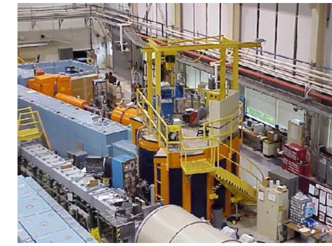
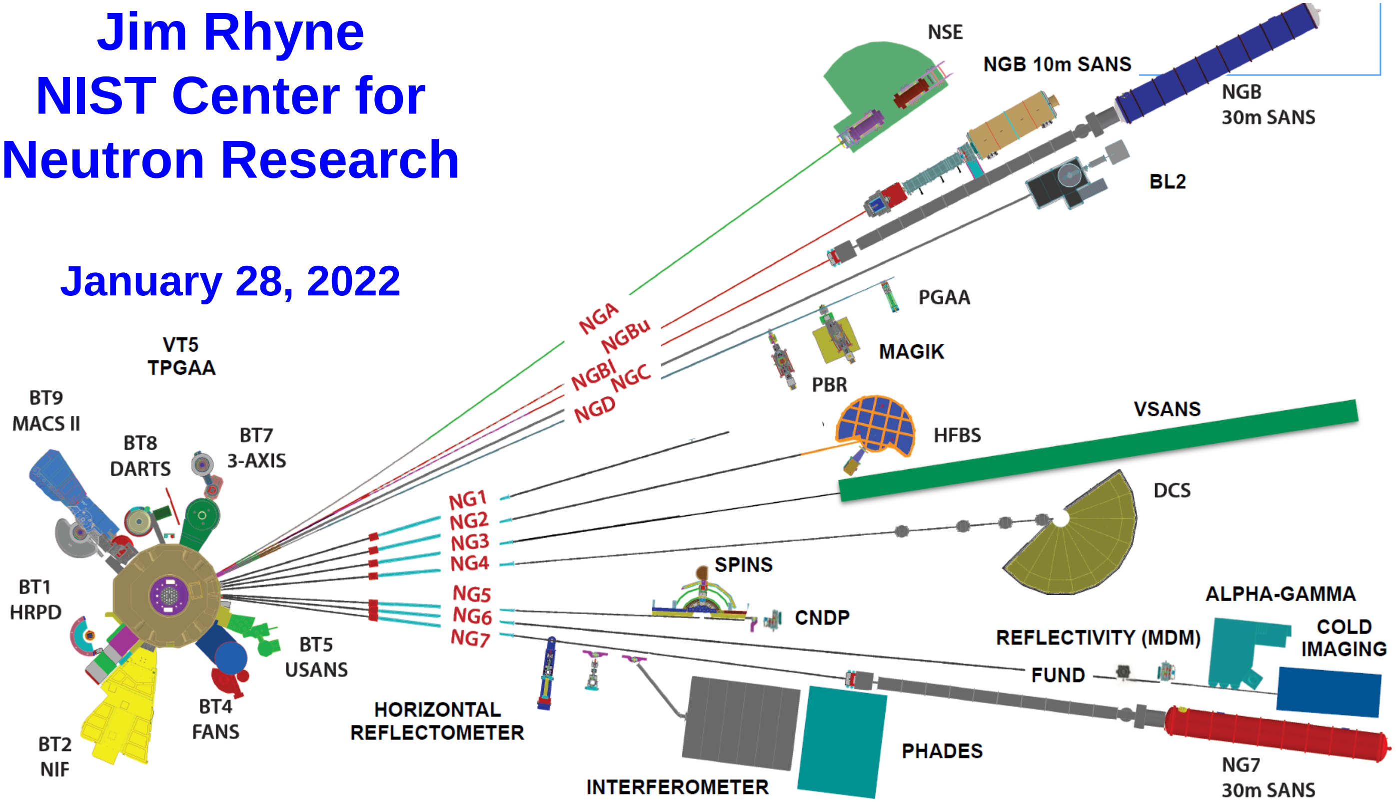
Choosing the Right Spectrometer



