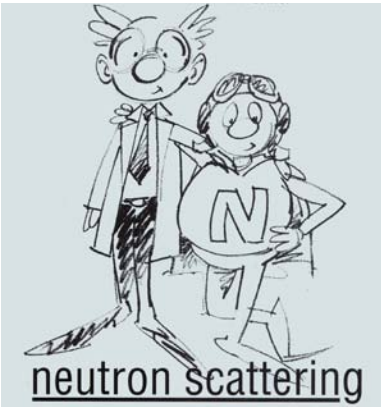
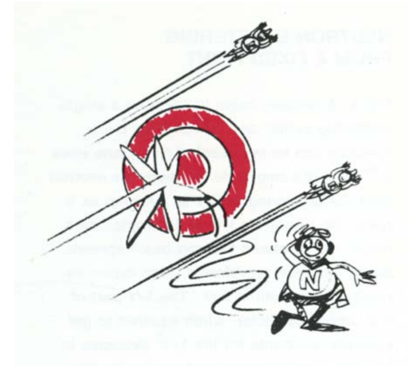




An Introduction to Neutron Scattering

by

Roger Pynn

Indiana University and Oak Ridge National Lab

With thanks to various friends for allowing me to use their materials

INDIANA UNIVERSITY

The next-generation neutron-scattering facility for the United States

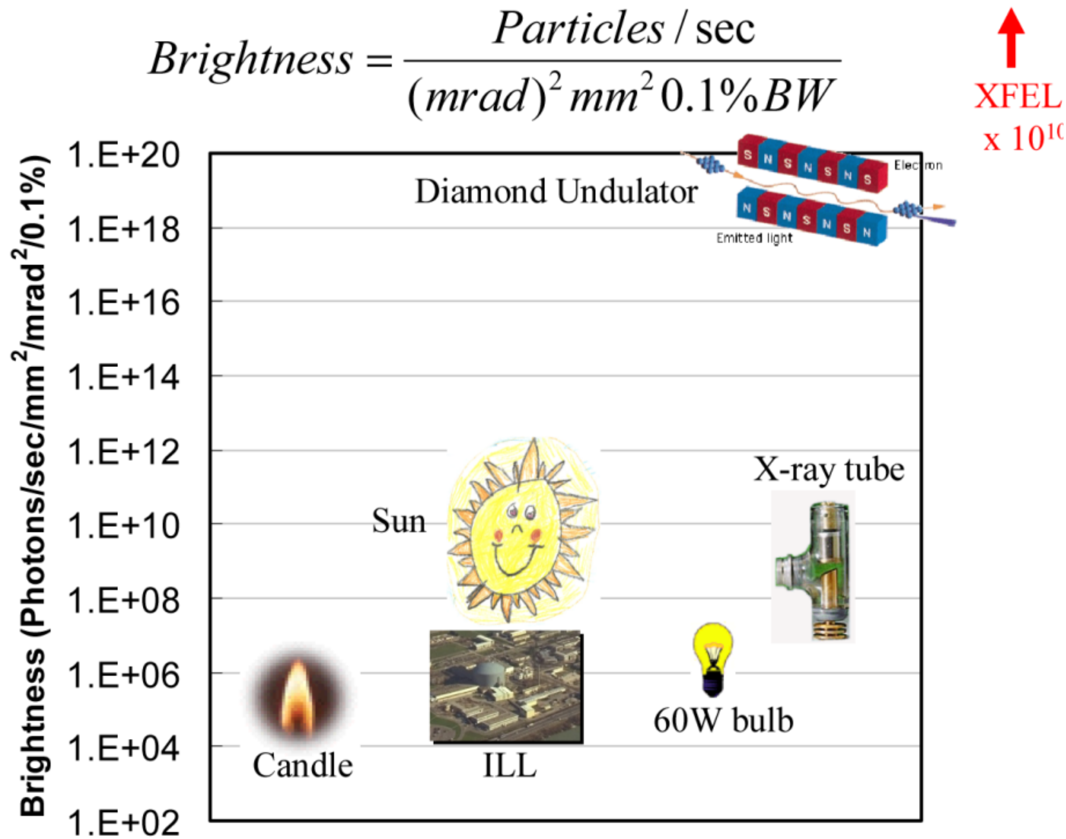
SNS

You Know All About X-Rays – Why Do Neutron Scattering?

- Not because neutron sources are bright!
- In fact, neutron scattering is signal-limited
- So why do it?

Source Brightness

$$Brightness = \frac{Particles / \sec}{(mrad)^2 mm^2 0.1\% BW}$$



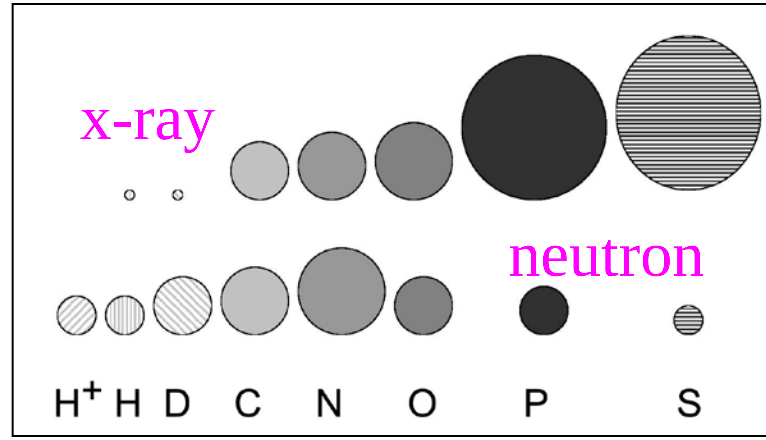
Brightness & Fluxes for Neutron & X-Ray Sources

	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}

Flux = brightness * divergence; brilliance = brightness / energy bandwidth

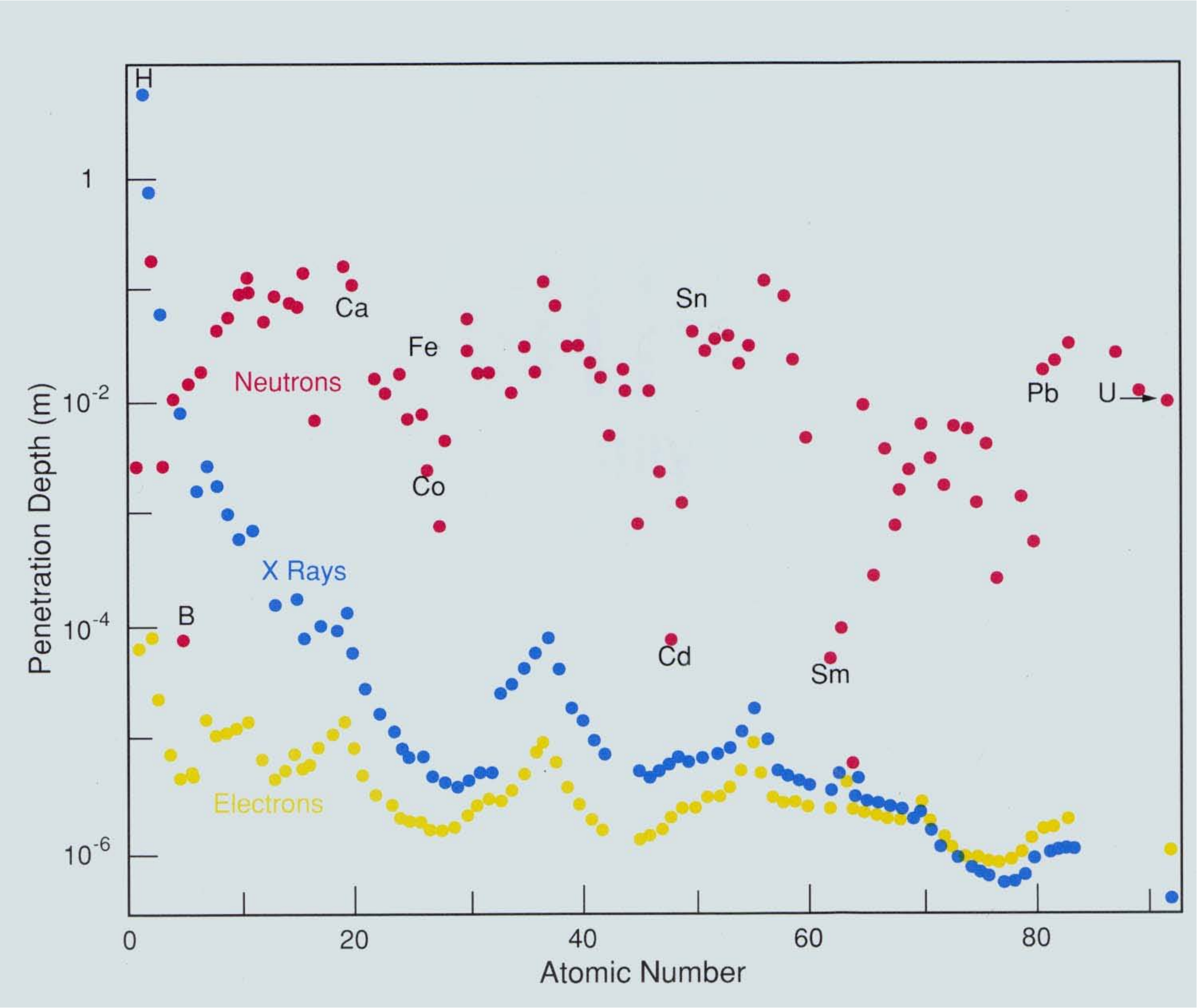
The Success of Neutron Scattering is Rooted in the Neutron's Interactions with Matter

- Interact with nuclei not electrons
- Isotopic sensitivity (especially D and H)
- Penetrates sample containment
- Sensitive to bulk and buried structure



- Simple interpretation – provides statistical averages, not single instances
- Wavelength similar to inter-atomic spacings
- Energy similar to thermal energies in matter
- Nuclear and magnetic interactions of similar strength

Thermal Neutrons, 8 keV X-Rays & Low Energy Electrons:- Penetration in Matter



Note for neutrons:

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

For x-rays:

- decreasing penetration as Z increases

Advantages & Disadvantages of Neutrons

- Advantages
  - λ similar to interatomic spacings
  - Penetrates bulk matter
  - Strong contrast possible
  - Energy similar to that of elementary excitations (phonons, magnons etc)
  - Scattering strongly by magnetic fields
  - Data interpretation is direct
- Disadvantages
  - Low brilliance of neutron sources
  - Some elements absorb neutrons strongly
  - Kinematic restrictions on Q for large energy transfers
  - Difficult to study excitations at high (eV) energies
  - Provides statistical averages rather than real space pictures

Some Neutron History

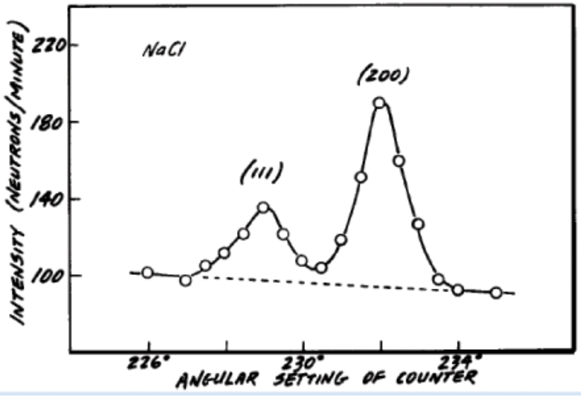
- 1932 – Chadwick discovers the neutron
- 1934 – thermalisation (Fermi)
- 1936 – scattering theory (Breit, Wigner)
- 1936 – wave interference (Mitchell, Powers)
- 1939 – fission

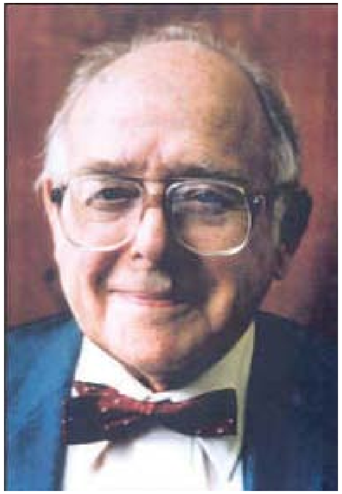
- 1945 – diffraction (Shull, Wollan), reflection, refraction
- 1948 – coherent & incoherent scattering (Shull, Wollan)
- 1948 – spallation
- 1949 – structure of AFM (Shull)

- 1951 – polarized neutrons (Shull & Wollan)
- 1955 – three axis spectrometer (Brockhouse)
- 1958 – rotons in helium (Palevsky, Otnes, Larsson)

- 1962 – Kohn anomalies
- 1960 – 79 – soft phonons & structural phase transitions
- 1969 – 79 – scaling and universality

- 1972 – conformation of polymers
-
- 1994 – Nobel Prize for Shull and Brockhouse

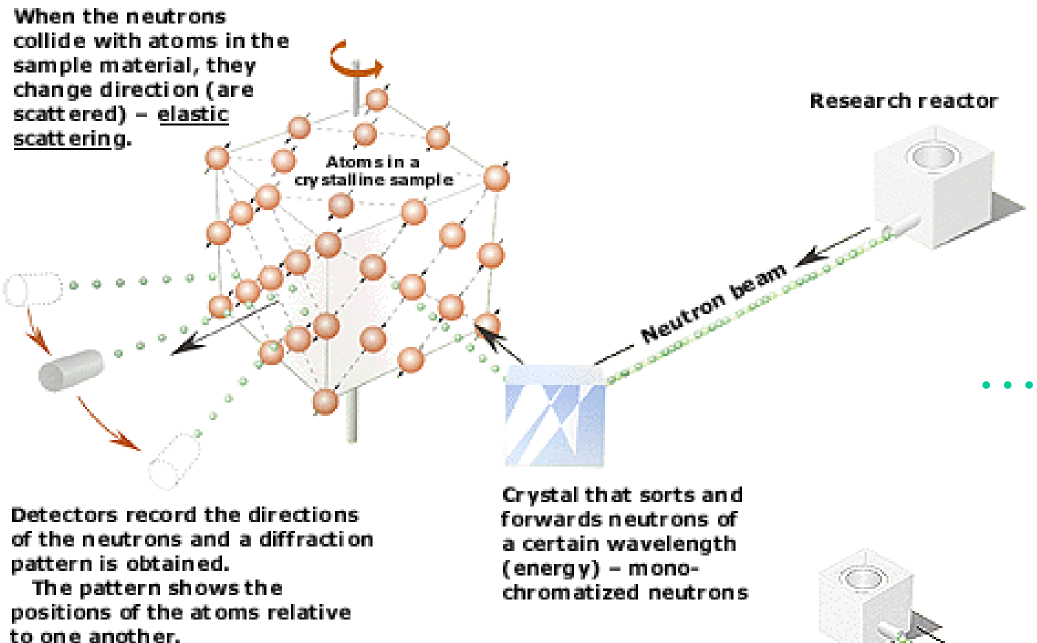




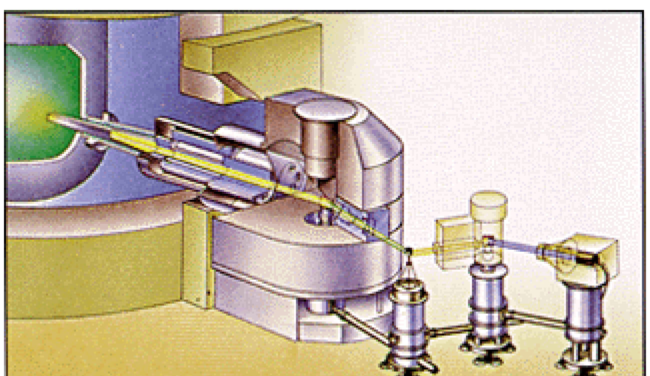
Cliff Shull (1915 – 2001)

