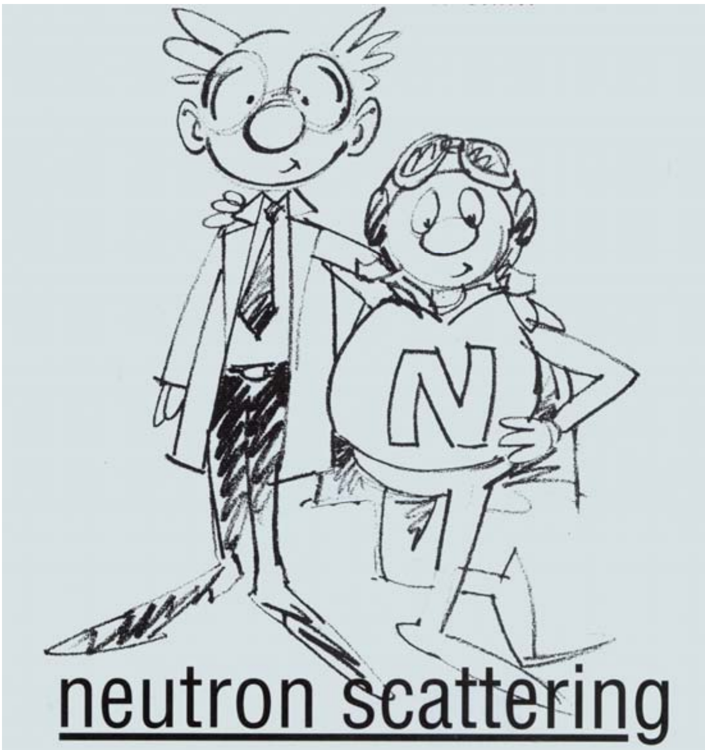


by

Roger Pynn

LECTURES 1 and 2: Theory of Nuclear Scattering of Neutrons

Overview

Introduction and theory of neutron scattering

- Advantages/disadvantages of neutrons for scattering measurements
- Neutron properties
- Comparison with other structural probes
- Definition of scattering cross sections
- Fermi pseudopotential
- Refractive index for neutrons – two different calculations
- Elastic scattering and definition of the structure factor, S(Q)
- Coherent & incoherent scattering
- Inelastic scattering
- References

Why do Neutron Scattering?

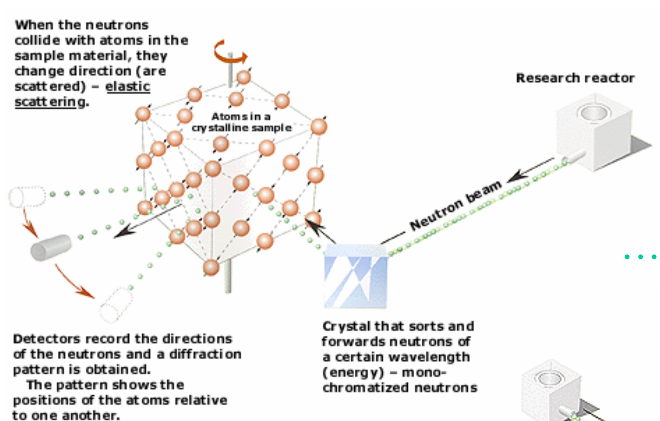
- To determine the positions and motions of atoms in condensed matter
  - 1994 Nobel Prize to Shull and Brockhouse cited these areas
    (see http://www.nobel.se/physics/educational/poster/1994/neutrons.html)

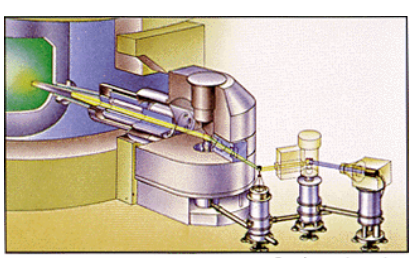
- Neutron advantages:
  - Wavelength comparable with interatomic spacings
  - Kinetic energy comparable with that of atoms in a solid
  - Penetrating => bulk properties are measured & sample can be contained
  - Weak interaction with matter aids interpretation of scattering data
  - Isotopic sensitivity allows contrast variation
  - Neutron magnetic moment couples to $\boldsymbol{B}$ => 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)

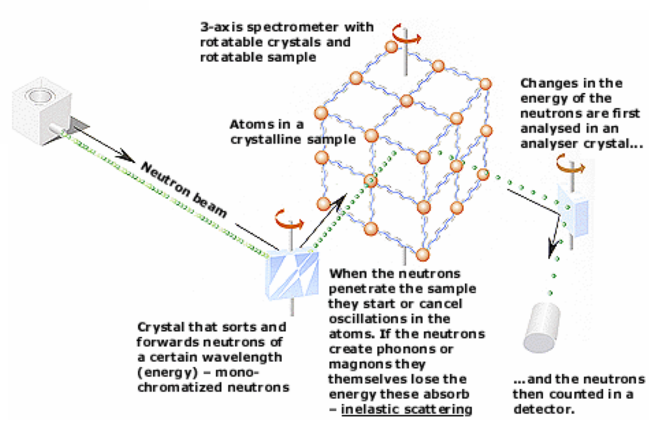
The 1994 Nobel Prize in Physics – Shull & Brockhouse

Neutrons show where the atoms are….





…and what the atoms do.