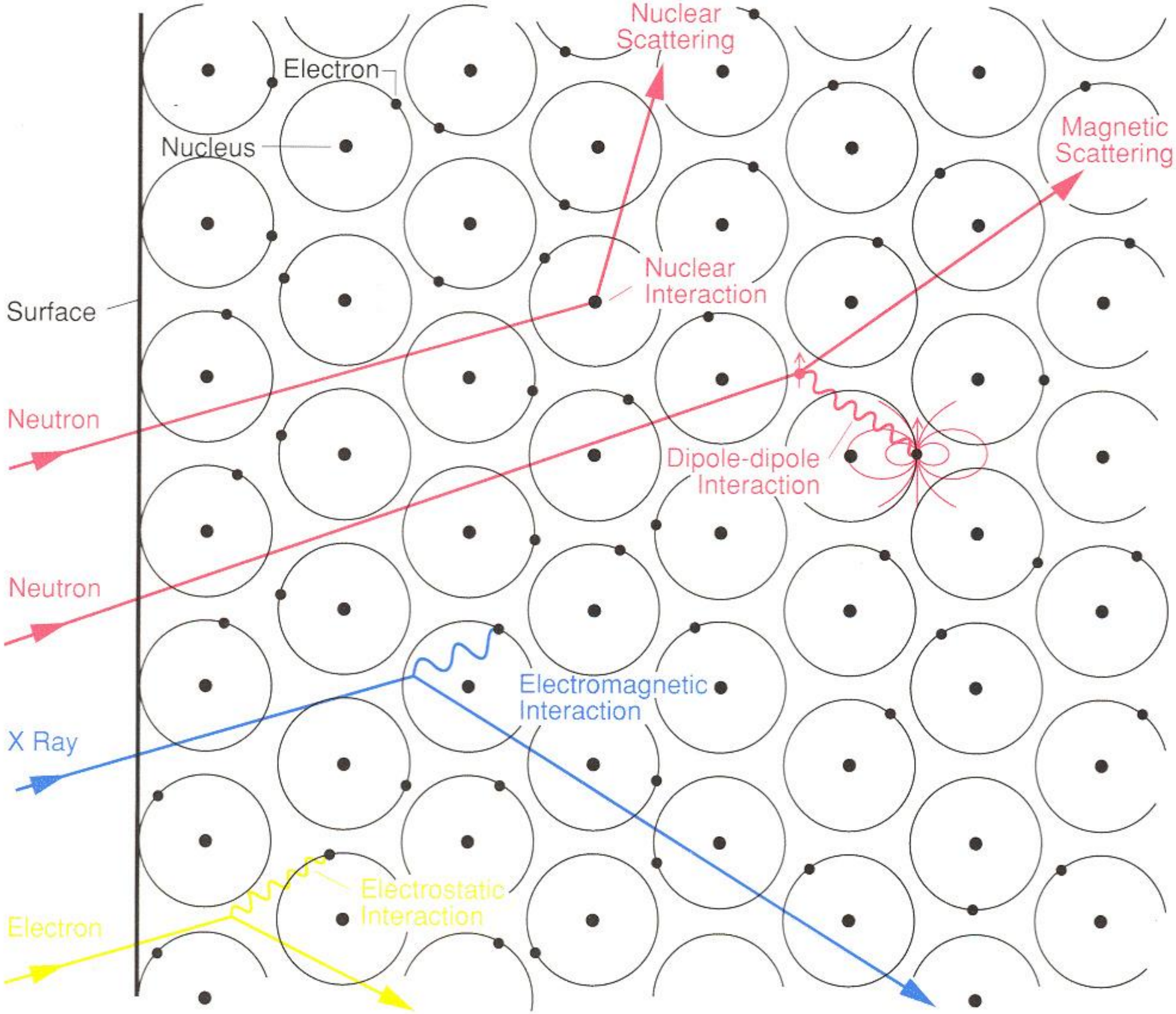
Jim Rhyne
NIST Center for Neutron Research

January 28, 2022



Thanks to Peter Gehring, Jeff Lynn, and Dan Neumann for preparing many of the slides