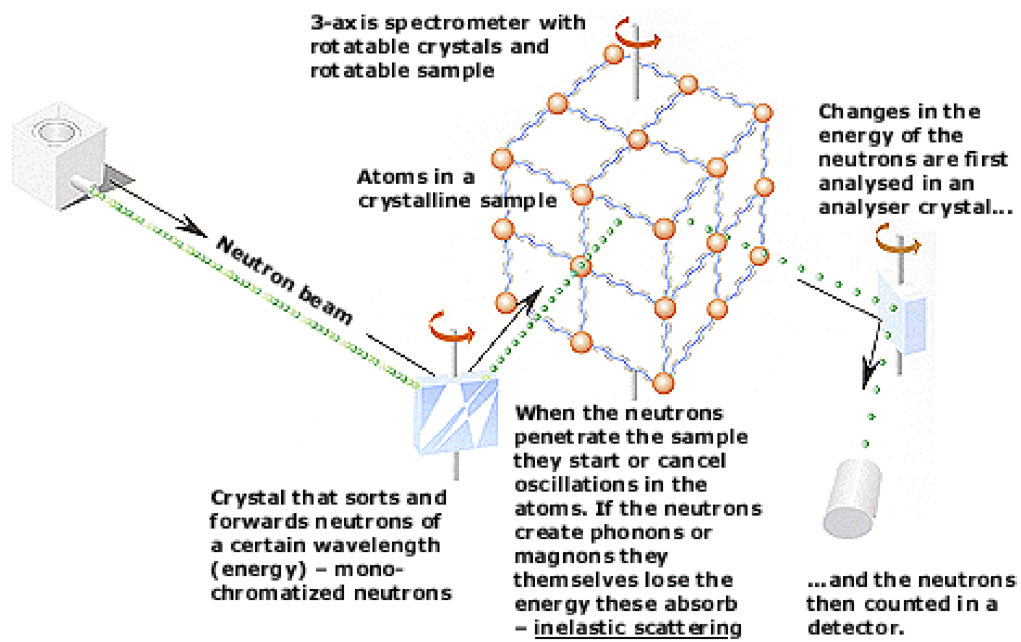
The 1994 Nobel Prize in Physics – Shull & Brockhouse

Neutrons show where the atoms are….

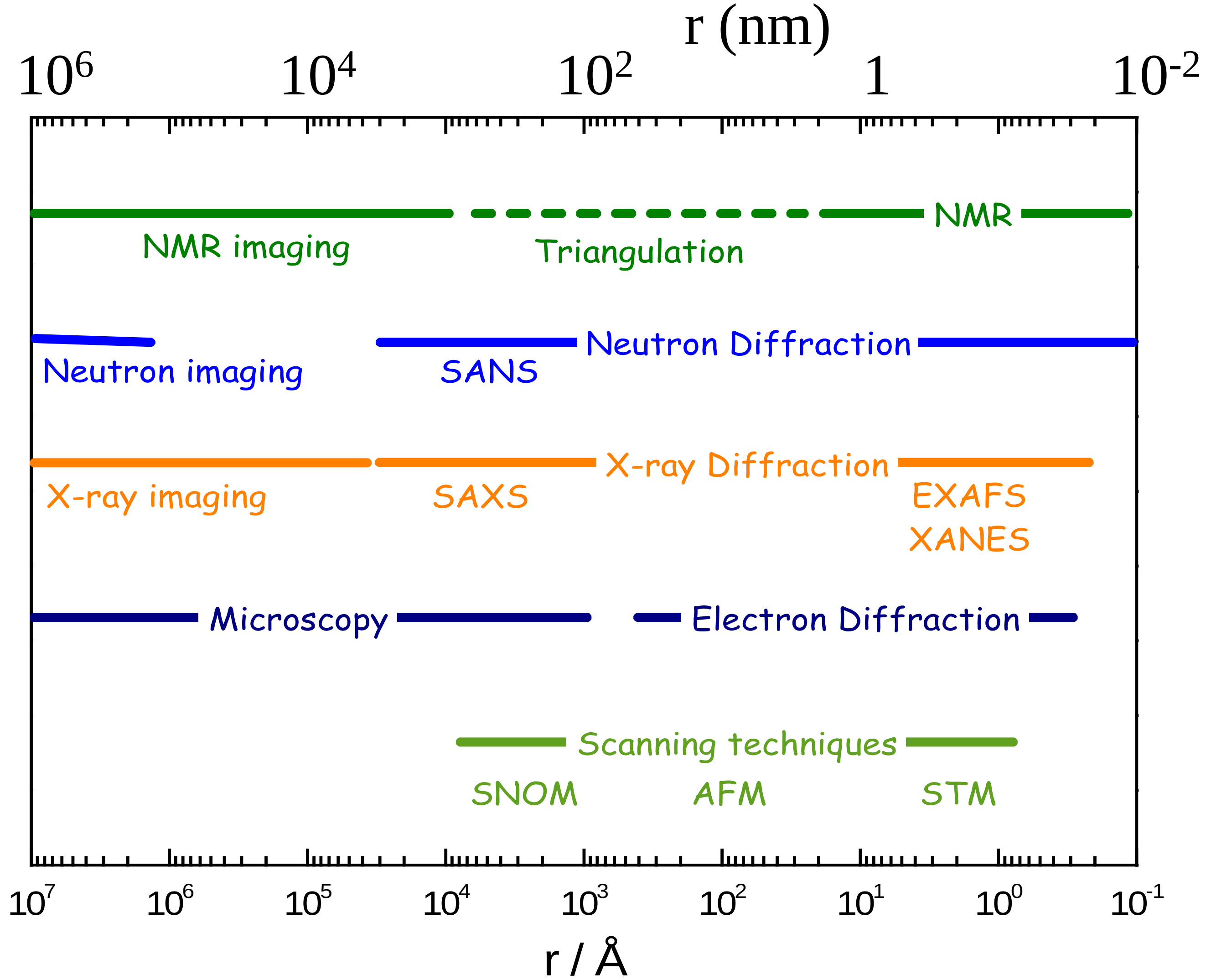


…and what the atoms do.

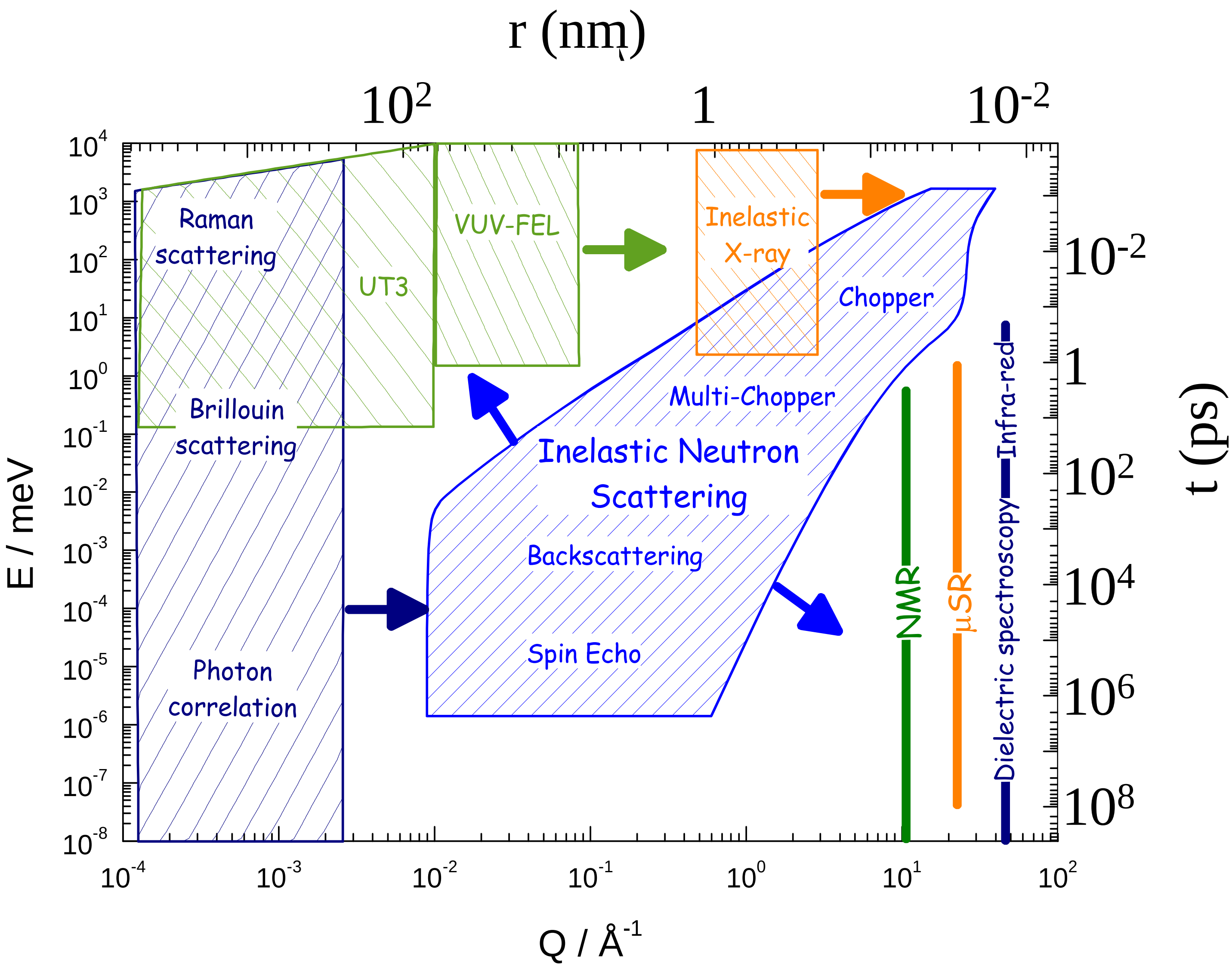

3-axis spectrometer



Neutron & X-ray Scattering Complement Other Techniques in Length Scale….



……and Time Scale



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

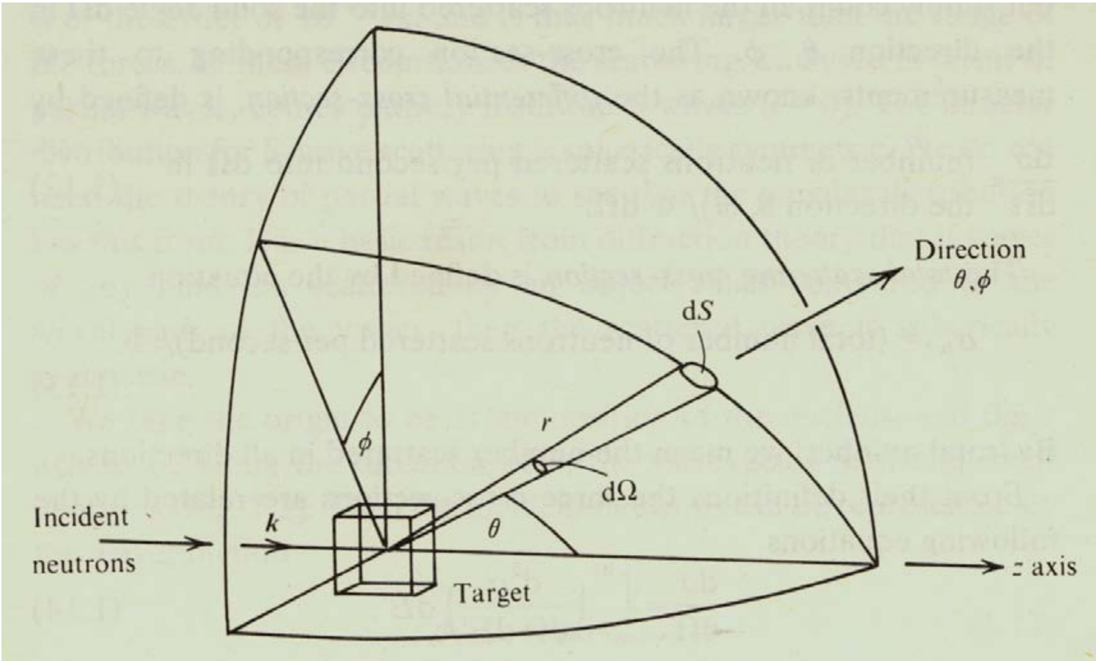
- Mass: $m_n = 1.675 \times 10^{-27}$ kg
- Charge = 0; Spin = ½
- Magnetic dipole moment: $\mu_n = - 1.913\ \mu_N$
- Nuclear magneton: $\mu_N = eh/4\pi m_p = 5.051 \times 10^{-27}$ J T^{-1}
- Velocity (v), kinetic energy (E), wavevector (k), wavelength (λ), temperature (T).
- $E = m_n v^2/2 = k_B T =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

	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})

Cross Sections



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!"

$$\Phi = \text{number of incident neutrons per cm}^2 \text{ per second}$$

$$\sigma = \text{total number of neutrons scattered per second} / \Phi$$

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

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

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

Adding up Neutrons Scattered by Many Nuclei

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

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

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

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

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

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

$$\boxed{\text{For x-rays: } \frac{d\sigma}{d\Omega} = r_0^2 \sum_{i,j} e^{i(\vec{k}_0-\vec{k}').(\vec{R}_i-\vec{R}_j)}\left\{\frac{1-\cos^2 2\theta}{2}\right\}}$$

where $\vec{R}_i$ are electron positions Note: we have assumed the scattering centers don't move

