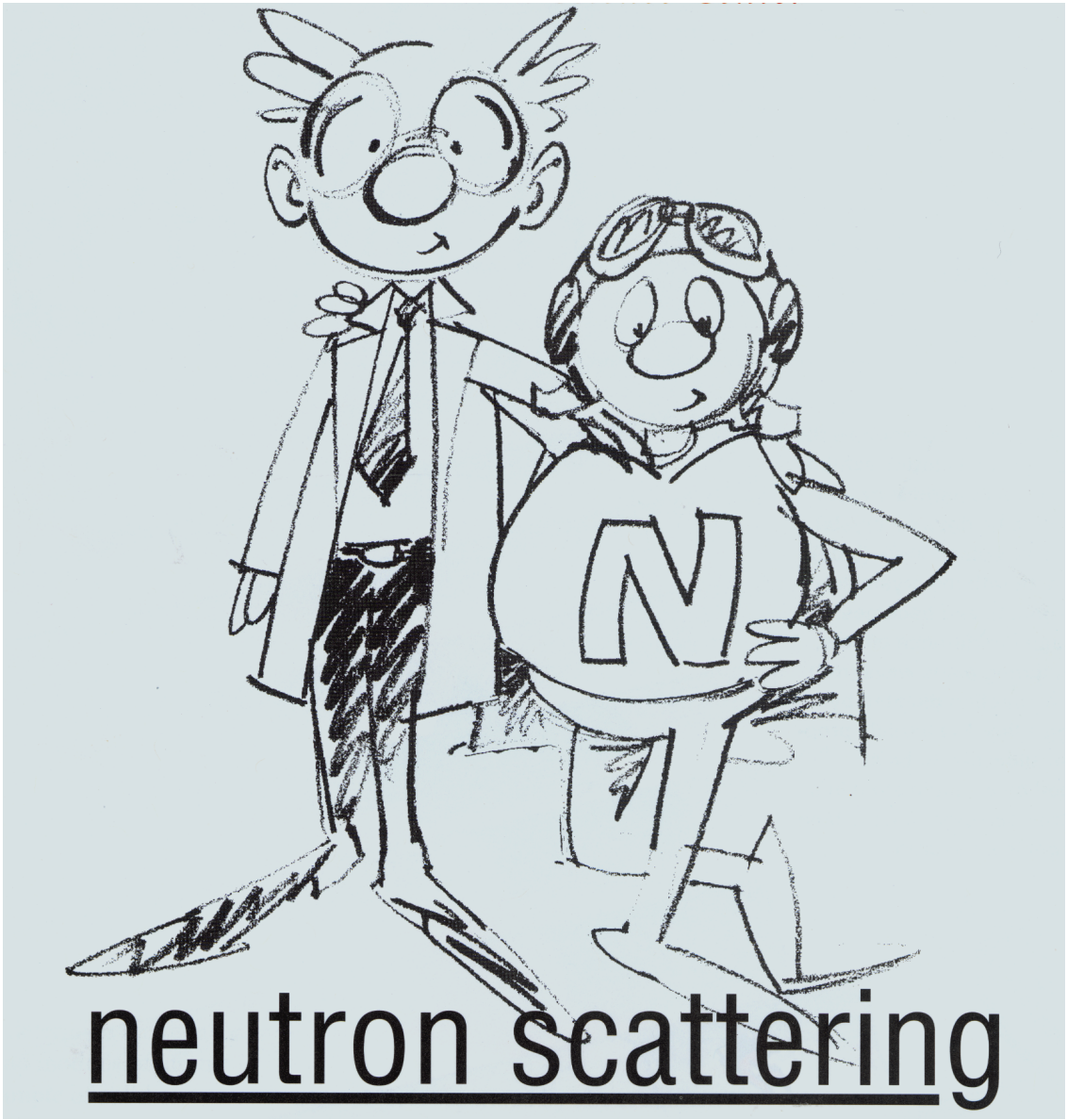

An Overview

By

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Overview

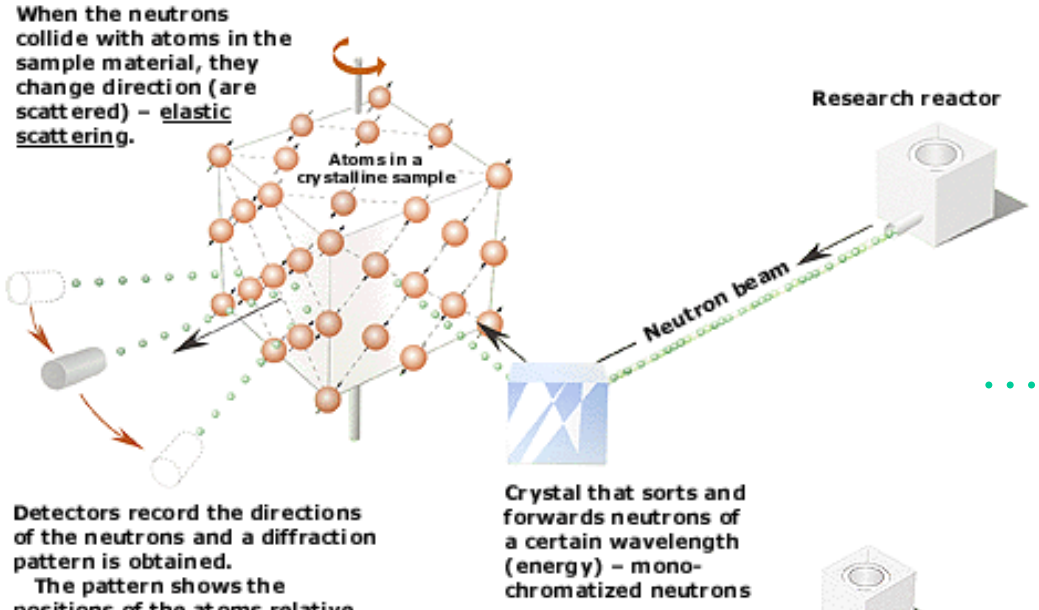
Introduction and theory of neutron scattering

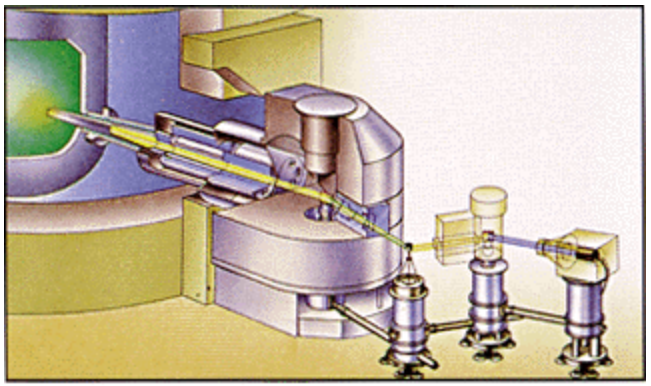
- Advantages/disadvantages of neutrons for scattering measurements
- Neutron properties
- Comparison with other structural probes
- What do we measure? The neutron scattering cross section
- Elastic and inelastic scattering
- Coherent and incoherent scattering

- Break

The 1994 Nobel Prize in Physics – Shull & Brockhouse

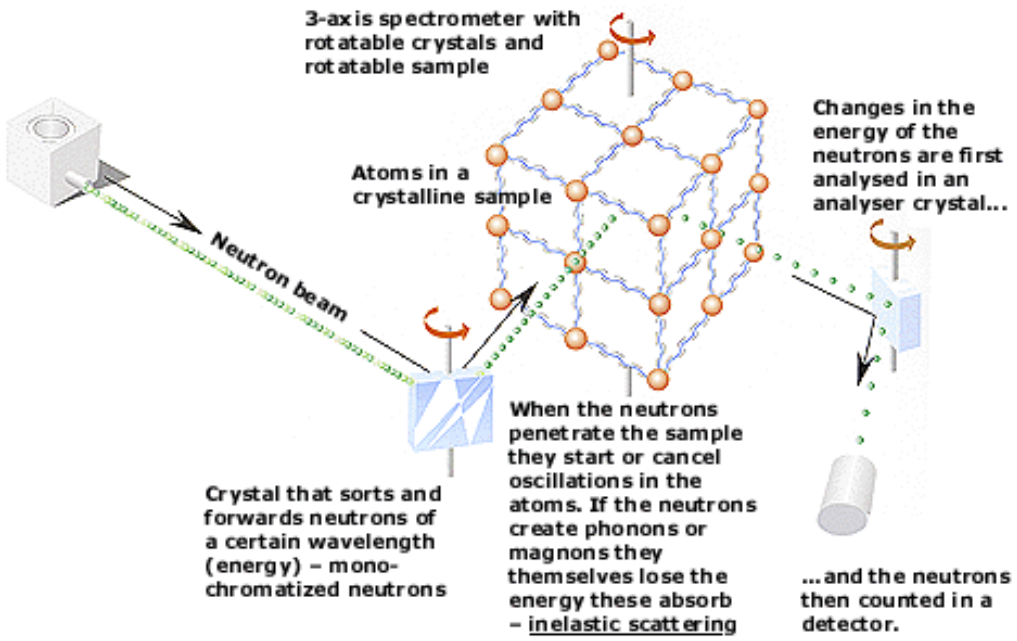
Neutrons show where the atoms are….





3-axis spectrometer

…and what the atoms do.



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 Neutron has Both Particle-Like and Wave-Like Properties

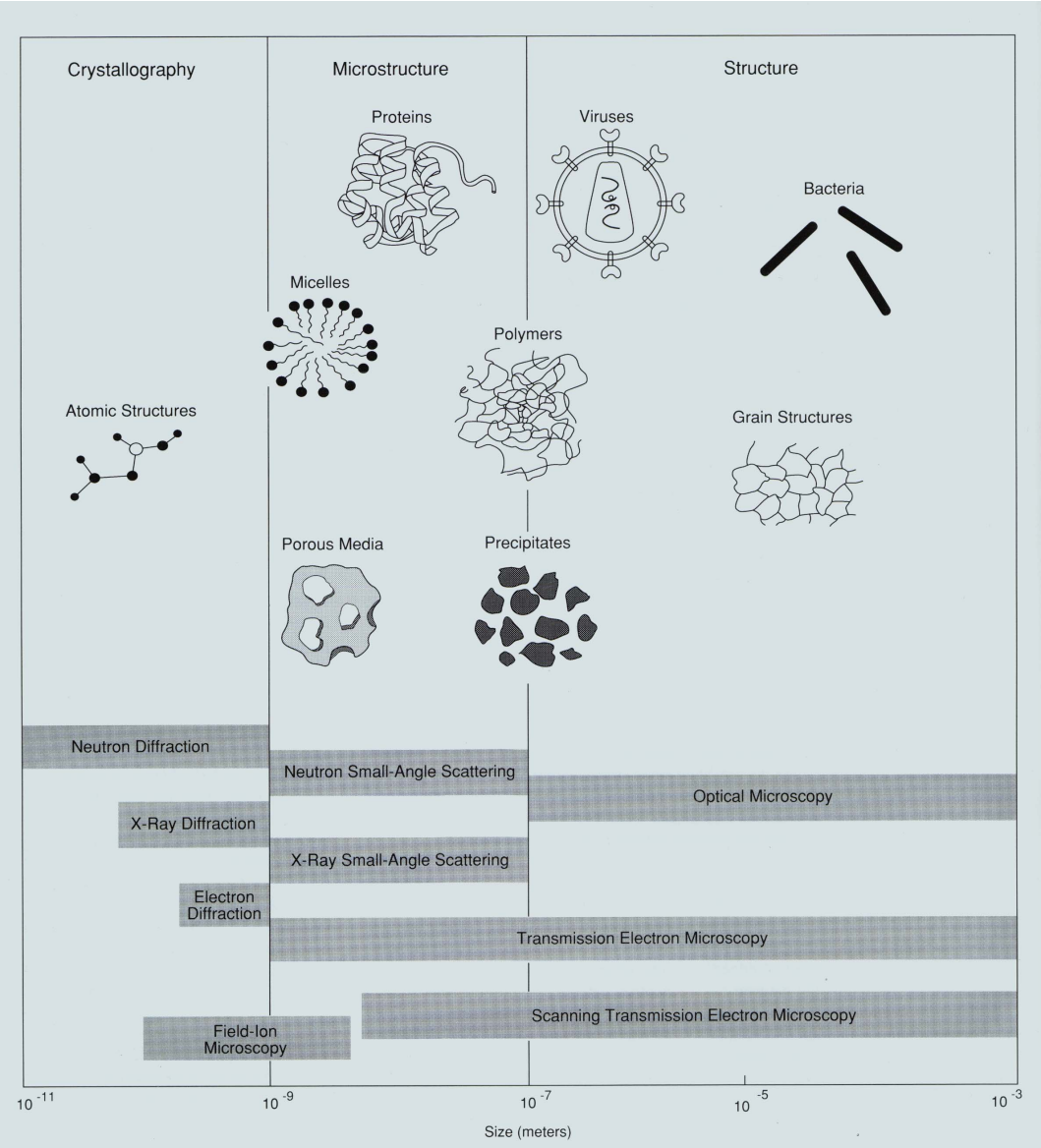
- Mass: $m_n = 1.675 \times 10^{-27}$ kg
- Charge = 0; Spin = ½
- Magnetic dipole moment: $\mu_n = -1.913\ \mu_N$
- Nuclear magneton: $\mu_N = eh/4\pi m_p = 5.051 \times 10^{-27}$ J T^{-1}
- Velocity (v), kinetic energy (E), wavevector (k), wavelength (λ), temperature (T).
- $E = m_n v^2/2 = k_B T =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

	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**

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

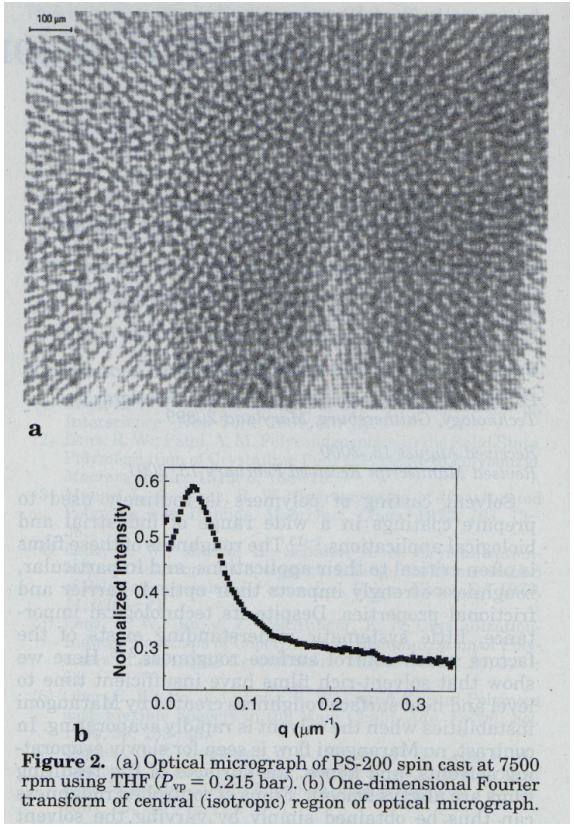


Figure 2. (a) Optical micrograph of PS-200 spin cast at 7500 rpm using THF (P_{vp} = 0.215 bar). (b) One-dimensional Fourier transform of central (isotropic) region of optical micrograph.