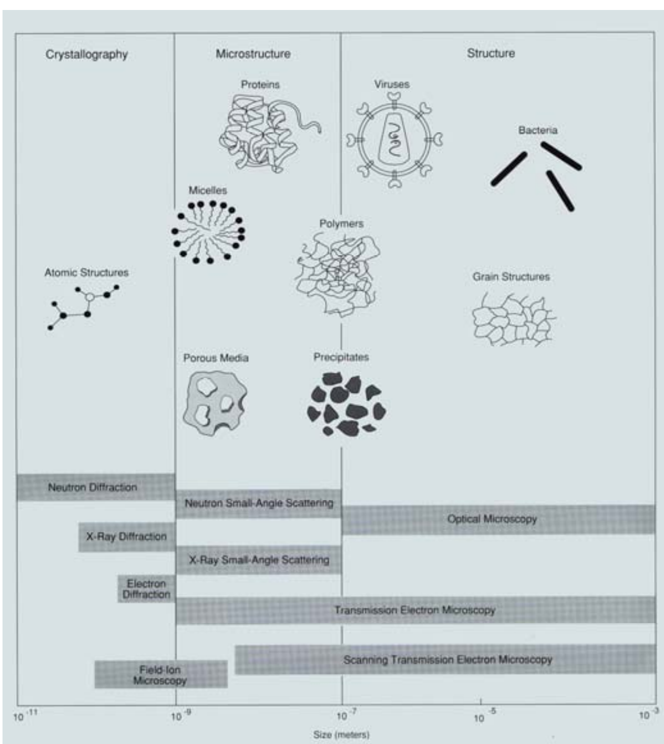
The Neutron has Both Particle-Like and Wave-Like Properties

- Mass: $m_n = 1.675 \times 10^{-27}$ kg
- Charge = 0; Spin = ½
- Magnetic dipole moment: $\mu_n = -1.913\ \mu_N$
- Nuclear magneton: $\mu_N = eh/4\pi m_p = 5.051 \times 10^{-27}\ \mathrm{J\ T^{-1}}$
- 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**

λ (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

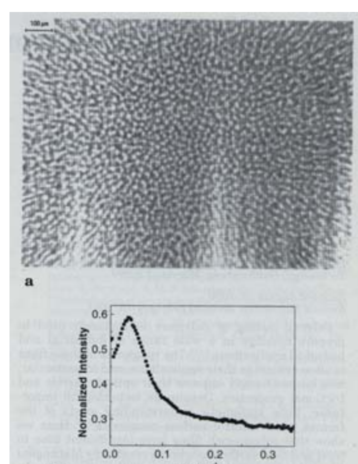
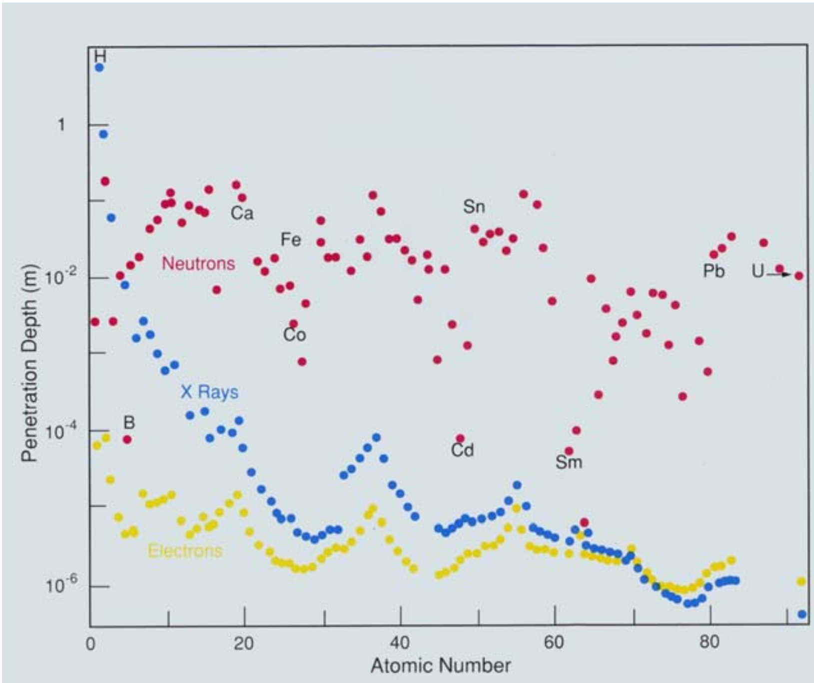


Figure 2. (a) Optical micrograph of PS-200 spin cast at 7500 rpm using THF (P_{vp} = 0.215 bar). (b) One-dimensional Fourier transform of central (isotropic) region of optical micrograph.

Macromolecules, 34, 4669 (2001)

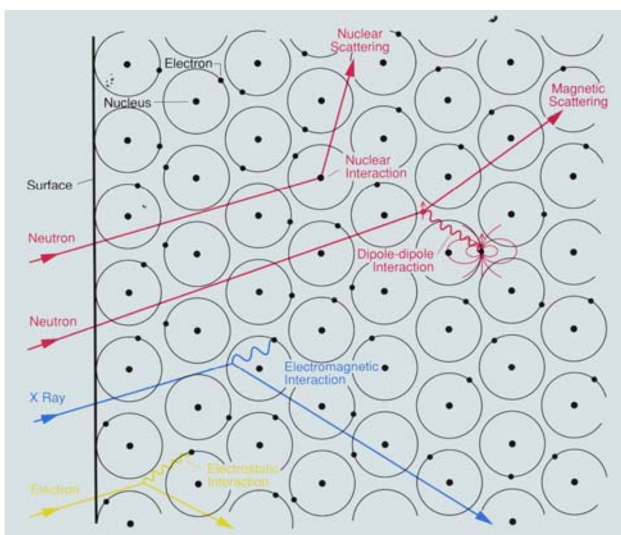
Thermal Neutrons, 8 keV X-Rays & Low Energy Electrons:- Absorption by Matter



Note for neutrons:

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

Interaction Mechanisms

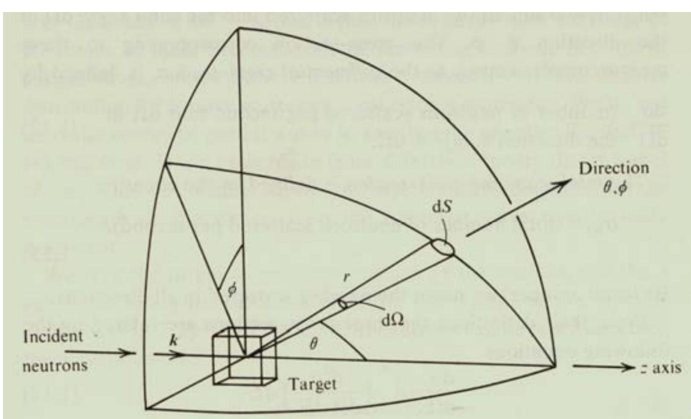


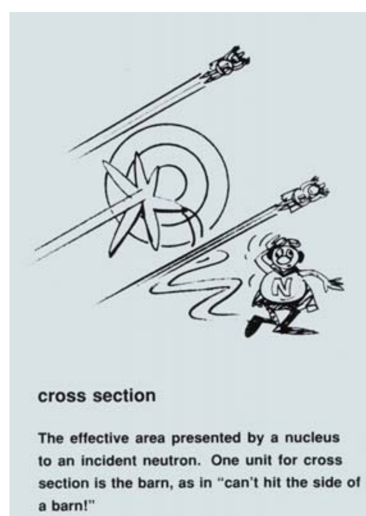
- Neutrons interact with atomic nuclei via very short range (~fm) forces.
- Neutrons also interact with unpaired electrons via a magnetic dipole interaction.

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

σ = total number of neutrons scattered per second / Φ

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

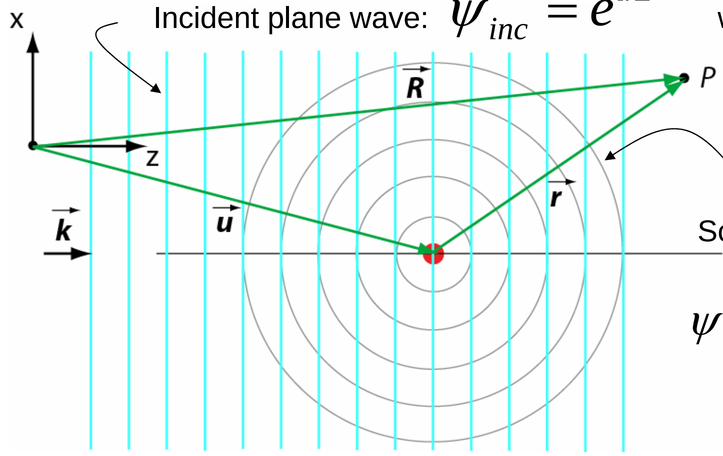
$$\frac{d^2\sigma}{d\Omega dE} = \frac{\text{number of neutrons scattered per second into } d\Omega \text{ \& } dE}{\Phi \, d\Omega \, dE}$$

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 nucleus

Incident plane wave: $\psi_{inc} = e^{ikz}$ where $k = \frac{2\pi}{\lambda}$



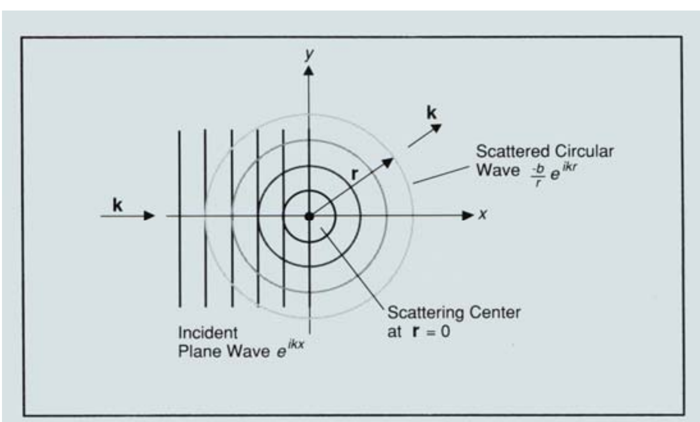
Scattered (circular) wave:

$$\psi_s = -\frac{\bar{b}}{r} e^{ikr} e^{ikz}$$

$$= -\frac{\bar{b}}{\left|\vec{R}-\vec{u}\right|} e^{ik\left|\vec{R}-\vec{u}\right|} e^{ikz}$$

Squires Eq. 1.18 generalized for the origin at any location.

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 :

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

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

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

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

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

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 $\vec{Q} = \vec{k}' - \vec{k}_0$

The Fermi Pseudo-Potential

$\frac{d\sigma}{d\Omega} = \frac{1}{\Phi}\frac{1}{d\Omega}\sum_{\vec{k}'\text{ in }d\Omega} W_{\vec{k}\to\vec{k}'}$ where the sum is over probabilities of all transitions

By Fermi's Golden Rule: $\sum_{\vec{k}'\text{ in }d\Omega} W_{\vec{k}\to\vec{k}'} = \frac{2\pi}{\hbar}\rho_{\vec{k}'}\left|\langle\vec{k}'|V|\vec{k}\rangle\right|^2 = \frac{2\pi}{\hbar}\rho_{\vec{k}'}\frac{1}{Y^2}\left|\int_Y V(\vec{r})e^{i(\vec{k}-\vec{k}').\vec{r}}d\vec{r}\right|^2$

where $\rho_{\vec{k}'}$ is # of momentum states in $d\Omega$, per unit energy, for neutrons in state $\vec{k}'$

Using standard "box normalization", the volume per k state is $(2\pi)^3/Y$ where Y = box volume

Final neutron energy is $E' = \frac{\hbar^2 k'^2}{2m} \Rightarrow dE' = \frac{\hbar^2 k' dk'}{m}$ so

$\rho_{k'} dE' =$ number of wavevector states in volume $k'^2 dk' d\Omega = \frac{Y}{(2\pi)^3}k'^2 dk' d\Omega$

i.e. $\rho_{\vec{k}'} = \frac{\text{number of wavevector states}}{dE'} = \frac{Y}{(2\pi)^3}k'\frac{m}{\hbar^2}d\Omega$