Coherent and Incoherent Scattering of Neutrons

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

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

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

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

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

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

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

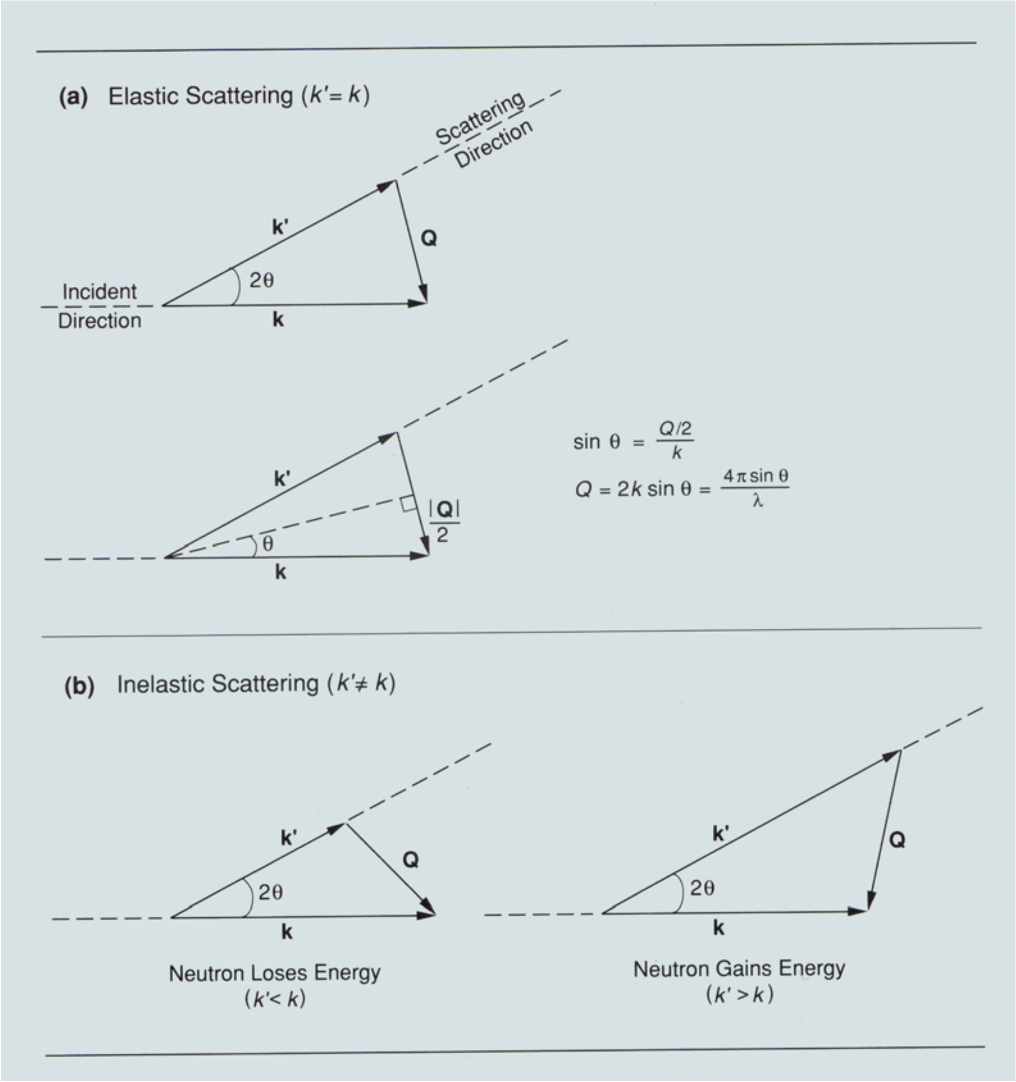
Values of σ_{coh} and σ_{inc}

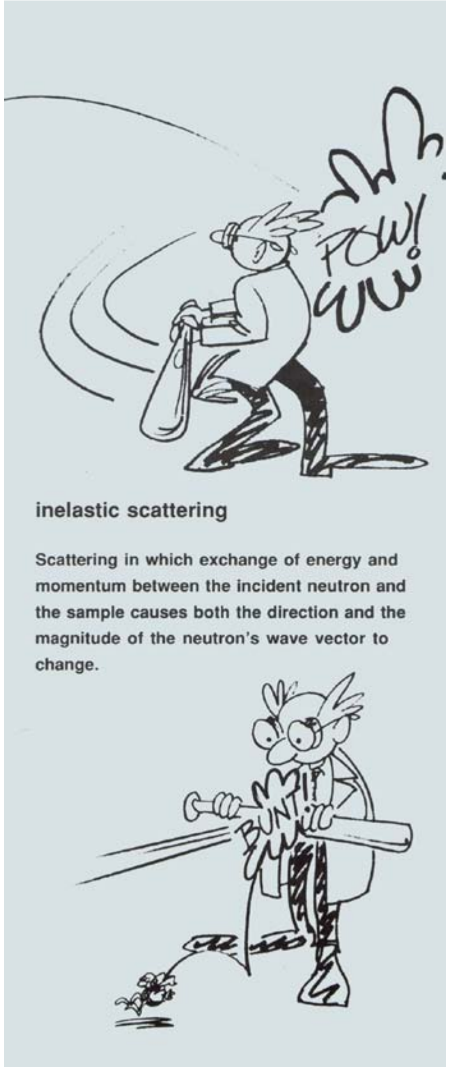
Nuclide	σ_{coh}	σ_{inc}	Nuclide	σ_{coh}	σ_{inc}
^{1}H	1.8	80.2	V	0.02	5.0
^{2}H	5.6	2.0	Fe	11.5	0.4
C	5.6	0.0	Co	1.0	5.2
O	4.2	0.0	Cu	7.5	0.5
Al	1.5	0.0	^{36}Ar	24.9	0.0

- 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/

Neutrons can also gain or lose energy in the scattering process: this is called inelastic scattering. The expressions for the cross sections are similar but more complicated, involving both space and time Fourier transforms



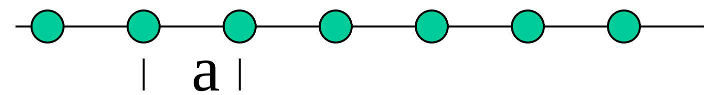


The Elastic & Inelastic Scattering Cross Sections Have an Intuitive Similarity

- The intensity of elastic, coherent neutron scattering is proportional to the spatial Fourier Transform of the Pair Correlation Function, G(r) I.e. the probability of finding a particle at position r if there is simultaneously a particle at r=0
- The intensity of inelastic coherent neutron scattering is proportional to the space ***and time*** Fourier Transforms of the ***time-dependent*** pair correlation function function, G(r,t) = probability of finding a particle at position r ***at time t*** when there is a particle at r=0 and ***t=0***.
- For inelastic ***incoherent*** scattering, the intensity is proportional to the space and time Fourier Transforms of the ***self-correlation*** function, $G_s(r,t)$ I.e. the probability of finding a particle at position r at time t when ***the same*** particle was at r=0 at t=0

Diffraction from a Frozen Wave

- Recall that $S(\vec{Q}) = \frac{1}{N}\left|\sum_k e^{i\vec{Q}.\vec{r}_k}\right|^2$

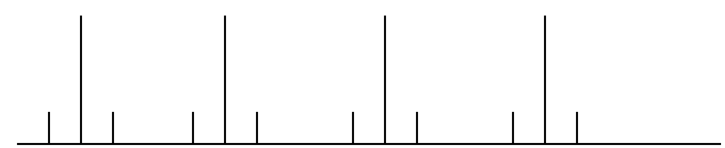


- We know that for a linear chain of “atoms” along the x axis $S(Q_x)$ is just a series of delta function reciprocal lattice planes at $Q_x = n2\pi/a$, where a is the separation of atoms

What happens if we put a "frozen" wave in the chain of atoms so that the atomic positions are $x_p = pa + u\cos kpa$ where p is an integer and u is small?

$$S(Q) = \left|\sum_p e^{iQpa} e^{iQu\cos kpa}\right|^2 \approx \left|\sum_p e^{iQpa}(1 + iQu[e^{ikpa} + e^{-ikpa}])\right|^2$$

$$\approx \left|\sum_p e^{iQpa} + iQu[e^{i(Q+k)pa} + e^{i(Q-k)pa}]\right|^2$$



so that in addition to the Bragg peaks we get weak satellites at $Q = G \pm k$

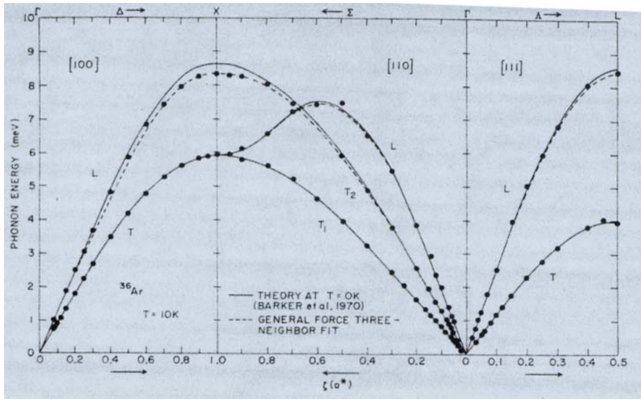
What Happens if the Wave Moves?

- If the wave moves through the chain, the scattering still occurs at wavevectors G + k and G – k but now the scattering is inelastic

- For quantized lattice vibrations, called phonons, the energy change of the neutron is $\hbar\omega$ where ω is the vibration frequency.

- In a crystal, the vibration frequency at a given value of $\vec{k}$ (called the phonon wavevector) is determined by interatomic forces. These frequencies map out the so-called phonon dispersion curves.

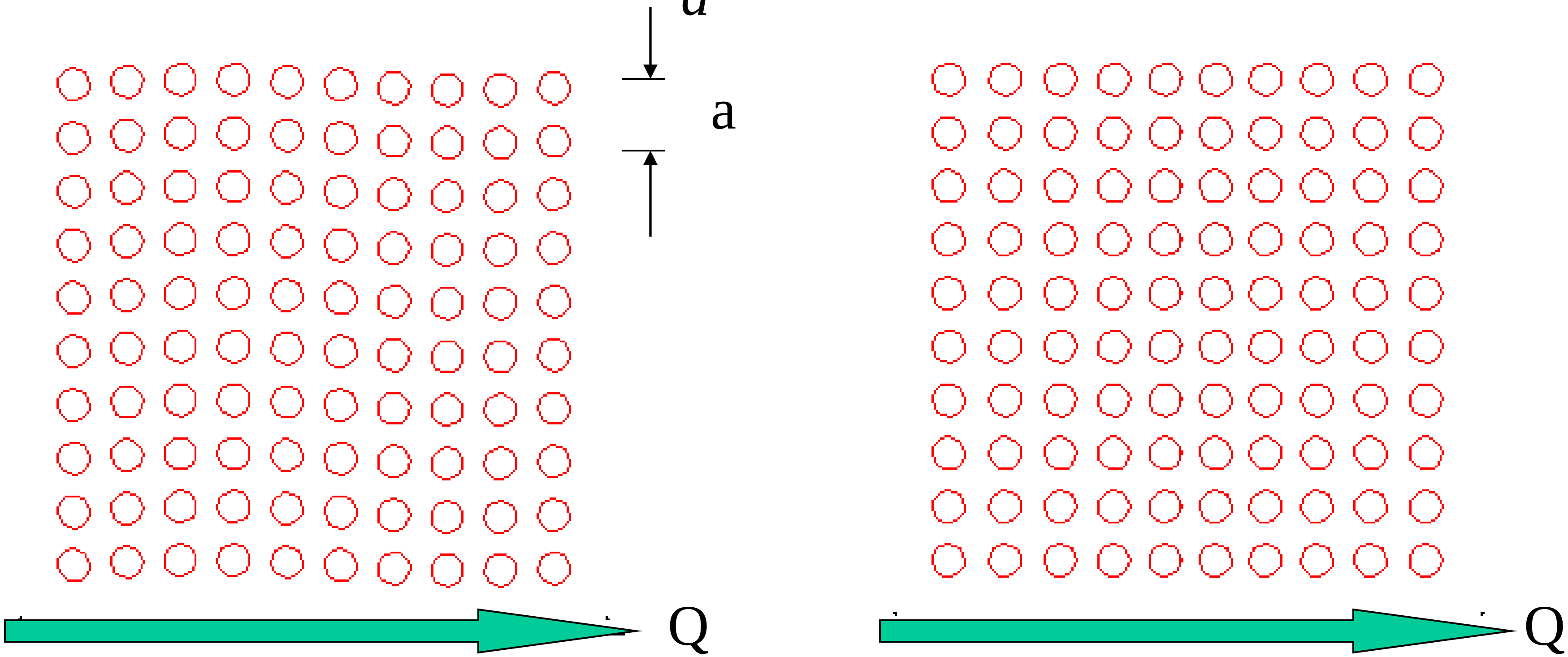
- Different branches of the dispersion curves correspond to different types of motion



phonon dispersion in ^{36}Ar

Atomic Motions for Longitudinal & Transverse Phonons

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



Transverse phonon

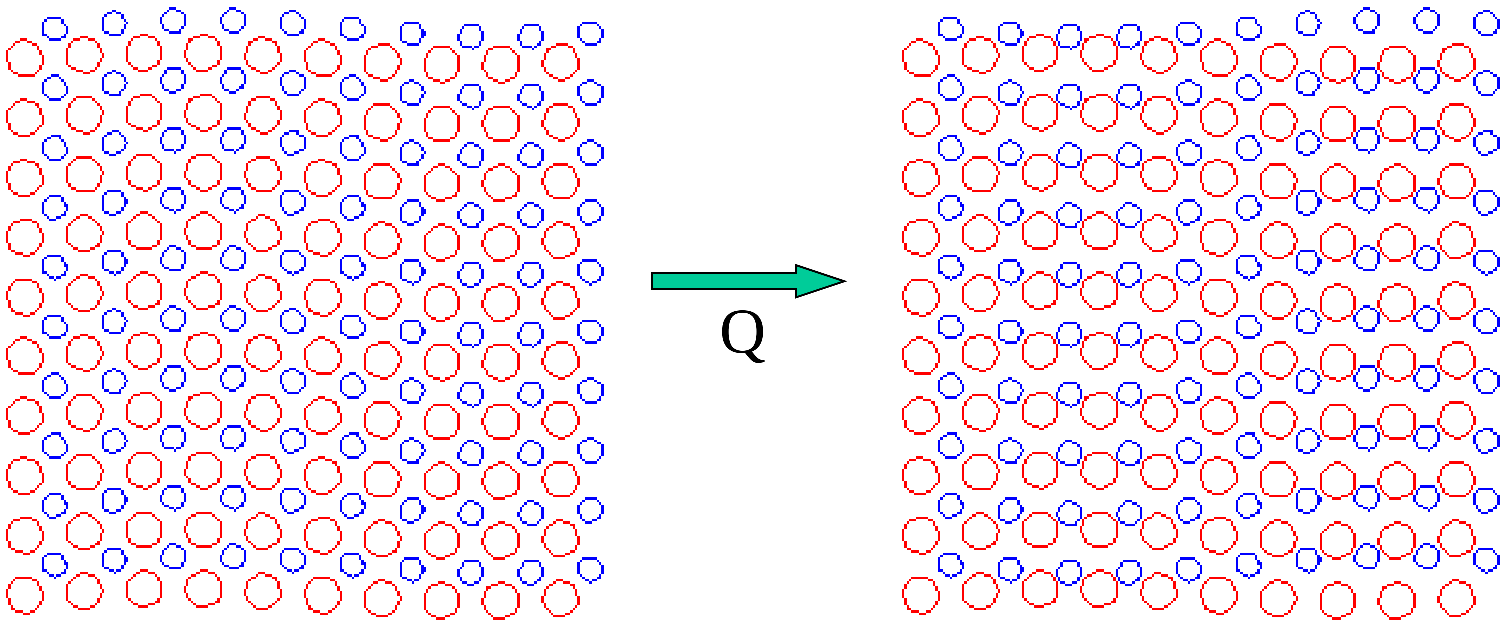
$$\vec{e}_T = (0,0.1,0)a$$

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

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

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

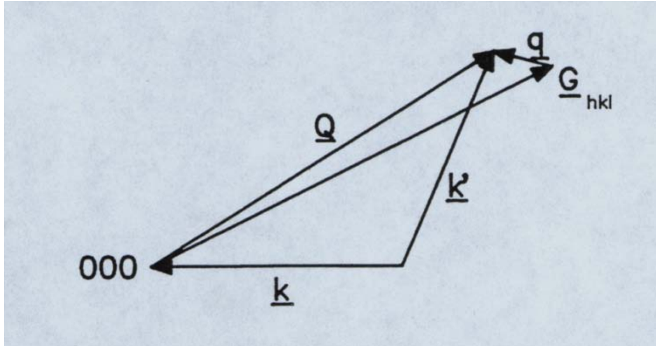
$$\vec{R}_{lk} = \vec{R}^0_{lk} + \vec{e}_s e^{i(\vec{Q}.\vec{R}_l - \omega t)}$$