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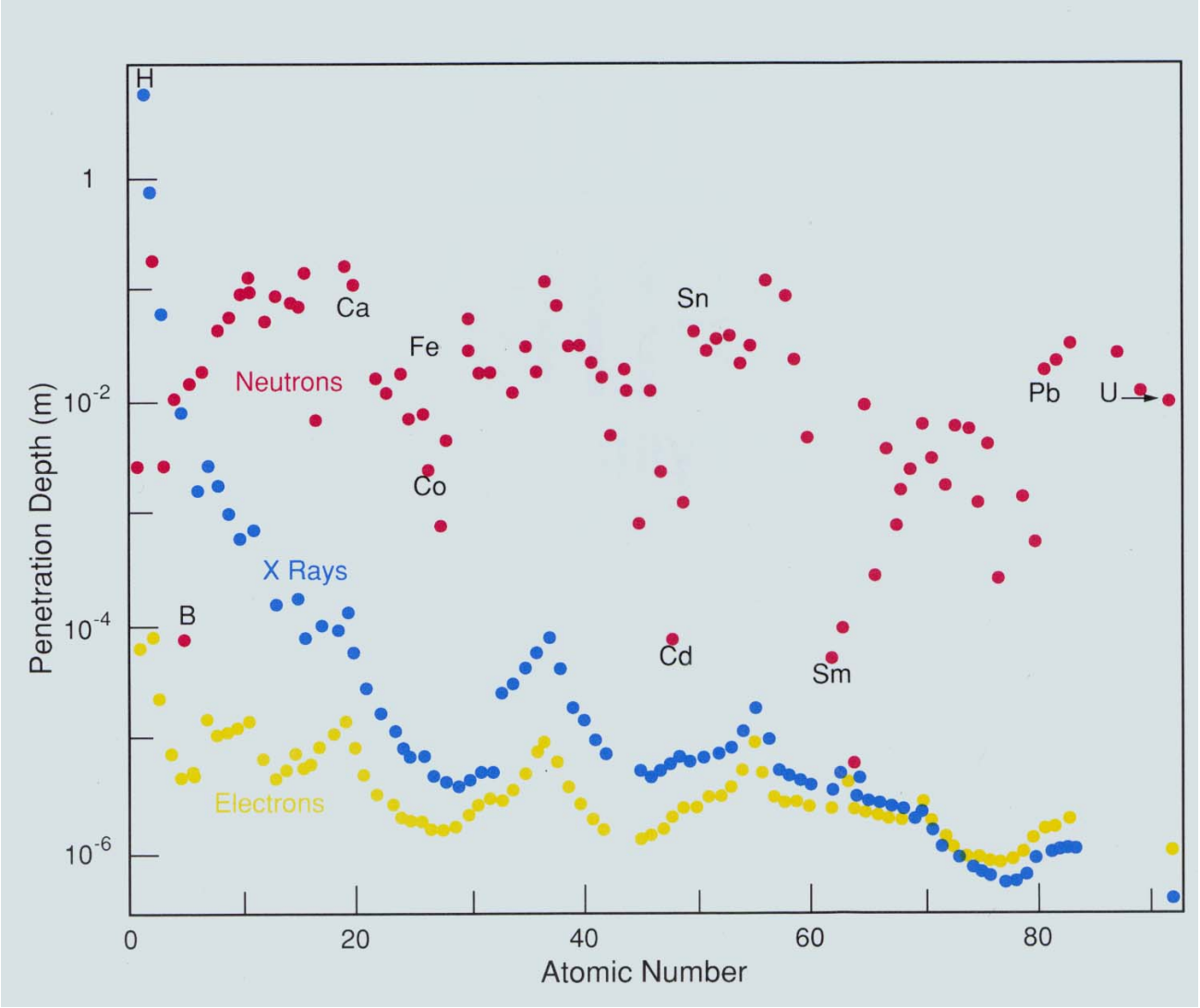
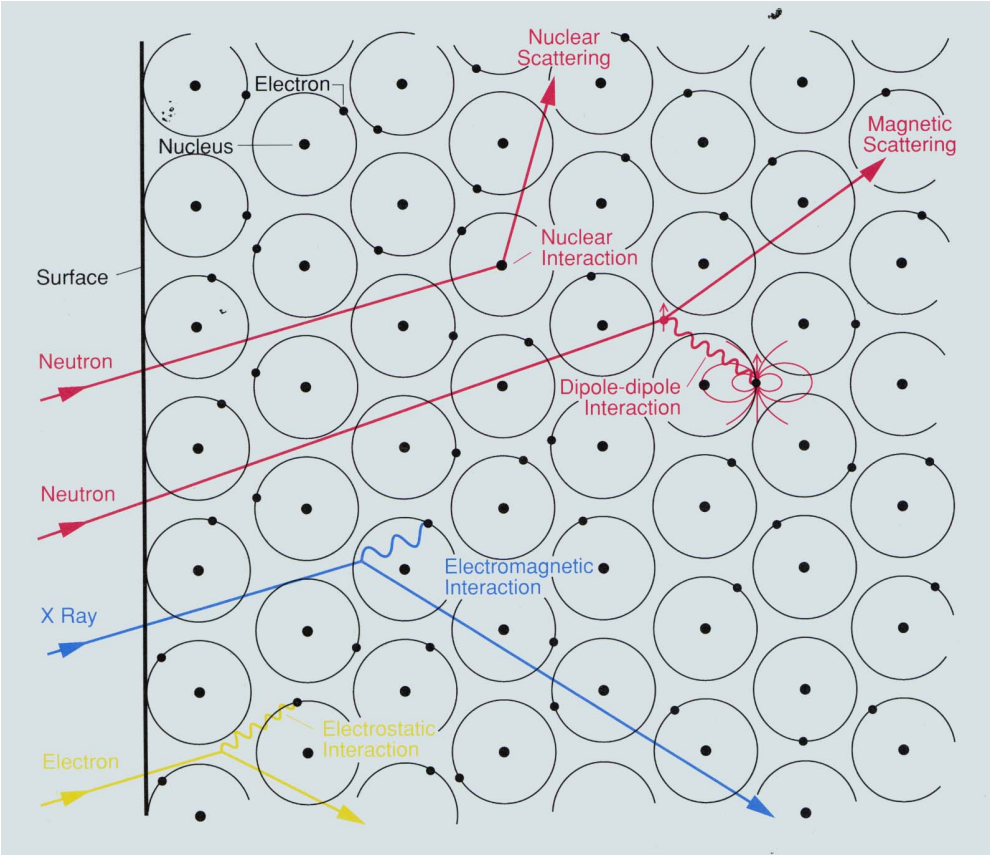
Thermal Neutrons, 8 keV X-Rays & Low Energy Electrons:- Absorption by Matter



Note for neutrons:

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

Interaction Mechanisms

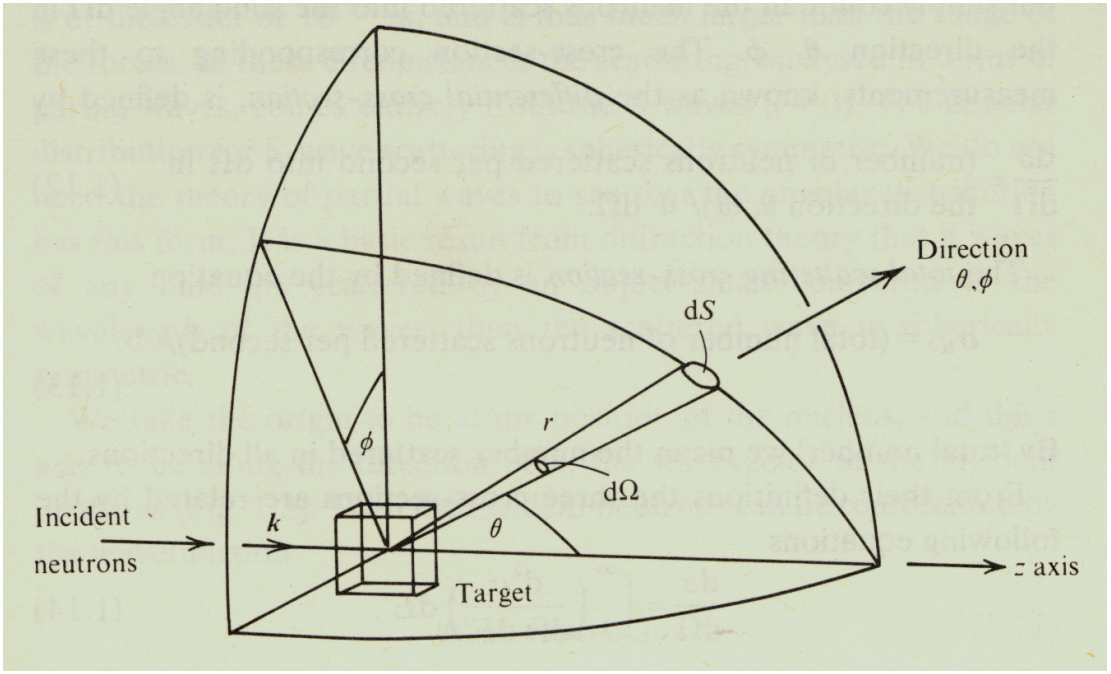


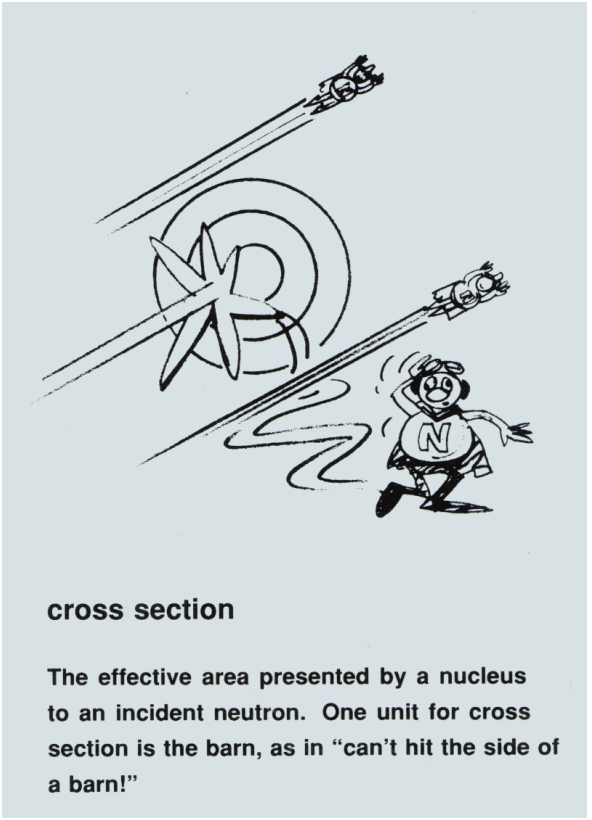
- Neutrons interact with atomic nuclei via very short range (~fm) forces.
- Neutrons also interact with unpaired electrons via a magnetic dipole interaction.

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

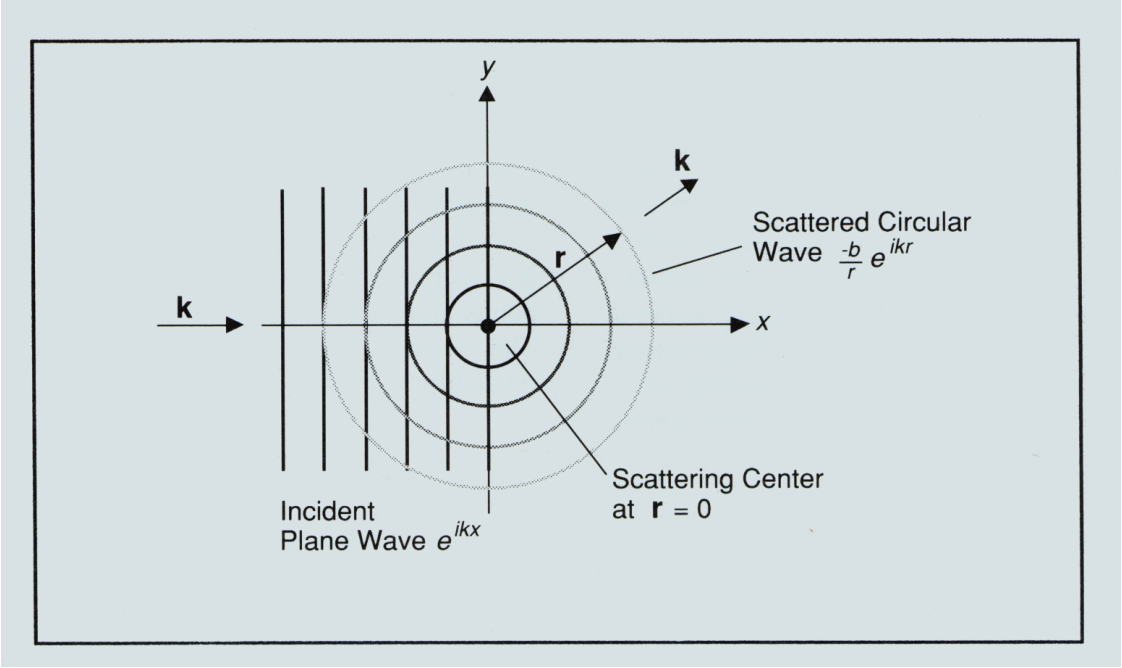
$$\frac{d\sigma}{d\Omega} = \frac{\text{number of neutrons scattered per second into } d\Omega}{\Phi\, d\Omega}$$

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

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 (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 areas is: $\Phi = v\left|\psi_{incident}\right|^2 = v$

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

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

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 : $\overline{V} = \frac{2\pi\hbar^2}{m}\rho$ 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 $\frac{\hbar^2 k^2}{2m} + \overline{V}$

Conservation of energy gives $\frac{\hbar^2 k_0^2}{2m} = \frac{\hbar^2 k^2}{2m} + \overline{V} = \frac{\hbar^2 k^2}{2m} + \frac{2\pi\hbar^2}{m}\rho$ 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 :

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

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

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:

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

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

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

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

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

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

Values of σ_{coh} and σ_{inc}

Nuclide	σ_{coh}	σ_{inc}	Nuclide	σ_{coh}	σ_{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

$\frac{d\sigma}{d\Omega} = \langle b \rangle^2 N.S(\vec{Q})$ 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 $\sum_i e^{-i\vec{Q}.\vec{R}_i} = \int d\vec{r}.e^{-i\vec{Q}.\vec{r}} \sum_i \delta(\vec{r} - \vec{R}_i) = \int d\vec{r}.e^{-i\vec{Q}.\vec{r}} \rho_N(\vec{r})$ where ρ_N is the nuclear number density

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

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

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

where $g(\vec{R}) = \sum_{i \neq 0} \langle \delta(\vec{R} - \vec{R}_i + \vec{R}_0) \rangle$ 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, 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.)

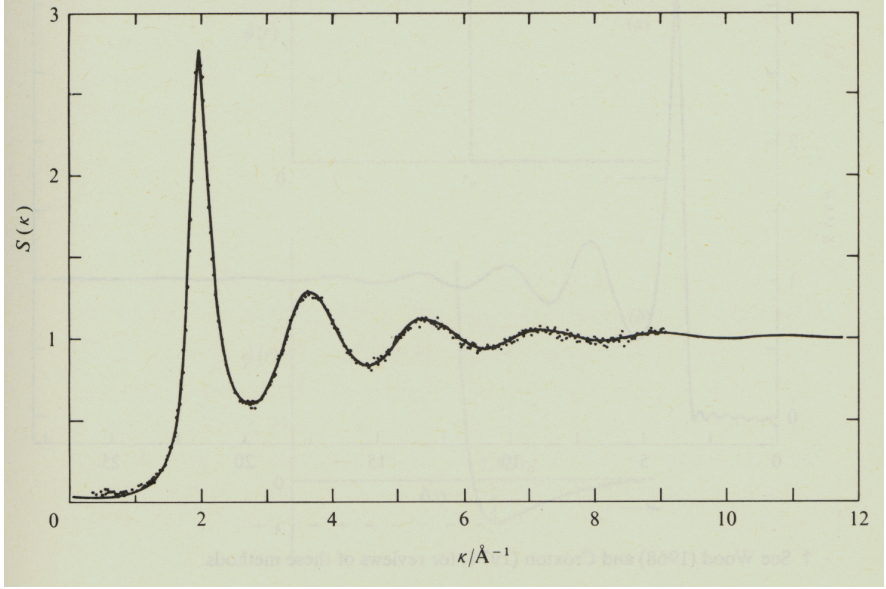
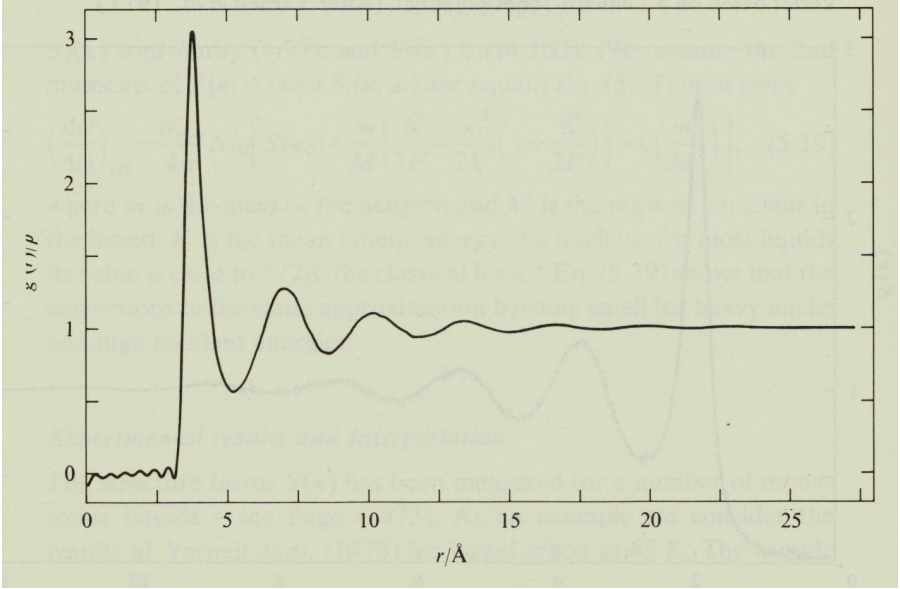
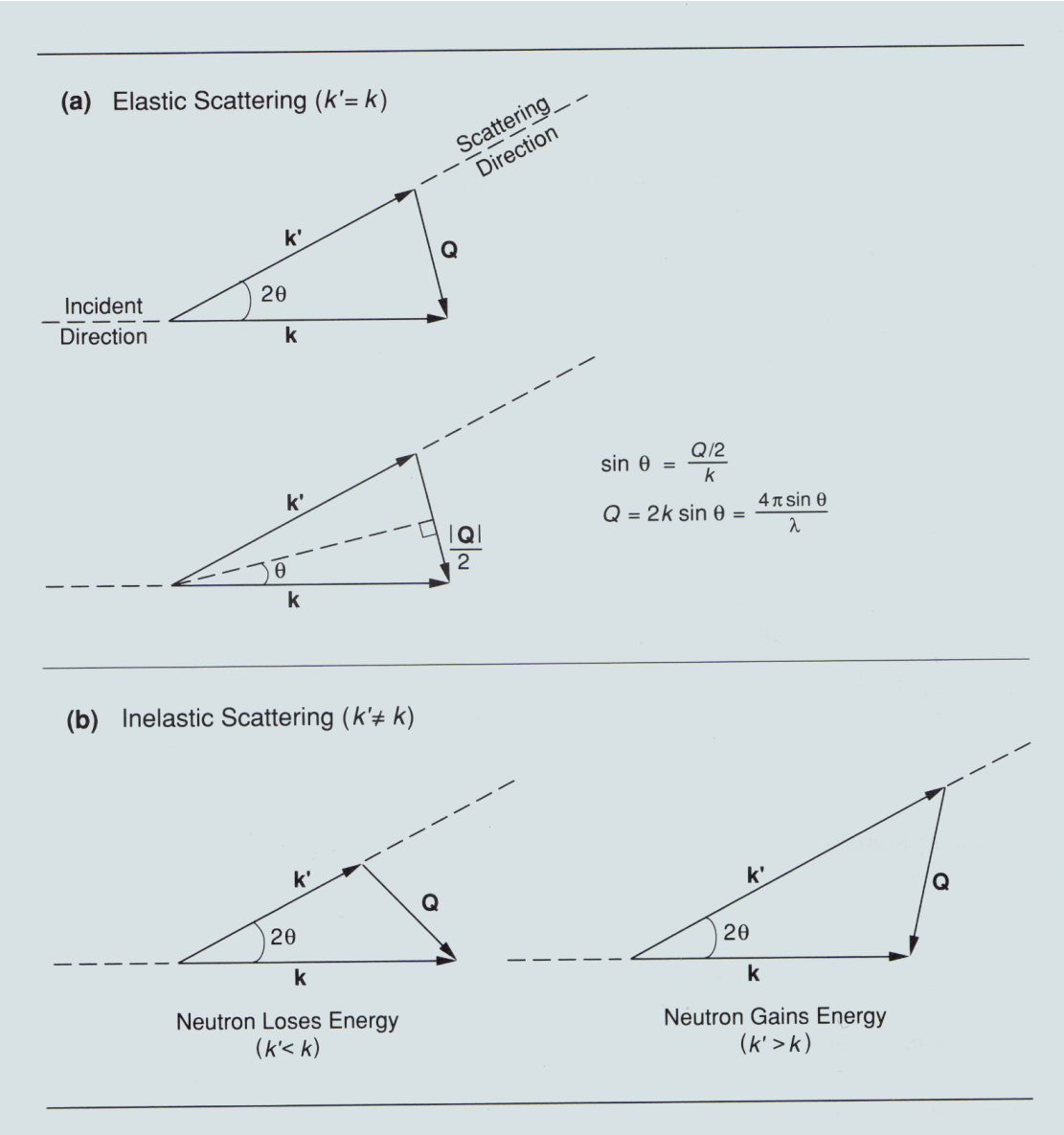
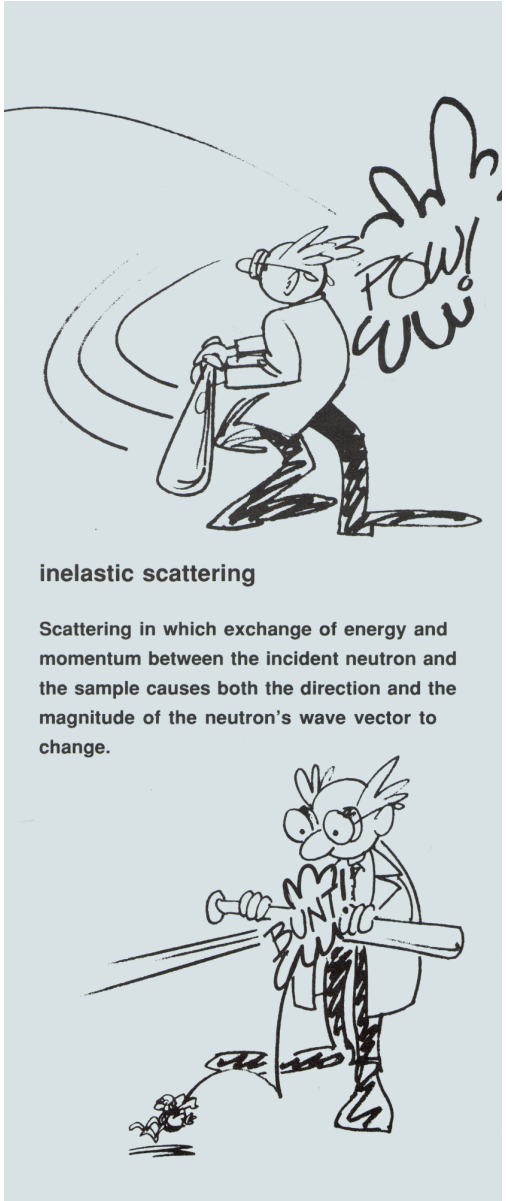