Interaction of radiation with materials

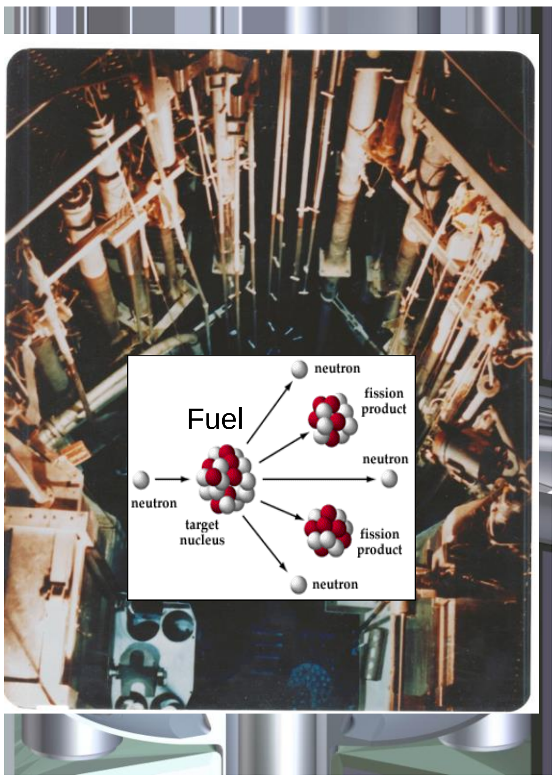


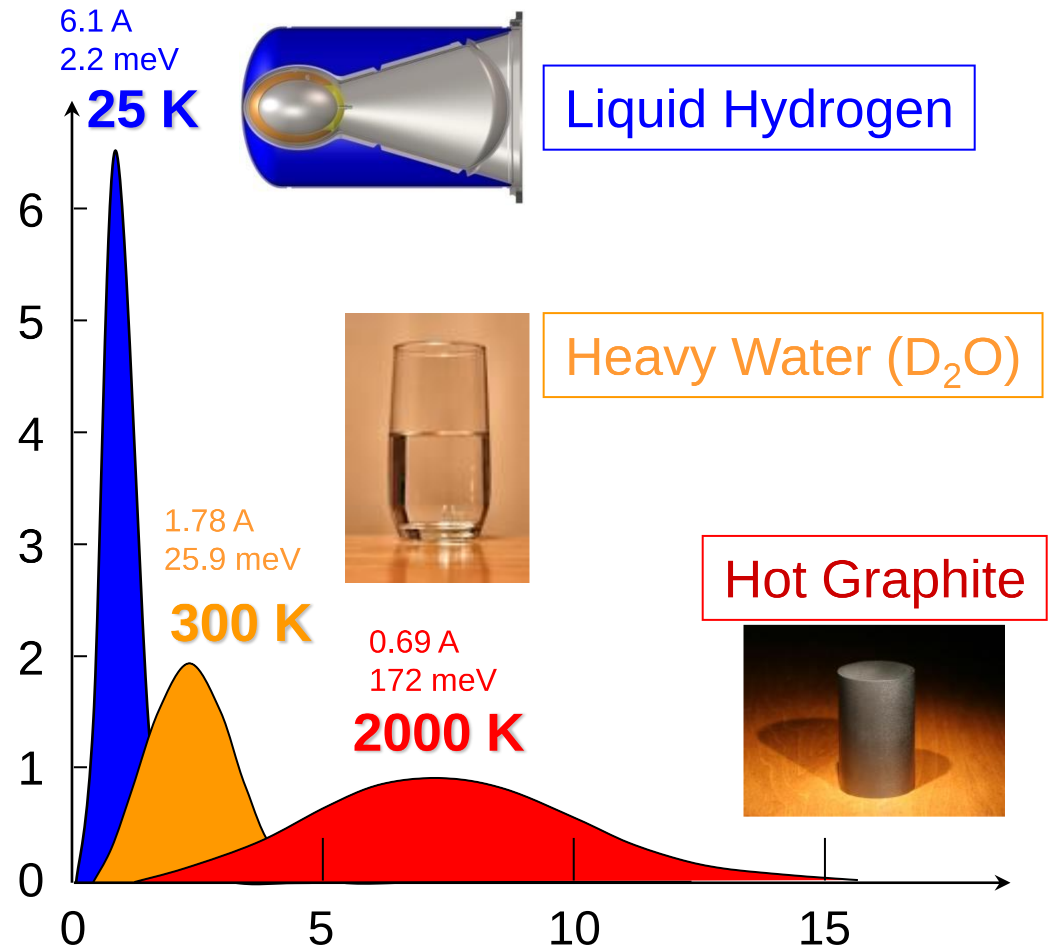
From Roger Pynn – *Neutron Primer*

Neutron Source: Moderation

Maxwellian Distribution

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





“Fast” neutrons: v = 20,000 km/sec

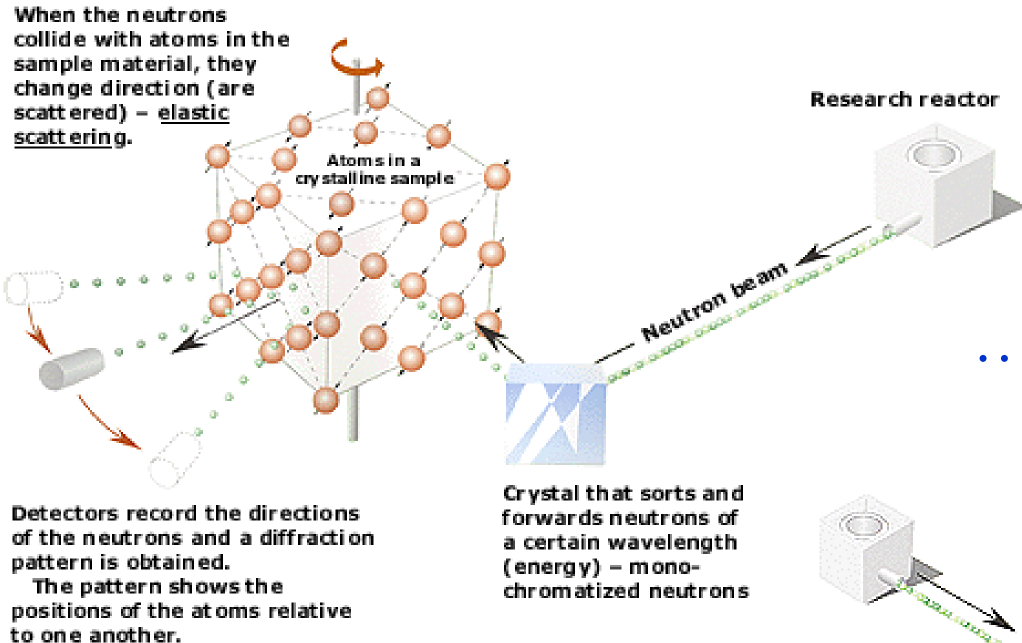
Neutron velocity v (km/sec)