Phonons – the Classical Use for Inelastic Neutron Scattering

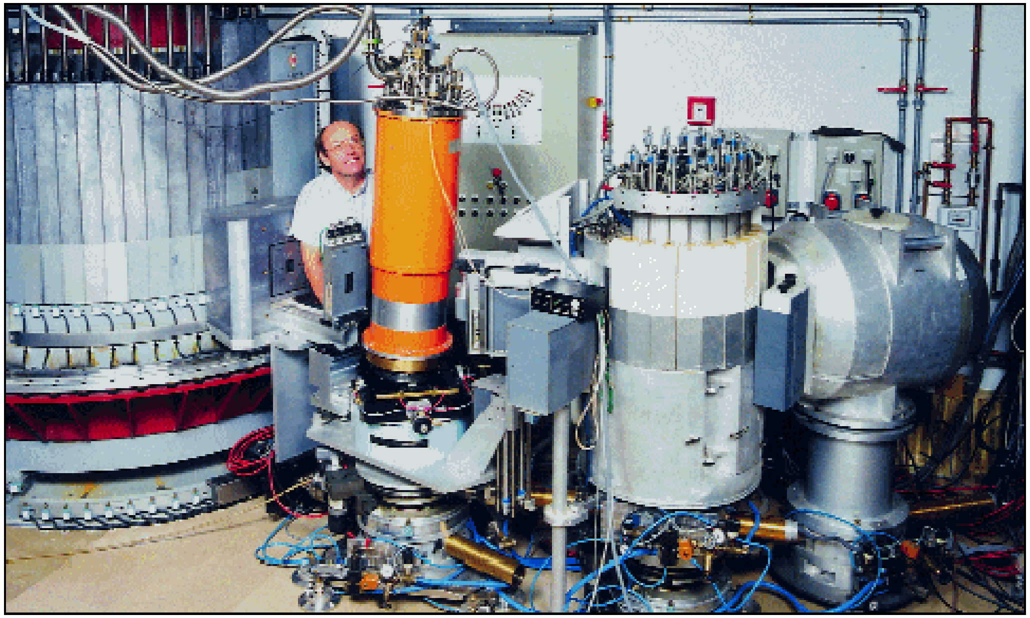
- Coherent scattering measures scattering from single phonons

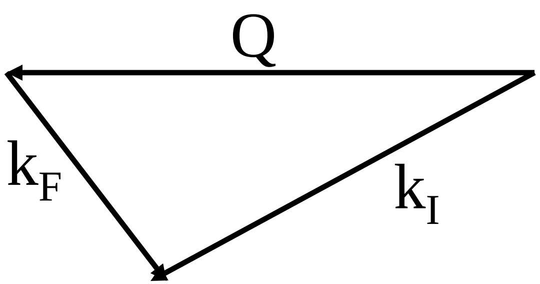
$$\left(\frac{d^2\sigma}{d\Omega dE}\right)_{coh\pm1} = \sigma_{coh}\frac{k'}{k}\frac{\pi^2}{MV_0}e^{-2W}\sum_s\sum_G\frac{(\vec{Q}.\vec{e}_s)^2}{\omega_s}(n_s+\frac{1\pm1}{2})\delta(\omega\mp\omega_s)\delta(\vec{Q}-\vec{q}-\vec{G})$$

- Note the following features:
  - Energy & momentum delta functions => see single phonons (labeled *s*)
  - Different thermal factors for phonon creation (n_s+1) & annihilation (n_s)
  - Can see phonons in different Brillouin zones (different recip. lattice vectors, ***G***)
  - Cross section depends on relative orientation of ***Q*** & atomic motions (***e***$_s$)
  - Cross section depends on phonon frequency (ω_s) and atomic mass (M)
  - In general, scattering by multiple excitations is either insignificant or a small correction (the presence of other phonons appears in the Debye-Waller factor, W)

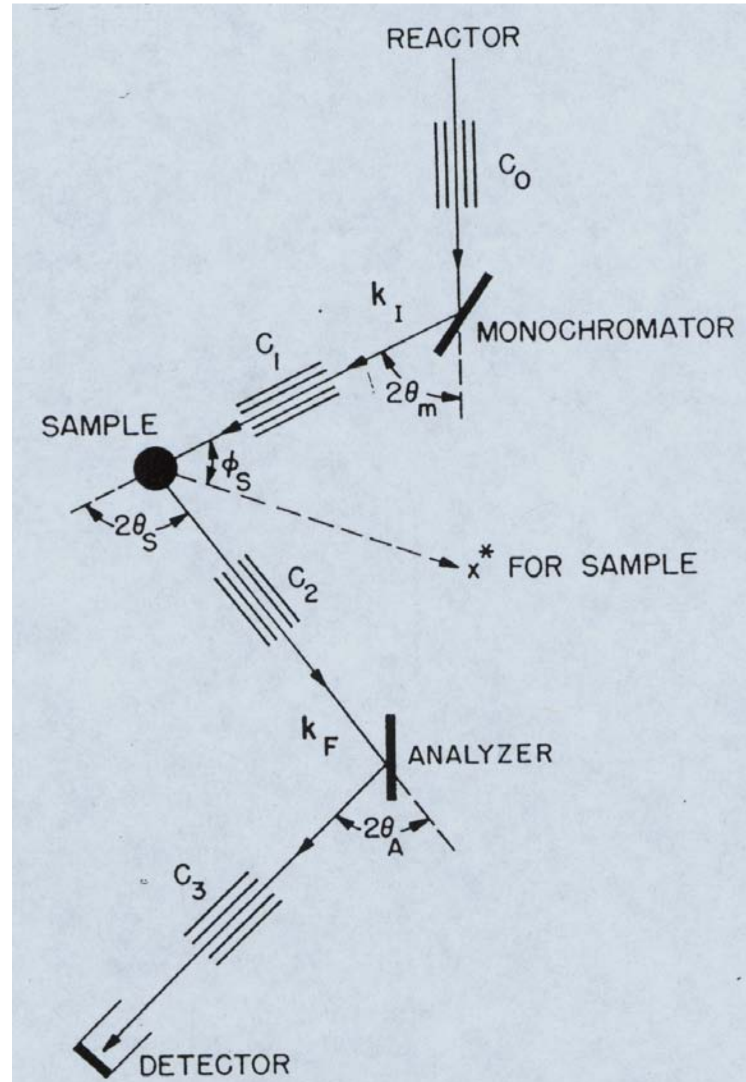


The Workhorse of Inelastic Scattering Instrumentation at Reactors Is the Three-axis Spectrometer





"scattering triangle"



What Use Have Phonon Measurements Been?

- Quantifying interatomic potentials in metals, rare gas solids, ionic crystals, covalently bonded materials etc

- Quantifying anharmonicity (I.e. phonon-phonon interactions)

- Measuring soft modes at 2nd order structural phase transitions

- Electron-phonon interactions including Kohn anomalies

- Roton dispersion in liquid He

- Relating phonons to other properties such as superconductivity, anomalous lattice expansion etc

Examples of $S(Q,\omega)$ and $S_s(Q,\omega)$

- Expressions for $S(Q,\omega)$ and $S_s(Q,\omega)$ can be worked out for a number of cases e.g:
  - Single phonons
  - Phonon density of states
  - Various models for atomic motions in liquids and glasses
  - Various models of atomic & molecular translational & rotational diffusion
  - Rotational tunneling of molecules
  - Transitions between crystal field levels
  - Spin waves and other magnetic excitations such as spinons
- Inelastic neutron scattering reveals details of the shapes of interaction potentials in materials

Magnetic Properties of the Neutron

- The neutron has a magnetic moment of -9.649 x 10^{-27} JT^{-1}

$$\vec{\mu}_n = -\gamma \mu_N \vec{\sigma}$$

where $\mu_N = e\hbar / 2m_p$ is the nuclear magneton,

$m_p =$ proton mass, $e =$ proton charge and $\gamma = 1.913$

$\vec{\sigma}$ is the Pauli spin operator for the neutron. Its eignevalues are ± 1

- Note that the neutron's spin and magnetic moment are antiparallel
- Because of its magnetic moment, the neutron feels a potential given by:

$$V_m(\vec{r}) = -\vec{\mu}_n.\vec{B}(\vec{r}) \quad \text{where} \quad \vec{B}(\vec{r}) = \mu_0 \mu \vec{H}(\vec{r}) = \mu_0 [\vec{H}(\vec{r}) + \vec{M}(\vec{r})]$$

- Thus the neutron senses the distribution of magnetization in a material
- Homework problems: What is the Zeeman energy in meV of a neutron in a 1 Tesla field? At what temperature is the Boltzmann energy equal to this Zeeman energy? What is the effective scattering length of a "point" magnetic moment of one Bohr magneton?

Magnetic Scattering of the Neutron

- For nuclear scattering, the matrix element that appears in the expression for the scattering cross section is: $\sum_j b_j e^{i\vec{Q}.\vec{R}_j}$

- The equivalent matrix element for magnetic scattering is:

$$\gamma r_0 \frac{1}{2\mu_B} \vec{\sigma}.\vec{M}_\perp(\vec{Q}) \quad \text{where } \mu_B = \frac{e\hbar}{2m_e} \text{ is the Bohr magneton } (9.27 \text{ x } 10^{-24} \text{ JT}^{-1})$$

$$\text{and } r_0 = \frac{\mu_0}{4\pi} \frac{e^2}{m_e} \text{ is classical radius of the electron } (2.818 \text{ x } 10^{-6} \text{ nm})$$

- Here $\vec{M}_\perp(\vec{Q})$ is the component of the Fourier transform of the magnetization that is perpendicular to the scattering vector $\vec{Q}$. This form arises directly from the dipolar nature of the magnetic interaction.

- Unlike the neutron-nucleus interaction, the magnetic interaction of the neutron with a scattering system specifically depends on neutron spin

What Happens to a Neutron's Spin When the Neutron is Scattered?

- The cross section for magnetic scattering that takes the neutron spin state from σ->σ' and the scattering system from λ->λ' is:

$$\left(\frac{d^2\sigma}{d\Omega.dE}\right)_{\sigma\lambda\to\sigma'\lambda'} = \left(\frac{\gamma r_0}{2\mu_B}\right)^2 \frac{k'}{k} \left|\left\langle \sigma'\lambda' \middle| \vec{\sigma}.\vec{M}_\perp(\vec{Q}) \middle| \sigma\lambda \right\rangle\right|^2 \delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

- One can show (see Squires) that if $|u\rangle, |v\rangle$ are the neutron spin eigenstates:

$$\langle u|\vec{\sigma}.\vec{M}_\perp|u\rangle = M_{\perp z};\ \ \langle v|\vec{\sigma}.\vec{M}_\perp|v\rangle = -M_{\perp z};\ \ \langle v|\vec{\sigma}.\vec{M}_\perp|u\rangle = M_{\perp x} + iM_{\perp y};\ \ \langle u|\vec{\sigma}.\vec{M}_\perp|v\rangle = M_{\perp x} - iM_{\perp y}$$

so, components of M_{perp} parallel to the neutron's magnetic moment (z) does not change the neutron spin, whereas perpendicular components of M_{perp} 'flip' the neutron's spin

- Homework: show that for a paramagnet (where $\left\langle S_i^\alpha S_j^\beta \right\rangle = \frac{1}{3}\delta_{ij}\delta_{\alpha\beta}S(S+1)$ for spins i and j)
  - If z is parallel to **Q**, the scattering is entirely spin flip
  - If z is perpendicular to **Q**, half the scattering is spin flip

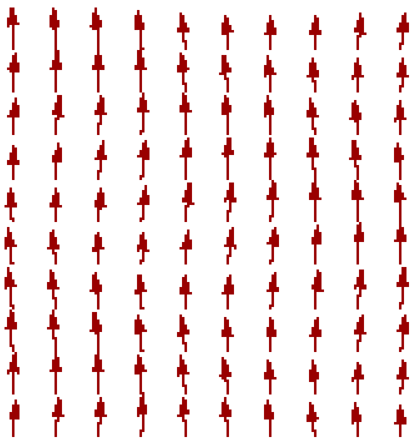
Inelastic Magnetic Scattering of Neutrons

- In the simplest case, atomic spins in a ferromagnet precess about the direction of mean magnetization

$$H = \sum_{l,l'} J(\vec{l} - \vec{l}')\vec{S}_l.\vec{S}_{l'} = H_0 + \sum_q \hbar\omega_q b_q^+ b_q$$

Heisenberg interaction

spin waves (magnons)

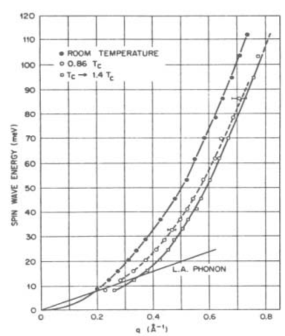


with

$$\hbar\omega_q = 2S(J_0 - J_q) \qquad \text{where} \quad J_q = \sum_l J(\vec{l})e^{i\vec{q}.\vec{l}}$$

$\hbar\omega_q = Dq^2$ is the dispersion relation for a ferromagnet

Fluctuating spin is perpendicular to mean spin direction



Inelastic Magnetic Neutron Scattering

- Conventional spin waves (FM and AFM) in systems with itinerant & localized electrons
- Stoner modes
- Crystal field excitations
- Excitations in low dimensional magnets
  - Effects of Hamiltonian symmetry (Ising, XY, Heisenberg)
  - Effects of spin value (e.g. Haldane gap for S=1 and not S=1/2 in 1d AFM)
- Non-linear magnetic excitations (solitons, spinons, beathers etc)
- Diffusive versus propagating excitations
- Magnetic critical scattering
- Spin glasses
- Frustrated magnets
- Singlet ground state systems
- High temperature superconductors
- Correlated electron systems
- Quantum fluctuations and QCPs

Polarized Neutrons: What you Need to Know

- When a polarized neutron beam is scattered by a sample, we can measure the polarization of the scattered neutrons