Fig. 5.2 The pair-distribution function $g(r)$ obtained from the experimental results in Fig. 5.1. The mean number density is $\rho = 2.13 \times 10^{28}$ atoms m^{-3}. (After Yarnell *et al.*, 1973.)



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





General Expression for $d^2\sigma/d\Omega dE$

- 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, $\langle \mathrm{A} \rangle = \sum_\lambda p_\lambda \langle \lambda | A | \lambda \rangle$

- 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

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

- 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 $\omega = 0$. Since it involves a $\delta(\omega)$, 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-> ∞)

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

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

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

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

Note – no truly elastic scattering for a liquid

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

The Static Approximation

Earlier we had :

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

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

- 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.

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

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)

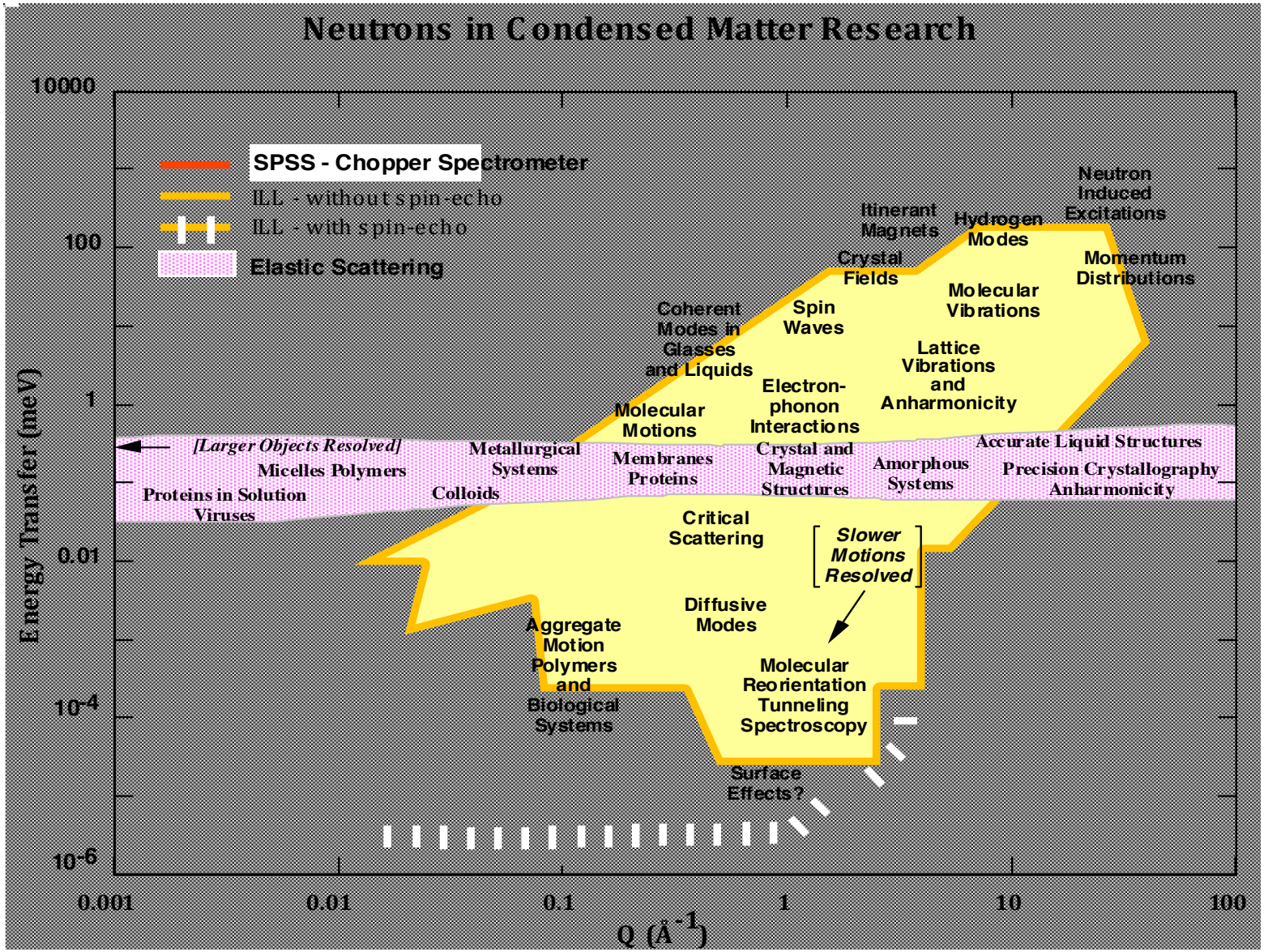
- It is not possible to derive exact expressions for I, G or S except for simple models. It is therefore useful to know the various analytical properties of these functions to ensure that models preserve them. Squires shows:

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

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

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

- 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.

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)

- Elements of Modern X-Ray Physics
  by Jens Als-Nielsen and Des McMorrow
  John Wiley and Sons
  ISBN 0471498580

Overview

Neutron scattering in practice

- Neutron sources
- Diffraction
- Small angle neutron scattering (SANS)
- Reflectometry
- Inelastic scattering

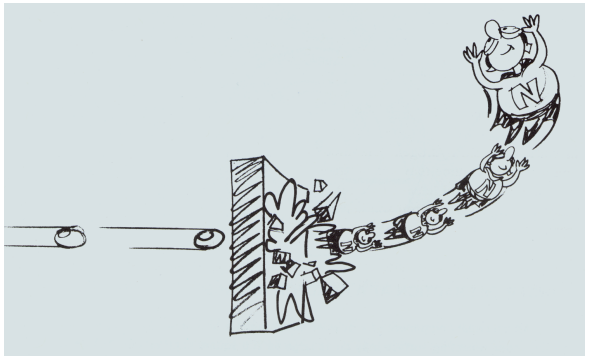
Neutron Scattering Requires Intense Sources of Neutrons

- Neutrons for scattering experiments can be produced either by nuclear fission in a reactor or by spallation when high-energy (~1 GeV) protons strike a heavy metal target (W, Ta, or U).
  - In general, reactors produce continuous neutron beams and spallation sources produce beams that are pulsed between 20 Hz and 60 Hz
  - The energy spectra of neutrons produced by reactors and spallation sources are different, with spallation sources producing more high-energy neutrons
  - Neutron spectra for scattering experiments are tailored by moderators – solids or liquids maintained at a particular temperature – although neutrons are not in thermal equilibrium with moderators at a short-pulse spallation sources

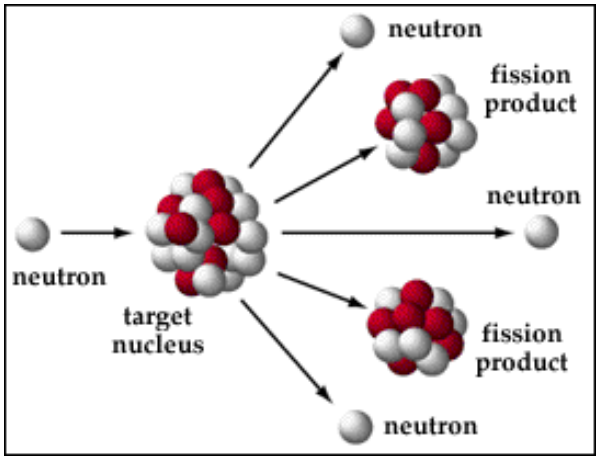
- Both reactors and spallation sources are expensive to build and require sophisticated operation.
  - SNS at ORNL will cost about $1.5B to construct & ~$140M per year to operate

- Either type of source can provide neutrons for 30-50 neutron spectrometers
  - Small science at large facilities

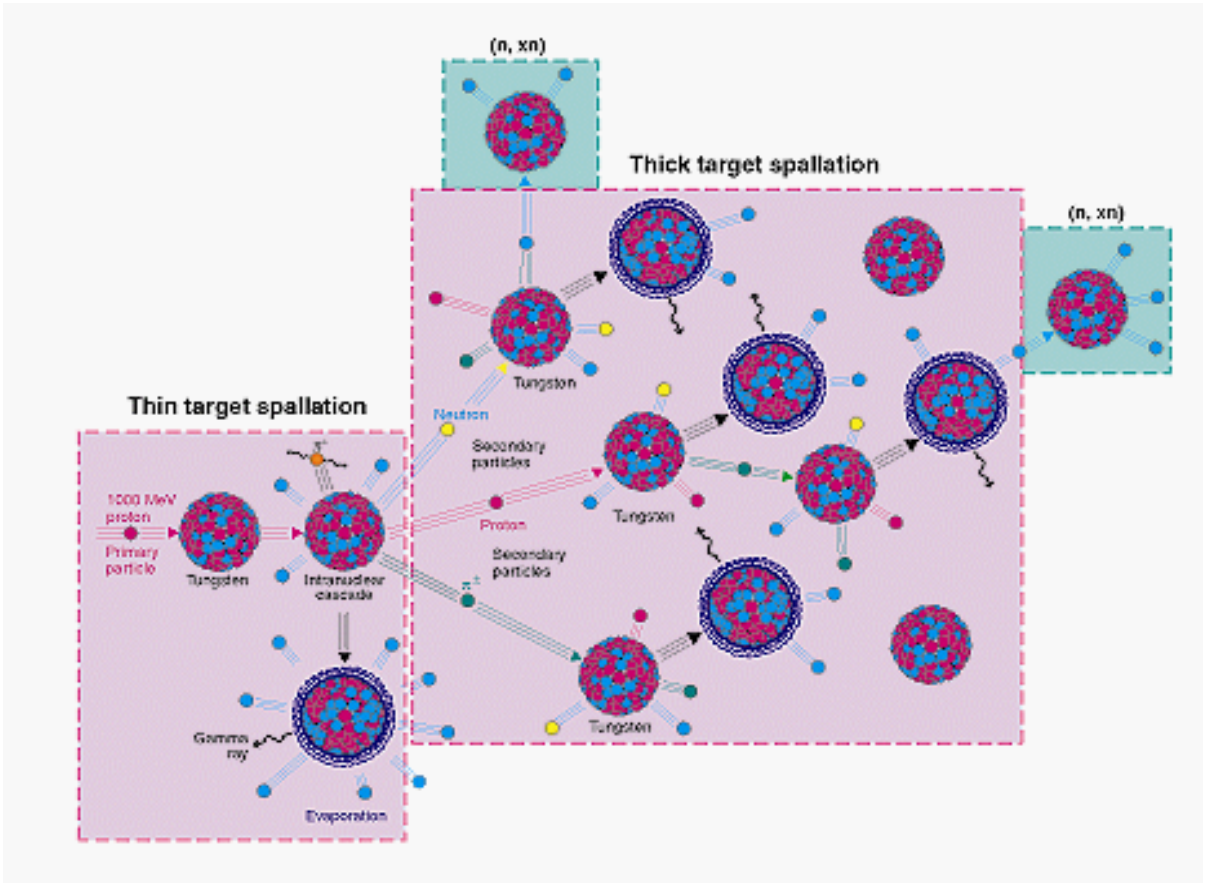
Nuclear Fission & Spallation are the Methods of Choice to Produce Neutrons for Scattering



Artist's view of spallation



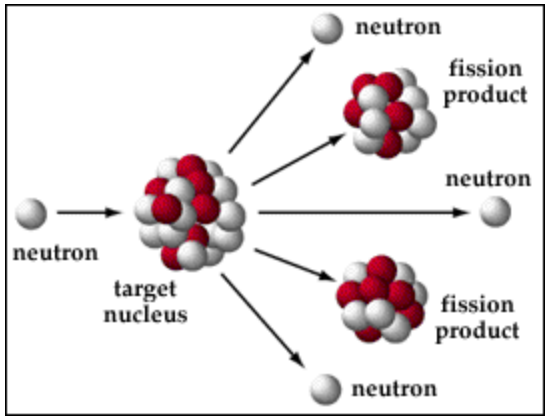
Nuclear Fission



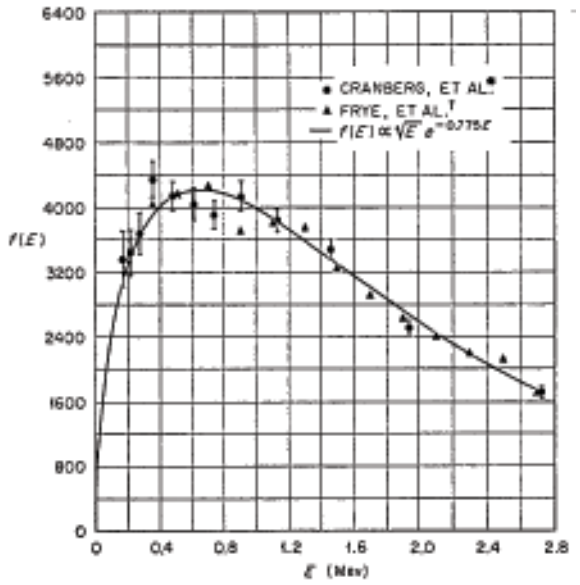
Spallation

Nuclear Fission

- Prompt neutrons "evaporate" from excited nuclei with energy ~ 2 MeV. About 2.5 neutrons produced/fission.
  - About 0.5% of neutrons are delayed by a few secs (decay chain).
- ~1 neutron from each fission is "useful"
  - 1 required to sustain reaction; ~0.5 lost to absorption
- Each fission event produces ~ 190 MeV (neutron & fragment KE, γ, β, ν)



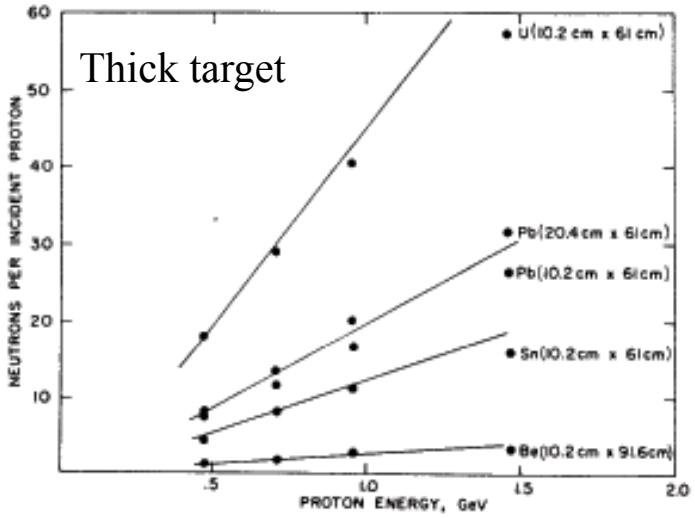
J. M. Carpenter
Neutron Production & Moderation
and the Characterization of Sources
http://www.neutron.anl.gov/NeutronProduction.pdf



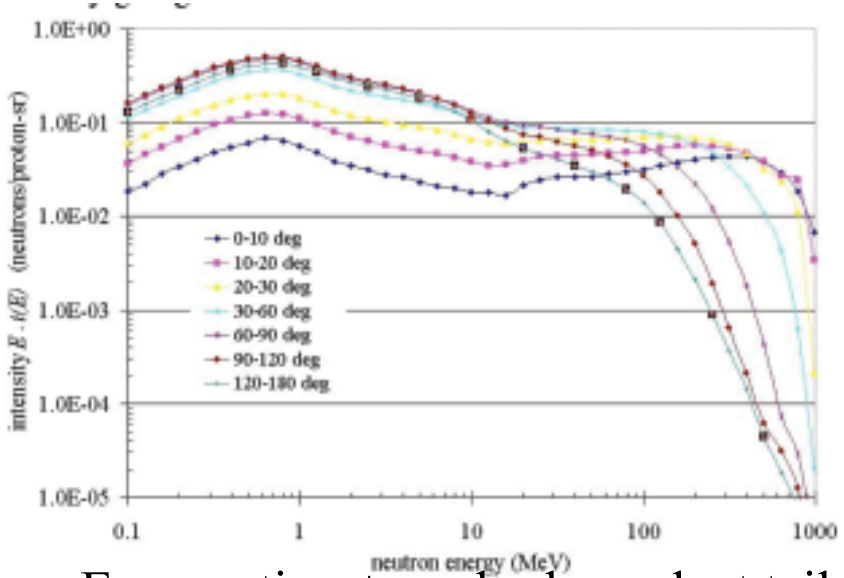
Energy distribution of prompt fission neutrons