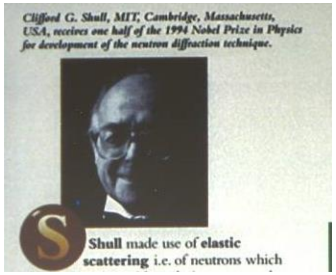
Neutron Scattering

The Nobel Prize in Physics 1994

CHRNS NIST

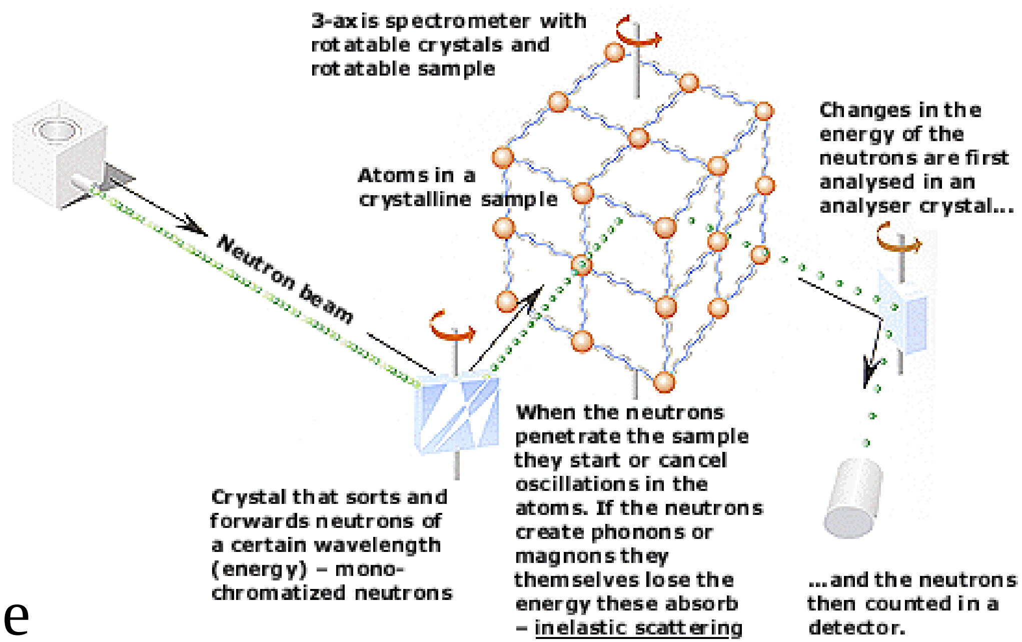
Neutrons show where the atoms are….

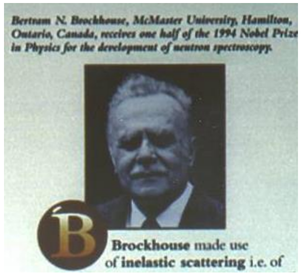




Cliff Shull

…and what the atoms do.





Bertram Brockhouse

Scattering of neutrons by nuclei

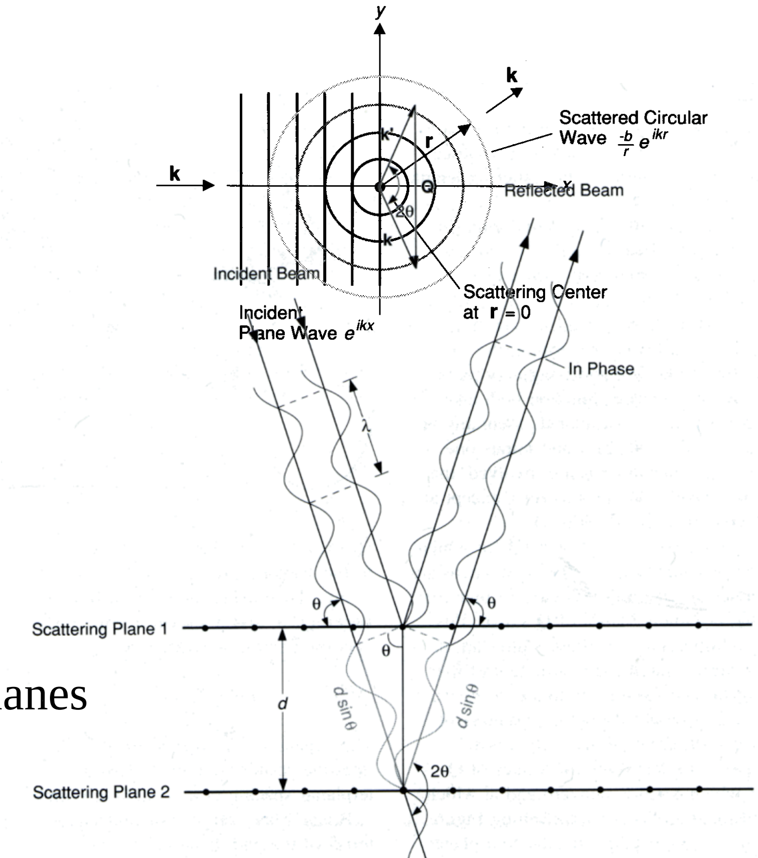
- A single isolated nucleus will scatter neutrons with an intensity (isotropic)
  - $I = I_0\,\sigma = I_0[4\pi b^2]$

    where I_0 = incident neutron intensity,
    b = scattering amplitude for nucleus
- What happens when we put nucleus (atom) in lattice?
  - Scattering from N neuclei can add up because they are on a lattice (constructive interference)
  - Adding is controlled by phase relationship between waves scattered from different lattice planes
  - Intensity is no longer isotropic

    Bragg law gives directional dependence

    $\sin\theta = m\,\lambda/2d$
  - Wave vector $|\mathbf{k}| = 2\pi/\lambda$

-- Intensity I(Q, or θ) is given by a scattering cross-section

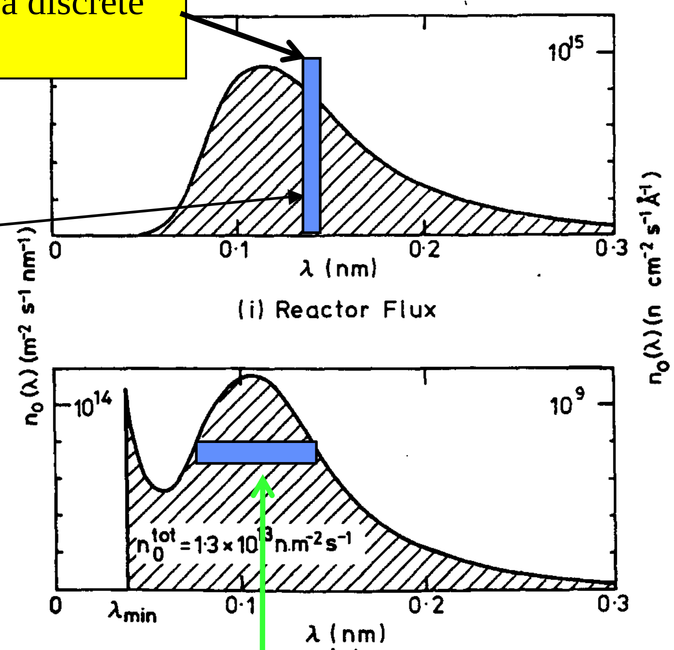


Path difference for neutrons (waves) scattered from two adjacent atomic planes
$= 2d\sin\theta = m\,\lambda$ for constructive interference to occur

