\vec{r} - \vec{R}_i) = \int d\vec{r}.e^{-i\vec{Q}.\vec{r}} r_N(\vec{r})$ where r_N is the nuclear number density

so $$S(\vec{Q}) = \frac{1}{N}\left\langle \left| \int d\vec{r}.e^{-i\vec{Q}.\vec{r}} r_N(\vec{r}) \right|^2 \right\rangle$$

or $S(\vec{Q}) = \frac{1}{N}\int d\vec{r}' \int d\vec{r}.e^{-i\vec{Q}.(\vec{r}-\vec{r}')} \langle r_N(\vec{r}) r_N(\vec{r}') \rangle = \frac{1}{N}\int d\vec{R} \int d\vec{r} e^{-i\vec{Q}.\vec{R}} \langle r_N(\vec{r}) r_N(\vec{r} - \vec{R}) \rangle$

ie $$S(\vec{Q}) = 1 + \int d\vec{R}.g(\vec{R}).e^{-i\vec{Q}.\vec{R}}$$

where $$g(\vec{R}) = \sum_{i \neq 0} \langle d(\vec{R} - \vec{R}_i + \vec{R}_0) \rangle$$ is a function of $\vec{R}$ only.

g(R) is known as the **static pair correlation function**. It gives the probability that there is an atom, i, at distance R from the origin of a coordinate system at time t, given that there is also a (different) atom at the origin of the coordinate system

S(Q) and g(r) for Simple Liquids

- Note that S(Q) and g(r)/ρ both tend to unity at large values of their arguments
- The peaks in g(r) represent atoms in “coordination shells”
- g(r) is expected to be zero for r < particle diameter – ripples are truncation errors from Fourier transform of S(Q)

Fig. 5.1 The structure factor $S(\kappa)$ for ^{36}Ar at 85 K. The curve through the experimental points is obtained from a molecular dynamics calculation of Verlet based on a Lennard-Jones potential. (After Yarnell *et al.*, 1973.)