Spallation

- Complex series of reactions when a high-energy (~1 GeV) particle hits a heavy nucleus
  - Incident particles collide with & excite nuclei which give off further particles that collide with nuclei…..
  - Excited nuclei reduce their energy by giving off (mostly) neutrons and (eventually) e^+ (because they have too many protons) and γ
- ~20 neutrons produced per incident proton
  - Most with (evaporation) energies ~2 MeV but some up to initial particle energy
- ~60% of initial proton energy deposited in target (as heat)



$Y(E,A) = 0.1(E_{GeV} - 0.12)(A + 20)$



Evaporation + angle-dependent tail

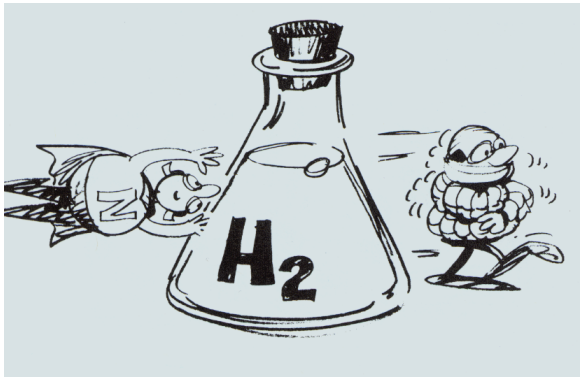
The Energy Cost of Various Neutron Sources

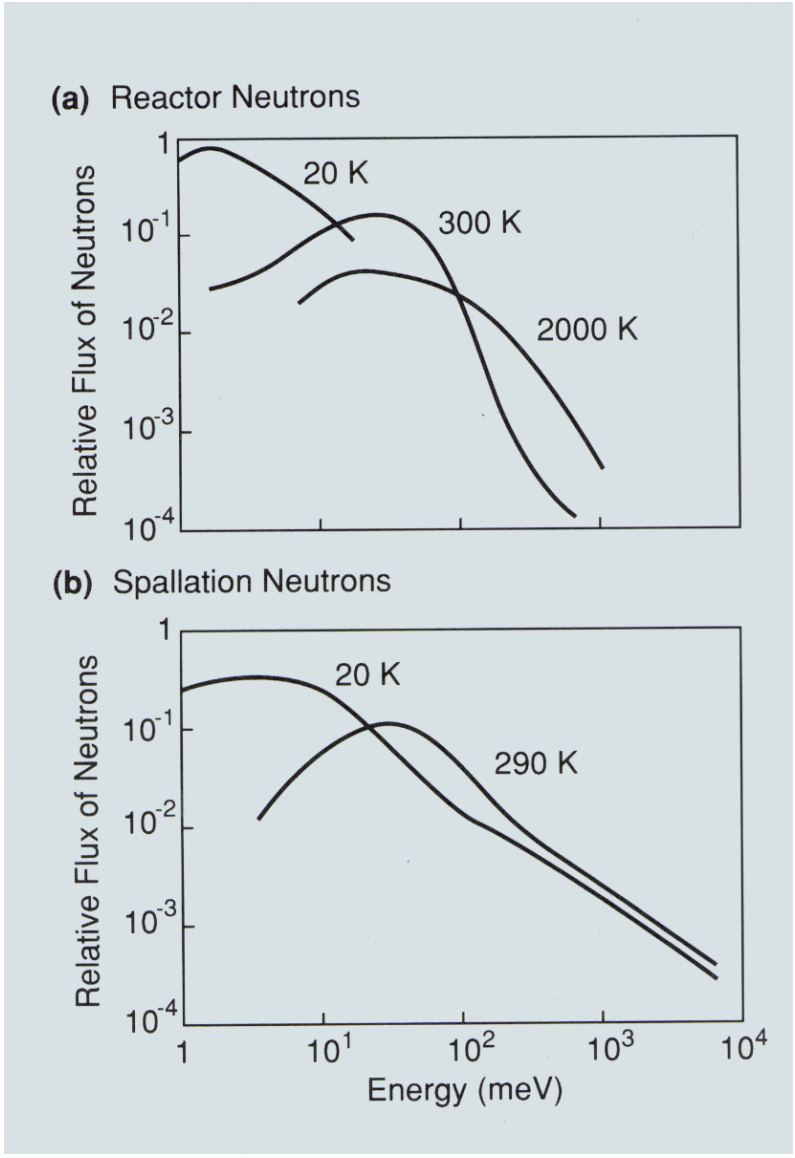
- For high-power sources the driving issue is heat removal => use spallation for high power sources
  - ~ 190 MeV per neutron for fission
  - ~ 25 MeV per neutron for spallation with protons (threshold at E_p~ 120 MeV)
  - ~ 1500 MeV per neutron for (n,p) on Be using 13 MeV protons
  - ~ 3000 MeV per neutron for electrons

- Driving issue for low-intensity sources is cost (electric power, regulatory, manpower etc)
  - Cost has to be kept "low" (i.e. construction ~$10-20M)
  - Cost/benefit is still the metric
  - Spallation and fission cost too much (absent a "killer app" money maker)
  - Use Be (p,n) or electrons on Ta

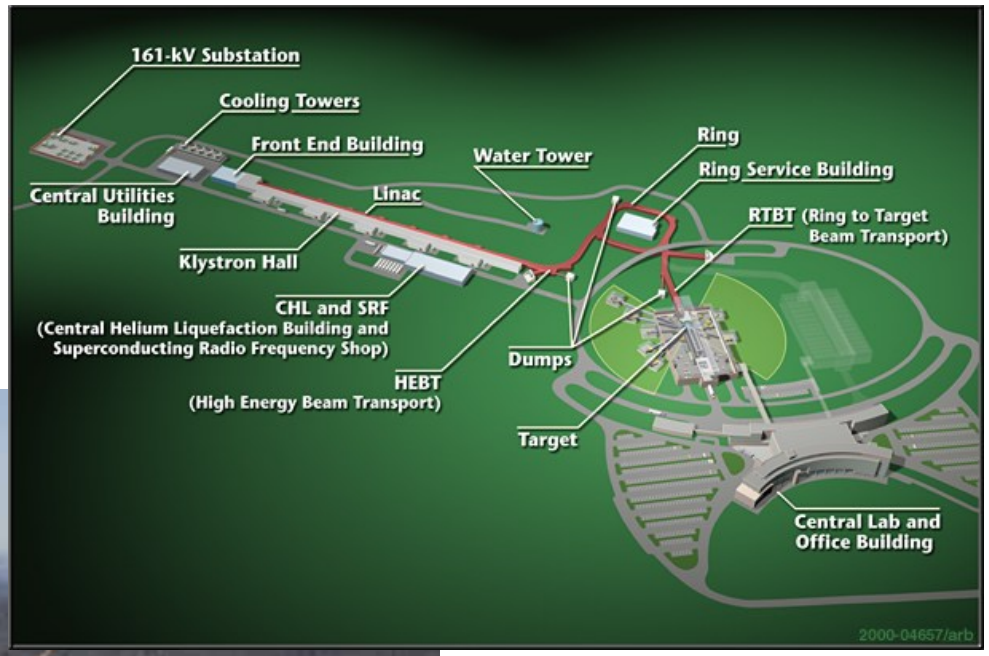
Neutrons From Reactors and Spallation Sources Must Be Moderated Before Being Used for Scattering Experiments

- Reactor spectra are Maxwellian
- Intensity and peak-width ~ $1/(E)^{1/2}$ at high neutron energies at spallation sources
- Cold sources are usually liquid hydrogen (though deuterium is also used at reactors & methane is sometimes used at spallation sources)
- Hot source at ILL (only one in the world) is graphite, radiation heated.





The Spallation Neutron Source



The ESRF* & ILL* With Grenoble & the Belledonne Mountains

*ESRF = European Synchrotron Radiation Facility; ILL = Institut Laue-Langevin

If we could measure the complex quantity F_{hkl} we could figure out the positions of all atoms. But we only measure $|F_{hkl}|^2$. In fact, we would be better off if diffraction measured phase of scattering rather than amplitude! Unfortunately, nature did not oblige us.

The Phase Problem

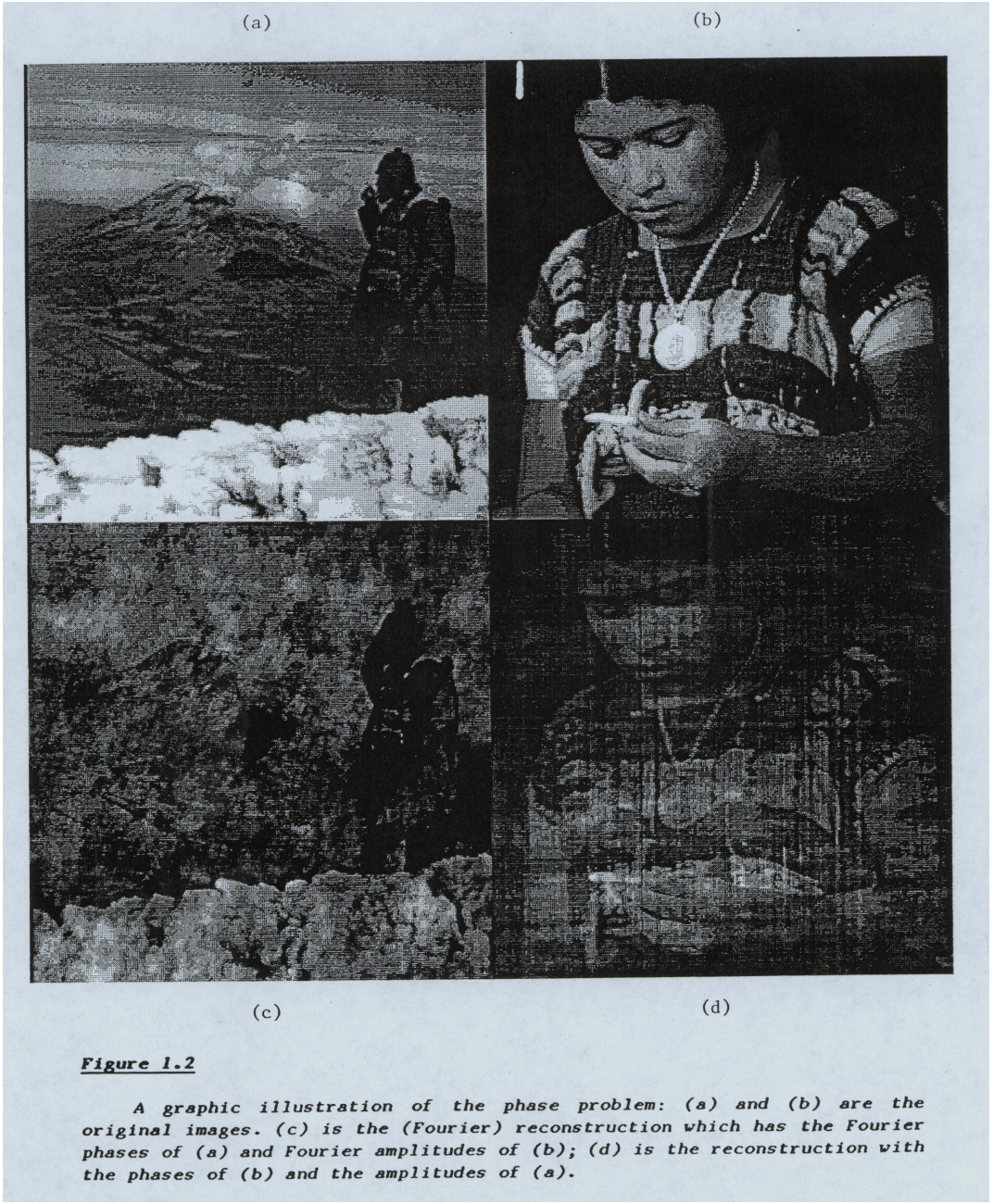
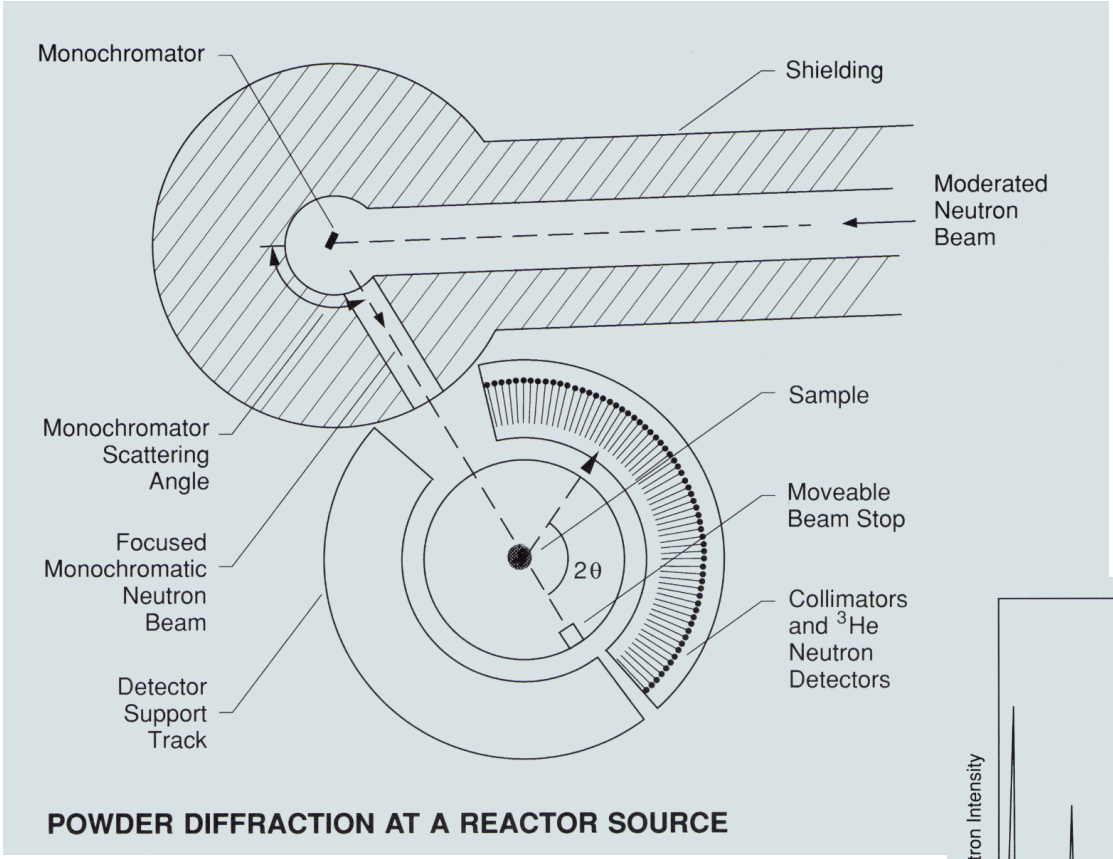


Figure 1.2

A graphic illustration of the phase problem: (a) and (b) are the original images. (c) is the (Fourier) reconstruction which has the Fourier phases of (a) and Fourier amplitudes of (b); (d) is the reconstruction with the phases of (b) and the amplitudes of (a).

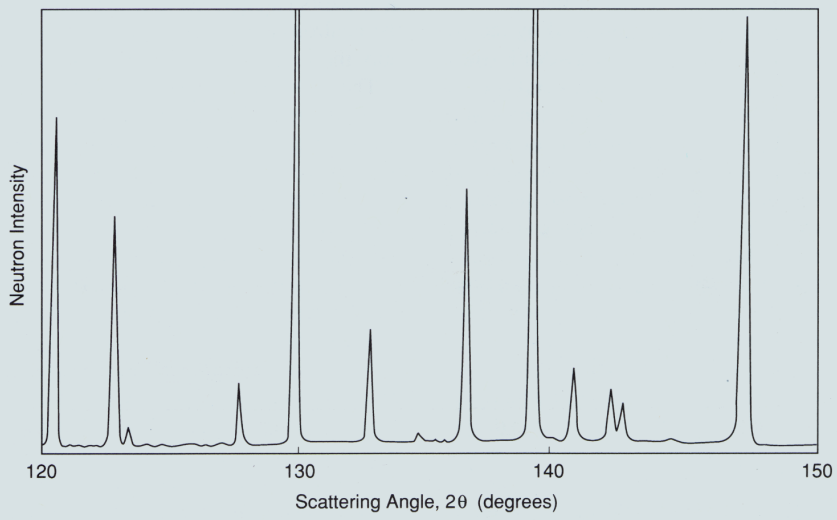
Picture by courtesy of D. Sivia

Measuring Neutron Diffraction Patterns with a Monochromatic Neutron Beam



POWDER DIFFRACTION AT A REACTOR SOURCE

Use a continuous beam of mono-energetic neutrons.