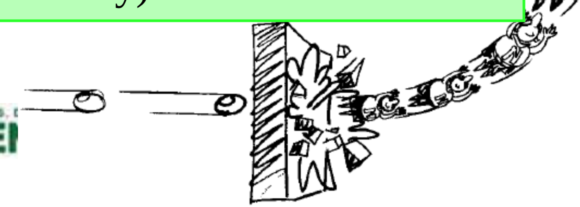
How do we find the wavelength to make the Bragg law work?

- **Reactor**
  - Fission of U^{235} produces neutrons
  - Fission spectrum moderated (slowed down) by either D_2O or H_20 (less effective moderator) and neutrons are extracted through beam tubes for spectrometers – fixed wavelength used
- **Spallation source**
  - High E protons (e.g., 800 MeV) impinge on target (W, Hg or U)
  - Nucleus of target is “exploded” by proton impact and emits 15 – 25 neutrons per proton with average E = 55 MeV (+ γs, neucleons and neutrinos)
  - Neutrons moderated by liquid H, H_20 or methane
  - Spallation sources generally operate in pulse mode – 60 Hz at SNS

Monochromator crystal is used to saw-out a discrete wavelength



Time of flight is used to sort out wavelengths
Short wavelength neutrons get to sample first (highest velocity)

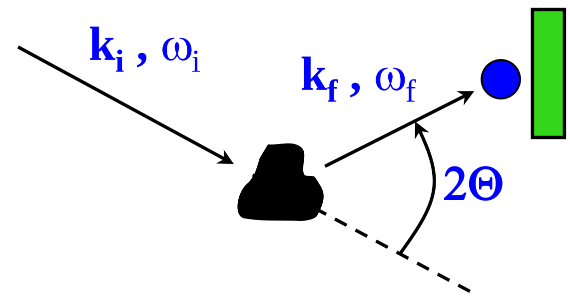


Los Alamos NATIONAL LABORATORY EST. 1943
LANSCE LUJAN CENTER Los Alamos National Laboratory

What and how do neutrons measure

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

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



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

(2) The expressions for the scattered neutron flux Φ (intensity) depend on the positions and motions of atomic nuclei or unpaired electron spins.

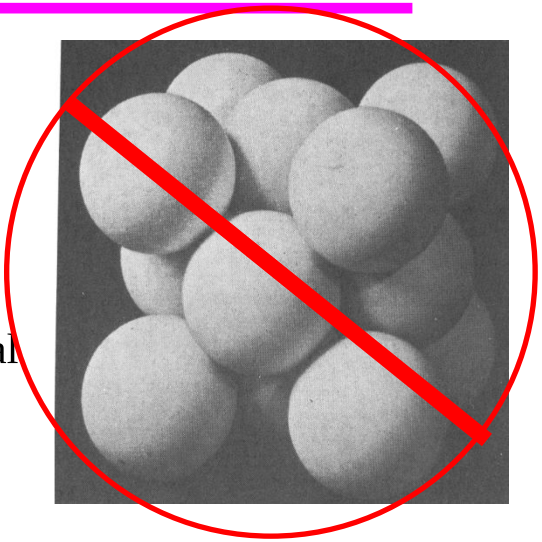
$$\boxed{\Phi = F\{\vec{r}_i(t), \vec{r}_j(t), \vec{S}_i(t), \vec{S}_j(t)\}}$$

Φ provides information about all of these quantities, but in reciprocal space!

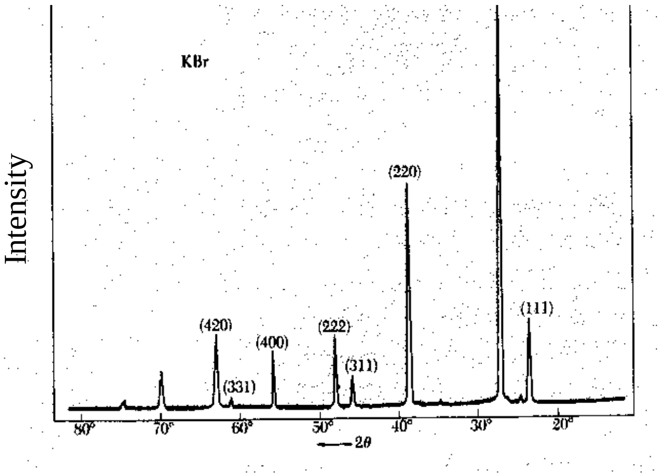
Golden Rule of Neutron Scattering

- We don't take pictures of atoms!

Atoms in fcc crystal



- Job security for neutron scatterers – we live in ***reciprocal space***



The Scattered Flux is Proportional to a Cross-section, One of Three Basic Cross-Sections

σ Total cross-section -- # of neutrons scattered per second -- Φ_i.