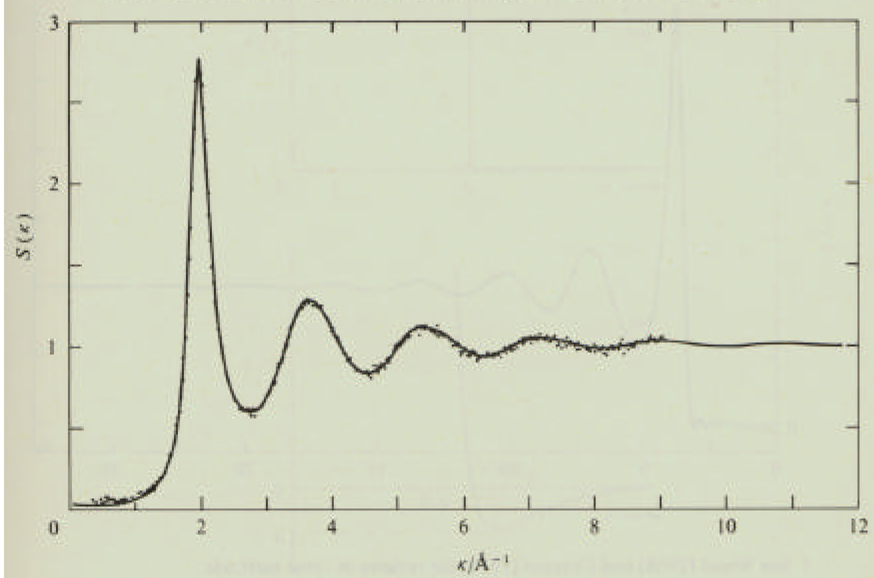
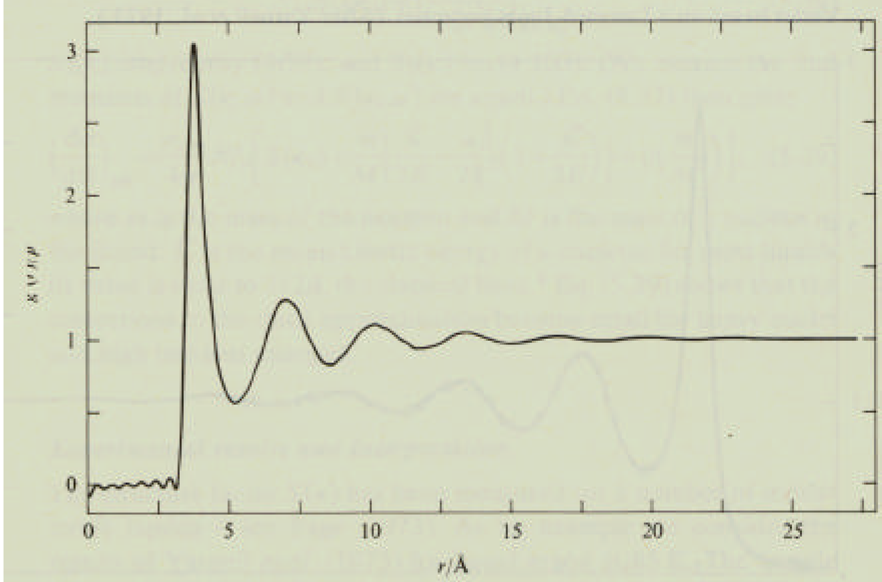