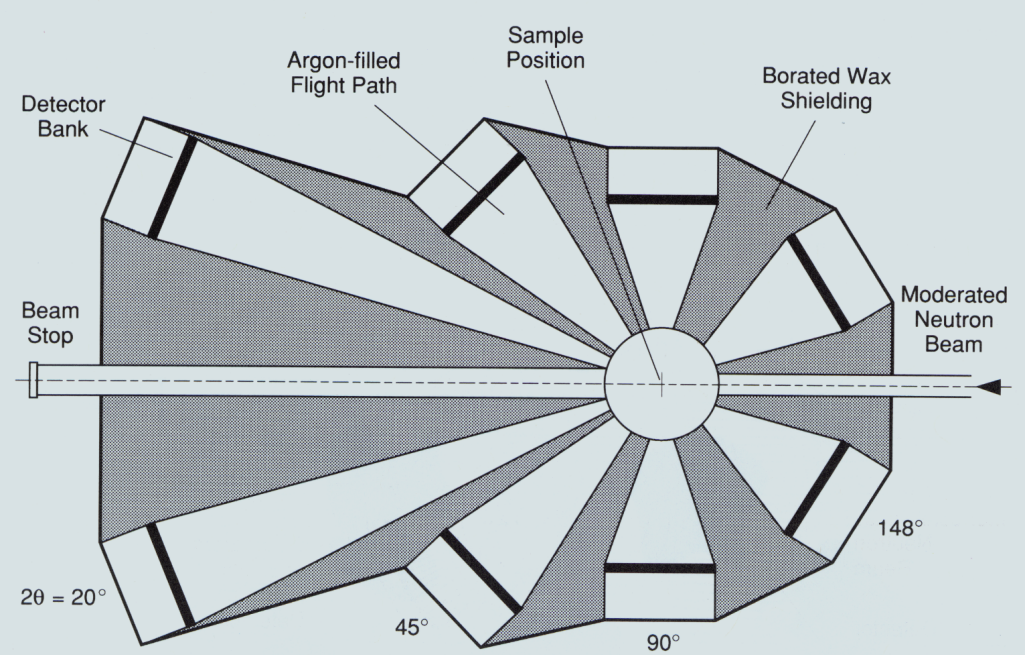


A POWDER DIFFRACTION PATTERN RECORDED AT A REACTOR

Since we know the neutron wavevector, k, the scattering angle gives G_{hkl} directly:

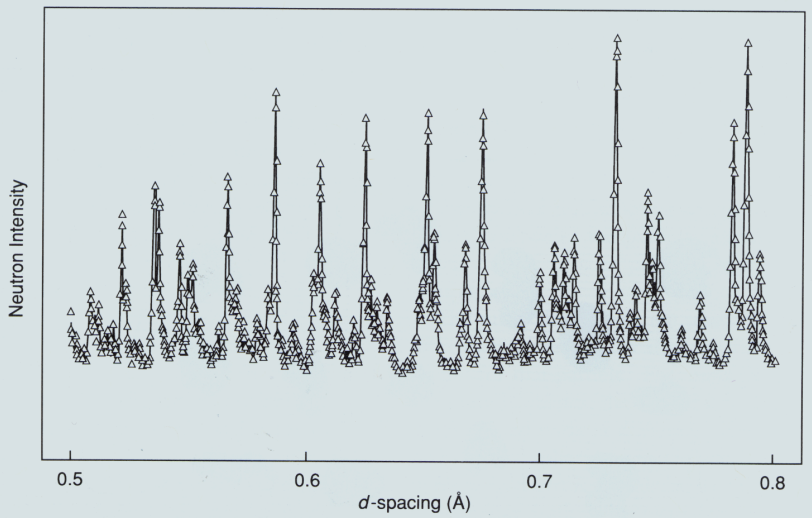
$G_{hkl} = 2\ k \sin \theta$

Time-of-Flight Powder Diffraction



POWDER DIFFRACTION AT A SPALLATION SOURCE

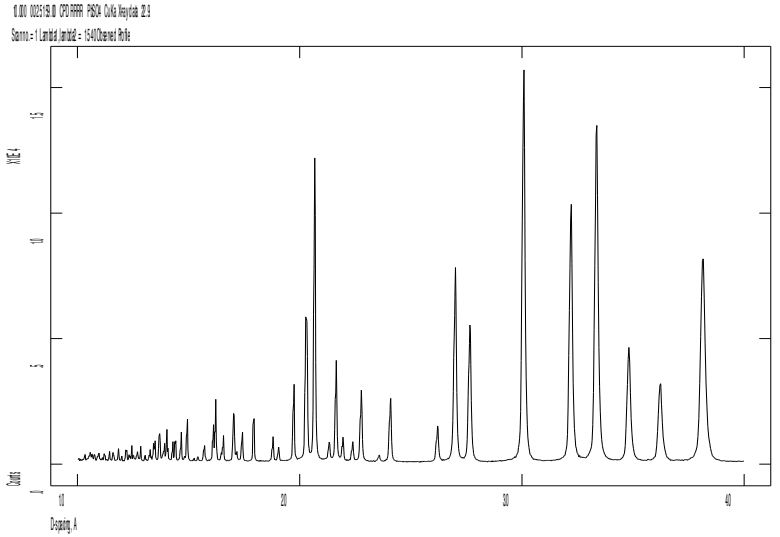
Use a pulsed beam with a broad spectrum of neutron energies and separate different energies (velocities) by time of flight.

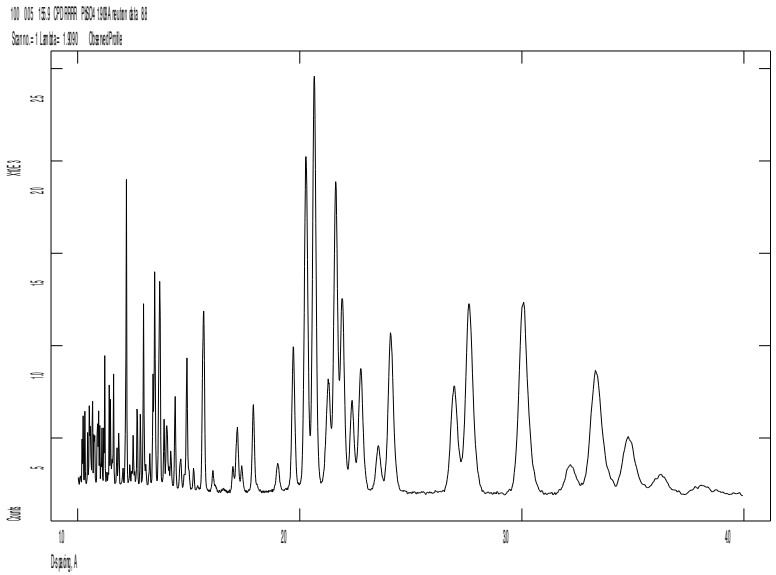


A POWDER DIFFRACTION PATTERN RECORDED AT A SPALLATION SOURCE

Compare X-ray & Neutron Powder Patterns

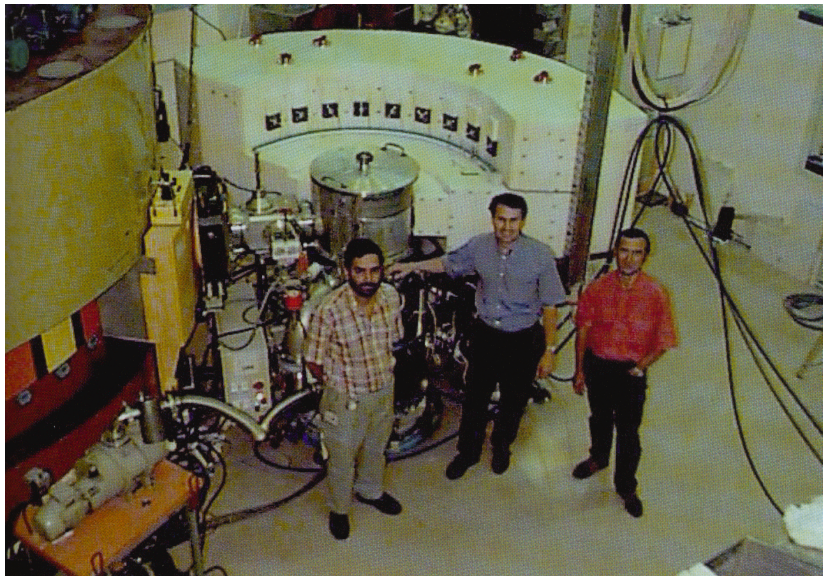
X-ray Diffraction - CuKa Phillips PW1710
- **Higher resolution**
- **Intensity fall-off at small d spacings**
- **Better at resolving small lattice distortions**

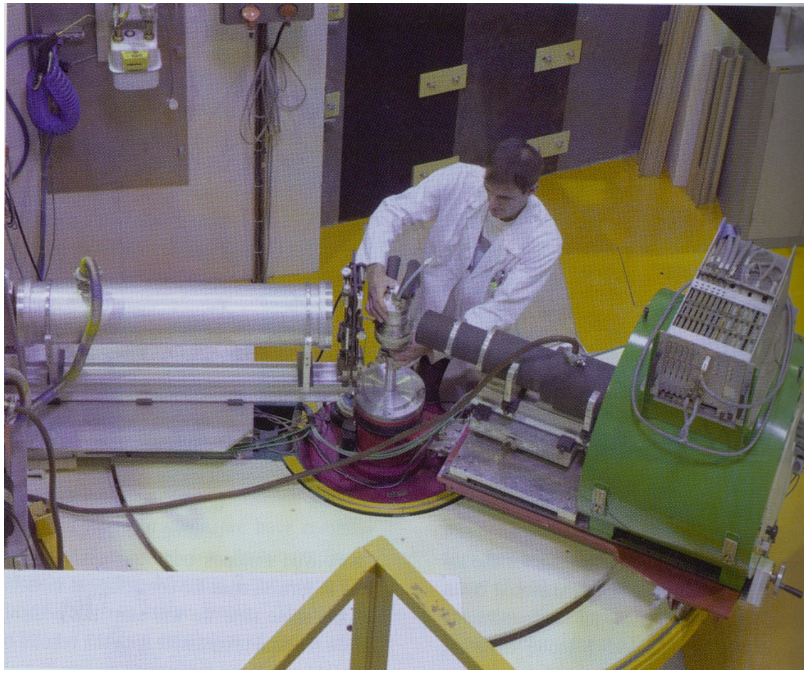


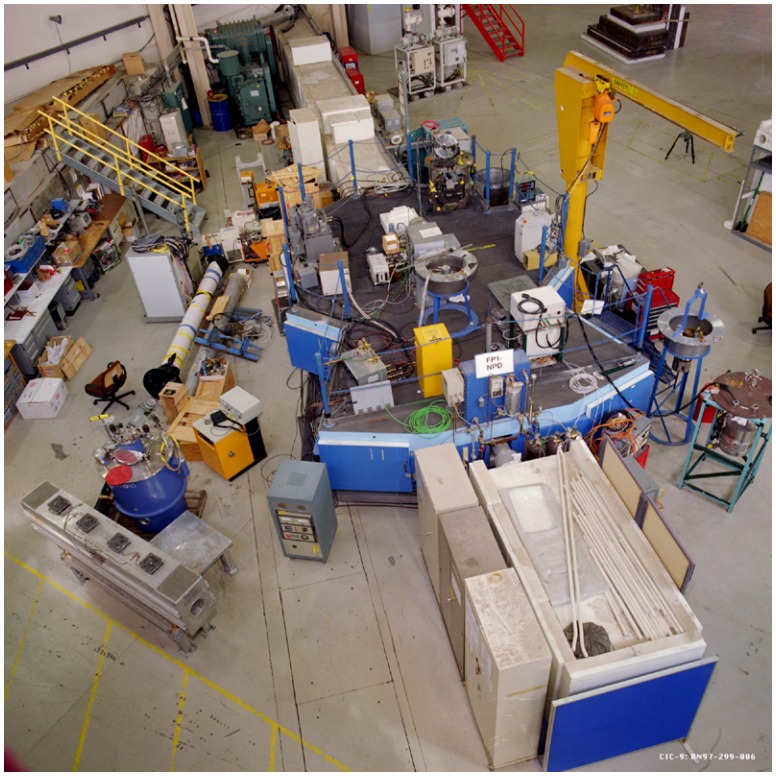


Neutron Diffraction - D1a, ILL λ=1.909 Å
- **Lower resolution**
- **Much higher intensity at small d-spacings**
- **Better atomic positions/thermal parameters**

What do Neutron Powder Diffractometers look like?







Note: relatively massive shielding; long flight paths for time-of-flight spectrometers; many or multi-detectors on modern instruments

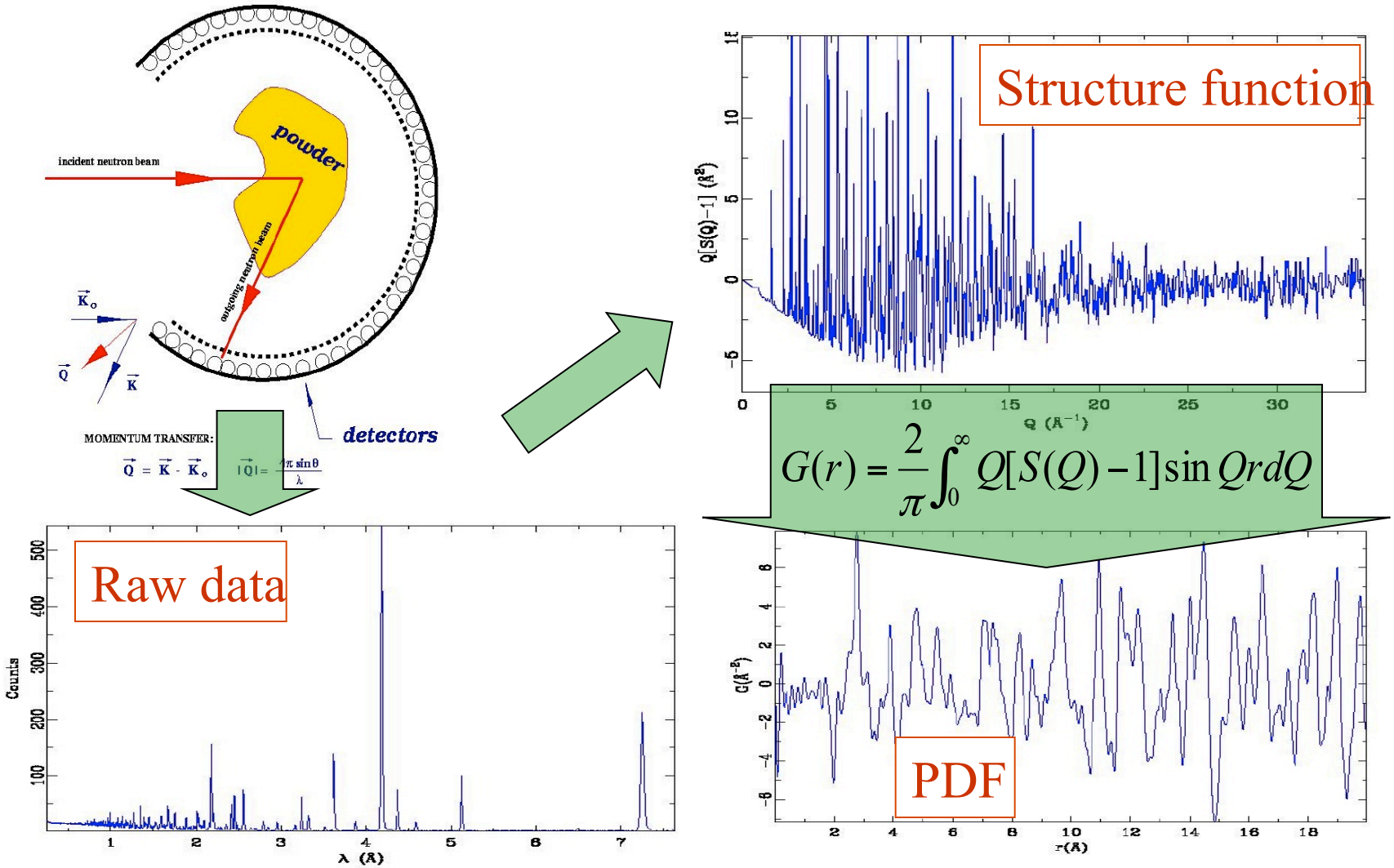
Examples of Science using Neutron Diffraction

- Refinement of structures of new materials
- Materials texture
- Strain measurements
- Laue instruments for neutron protein crystallography
- Pair distribution functions (PDF)

Pair Distribution Functions

- Modern materials are often disordered.
- Standard crystallographic methods lose the aperiodic (disorder) information.
- We would like to be able to sit on an atom and look at our neighborhood.
- The PDF method allows us to do that (see next slide):
  - First we do a neutron or x-ray diffraction experiment
  - Then we correct the data for experimental effects
  - Then we Fourier transform the data to real-space

Obtaining the Pair Distribution Function*



* See http://www.pa.msu.edu/cmp/billinge-group/

Small Angle Neutron Scattering (SANS) Is Used to Measure Large Objects (~10 nm to ~1 μm)

Recall that :