Further, Φ = incident flux = density x velocity $= \frac{1}{Y}\frac{\hbar}{m}k$

So, $\frac{d\sigma}{d\Omega} = \frac{Y}{k}\frac{m}{\hbar}\frac{1}{d\Omega}\frac{2\pi}{\hbar}\frac{Y}{(2\pi)^3}k'\frac{m}{\hbar^2}d\Omega\left|\langle\vec{k}'|V|\vec{k}\rangle\right|^2 = \left(\frac{m}{2\pi\hbar^2}\right)^2\left|\int V(\vec{r})e^{i(\vec{k}-\vec{k}').\vec{r}}d\vec{r}\right|^2$ so $V(\vec{r}) = \frac{2\pi\hbar^2}{m}b\delta(\vec{r})$

Fermi pseudopotential

Use V(r) to Calculate the Refractive Index for Neutrons

The nucleus - neutron potential is given by : $V(\vec{r}) = \frac{2\pi\hbar^2}{m} b\delta(\vec{r})$ for a single nucleus.

So the average potential inside the medium is : $\bar{V} = \frac{2\pi\hbar^2}{m}\rho$ where $\rho = \frac{1}{volume}\sum_i b_i$

ρ is called the nuclear Scattering Length Density (SLD) - used for SANS & reflectometry

The kinetic (and total) energy of neutron in vaccuum is $E = \frac{\hbar^2 k_0^2}{2m}$

Inside the medium the total energy is $\frac{\hbar^2 k^2}{2m} + \bar{V}$

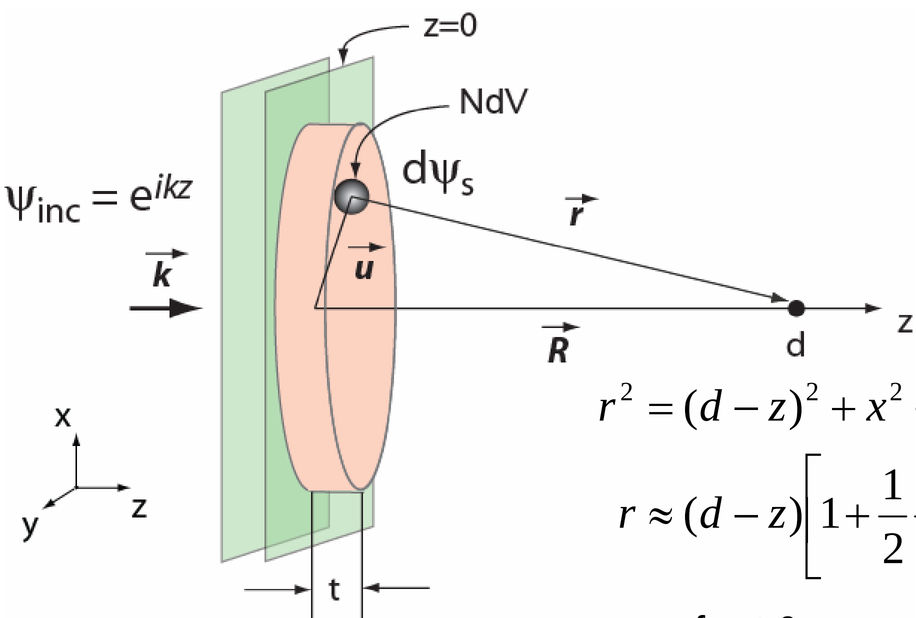
Conservation of energy gives $\frac{\hbar^2 k_0^2}{2m} = \frac{\hbar^2 k^2}{2m} + \bar{V} = \frac{\hbar^2 k^2}{2m} + \frac{2\pi\hbar^2}{m}\rho$ or $k_0^2 - k^2 = 4\pi\rho$

Since $k/k_0 = n =$ refractive index (by definition), and ρ is very small ($\sim 10^{-6}$ A^{-2}) we get :

$$n = 1\text{-}\lambda^2\rho/2\pi$$

Since generally n < 1, neutrons are externally reflected from most materials.

Now let's do it the Hard Way – Calculate the Scattered Wavefunction….



$$r^2 = (d-z)^2 + x^2 + y^2$$

$$r \approx (d-z)\left[1 + \frac{1}{2}\frac{x^2+y^2}{(d-z)^2}\right]$$

for *t* & *u* << *d*

Viewgraph from M. R. Fitzsimmons

$$d\psi_s = -\frac{\bar{b}}{\left|\vec{R}-\vec{u}\right|} e^{ik\left|\vec{R}-\vec{u}\right|} e^{ikz} N dV$$

$$\approx -\frac{N\bar{b}}{d-z} e^{ikd} e^{ik\frac{x^2+y^2}{2(d-z)}} dV$$

$$\psi_s = -\rho e^{ikd} \int_0^t dz \frac{1}{d-z} \int e^{ik\frac{y^2}{2(d-z)}} dy \int e^{ik\frac{x^2}{2(d-z)}} dx$$

A function with interesting consequences.