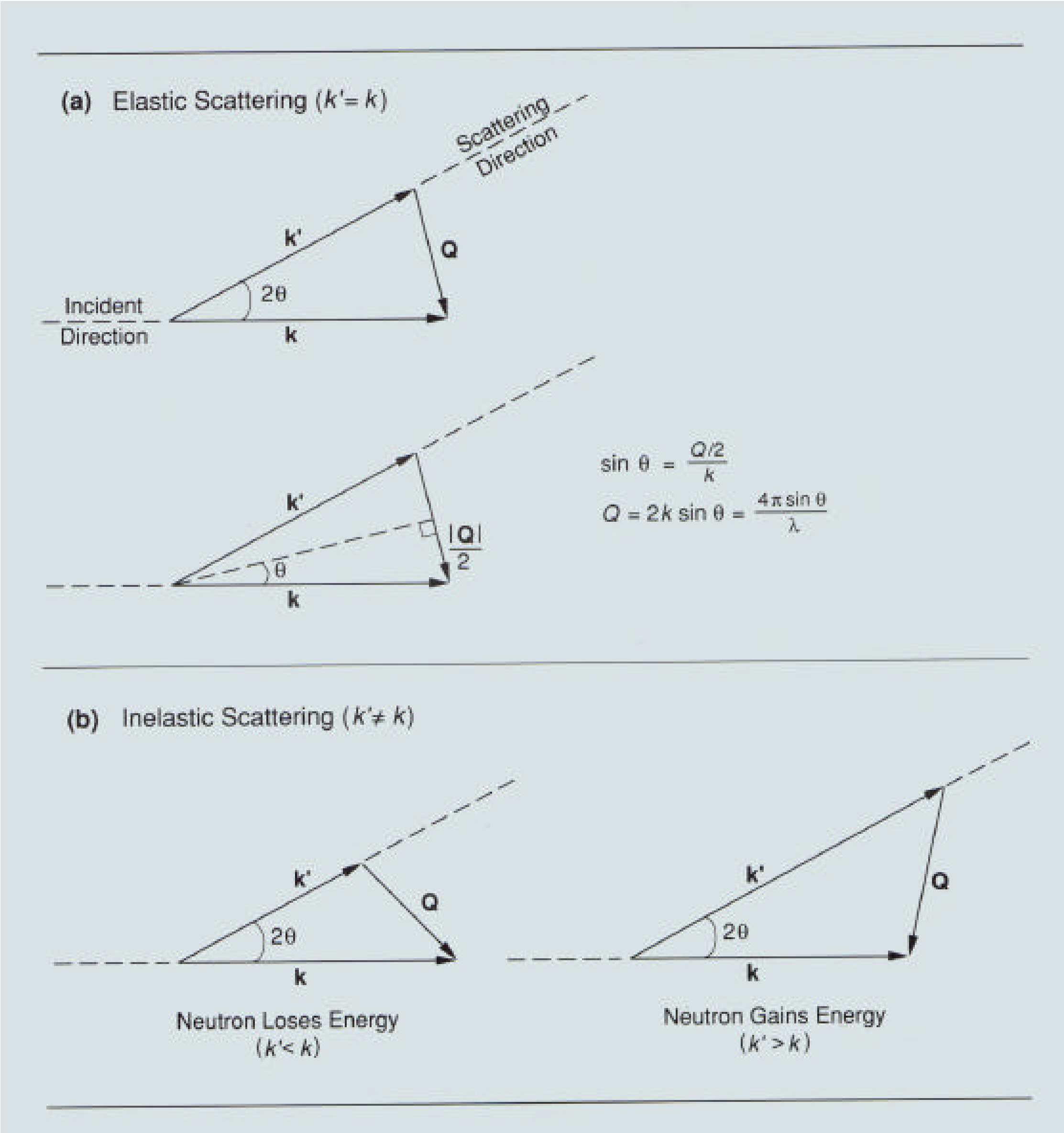
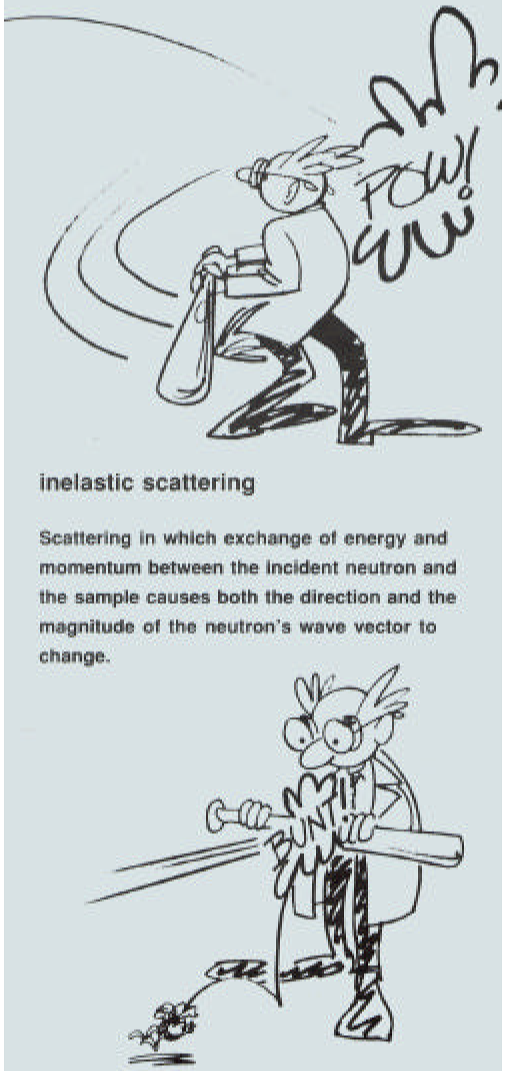