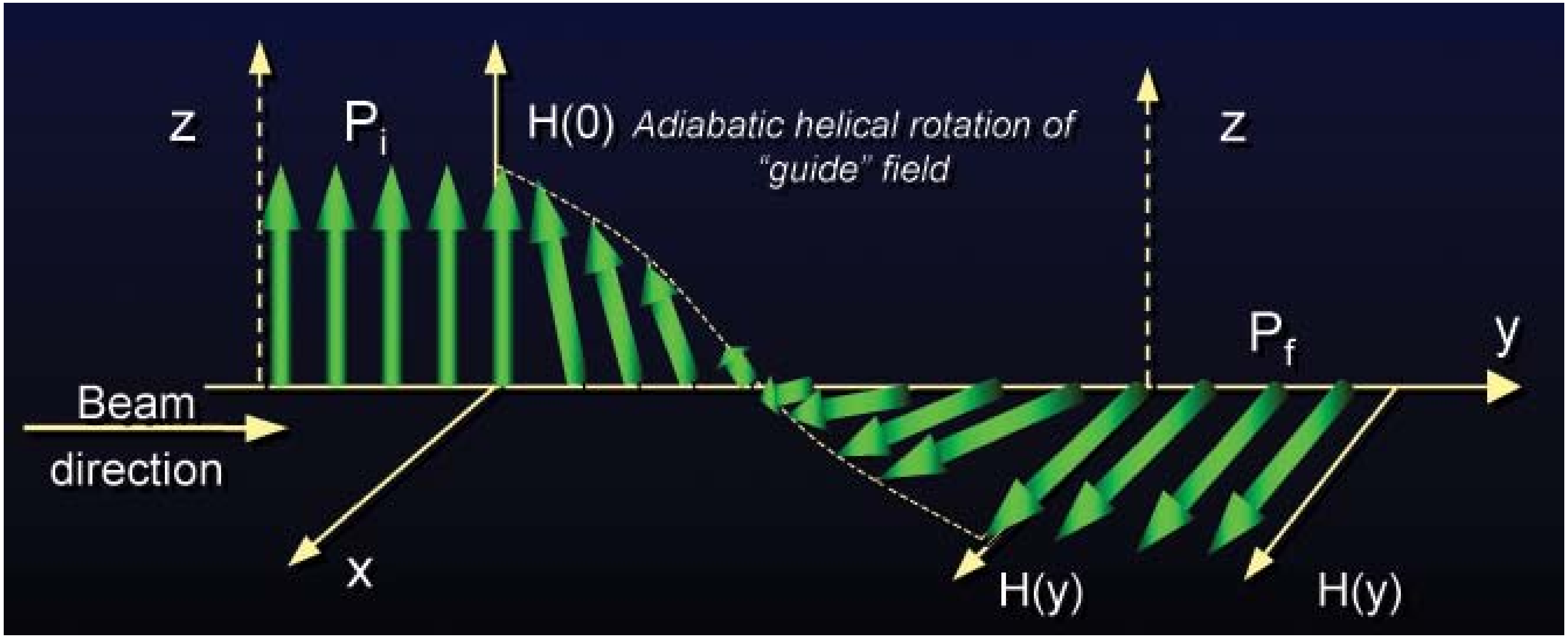
Type of scattering	Non-spin-flip	Spin-flip
Coherent	1	0
Isotopic incoherent	1	0
Nuclear spin incoherent	1/3	2/3
M fluctuation parallel to H	1	0
M fluctuation perpendicular to H	0	1

Production of Polarized Neutron Beams

- Polarizing filters
  - Usually ^{3}He these days, although pumped protons and rare earths have been tried
  - Good for polarizing large, divergent neutron beams
  - Depend on good polarization of filter material
- Polarizing monochromators
  - Mainly Heusler alloy (Cu_2MnAl) these days
  - Good when beam monochromatization is also required (e.g. TAS)
- Supermirrors
  - Very efficient, broad-band polarizers
  - "cavities" or "benders" are excellent for preparing polarized beams
  - Disadvantage is that wide angular beam divergence requires devices with non-uniform transmission

Guiding the Neutron Polarization

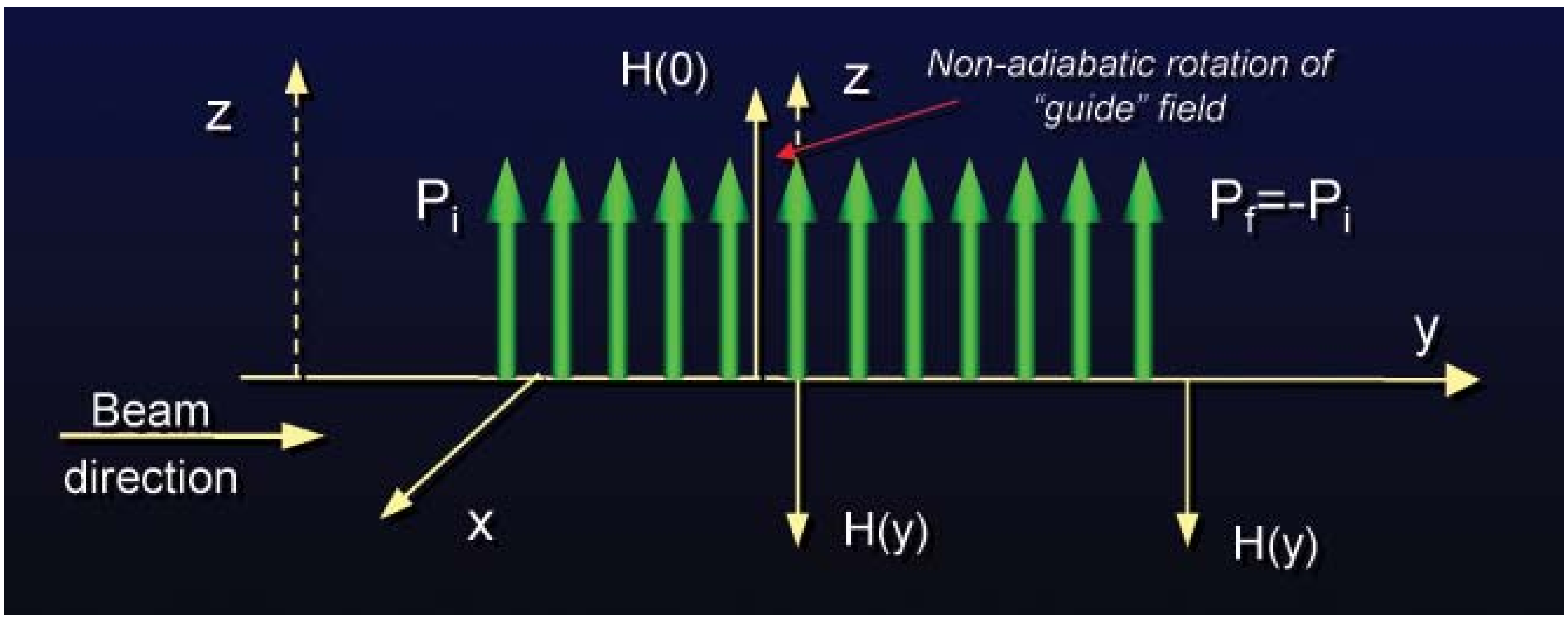
- If the direction of a magnetic field varies sufficiently slowly in space, the component of neutron polarization parallel to the applied field is preserved. This is adiabatic polarization rotation.



Drawing from Bob Cywinski

Non-Adiabatic Transitions

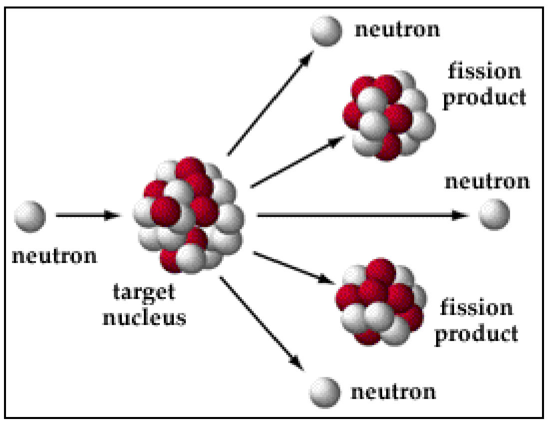
- If the guide field direction is suddenly changed (i.e. the adiabaticity parameter tan δ □ □ , the neutron polarization vector will precess about the new field direction.
- If the field is reversed, the neutron polarization is flipped with respect to the field

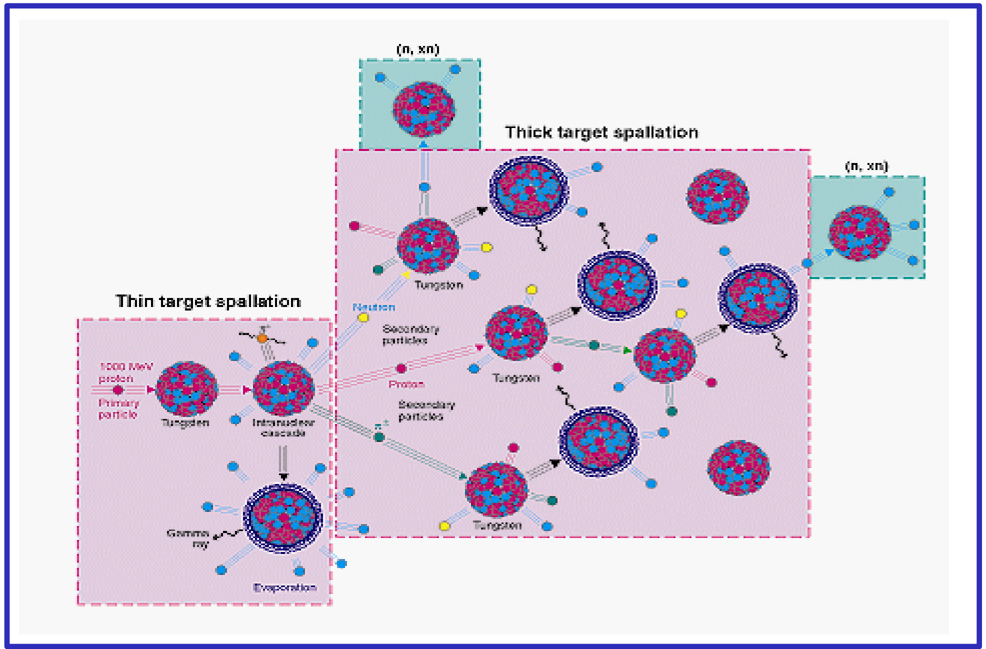


Graphic courtesy of Bob Cywinski

Neutron Sources

- Neutrons are produced by fission in nuclear reactors or by nuclear reactions when accelerated protons hit heavy nuclei.
  - Each neutron “costs” energy to produce – ~ 190 MeV/neutron for fission and ~25 MeV/neutron for spallation.
  - The fundamental limit to producing intense neutron sources is cooling
- Usually, accelerator-based sources produce pulsed beams & reactors produce CW beams





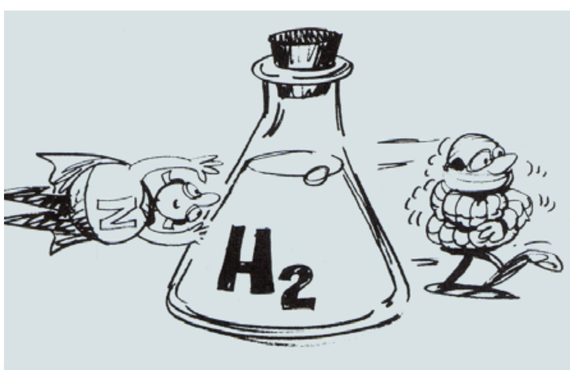
The Energy Cost of Various Neutron Sources

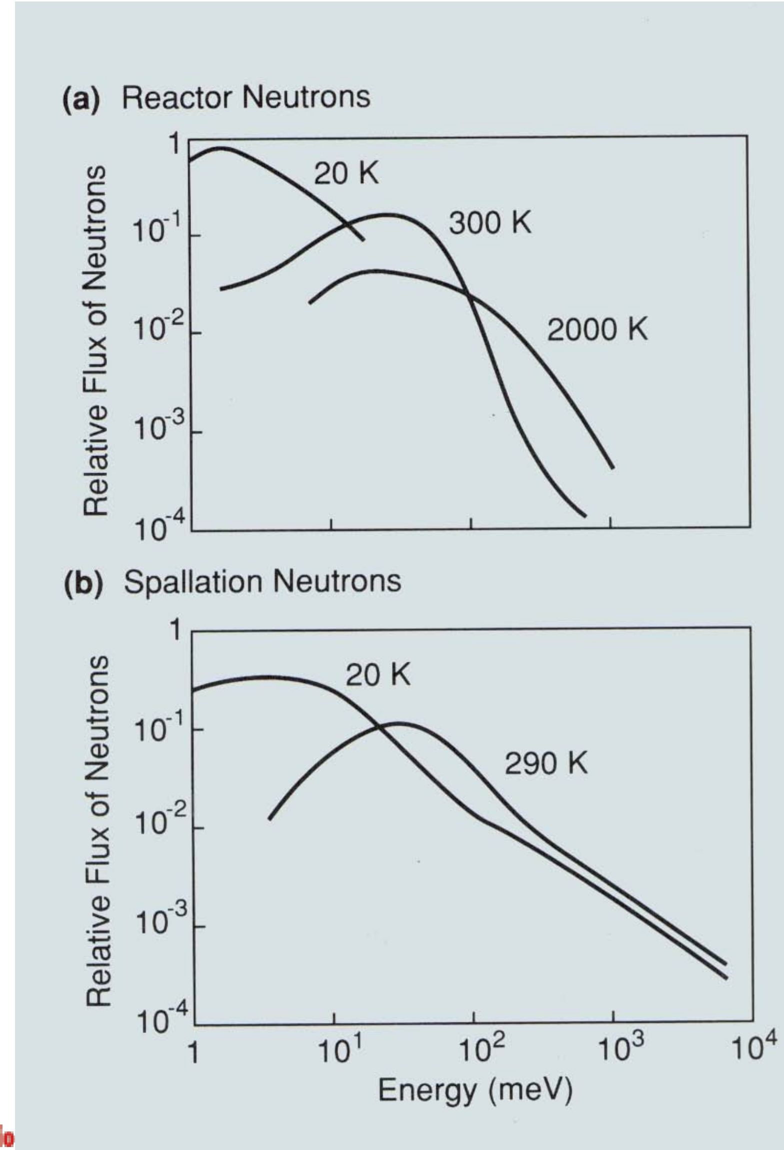
- For high-power sources the driving issue is heat removal => use spallation for high power sources
  - ~ 190 MeV per neutron for fission
  - ~ 25 MeV per neutron for spallation with protons (threshold at E_p~ 120 MeV)
  - ~ 1500 MeV per neutron for (n,p) on Be using 13 MeV protons
  - ~ 3000 MeV per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



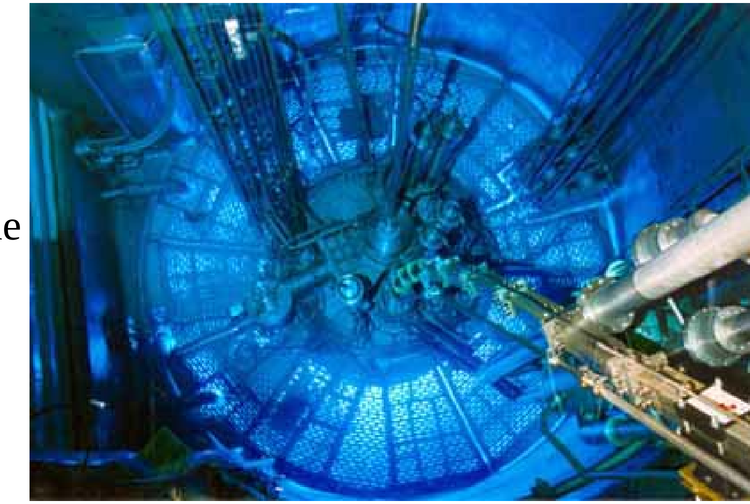


The Institut Laue Langevin Reactor

1. Safety rod
2. Neutron guide pool
3. Reflector tank
4. Double neutron guide
5. Vertcial cold source
6. Reactor core
7. Horiz cold source
8. Control rod