$$= -\rho e^{ikd} \int_0^t dz \frac{1}{d-z} \frac{i2\pi(d-z)}{k}$$

p.66 of Als-Nielsen & McMorrow

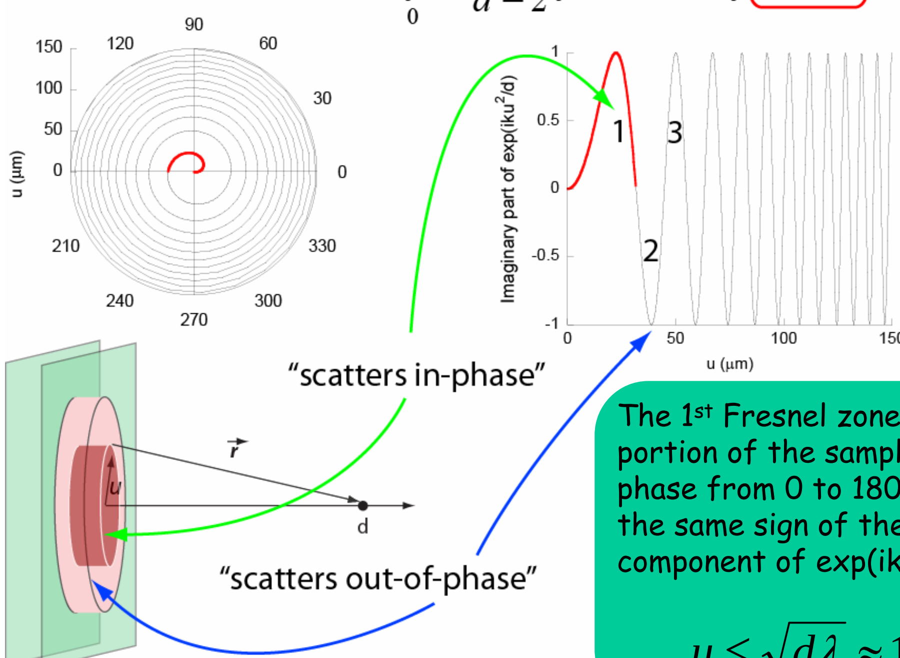
$$\psi_s = -i\rho\lambda t e^{ikd}$$

The scattered wave function at "d" from a plate of material illuminated by a plane wave.

Viewgraph from M. R. Fitzsimmons

Definition of the 1st Fresnel zone

Recall: $\psi_s = -\rho e^{ikd} \int_0^t dz \frac{1}{d-z} \int e^{ik\frac{y^2}{2(d-z)}} dy \int e^{ik\frac{x^2}{2(d-z)}} dx$



d = 1 m
λ = 1 nm

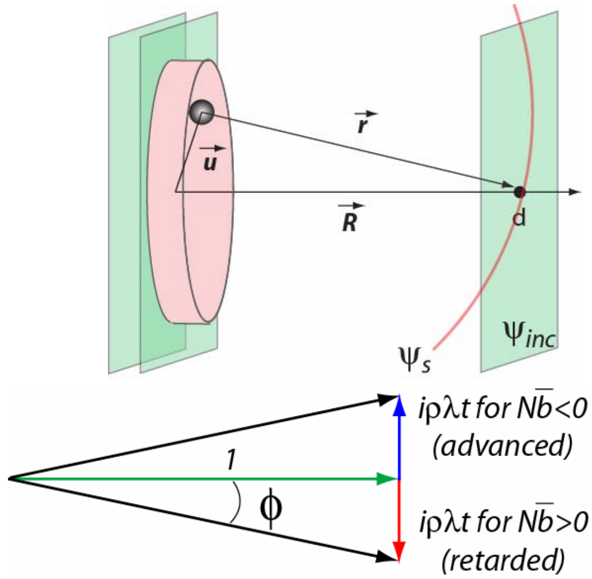
The 1st Fresnel zone is the portion of the sample yielding phase from 0 to 180°, i.e., having the same sign of the imaginary component of exp(iku²/d).

$$u \le \sqrt{d\lambda} \approx 10\mu\text{m}$$

Viewgraph from M. R. Fitzsimmons

$$\psi_s(d) = -i\rho\lambda t e^{ikd}$$
$$\psi_t(d) = \psi_{inc} + \psi_s$$
$$= (1 - i\rho\lambda t)e^{ikd}$$
$$\psi_t(d) \approx e^{-i\rho\lambda t} e^{ikd}$$

(scattering approach)

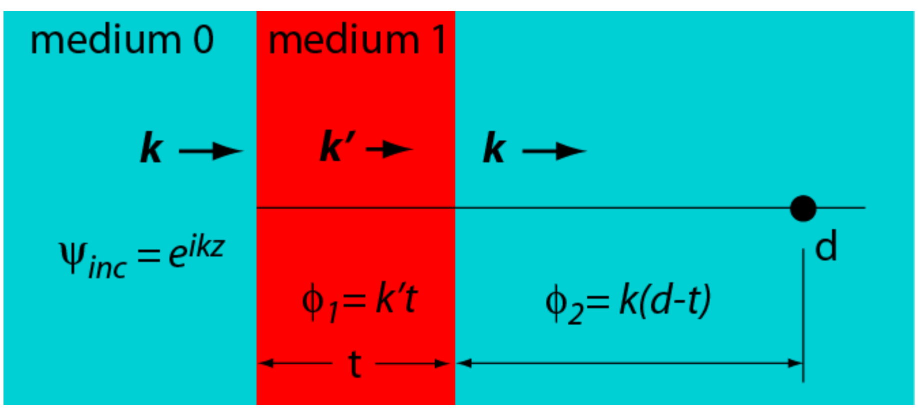


(1) The phase of the wave function in the forward direction ψ_t is related to the average (macroscopic) properties of the material, i.e., ρt, and not its atomic structure, e.g., crystalline, liquid, etc.

(2) The phase can be retarded or advanced depending upon the sign of $\bar{b}$, since ρ = Nb.

Viewgraph from M. R. Fitzsimmons

Let's Calculate $\psi_t(d)$ using a Macroscopic Approach



Relative to the front surface of Medium 1, the accumulated phase at **d**, ϕ(d) is:

$$\phi = \phi_1 + \phi_2$$
$$= k't + k(d-t)$$

Viewgraph from M. R. Fitzsimmons

From optics theory (Snell's law), the perpendicular components of the wave vectors across an interface are related by the index of refraction *n*:

$$k'_{\perp} = nk_{\perp}$$

so

$$\phi = k't + k(d-t)$$
$$= nkt + k(d-t)$$
$$\phi = (n-1)kt + kd$$
$$\Rightarrow \psi_t = e^{i(n-1)kt} e^{ikd}$$

This is the result obtained from the macroscopic (optics) approach

Viewgraph from M. R. Fitzsimmons

Equating the Scattering & Macroscopic Approaches…

$$\psi_t = e^{-i\rho\lambda t} e^{ikd}$$ (scattering approach)

$$\psi_t = e^{i(n-1)kt} e^{ikd}$$ (macroscopic approach)

$$(n-1)kt = \rho\lambda t$$

$$\Rightarrow \boxed{n = 1 - \frac{1}{2\pi}\rho\lambda^2}$$

Index of refraction – same result as before

	N (atoms/Å^3)	b (x10^{-5} Å)	n	
Ni	0.09	10.3	1-2x10^{-5}	Phase retarded, $\lambda_1>\lambda_0$
Mn	0.08	-3.7	1+0.8x10^{-5}	Phase advanced, $\lambda_1<\lambda_0$

Viewgraph from M. R. Fitzsimmons

Why do we Care about the Refractive Index?