$\vec{Q} = \vec{k}' - \vec{k}_0 = 2k_0 \sin\theta$ for elastic scattering

and that

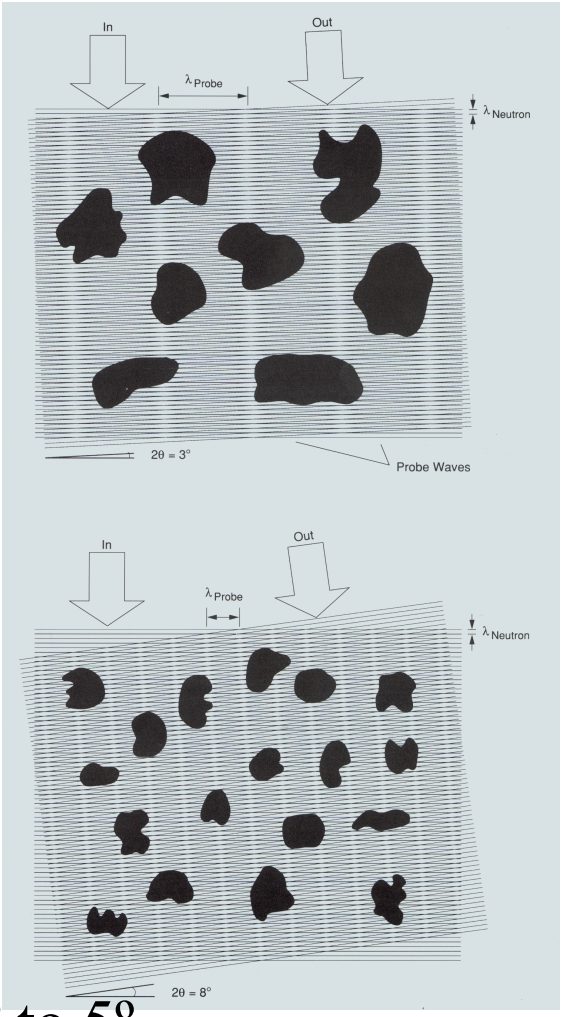
$\lambda = 2\pi / k = 2\pi /(Q / 2\sin\theta) = 4\pi \sin\theta / Q$

so we can rewrite Bragg's law $\lambda = 2d \sin\theta$ as

$d = 2\pi/Q$ or for small θ $d \approx \lambda/2\theta$

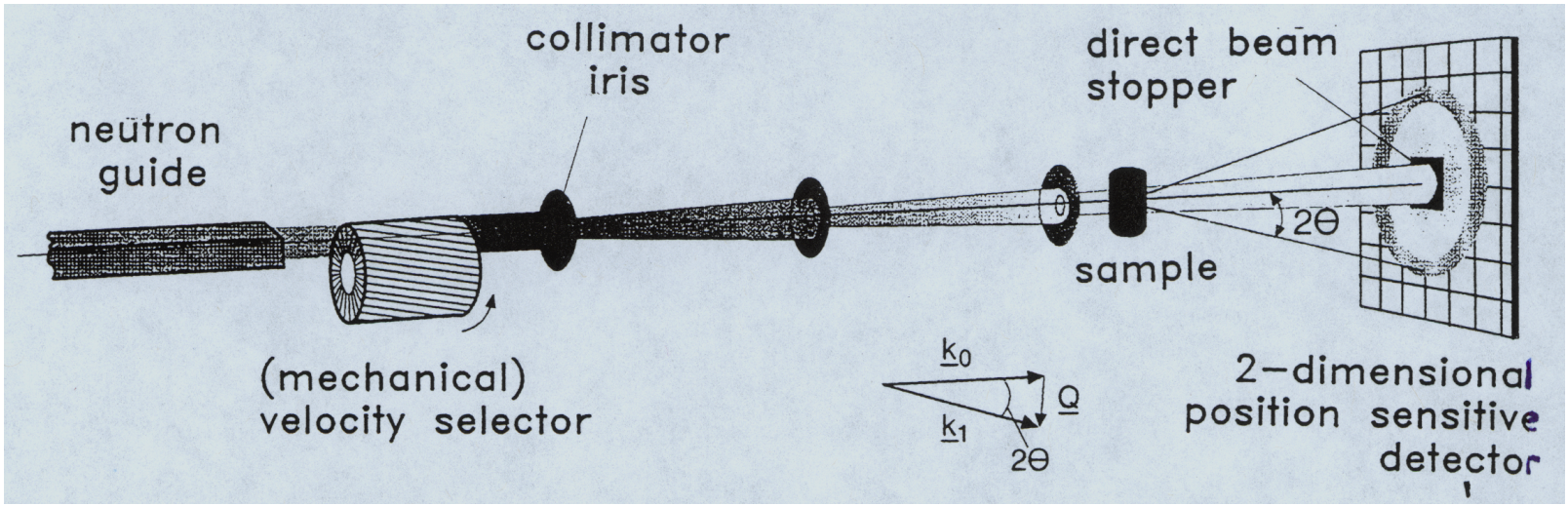
i.e. small Q => large length scales

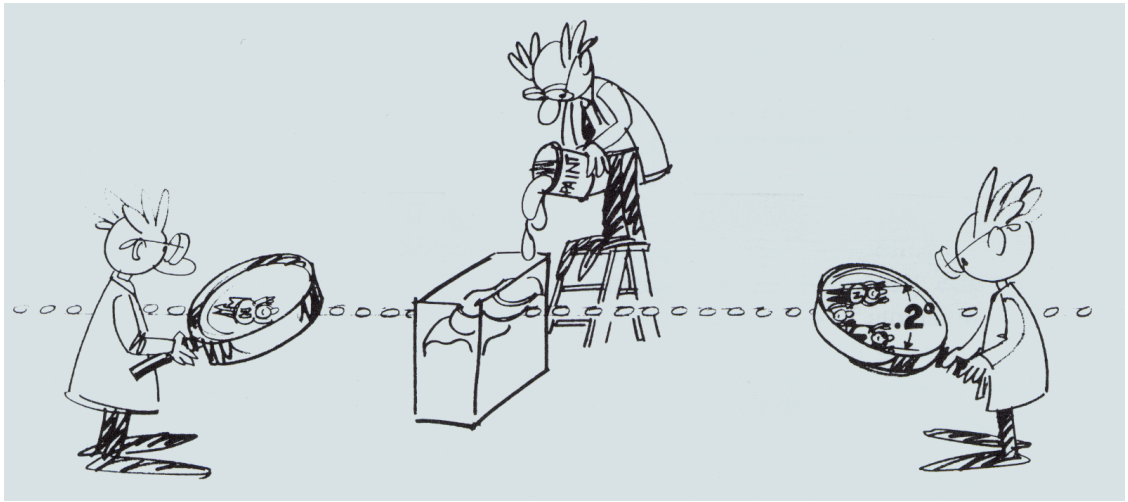
Scattering at small angles probes large length scales



Typical scattering angles for SANS are ~ 0.3º to 5º

Two Views of the Components of a Typical Reactor-based SANS Diffractometer

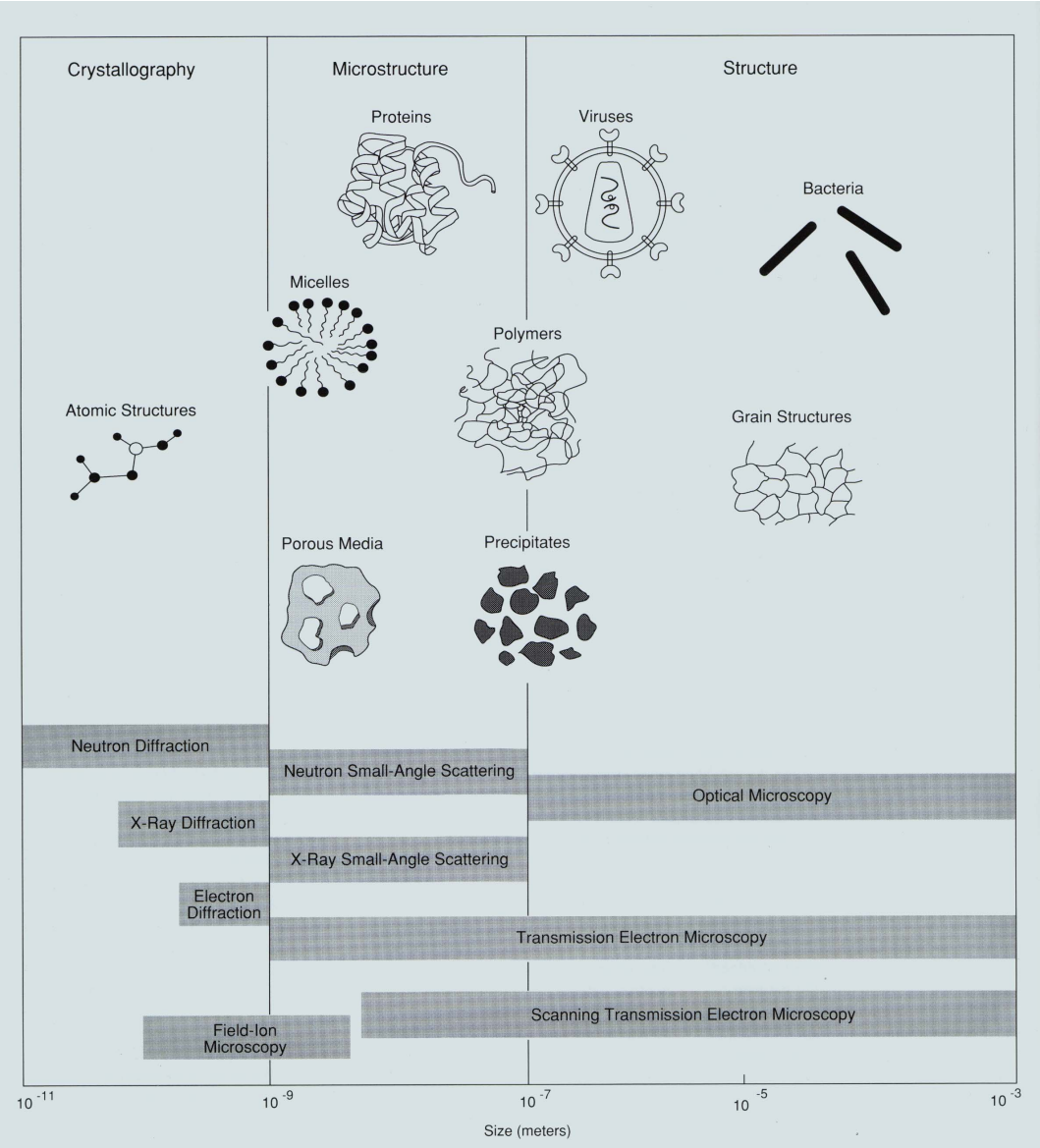




Note that SANS, like other diffraction methods, probes material structure in the direction of the wavevector transfer, Q

The NIST 30m SANS Instrument Under Construction

Where Does SANS Fit As a Structural Probe?



- SANS resolves structures on length scales of 1 – 1000 nm
- Neutrons can be used with bulk samples (1-2 mm thick)
- SANS is sensitive to light elements such as H, C & N
- SANS is sensitive to isotopes such as H and D

Typical SANS Applications

- Biology
  - Organization of biomolecular complexes in solution
  - Conformation changes affecting function of proteins, enzymes, protein/DNA complexes, membranes etc
  - Mechanisms and pathways for protein folding and DNA supercoiling
- Polymers
  - Conformation of polymer molecules in solution and in the bulk
  - Structure of microphase separated block copolymers
  - Factors affecting miscibility of polymer blends
- Chemistry
  - Structure and interactions in colloid suspensions, microemeulsions, surfactant phases etc
  - Mechanisms of molecular self-assembly in solutions

Verification of of the Gaussian Coil Model for a Polymer Melt

- One of the earliest important results obtained by SANS was the verification of that $r_g \sim N^{-1/2}$ for polymer chains in a melt

- A better experiment was done 3 years later using a small amount of H-PMMA in D-PMMA (to avoid the large incoherent background) covering a MW range of 4 decades

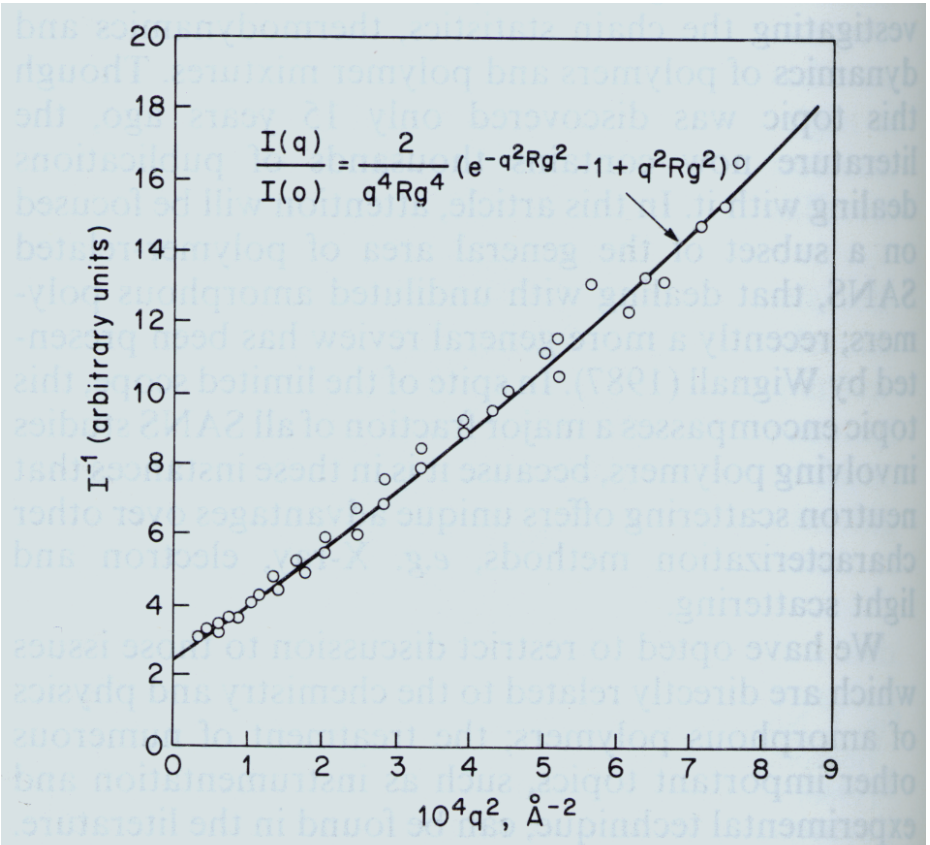
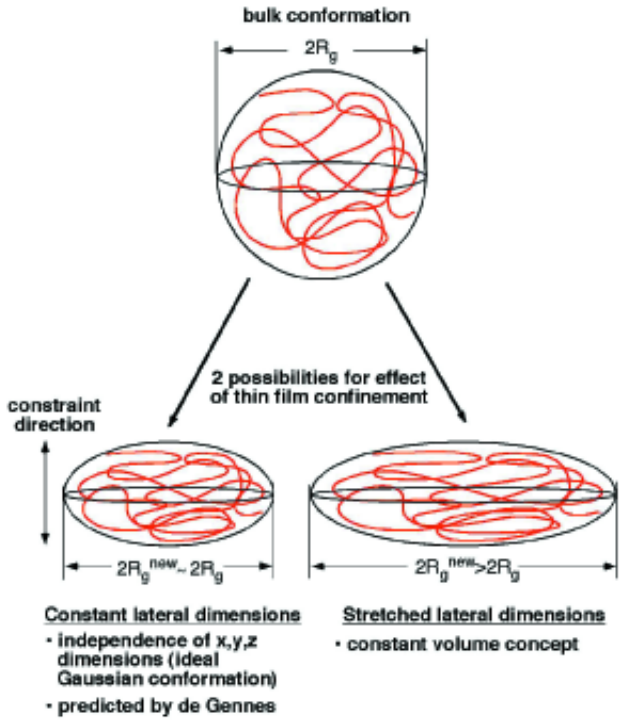
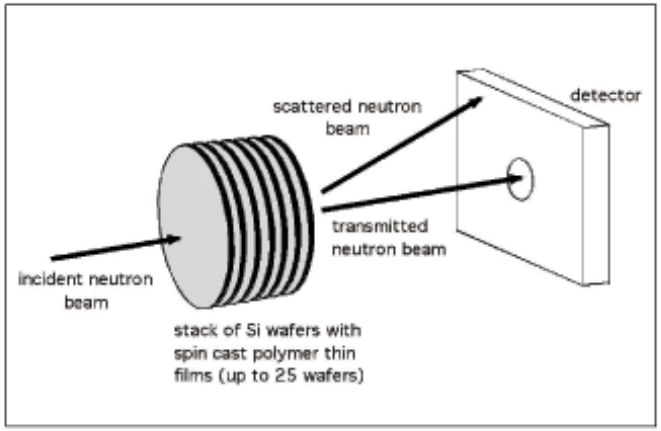


Fig. 1. SANS results obtained by Kirste, Kruse & Schelten (1972) for 1·2% deuterated poly(methyl methacrylate) (PMMA) in normal PMMA (mol. wt of 250 000) plotted in Ornstein–Zernike form. The solid curve represents a Debye function [equation (1)]. This was one of the first quantitative demonstrations of Gaussian coil behavior for bulk polymers.

Probing Chain Conformation in Thin Films

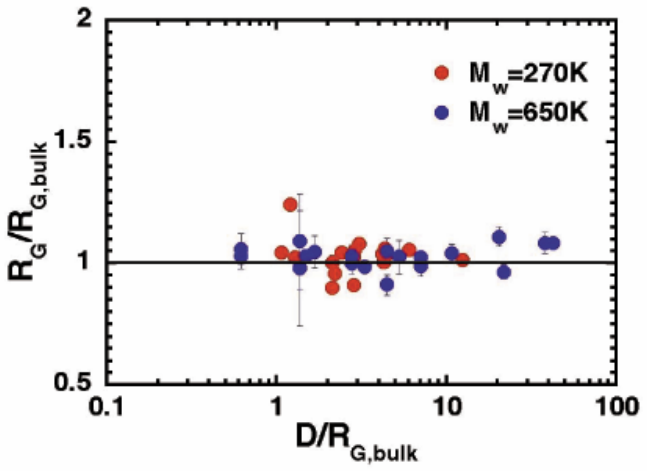


R.L. Jones, S.K. Kumar, D.L. Ho, R.M. Briber, T.P. Russell, *Nature*, 1999, 400, 146

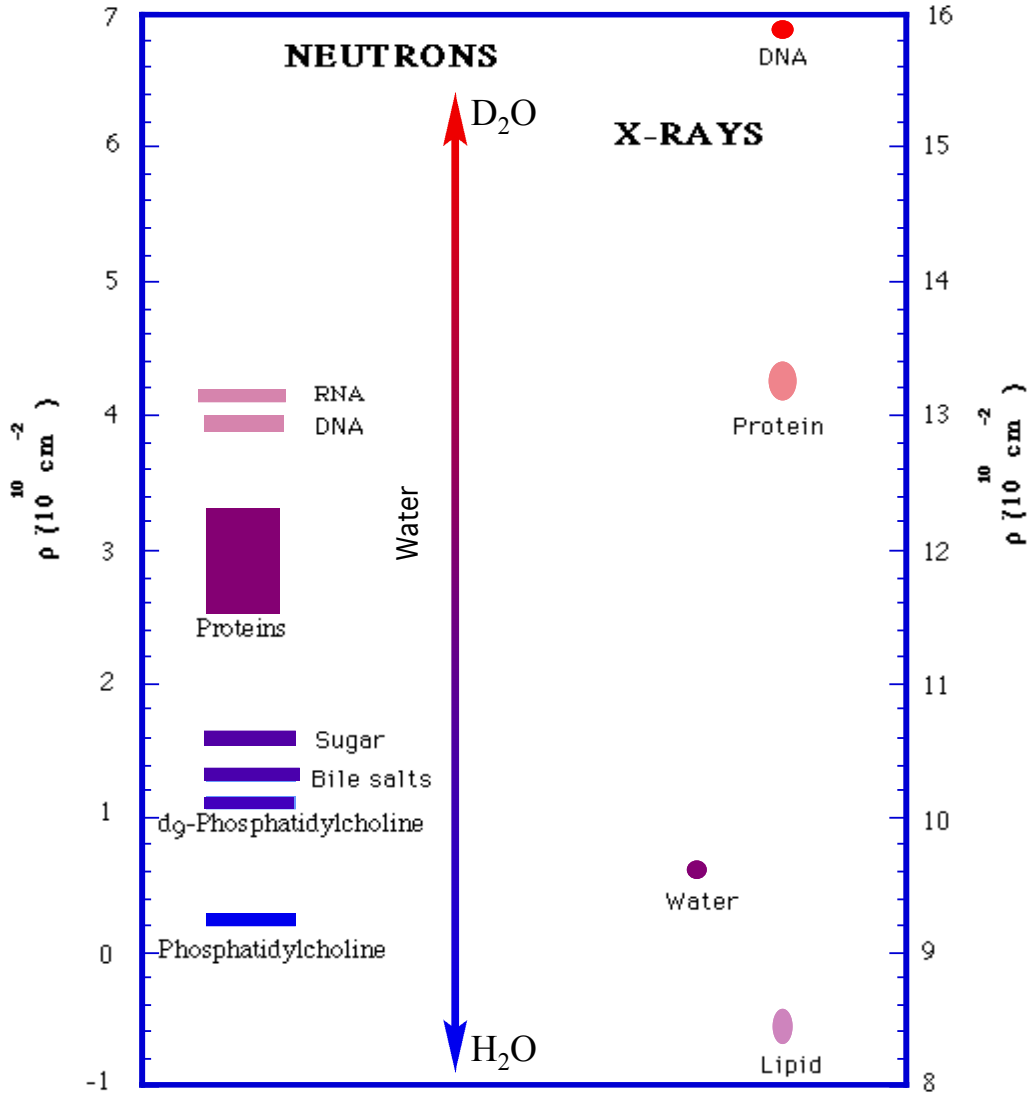


Thin films of 25% d-PS & 75% PS spun on to Si wafers. 25 wafers => 10 nm total polymer thickness – 0.2 mg