Fig. 5.2 The pair-distribution function $g(r)$ obtained from the experimental results in Fig. 5.1. The mean number density is $\rho = 2.13 \times 10^{28}$ atoms m^{-3}. (After Yarnell *et al.*, 1973.)



Neutrons can also gain or lose energy in the scattering process: this is called inelastic scattering





inelastic scattering

Scattering in which exchange of energy and momentum between the incident neutron and the sample causes both the direction and the magnitude of the neutron's wave vector to change.

Inelastic neutron scattering measures atomic motions

The concept of a pair correlation function can be generalized:

G(r,t) = probability of finding a nucleus at (r,t) given that there is one at r=0 at t=0

G_s(r,t) = probability of finding a nucleus at (r,t) if the *same* nucleus was at r=0 at t=0

Then one finds:

$$\left(\frac{d^2 s}{d\Omega . dE}\right)_{coh} = b_{coh}^2 \frac{k'}{k} N S(\vec{Q}, w)$$

$$\left(\frac{d^2 s}{d\Omega . dE}\right)_{inc} = b_{inc}^2 \frac{k'}{k} N S_i(\vec{Q}, w)$$

(h/2π)**Q** & (h/2π)ω are the momentum & energy transferred to the neutron during the scattering process

where

$$S(\vec{Q}, w) = \frac{1}{2p\hbar} \iint G(\vec{r}, t) e^{i(\vec{Q}.\vec{r} - wt)} d\vec{r} dt \quad \text{and} \quad S_i(\vec{Q}, w) = \frac{1}{2p\hbar} \iint G_s(\vec{r}, t) e^{i(\vec{Q}.\vec{r} - wt)} d\vec{r} dt$$

Inelastic coherent scattering measures *correlated* motions of atoms

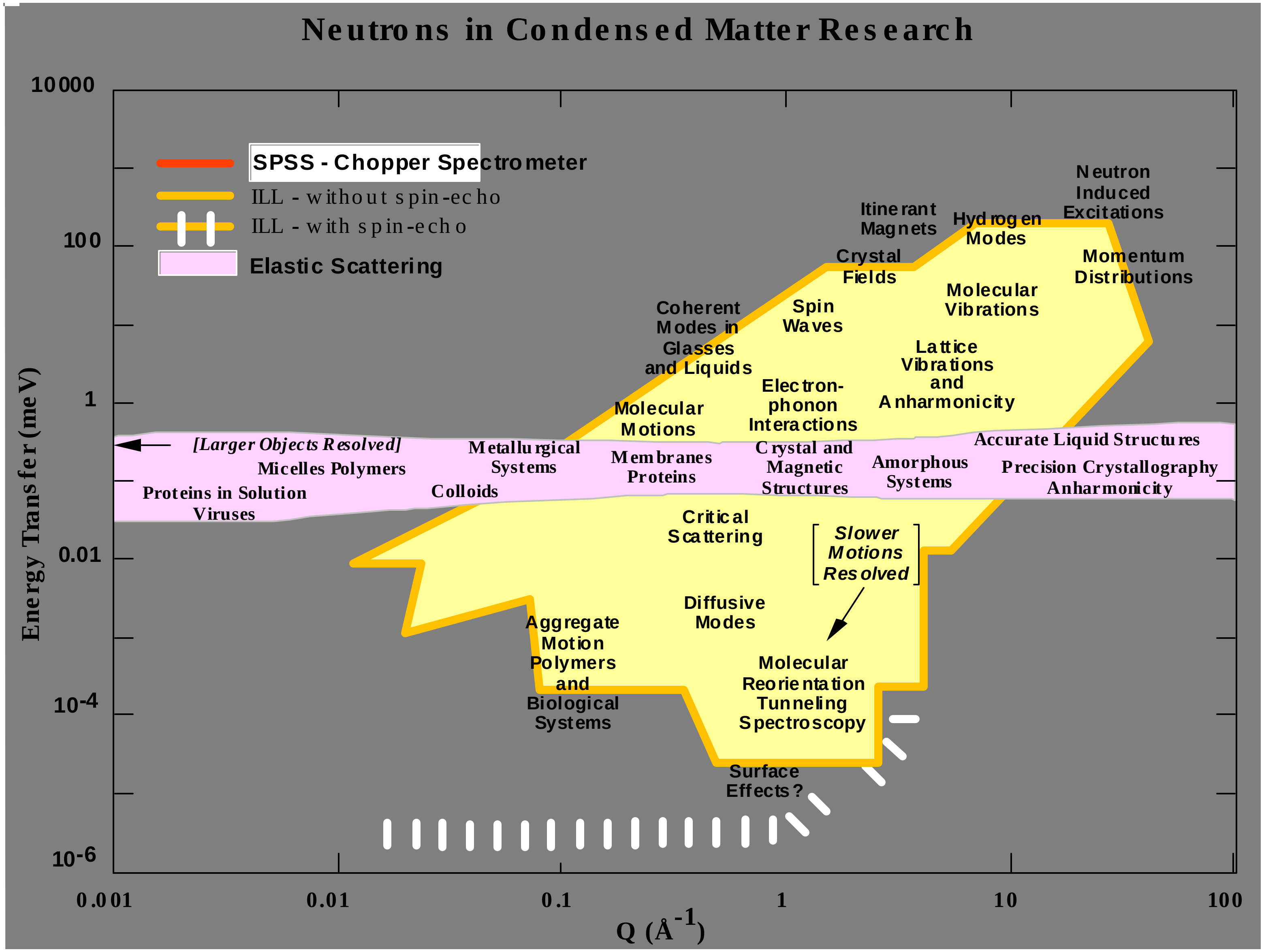
Inelastic incoherent scattering measures *self-correlations* e.g. diffusion