Cross section of reactor

Fuel element: top view



Reactor pool; light water

Spent fuel pool →

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

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

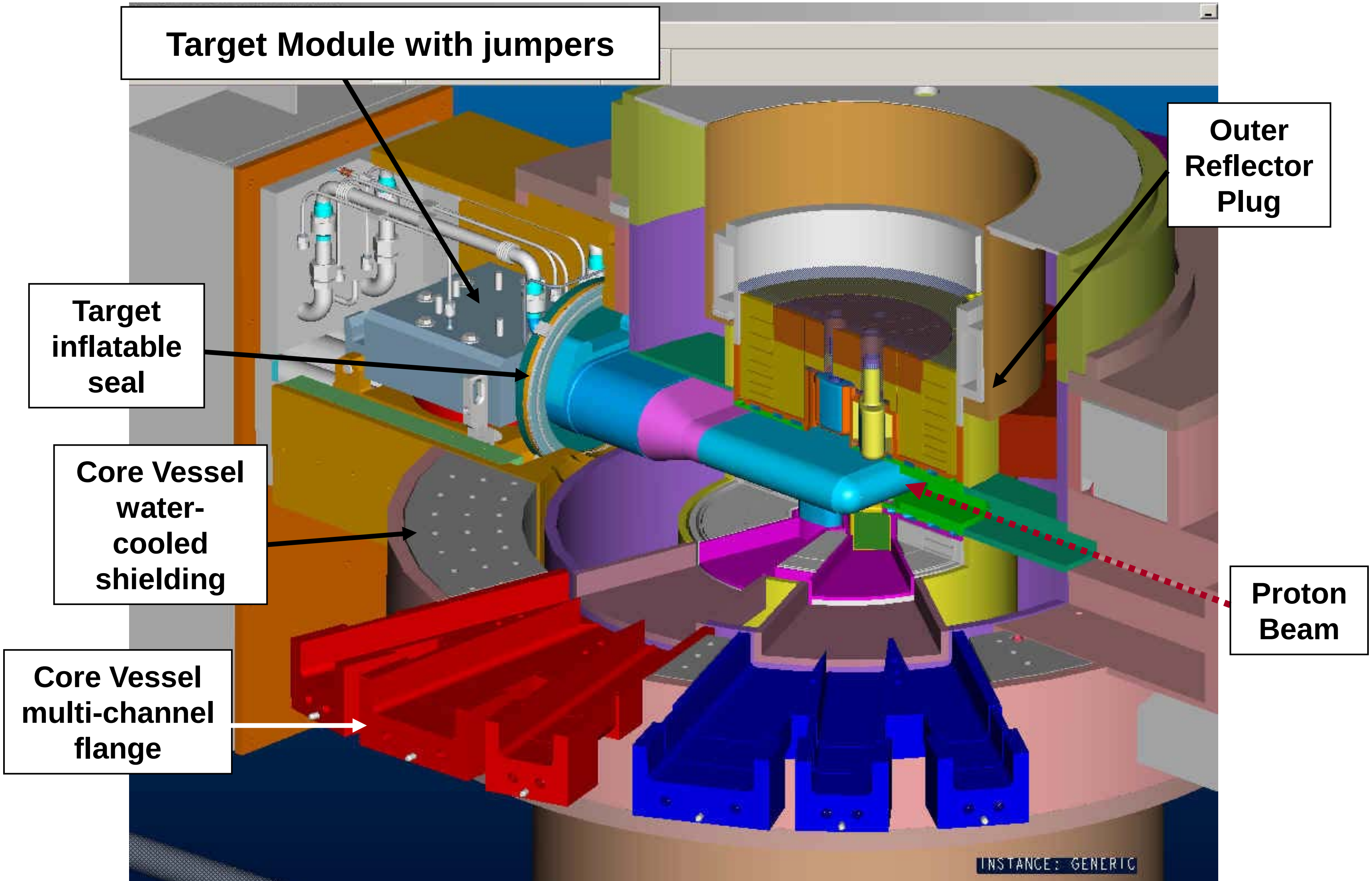
Neutron Scattering Facilities

- Most are large, expensive, centralized facilities because of the difficulty of producing intense neutron beams
  - Some exceptions (e.g. JEEP II at Kjeller is 2 MW reactor – ILL is 57 MW; LENS at Indiana Univ is a weak pulsed neutron source – 20 kW compared to 1 MW at SNS)
- Many operate either as national or multi-national user facilities open for peer-reviewed access by any qualified researcher

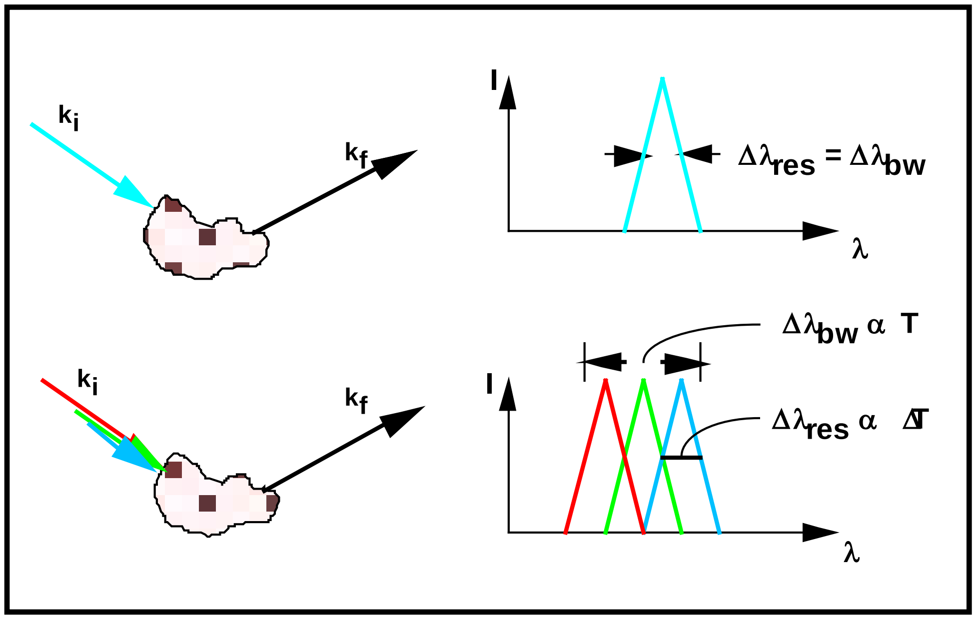

The neutron guide hall at the Institut Laue-Langevin in Grenoble, France

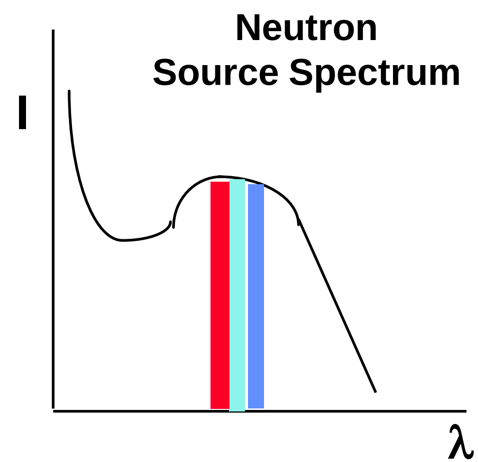

The Spallation Neutron Source in Oak Ridge Tennessee.

Producing Neutrons at SNS --Target/Moderators



Simultaneously Using Neutrons With Many Different Wavelengths Enhances the Efficiency of Neutron Scattering Experiments





Potential Performance Gain relative to use of a Single Wavelength is the Number of Different Wavelength Slices used

A Comparison of Reactors & Short Pulse Spallation Sources

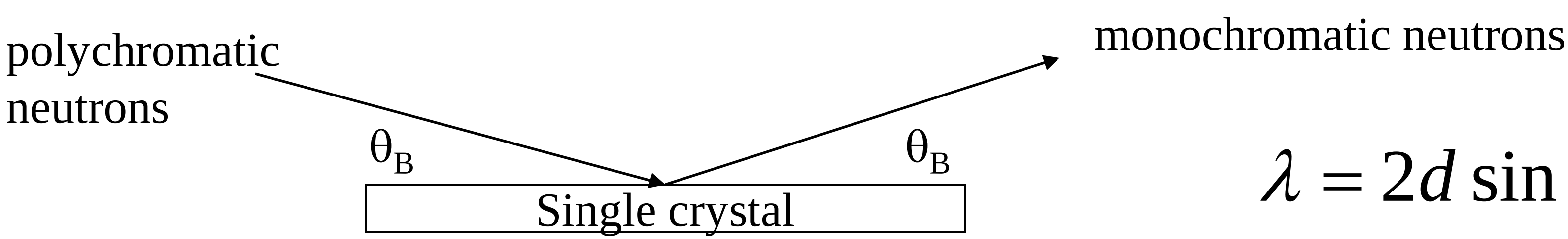
Short Pulse Spallation Source	Reactor
Energy deposited per useful neutron is ~20 MeV	Energy deposited per useful neutron is ~ 180 MeV
Neutron spectrum is “slowing down” spectrum – preserves short pulses	Neutron spectrum is Maxwellian
Constant, small $\delta\lambda/\lambda$ at large neutron energy => excellent resolution especially at large Q and E	Resolution can be more easily tailored to experimental requirements
Copious “hot” neutrons=> very good for measurements at large Q and E	Large flux of cold neutrons => very good for measuring large objects and slow dynamics
Low background between pulses => good signal to noise	Pulse rate for TOF can be optimized independently for different spectrometers
Single pulse experiments possible	Neutron polarization has been easier

Components of Neutron Scattering Instruments

- Monochromators (crystals, velocity selectors, choppers)
  - Select the energy of a neutron beam by various means
- Choppers
  - Define a short pulse or pick out a small band of neutron energies
- Collimators
  - Define the direction of travel of the neutron
- Guides
  - Allow neutrons to travel large distances without suffering intensity loss
- Filters (Be, benders, S-bend guides)
  - Reject unwanted neutron wavelengths
- Polarizers
  - Create or analyze a polarized neutron beam
- Detectors
  - Most commonly, neutrons are absorbed by ^{3}He and the gas ionization caused by recoiling particles is detected
- Shielding
  - Minimize background and radiation exposure to users

Wavelength Selection

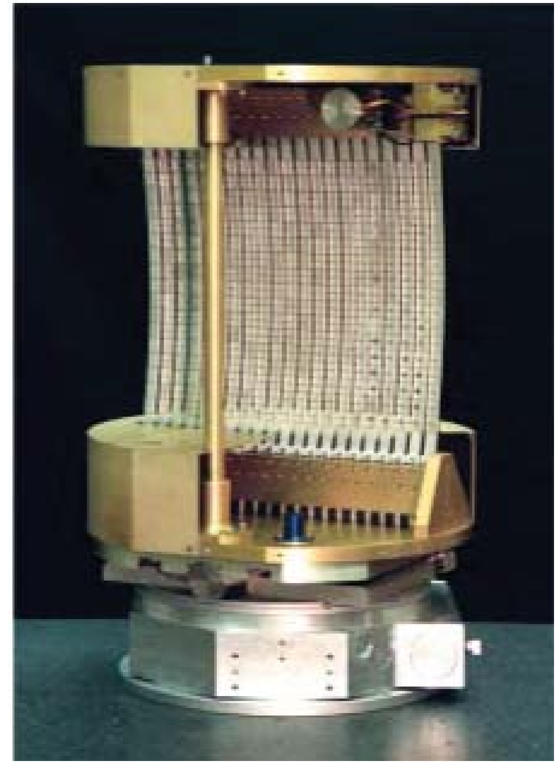
- Crystal monochromators are often used at CW neutron sources for diffractometers & three-axis machines
  - Bragg scattering from mosaic crystals defines the neutron wavelength
  - Crystal mosaic in the horizontal scattering plane controls intensity and $\delta\lambda/\lambda$
  - Crystal mosaic in vertical plane determines image beam size at the sample when focusing is used
  - Neutrons are often focused vertically – does not affect $\delta\lambda/\lambda$ but does worsen vertical Q resolution (often unimportant)
  - Neutrons are sometimes focused horizontally – degrades $\delta\lambda/\lambda$ and Q resolution in the scattering plane, but gains intensity



$$\lambda = 2d\sin\theta_B$$

Neutron Monochromators

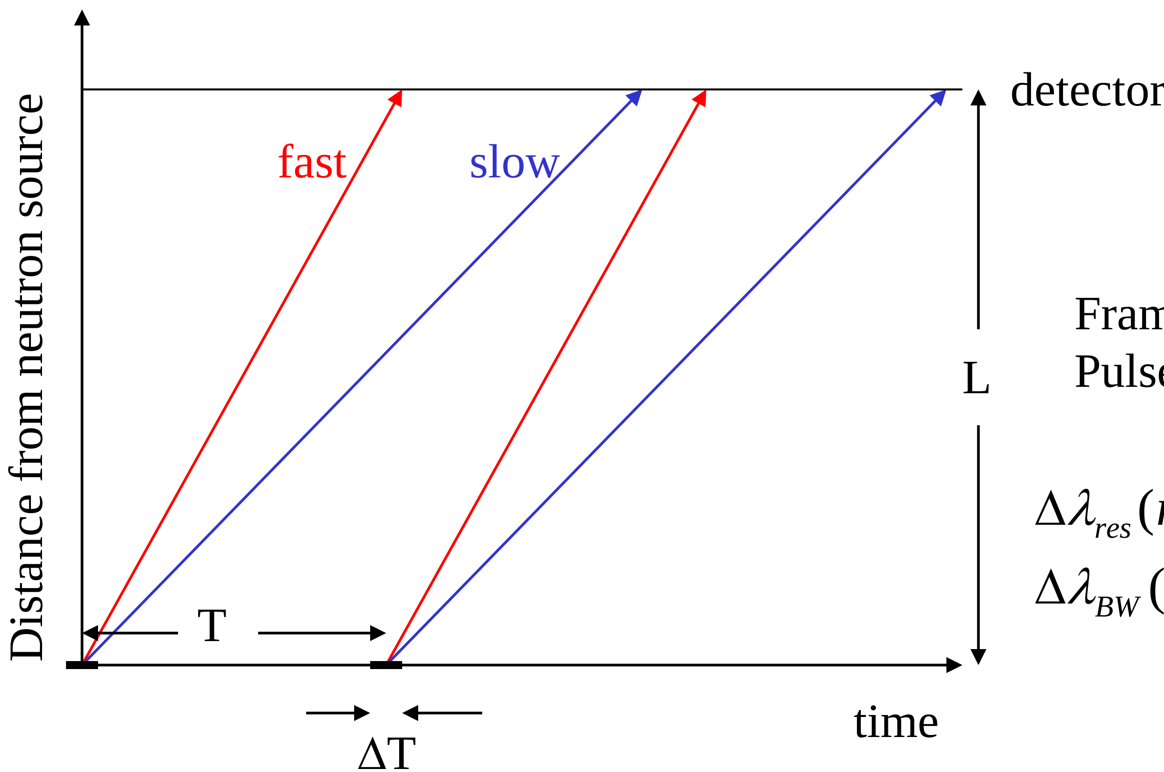
A simple, vertically focusing monochromator produced by Riso National Lab in Denmark comprised of 15 single crystals



A vertical and horizontally focusing monochromator fabricated by a Johns Hopkins team for the NCNR. See: www.pha.jhu.edu/~broholm/homepage/talks/MACSmonochromatoracns.pdf

Time of Flight

- At pulsed neutron sources (or with a chopped beam at a reactor), the neutron's TOF is used to determine it's speed (and, hence, wavelength)
- For elastic scattering (diffraction, SANS, reflectometry) no neutron monochromatization is needed