- When the wavevector transfer **Q** is small, the phase factors in the cross section do not vary much from nucleus to nucleus & we can use a continuum approximation

- We can use all of the apparatus of optics to calculate effects such as:
  - External reflection from single surfaces (for example from guide surfaces)
  - External reflection from multilayer stacks (including supermirrors)
  - Focusing by (normally) concave lenses or Fresnel lenses
  - The phase change of the neutron wave through a material for applications such as interferometry or phase radiography
  - Fresnel edge enhancement in radiography

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:

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

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

$$\text{but } \langle \delta b \rangle = 0 \text{ and } \langle \delta b_i \delta b_j \rangle \text{ vanishes unless i = j}$$

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

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

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

Incoherent Scattering
(scattering is uniform in all directions)

Note: N = number of atoms in scattering system

Nuclear Spin Incoherent Scattering

Consider a single isotope with spin I. The spin of the nucleus - neutron system can be $(I+1/2)$ or $(I-1/2)$.

The number of states with spin $(I+1/2)$ is $2(I+1/2)+1=2I+2$

The number of states with spin $(I-1/2)$ is $2(I-1/2)+1=2I$

If the neutrons and the nuclear spins are unpolarized, each spin state has the same *a priori* probability.

The frequency of occurence of b^+ state is $f^+=(2I+2)/(4I+2)$

The frequency of occurence of b^- state is $f^-=(2I)/(4I+2)$

Thus $\langle b\rangle=\frac{1}{2I+1}\left[(I+1)b^+ + Ib^-\right]$ and $\langle b^2\rangle=\frac{1}{2I+1}\left[(I+1)(b^+)^2+I(b^-)^2\right]$

Values of σ_{coh} and σ_{inc}

Nuclide	σ_{coh}	σ_{inc}	Nuclide	σ_{coh}	σ_{inc}
^1H	1.8	80.2	V	0.02	5.0
^2H	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

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

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

so

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

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

ie

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

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

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, given that there is also a (different) atom at the origin of the coordinate system at the same instant in time.

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.)

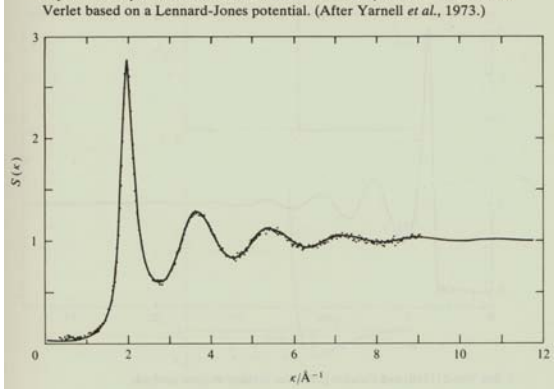
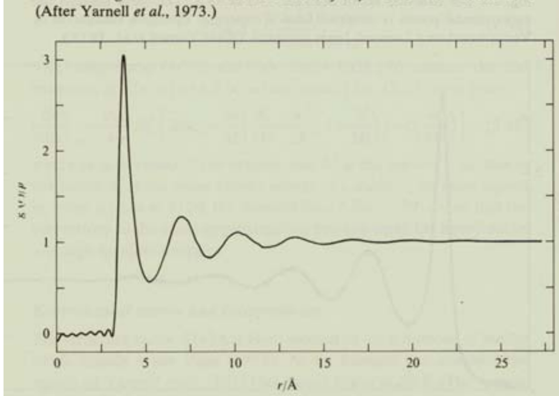
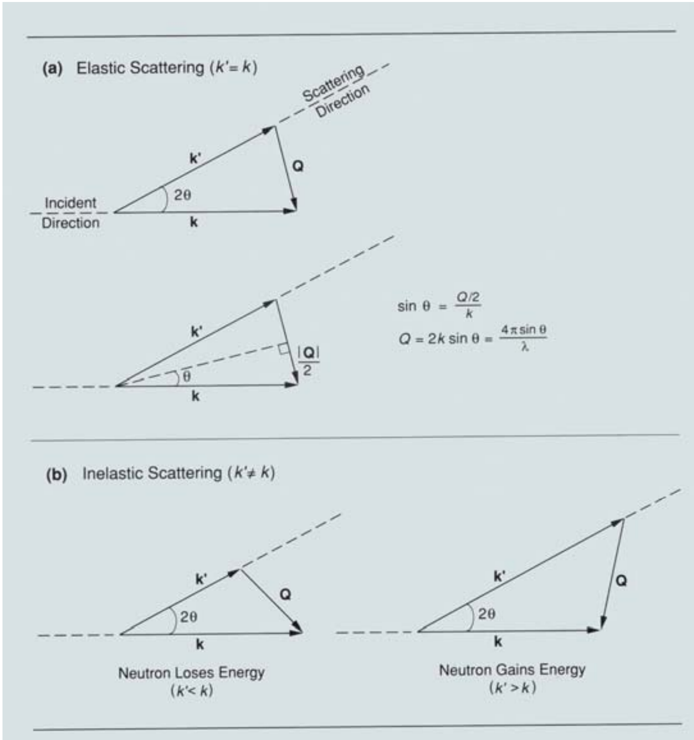
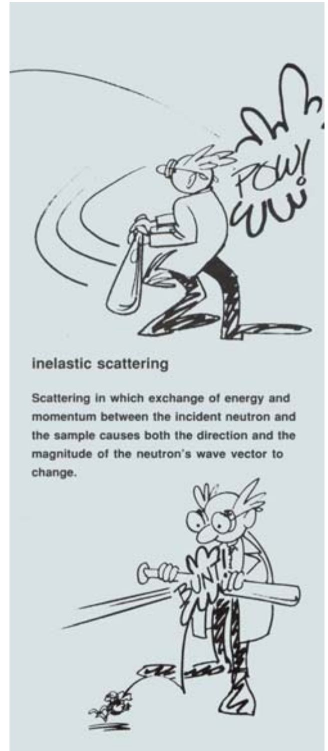