Magnetic Scattering

- The magnetic moment of the neutron interacts with *B* fields caused, for example, by unpaired electron spins in a material
  - Both spin and orbital angular momentum of electrons contribute to *B*
  - Expressions for cross sections are more complex than for nuclear scattering
    - Magnetic interactions are long range and non-central
  - Nuclear and magnetic scattering have similar magnitudes
  - Magnetic scattering involves a form factor – FT of electron spatial distribution
    - Electrons are distributed in space over distances comparable to neutron wavelength
    - Elastic magnetic scattering of neutrons can be used to probe electron distributions
  - Magnetic scattering depends *only* on component of *B* perpendicular to Q
  - For neutrons spin polarized along a direction z (defined by applied H field):
    - Correlations involving B_z do not cause neutron spin flip
    - Correlations involving B_x or B_y cause neutron spin flip
  - Coherent & incoherent nuclear scattering affects spin polarized neutrons
    - Coherent nuclear scattering is non-spin-flip
    - Nuclear spin-incoherent nuclear scattering is 2/3 spin-flip
    - Isotopic incoherent scattering is non-spin-flip

Magnetic Neutron Scattering is a Powerful Tool

- In early work Shull and his collaborators:
  - Provided the first direct evidence of antiferromagnetic ordering
  - Confirmed the Neel model of ferrimagnetism in magnetite (Fe_3O_4)
  - Obtained the first magnetic form factor (spatial distribution of magnetic electrons) by measuring paramagnetic scattering in Mn compounds
  - Produced polarized neutrons by Bragg reflection (where nuclear and magnetic scattering scattering cancelled for one neutronspin state)
  - Determined the distribution of magnetic moments in 3d alloys by measuring diffuse magnetic scattering
  - Measured the magnetic critical scattering at the Curie point in Fe
- More recent work using polarized neutrons has:
  - Discriminated between longitudinal & transverse magnetic fluctuations
  - Provided evidence of magnetic solitons in 1-d magnets
  - Quantified electron spin fluctuations in correlated-electron materials
  - Provided the basis for measuring slow dynamics using the neutron spin-echo technique…..etc



Neutron scattering experiments measure the number of neutrons scattered at different values of the wavevector and energy transfered to the neutron, denoted Q and E. The phenomena probed depend on the values of Q and E accessed.

Next Lecture

2. Neutron Scattering Instrumentation and Facilities – how is neutron scattering measured?
   1. Sources of neutrons for scattering – reactors & spallation sources
      1. Neutron spectra
      2. Monochromatic-beam and time-of-flight methods
   2. Instrument components
      1. Crystal monochromators and analysers
      2. Neutron guides
      3. Neutron detectors
      4. Neutron spin manipulation
      5. Choppers
      6. etc
   3. A zoo of specialized neutron spectrometers

References

- Introduction to the Theory of Thermal Neutron Scattering
  by G. L. Squires
  Reprint edition (February 1997)
  Dover Publications
  ISBN 048669447

- Neutron Scattering: A Primer
  by Roger Pynn
  Los Alamos Science (1990)
  (see www.mrl.ucsb.edu/~pynn)