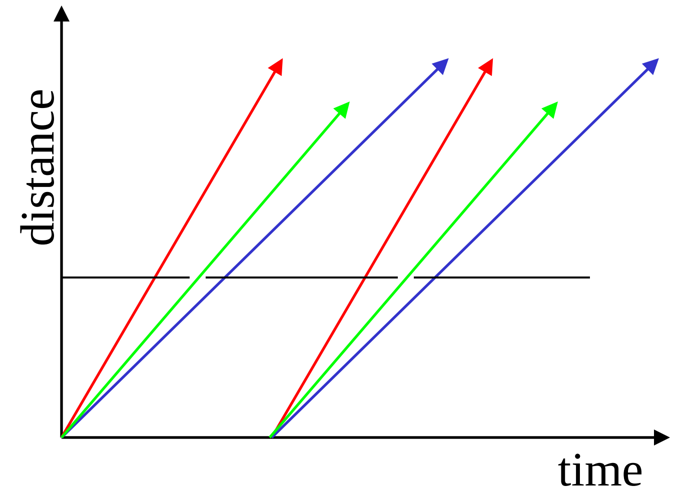
Frame length, T
Pulse length, ΔT

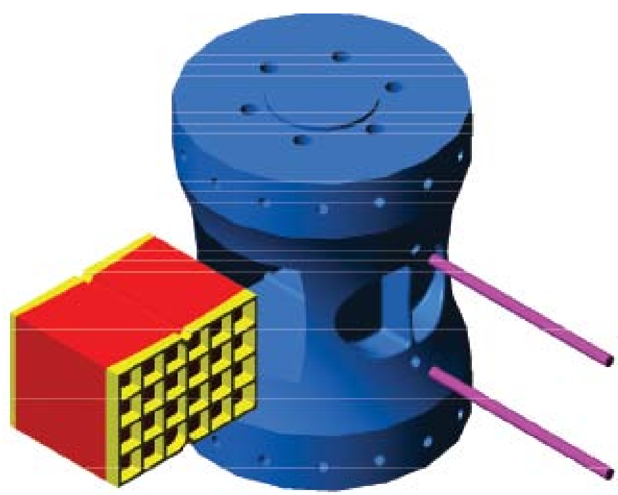
$$\Delta\lambda_{res}(nm) \approx 0.4\Delta T(ms)/L(m)$$

$$\Delta\lambda_{BW}(nm) \approx 0.4T(ms)/L(m)$$

For Inelastic Scattering at a Pulsed Source, Choppers are used as Monochromators

- Fermi chopper – rapidly rotating collimator



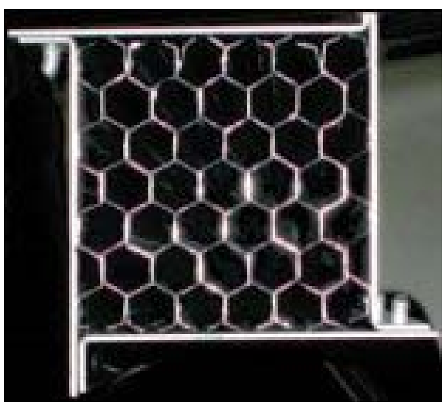


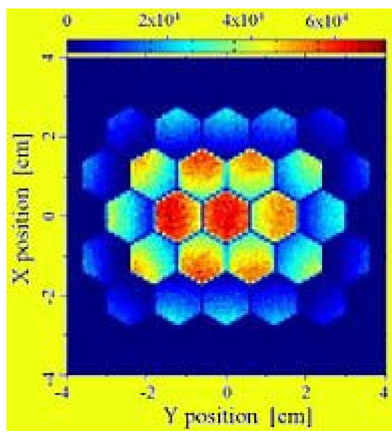
ARCS Fermi chopper housing and slit package (courtesy B. Fultz)

Collimators Define the Direction of Travel of the Neutrons

- A Soller collimator is a set of parallel neutron-absorbing plates that define the direction of the neutron beam
- More sophisticated arrangements are possible

Mylar collimator by Jens Linderholm, Denmark



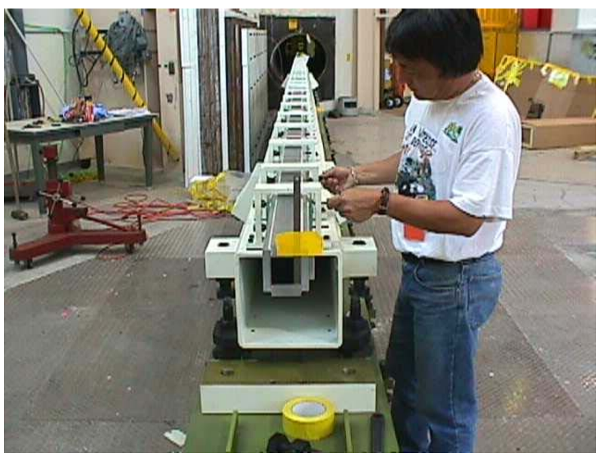


Honeycomb collimator for the Brillouin scattering instrument BRISP at ILL

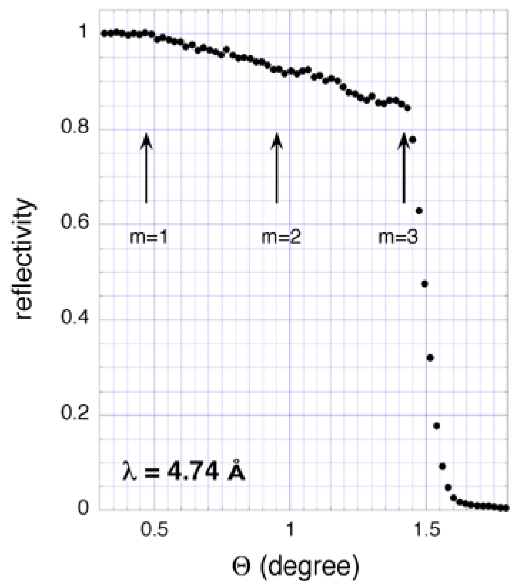
Radial collimator for the ARCS spectrometer to be installed at SNS

Neutron Guides Transport Neutrons over Long Distances with Little Loss

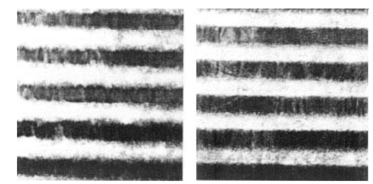
- Critical external reflection of neutrons for $\theta_c = \lambda\sqrt{\rho/\pi}$
  - Critical angle for Ni in degrees ~ 0.1 x neutron wavelength in Å
  - Guides can be coated with supermirror (e.g. NiTi) to extend the critical angle by a factor of up to ~7 (8000 layers) but reflectivity drops



Neutron guide being installed on SMARTS at the Lujan Center

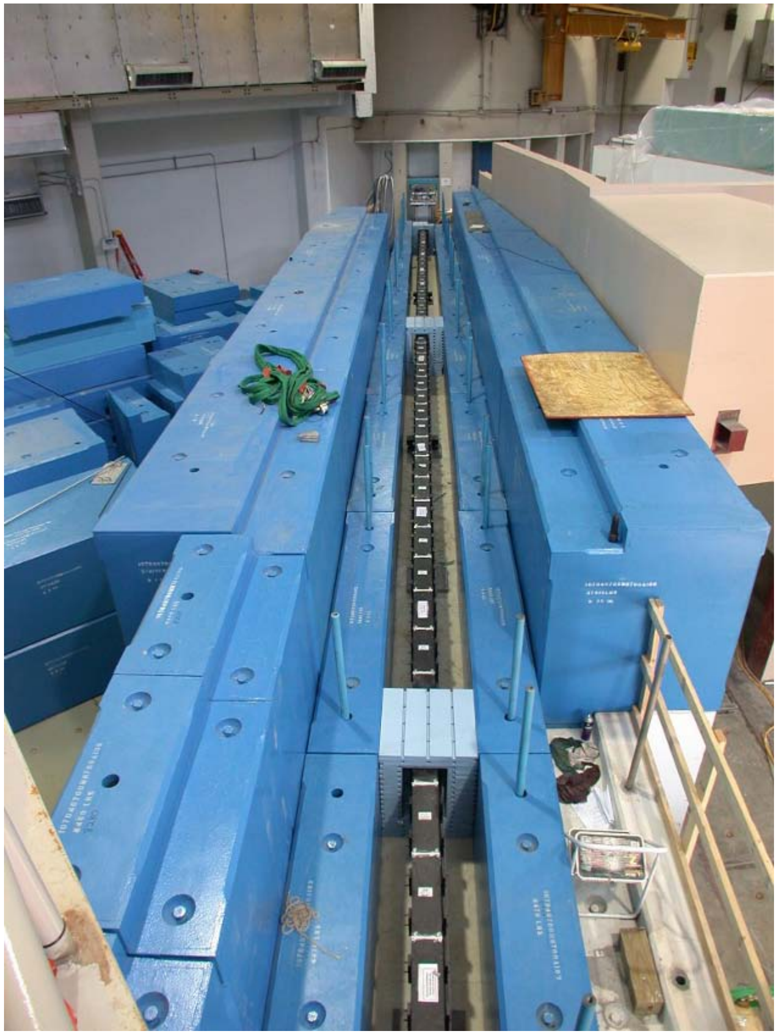


Swiss Neutronics SM guide reflectivity for m=3



TEM of unpolished & ion-polished NiTi layered supermirrors produced at JAEA in Japan – polishing allows high critical angle to be achieved

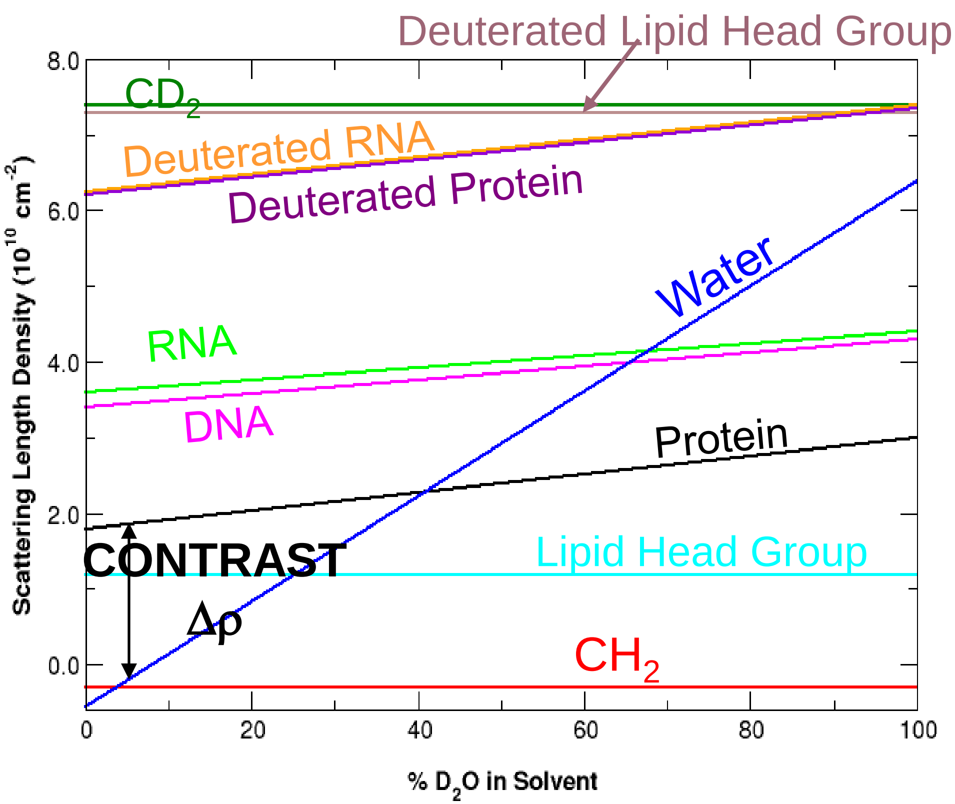
Guides at Spallation Sources need Heavy Shielding Close to the Neutron Source

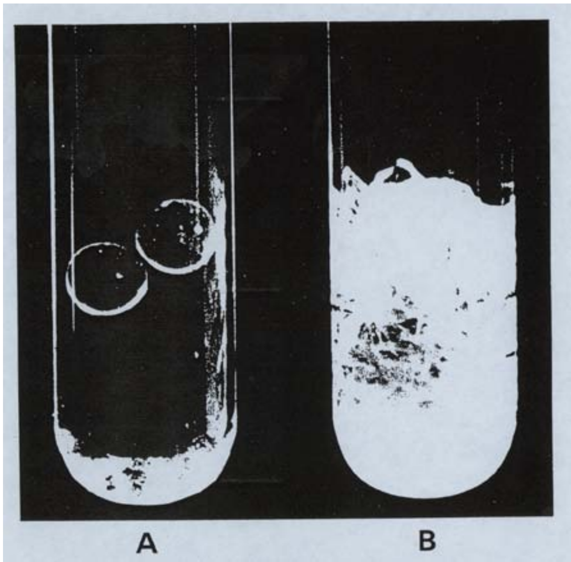


Backscattering spectrometer at SNS. Viewgraph courtesy of Kent Crawford

Contrast Variation is a Key Neutron Technique

- In many cases, the difference between neutron scattering by hydrogen and deuterium is exploited.



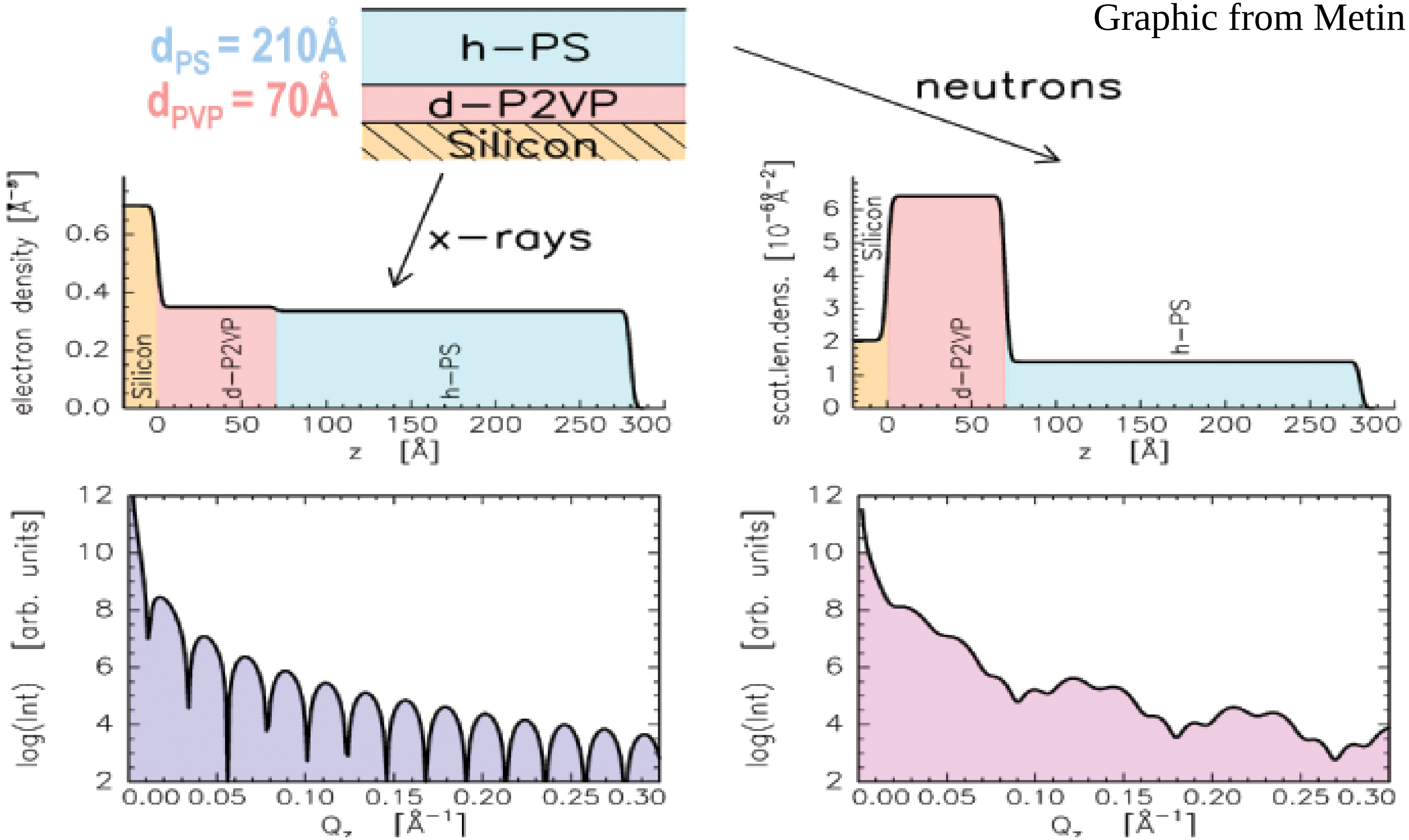


Both tubes contain borosilicate beads + pyrex fibers + solvent. (A) solvent refractive index matched to pyrex;. (B) solvent index different from both beads and fibers – scattering from fibers dominates