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

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

It's all about Conservation of Momentum and Energy

$$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$$

Wave vector transfer = vector difference k_i and k_f

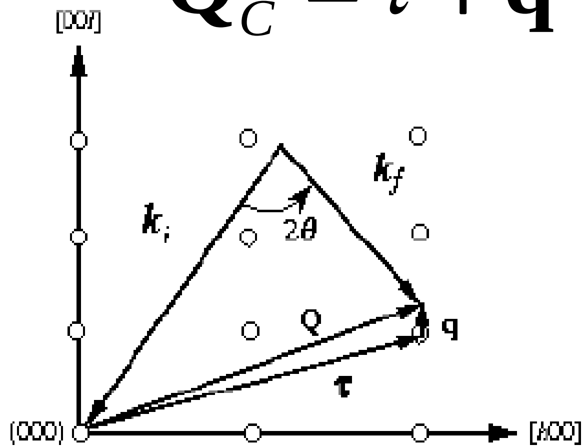
$$\Delta E = \frac{\hbar^2 k_i^2}{2m} - \frac{\hbar^2 k_f^2}{2m}$$

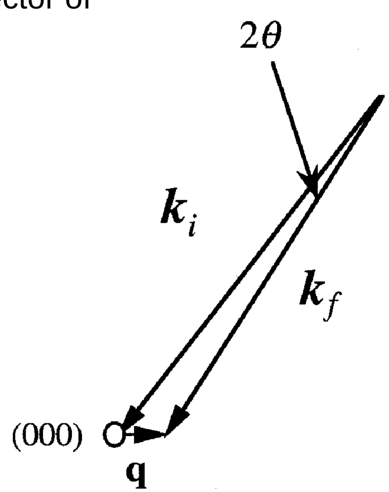
Energy transfer to/from neutron to sample excitation

Reciprocal lattice vector

$$\mathbf{Q}_C = \tau + \mathbf{q}$$

Momentum vector of excitation

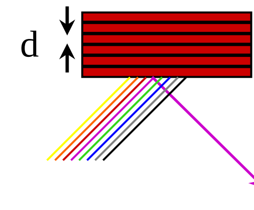




Methods of Specifying and Measuring $\vec{k}_i$ and $\vec{k}_f$ for various instruments

1. Bragg Diffraction

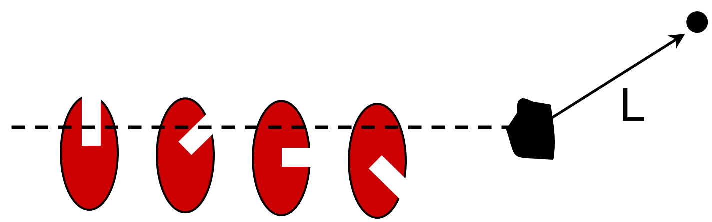
   BT7, MACS, HFBS



$n\lambda = 2d\sin\theta$

$k = \pi/(d \sin\theta)$

2. Time-of-Flight (TOF)



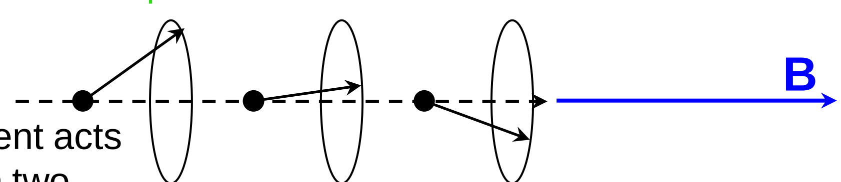
$v = L/t$

$k = m_n L/(\hbar t)$

DCS [TOF] uses a synchronized array of 7 choppers to produce clean beam of k_i

HFBS uses a Doppler drive to vary k_i with k_f fixed

3. Larmor Precession



NSE

Larmor precession of neutron mag moment acts as a clock to time neutron transit through two solenoids – Sample introduces $\Delta E \neq 0$ and changes time