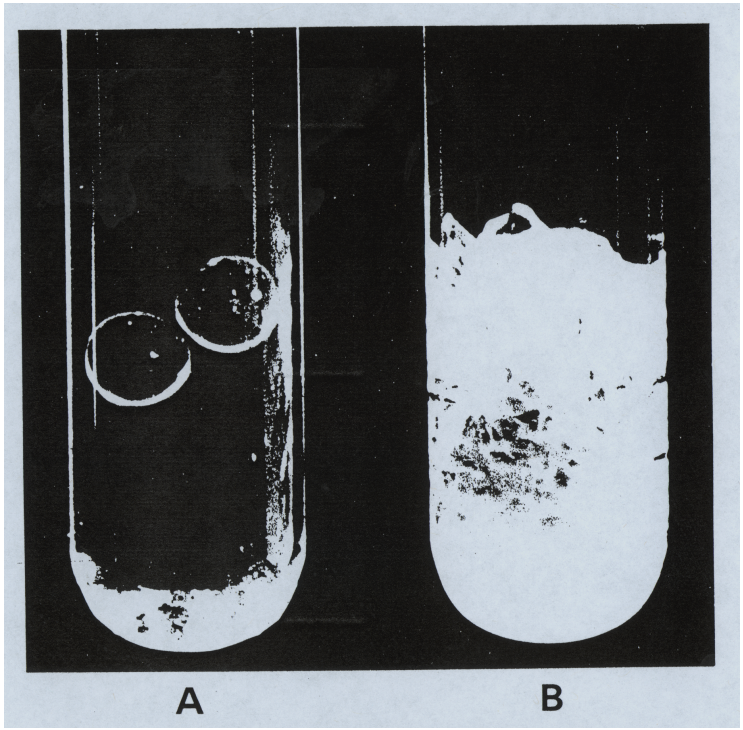
R_g in the plane of the film is unchanged down to film thickness of $R_g/2$



Contrast & Contrast Matching

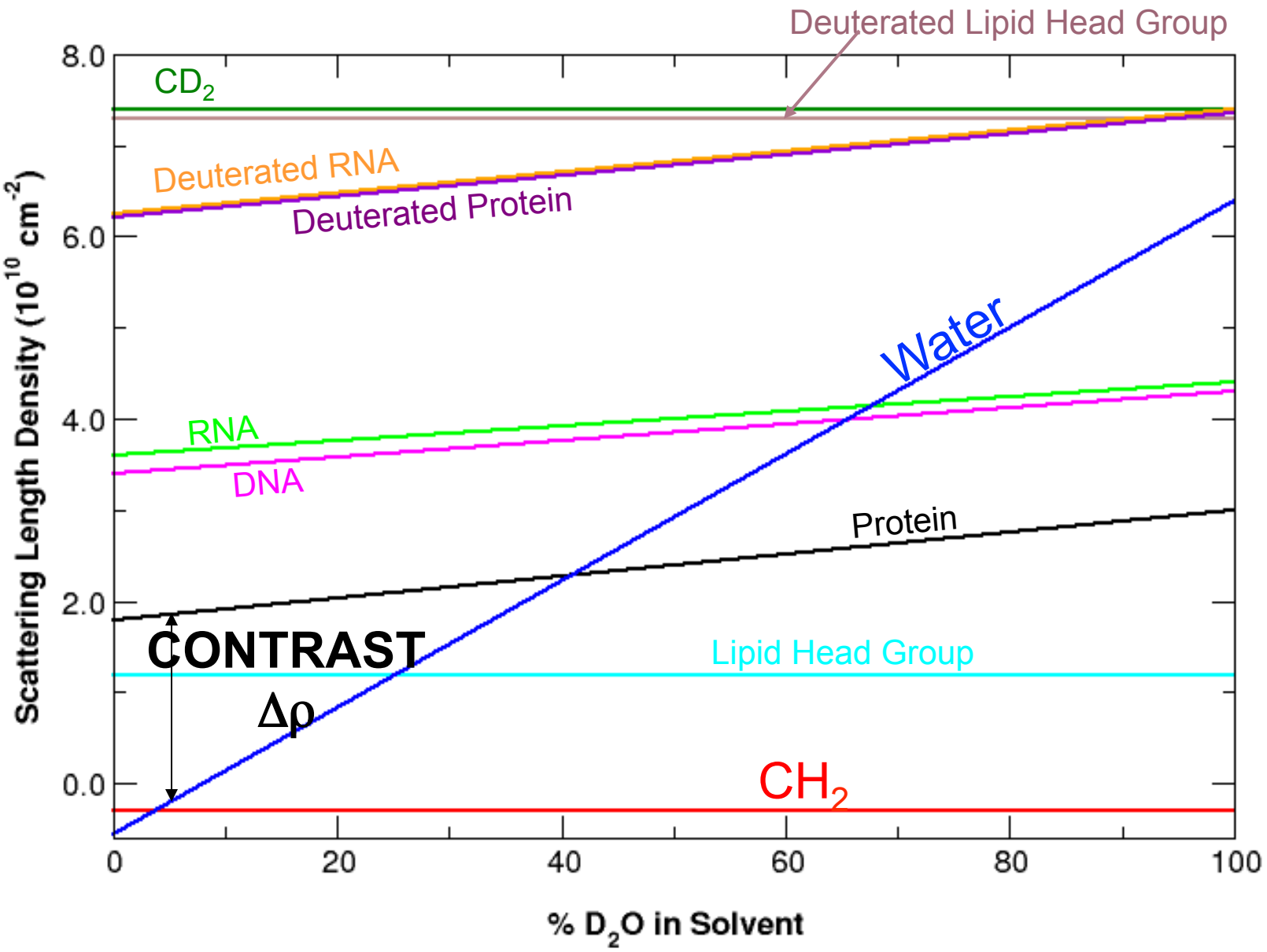


* Chart courtesy of Rex Hjelm

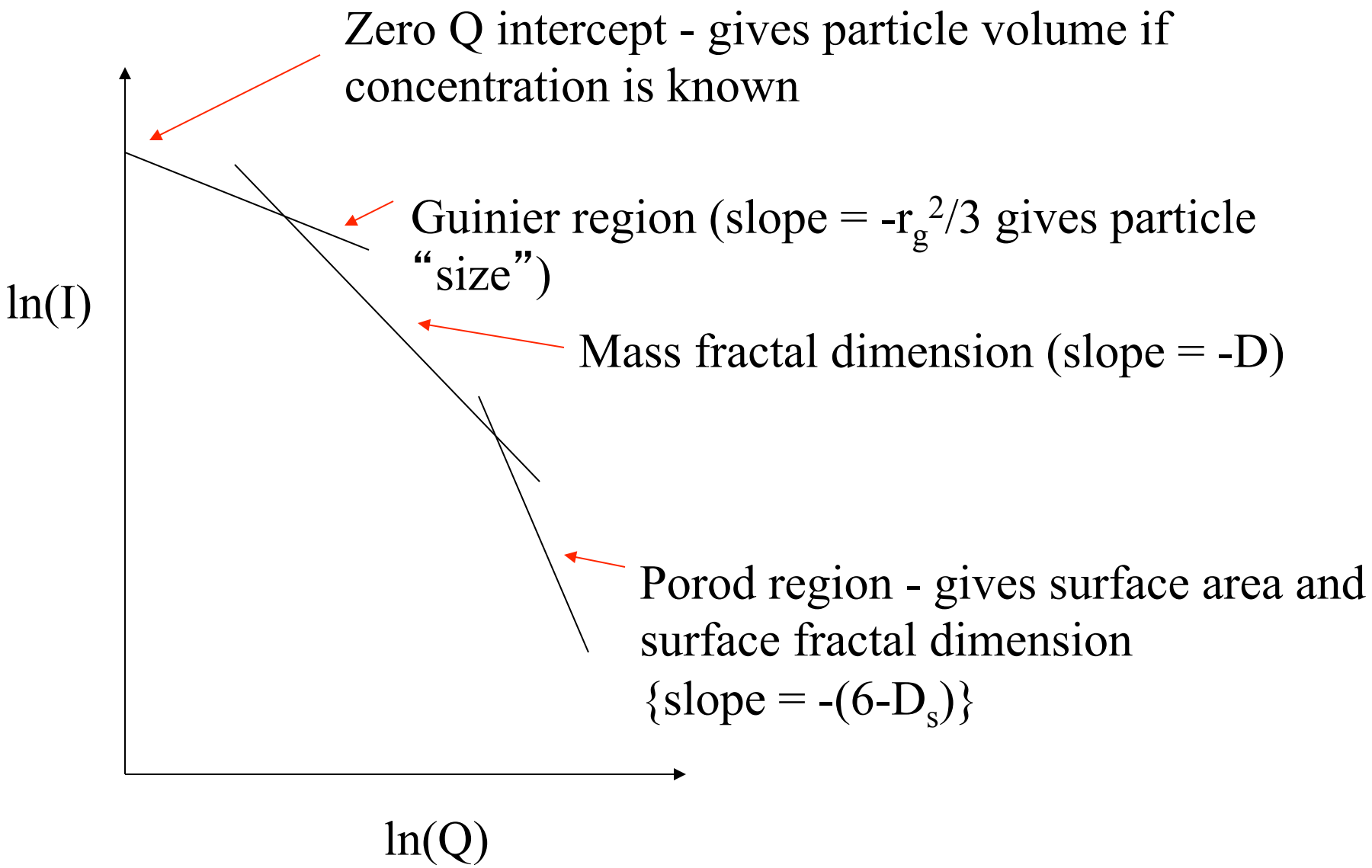


Both tubes contain borosilicate beads + pyrex fibers + solvent. (A) solvent refractive index matched to pyrex;. (B) solvent index different from both beads and fibers – scattering from fibers dominates

Contrast Variation



Typical Intensity Plot for SANS From Disordered Systems



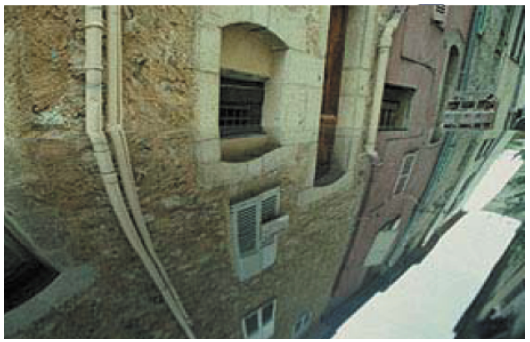
Surface Reflection Is Very Different From Most Neutron Scattering

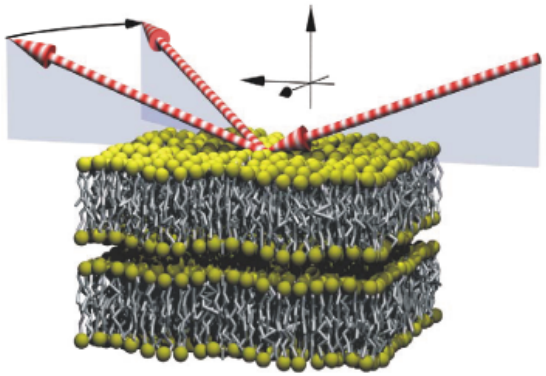
- We worked out the neutron cross section by adding scattering from different nuclei
  - We ignored double scattering processes because these are usually very weak
- This approximation is called the Born Approximation

- Below an angle of incidence called the critical angle, neutrons are perfectly reflected from a smooth surface
  - This is NOT weak scattering and the Born Approximation is not applicable to this case
- Specular reflection is used:
  - In neutron guides
  - In multilayer monochromators and polarizers
  - To probe surface and interface structure in layered systems

Why Use Neutron Reflectivity?

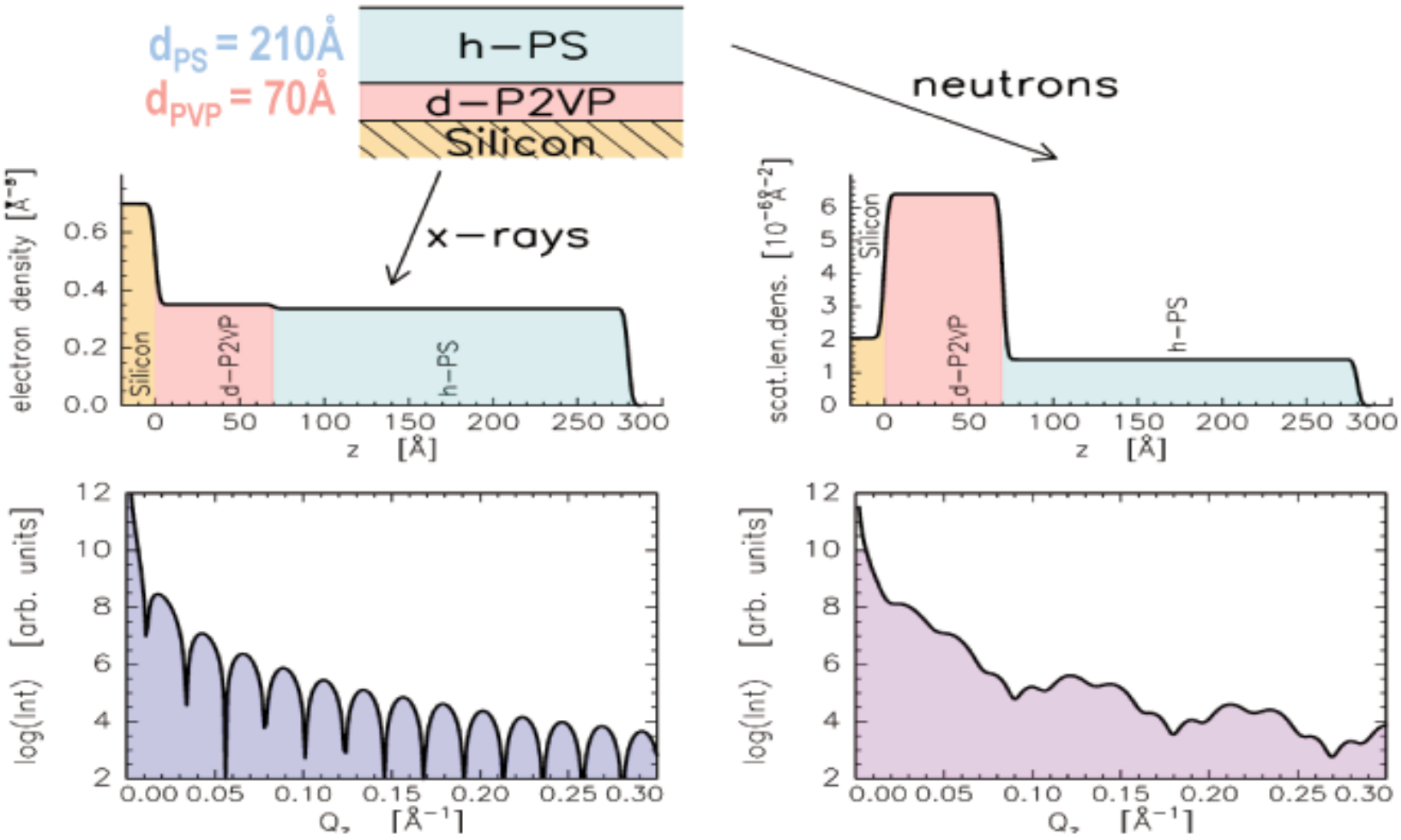
- Neutrons are reflected from most materials at grazing angles
- If the surface is flat and smooth the reflection is specular
  - Perfect reflection below a critical angle
  - Above the critical angle reflectivity is determined by the variation of scattering length density perpendicular to the surface
  - i.e. we can determine the "average" density profile normal to the surface of a film on the surface





Images courtesy of M. Tolan & T. Salditt

Comparison of Neutron and X-Ray Reflectivity



Neutrons often provide better contrast and don't damage samples
X-rays provide better Q resolution and higher Q values