Fig. 5.2 The pair-distribution function $g(r)$ obtained from the experimental results in Fig. 5.1. The mean number density is $\rho = 2.13 \times 10^{28}$ atoms m^{-3}. (After Yarnell *et al.*, 1973.)



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





General Expression for d²σ/dΩdE

- Squires (eqn 2.59) derives the following expression:

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{i,i'} b_i b'_{i'} \int_{-\infty}^{\infty} \left\langle e^{-i\vec{Q}.\vec{R}_{i'}(0)} e^{i\vec{Q}.\vec{R}_i(t)} \right\rangle e^{-i\omega t} dt$$

where $\vec{R}_i(t)$ is a Heisenberg operator i.e.

$e^{-i\vec{Q}.\vec{R}_i(t)} = e^{iHt/\hbar} e^{-i\vec{Q}.\vec{R}_i} e^{-iHt/\hbar}$ where H is the Hamiltonian of the scatterer

and $\langle \ \rangle$ denotes a thermal average over the possible states, λ, of the

scatterer -- i.e. for any operator, $\langle A \rangle = \sum_\lambda p_\lambda \langle \lambda | A | \lambda \rangle$

- Note that, because of the operators and the average over the states of the system, this expression is not easy to evaluate in the general case

- Note also that the exponential operators do not commute – each contains H and therefore p, and p and R do not commute.

Correlation Functions

Suppose we define : $G(\vec{r},t) = \frac{1}{(2\pi)^3}\frac{1}{N}\int e^{-i\vec{Q}'.\vec{r}}\sum_{j,j'}\left\langle e^{-i\vec{Q}'.\vec{R}_{j'}(0)}e^{i\vec{Q}'.\vec{R}_j(t)}\right\rangle d\vec{Q}'$

and $S(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\int G(\vec{r},t)e^{i(\vec{Q}.\vec{r}-\omega t)}d\vec{r}.dt$ then we find

$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{coh} = b_{coh}^2\frac{k'}{k}NS(\vec{Q},\omega)$ provided there is only one type of atom

Squires (eqn 4.14 to 4.17) shows that

$$G(\vec{r},t) = \frac{1}{N}\sum_{j,j'}\int\left\langle \delta\{\vec{r}'-\vec{R}_{j'}(0)\}\delta\{\vec{r}'+\vec{r}-\vec{R}_j(t)\}\right\rangle d\vec{r}'$$

- Note again that the operators do not commute. If we ignore this fact, we can do the integration and obtain

$$G_{classical}(\vec{r},t) = \frac{1}{N}\sum_{j,j'}\left\langle \delta\{\vec{r}-\vec{R}_j(t)+\vec{R}_{j'}(0)\}\right\rangle$$

Correlation Functions (cont'd)

$$G_{classical}(\vec{r},t) = \frac{1}{N}\sum_{j,j'}\left\langle \delta\{\vec{r}-\vec{R}_j(t)+\vec{R}_{j'}(0)\}\right\rangle$$

- We expressed the coherent scattering cross section in terms of G(**r**,t)
- If we use the classical variant given above, there is a clear physical meaning – G(**r**,t) is the probability that if particle j' is at the origin at time zero, particle j will be at position **r** at time t.
- We can do the same thing with the incoherent scattering and express it in terms of a self-correlation function whose classical version is

$$G_{classical}^{self}(\vec{r},t) = \left\langle \delta\{\vec{r}-R_j(t)+R_j(0)\}\right\rangle$$

- This says that the incoherent scattering is related to the probability that if a particle is at the origin at time zero, *the same* particle will be at position **r** at time t.

Inelastic Neutron Scattering Measures Atomic Motions

In term of the pair correlation functions, one finds

$$\left(\frac{d^2\sigma}{d\Omega.dE}\right)_{coh} = b_{coh}^2 \frac{k'}{k} NS(\vec{Q},\omega)$$

$$\left(\frac{d^2\sigma}{d\Omega.dE}\right)_{inc} = b_{inc}^2 \frac{k'}{k} NS_s(\vec{Q},\omega)$$

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

where

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

- Inelastic coherent scattering measures *correlated* motions of different atoms
- Inelastic incoherent scattering measures *self-correlations* e.g. diffusion

Elastic Scattering as the t⟶ ∞ Limit of G(**r**,t)

- Elastic scattering occurs at ω = 0. Since it involves a δ(ω), only the part of G(**r**,t) which is constant contributes
- G(**r**,t) decays as t increases, so the constant part is G(**r**,∞)
- Since we only need the part of the correlation that is time-independent, we can write (noting that the correlation between the positions of j and j' are independent of t as t-> ∞)

$$G(\vec{r},t) = \frac{1}{N}\sum_{j,j'}\int \left\langle \delta\{\vec{r}'-\vec{R}_{j'}(0)\}\delta\{\vec{r}'+\vec{r}-\vec{R}_j(t)\}\right\rangle d\vec{r}'$$

$$G(\vec{r},\infty) = \frac{1}{N}\sum_{j,j'}\int \left\langle \delta\{\vec{r}'-\vec{R}_{j'}\}\right\rangle\left\langle \delta\{\vec{r}'+\vec{r}-\vec{R}_j\}\right\rangle d\vec{r}'$$

$$= \frac{1}{N}\int \langle \rho(\vec{r}')\rangle\langle \rho(\vec{r}'+\vec{r})\rangle d\vec{r}'$$