Energy, wave vector, and wavelength relations for various probes

$$E_{neutron}(meV) = 2.0719k^2 = 81.7968/\lambda^2$$

$$E_{photon}(keV) = 2.0k = 12.4/\lambda$$

$$E_{electron}(eV) = 3.8k^2 = 150/\lambda^2$$

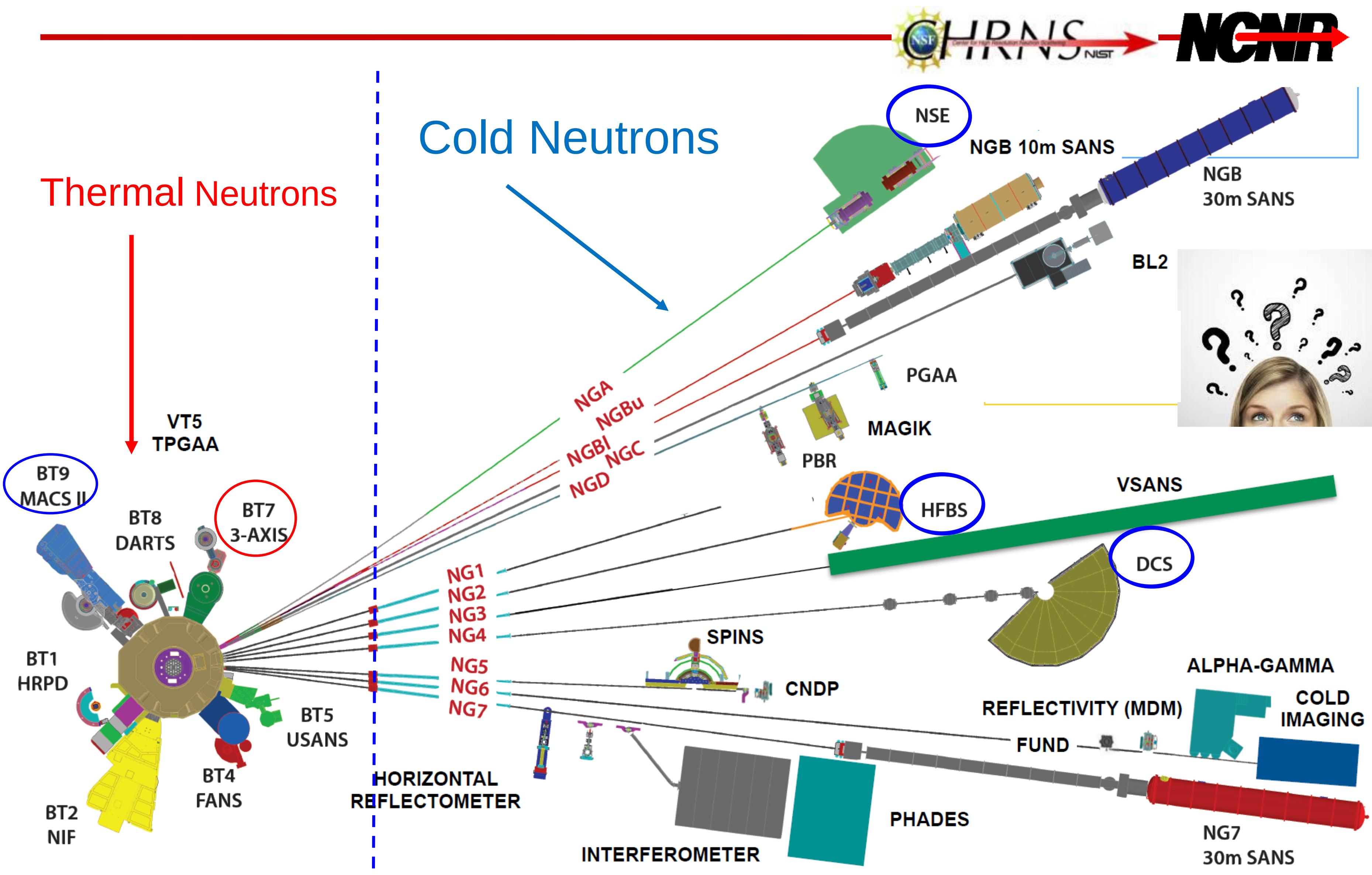
$$1\,meV = 11.6\,K \quad (k_B T)$$

$$1\,meV = 8.06\,cm^{-1} \quad (E/hc)$$

$$1\,meV = 0.2418\,THz \quad (E/h)$$

$$1\,meV/\mu_B = 17.3\,T \quad (E/\mu_B)$$

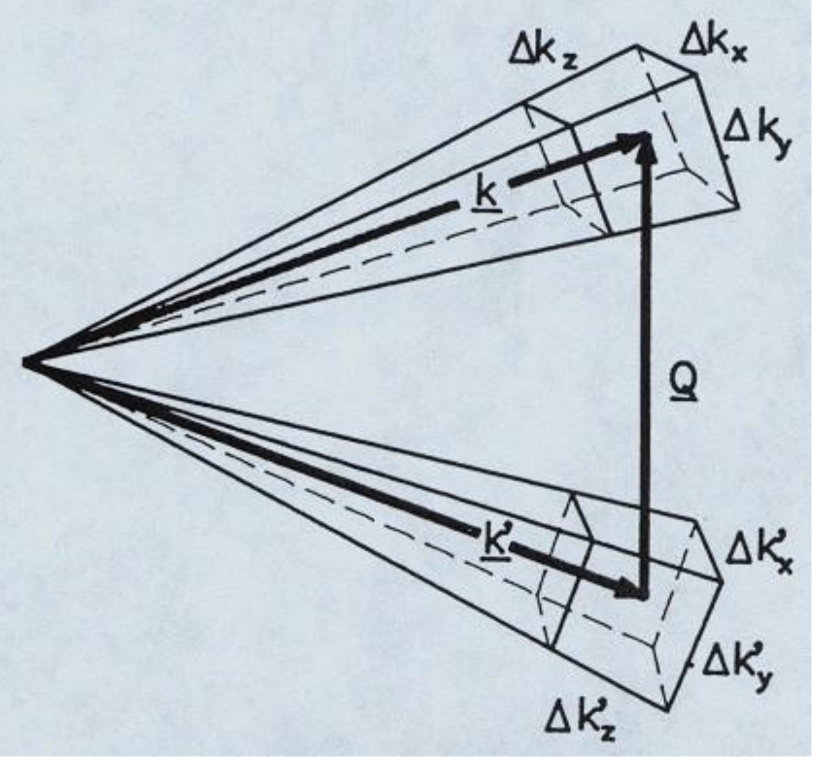
The NCNR Instrument Zoo



Why So Many Different Spectrometers?

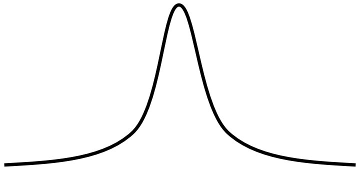
Because neutron scattering is an intensity-limited technique. Thus detector coverage and resolution MUST be tailored to the science.