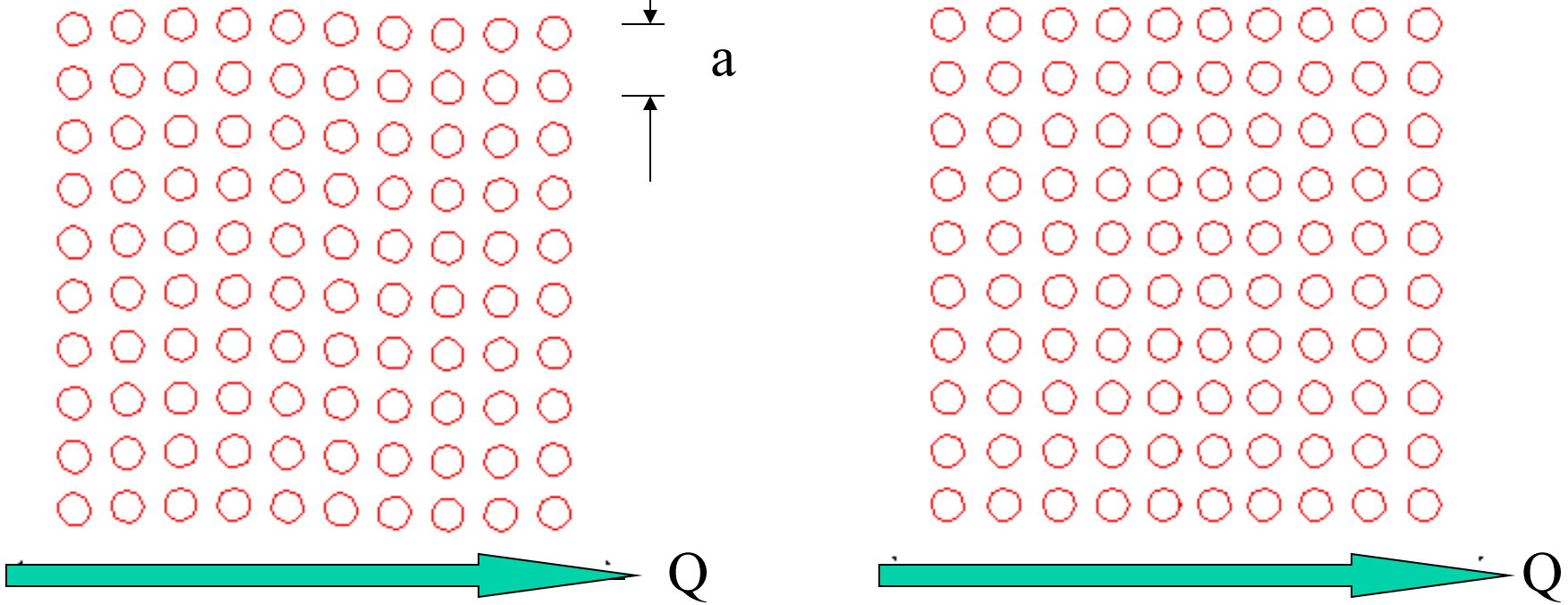
Viewgraph courtesy of M. Tolan

The Goal of Reflectivity Measurements Is to Infer a Density Profile Perpendicular to a Flat Interface

- In general the results are not unique, but independent knowledge of the system often makes them very reliable
- Frequently, layer models are used to fit the data
- Advantages of neutrons include:
  - Contrast variation (using H and D, for example)
  - Low absorption – probe buried interfaces, solid/liquid interfaces etc
  - Non-destructive
  - Sensitive to magnetism
  - Thickness length scale 10 – 5000 Å
- Issues include
  - Generally no unique solution for the SLD profile (use prior knowledge)
  - Large samples (~10 cm^2) with good scattering contrast are needed

Atomic Motions for Longitudinal & Transverse Phonons

$$\vec{Q} = \frac{2\pi}{a}(0.1,0,0)$$



Transverse phonon

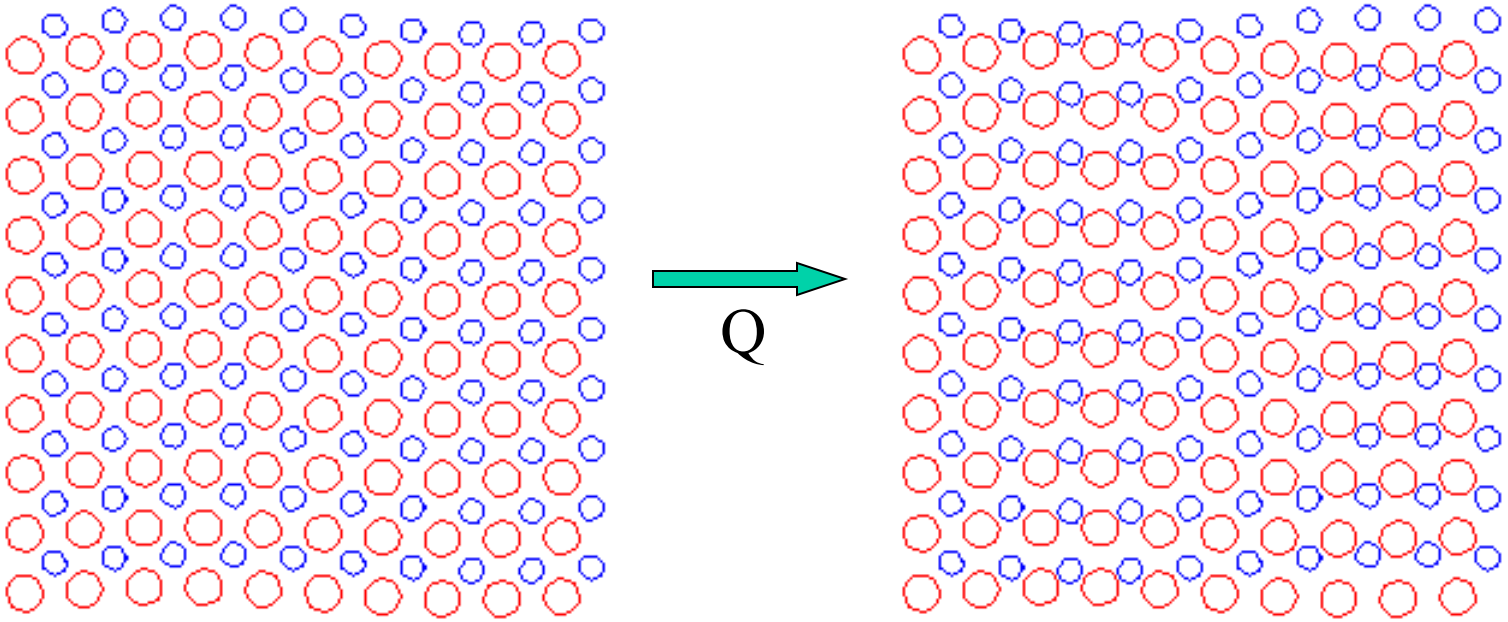
$$\vec{e}_T = (0,0.1,0)a$$

Longitudinal phonon

$$\vec{e}_L = (0.1,0,0)a$$

$$\vec{R}_l = \vec{R}_{l0} + \vec{e}_s e^{i(\vec{Q}.\vec{R}_l - \omega t)}$$

Transverse Optic and Acoustic Phonons



Acoustic

$\vec{e}_{red} = (0,0.1,0)a$

$\vec{e}_{blue} = (0,0.14,0)a$

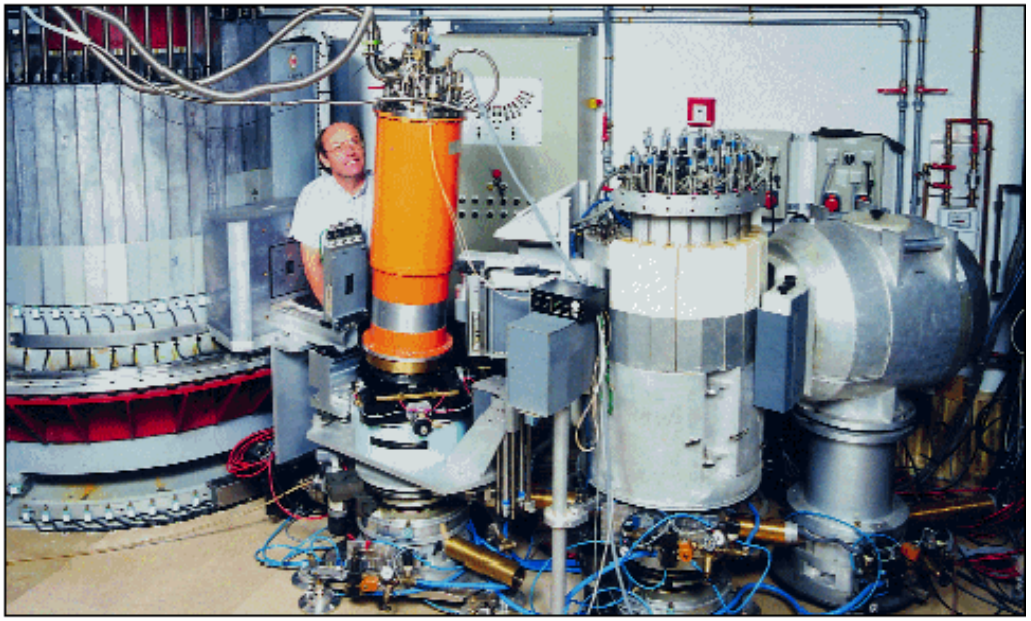
Optic

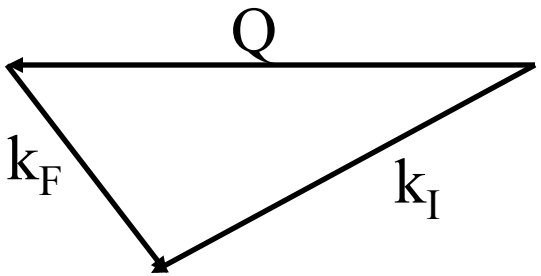
$\vec{e}_{red} = (0,0.1,0)a$

$\vec{e}_{blue} = (0,-0.14,0)a$

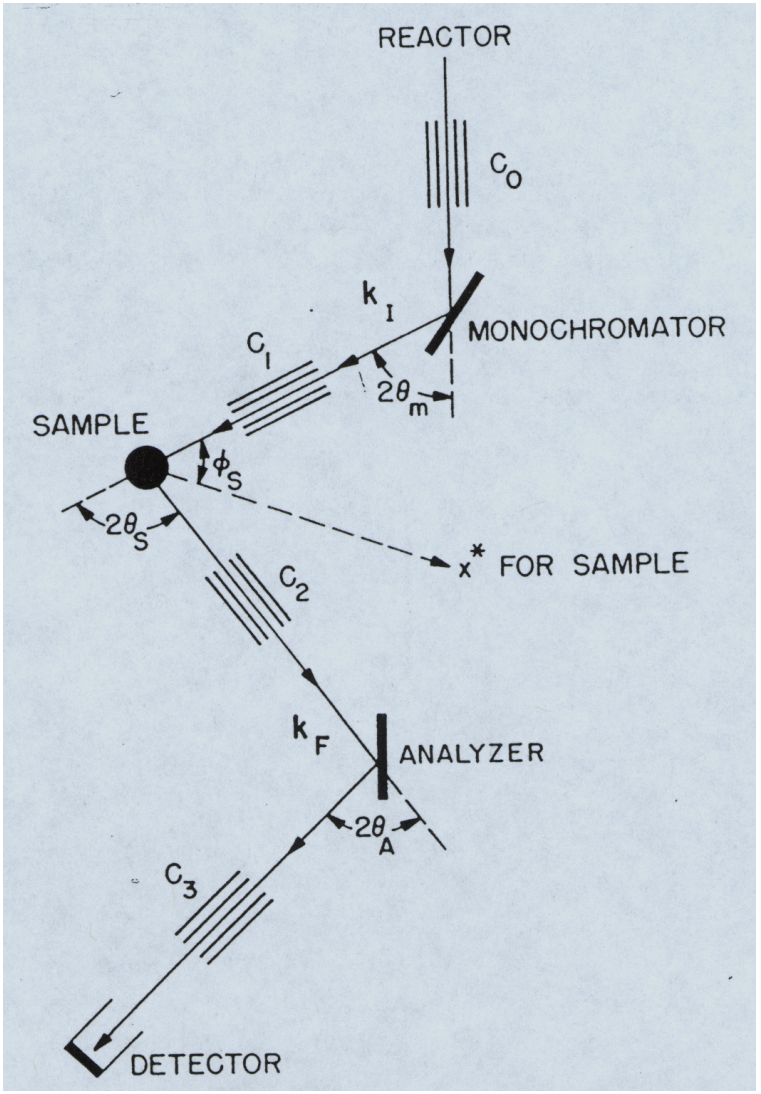
$$\vec{R}_{lk} = \vec{R}^0_{lk} + \vec{e}_s e^{i(\vec{Q}.\vec{R}_l - \omega t)}$$

The Workhorse of Inelastic Scattering Instrumentation at Reactors Is the Three-axis Spectrometer



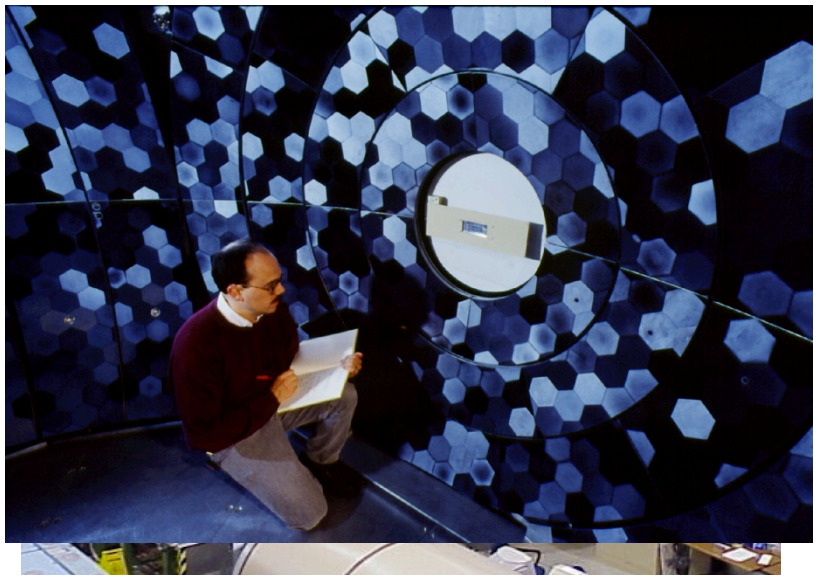


"scattering triangle"



Spectrometers for Measuring Quasielastic Scattering

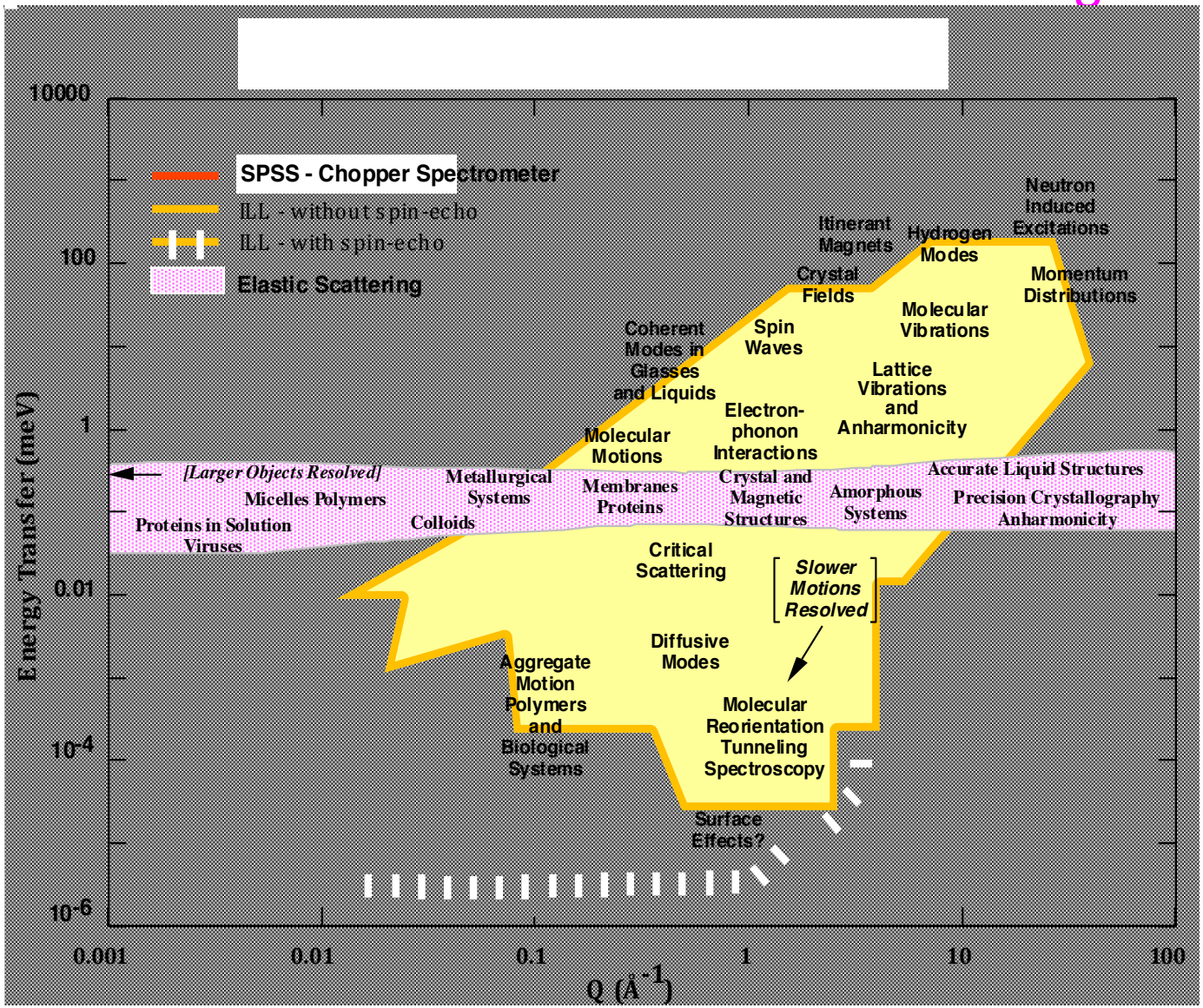
Chopper spectrometer with pulsed monochromatic incident neutron beam and time-of-flight energy analysis
$0.01 < \Delta E < 0.1$ meV for cold neutrons
$1 < \Delta E < 10$ meV for thermal neutrons



Backscattering spectrometer with polychromatic incident beam and energy analysis by crystal analyzer
$0.001 < \Delta E < 0.1$ meV for cold neutrons

Note (1) that the value of the energy resolution, ΔE, sets the minimum observed width of spectral line and (2) that the good energy resolution of backscattering is obtained at the expense of poor Q resolution

Much of the Scientific Impact of Neutron Scattering Has Involved the Measurement of Inelastic Scattering



Energy & Wavevector Transfers accessible to Neutron Scattering