Note – no truly elastic scattering for a liquid

where $\rho(\vec{r}')$ is the particle density operator at any time

G(**r**,∞) is called the Patterson function

The Static Approximation

Earlier we had :

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{coh} = b_{coh}^2 \frac{k'}{k} N \frac{1}{2\pi\hbar} \int G(\vec{r},t) e^{i(\vec{Q}.\vec{r}-\omega t)} d\vec{r}.dt$$

where $G(\vec{r},t) = \frac{1}{(2\pi)^3} \frac{1}{N} \int e^{-i\vec{Q}.\vec{r}} \sum_{j,j'} \left\langle e^{-i\vec{Q}.\vec{R}_{j'}(0)} e^{i\vec{Q}.\vec{R}_j(t)} \right\rangle$

- In diffraction measurements, we measure scattered neutron intensity in a particular direction, independent of the change in neutron energy – i.e. we integrate the cross section over E = hω/2π. This is the Static Approximation.

Because $\hbar\delta(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\omega t} d(\hbar\omega)$

the integral over ω picks out the t = 0 value of G to give

$$\left(\frac{d\sigma}{d\Omega}\right)_{coh}^{static} = b_{coh}^2 N \int G(\vec{r},t) e^{i(\vec{Q}.\vec{r}-\omega t)} d\vec{r}.dt.d\omega$$

$$= b_{coh}^2 N \int G(\vec{r},0) e^{i\vec{Q}.\vec{r}} d\vec{r}$$

Comparison of Elastic Scattering and the Static Approximation

$$\left(\frac{d\sigma}{d\Omega}\right)_{coh}^{static} = b_{coh}^2 N \int G(\vec{r},0) e^{i\vec{Q}.\vec{r}} d\vec{r}$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{coh}^{elastic} = b_{coh}^2 N \int G(\vec{r},\infty) e^{i\vec{Q}.\vec{r}} d\vec{r}$$

- These are not the same, except in an (unreal) system with no motion
- The elastic scattering cross section gives the *true* elastic scattering that results when the positions of different atoms are correlated for all times, as they are in a crystalline solid, even when phonons are present
- The static approximation, as its name suggests, gives the scattering for a system that is frozen in time

The Intermediate Scattering Function

- Another function that is often useful is the Intermediate Scattering Function defined as

$$I(\vec{Q},t) = \int G(\vec{r},t) e^{i\vec{Q}.\vec{r}} d\vec{r}$$

This is the quantity measured with Neutron Spin Echo (NSE)

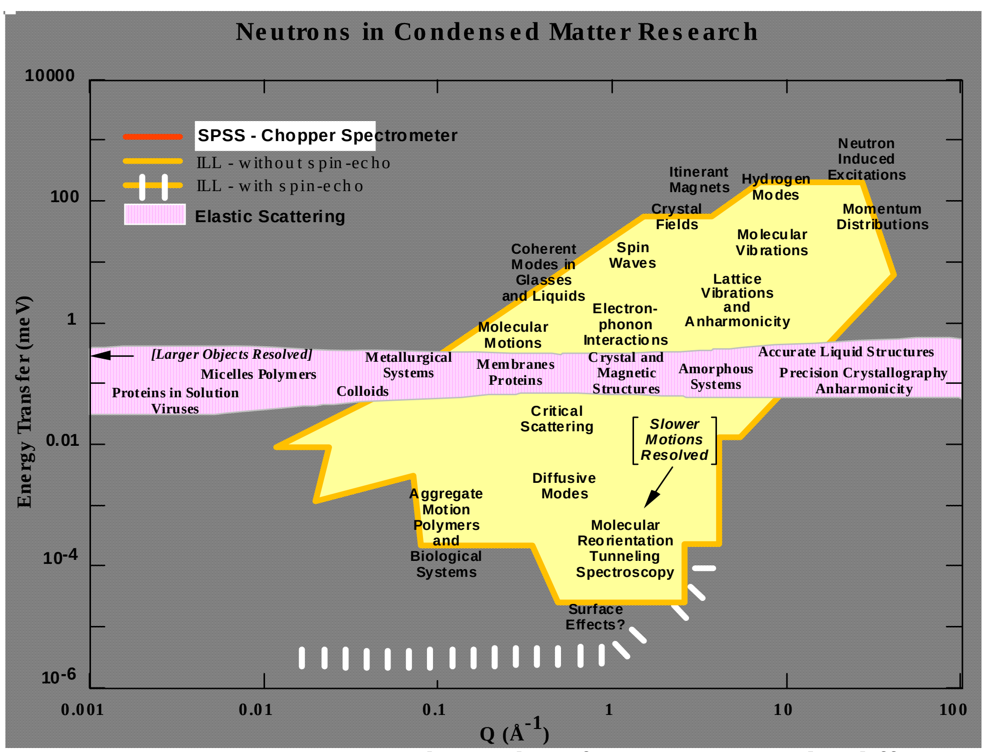
- It is not possible to derive exact expressions for I, G or S except for simple models. It is therefore useful to know the various analytical properties of these functions to ensure that models preserve them. Squires shows:

$$I(\vec{Q},t) = I^*(\vec{Q},-t); \qquad I(\vec{Q},t) = I(-\vec{Q},-t+i\hbar/k_B T)$$

$$G(\vec{r},t) = G^*(-\vec{r},-t); \qquad G(\vec{r},t) = G(-\vec{r},-t+i\hbar/k_B T)$$

$$S(\vec{Q},\omega) = S^*(\vec{Q},\omega); \qquad S(\vec{Q},\omega) = e^{\hbar\omega/k_B T} S(-\vec{Q},-\omega)$$

- There are also various sum & moment rules on these quantities that are sometimes useful (see Squires for details)



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.

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