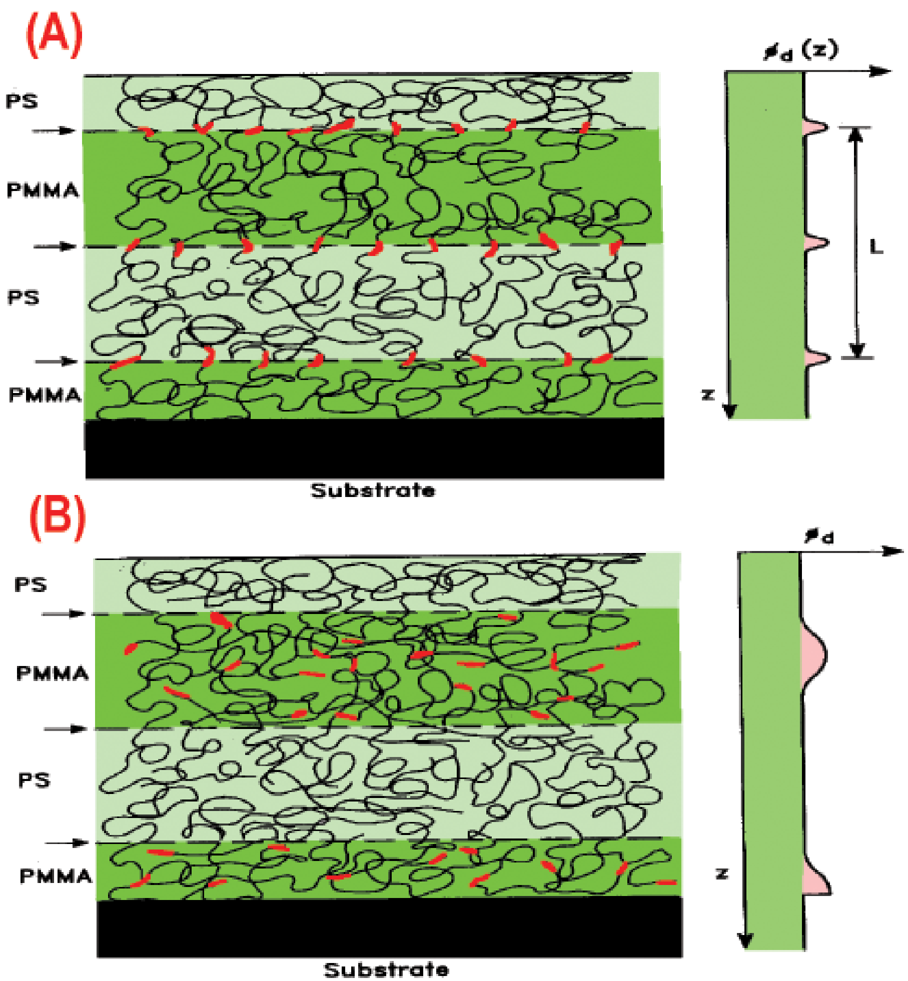
Structures of Thin Films using Reflectometry

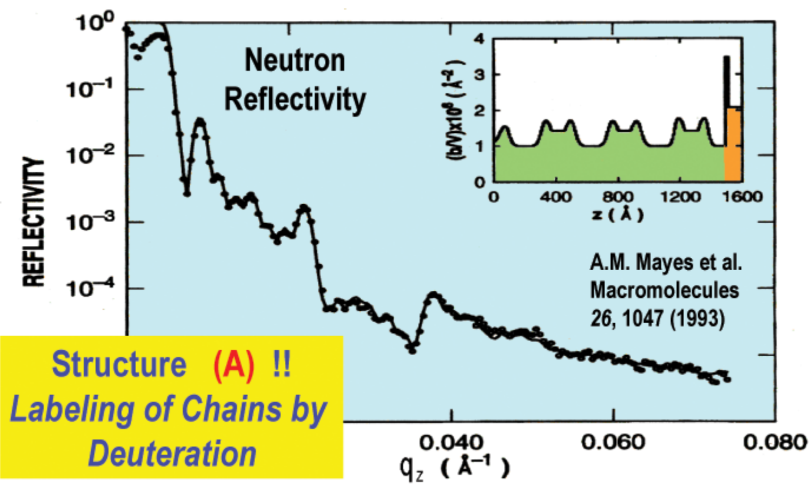
- The contrast between deuterated and protonated polymers allows them to be distinguished by neutron reflectometry

Graphic from Metin Tolan



Where are the Chain Ends & Junctions in an Annealed Diblock Copolymer Film?

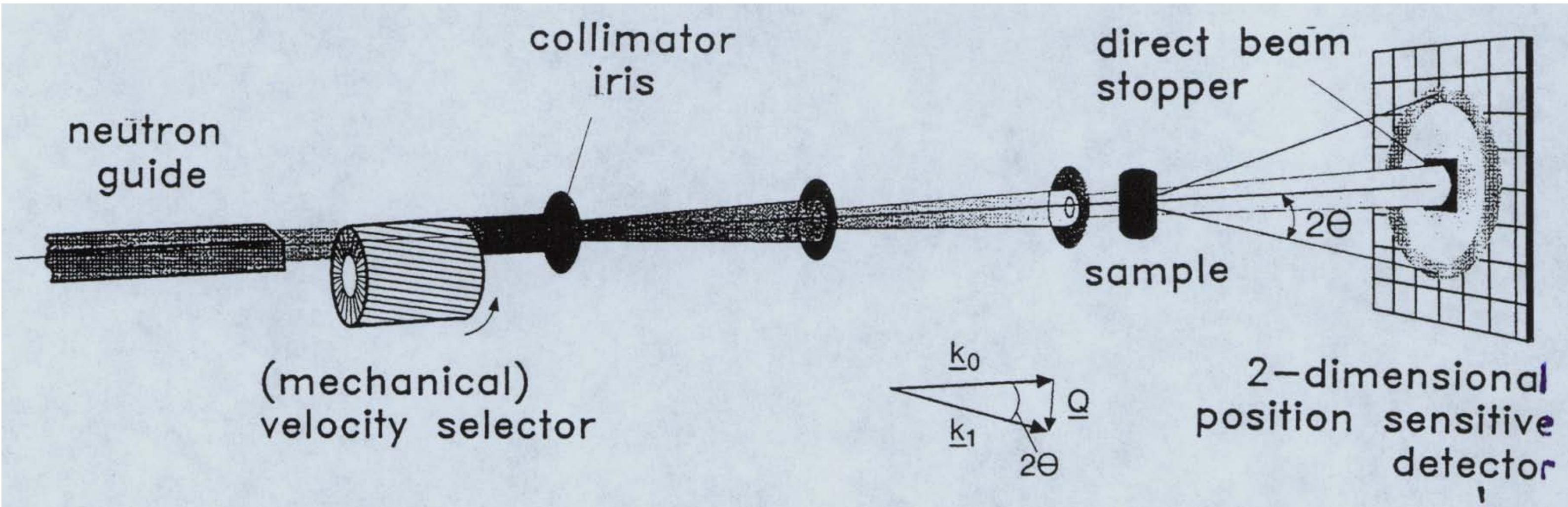




Junctions segregate to the interface between domains; ends are distributed throughout the corresponding domain

Another Simple Technique has Provided Much Information about Complex Nano-Structures

- Small Angle Neutron Scattering (SANS)



  - Neutron wavelength (typically ~ 1 nm for SANS) is small compared to the size of scattering objects (typically 10 – 100 nm) so scattering angles are small (think of Bragg's law)
  - Q-dependence of the scattered neutron intensity depends on the shape and size of the scattering objects and the correlations between them.

SANS Has Been Used to Study Bio-machines

- Capel and Moore (1988) used the fact that prokaryotes can grow when H is replaced by D to produce reconstituted ribosomes with various pairs of proteins (but not rRNA) deuterated
- They made 105 measurements of inter-protein distances involving 93 30S protein pairs over a 12 year period. They also measured radii of gyration
- Measurement of inter-protein distances is done by Fourier transforming the form factor to obtain G(R)
- They used these data to solve the ribosomal structure, resolving ambiguities by comparison with electron microscopy

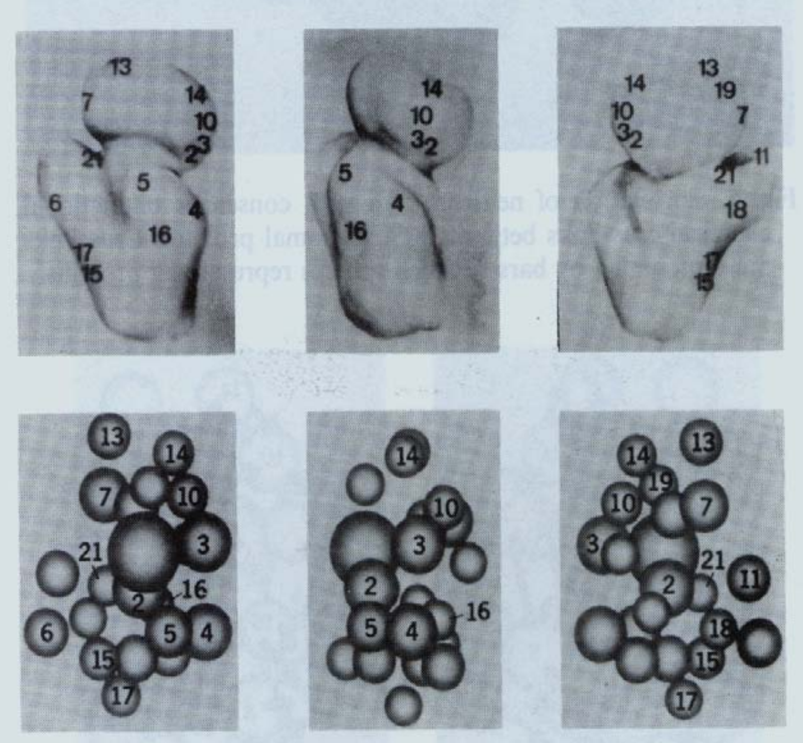
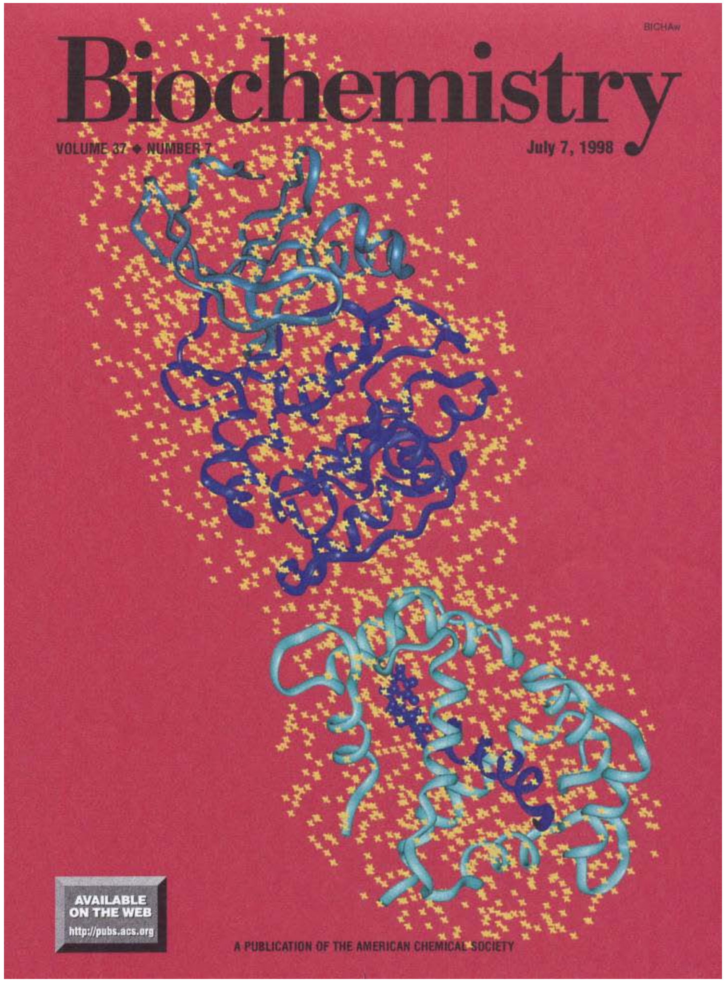


Fig. 4. Comparison of neutron map with a mapping of surface-exposed antigenic sites of ribosomal proteins of the 30S subunit obtained by immune-electron microscopy (Stoeffler & Stoeffler-Meilicke, 1986).

Often it is Necessary to use a Combination of Tools

Crystallography – structure of the catalytic core of the enzyme and reveals the location of the catalytic cleft.

High field NMR with isotope labeling – high resolution solution structure of calmodulin complexed with its binding domain from the enzyme.



Neutron scattering with isotope labeling – shapes and positions of the Myosin Light Chain Kinase enzyme and calmodulin in the Ca^{2+}-calmodulin activated complex.

Use computational modeling based on crystallographic data to determine molecular shapes under various binding conditions

Krueger *et al.*, 1997 **Biochemistry** *36:* 6017.

Early (pre 1970) Neutron Scattering Experiments Provided Underpinnings of Modern Understanding

- Localization of hydrogen in crystal structures
- Neel state of antiferromagnets & ferrimagnets
- Electronic distributions around atoms (form factors)
- Interatomic potentials in metals, semiconductors, rare gases, ionic crystals etc deduced from phonon dispersion curves
- Roton excitations in liquid ^{4}He
- Structural phase transitions (soft modes, central peaks)

Can it help us understand modern complex materials?

Recent Examples Where Neutron Scattering has been Helpful

- Glasses (spin and real) & amorphous materials
- Membranes (mostly artificial) & thin films
- Polymers and colloids
- Percolation
- Absorption and diffusion in porous media
- Strain distributions (~1 mm length scale)
- Critical exponents
- Protein structures (esp. position of protons)
- Correlated electron materials
- Wetting
- ……..

The Science Addressed by Neutron Scattering Covers Many Fields