To get a usable signal, the neutron wavelength and energy can only defined with finite precision and likewise for **Q**.

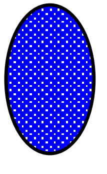


Courtesy of R. Pynn

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



$\propto$



How do I Choose the Right Spectrometer?

Two basic considerations:

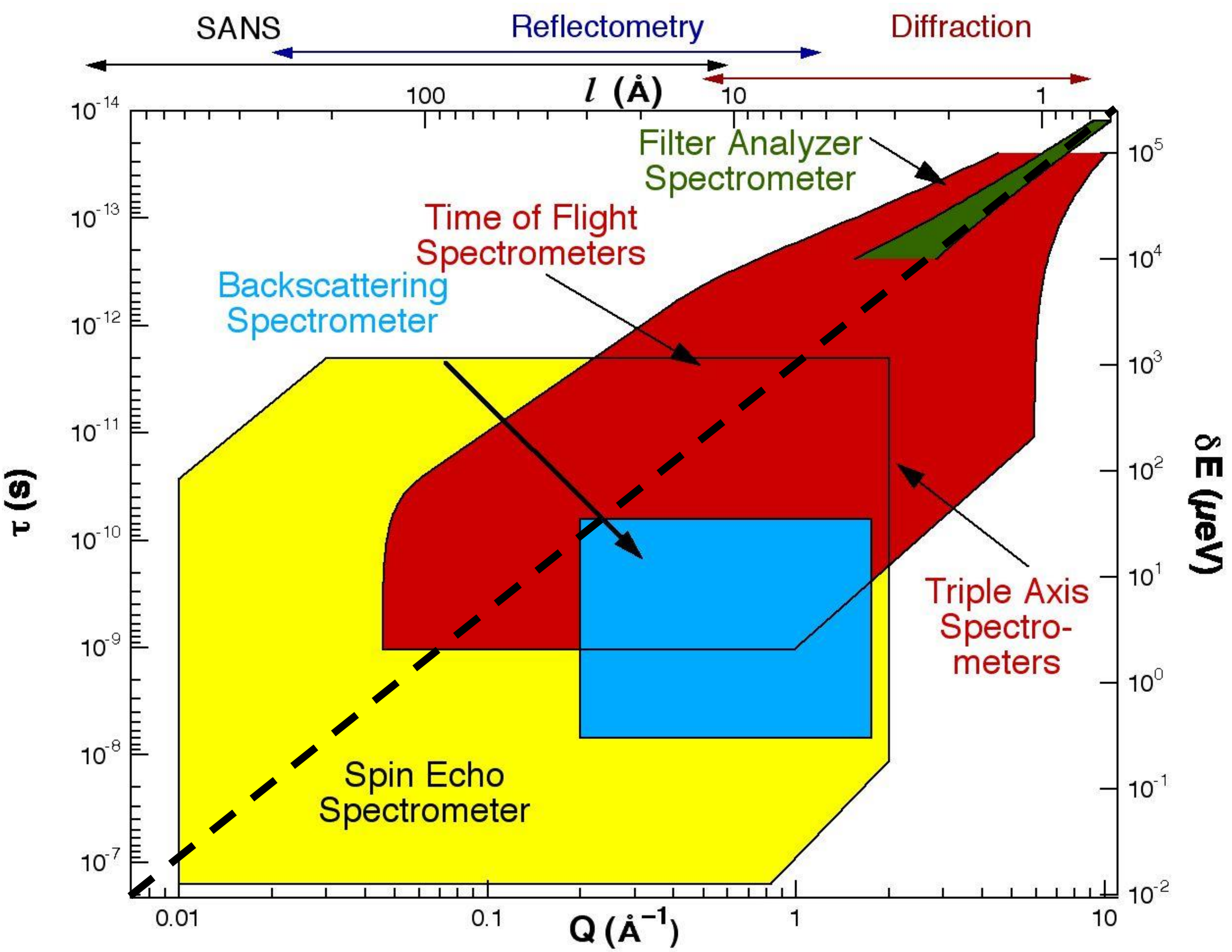
1. What are the time scales ($\hbar\omega$) of interest?
2. What are the length scales (Q) of interest?

(Some spectrometers overlap →
the choice may boil down to one of resolution)

Resolution considerations:

1. What energy resolution ($\Delta\hbar\omega$) is required?
2. What momentum resolution (ΔQ) is required?

Different Spectrometers Cover Different Regions of Phase Space



Inelastic Spectrometers

	Instrument	Approx. Resol.
	Thermal triple-axis instruments (BT-7) (BT-4)	1 meV
	Cold neutron triple-axis instrument (MACS) (SPINS)	
*S(**Q**,E)*	Disk chopper time-of-flight spectrometer (DCS)	250 μeV
	High flux backscattering spectrometer (HFBS)	1 μeV
*S(**Q**,t)*	Spin-echo spectrometer (NSE)	δt → ~50 neV

Remember – **Intensity** ↓

Resolution ↑

All these different spectrometers are designed differently to optimize intensity and resolution for different measurement requirements

Rules of Thumb

1. What are the energies ($\hbar\omega$), i.e. time scales ($\Delta t \sim 1/\omega$), of interest?

$\hbar\omega \approx$ 1-100 meV - use a thermal triple-axis spectrometer like BT7.

$\hbar\omega \approx$ 20-30 μeV - use HFBS or NSE.

In between - use MACS or DCS or a cold-neutron triple-axis spectrometer like SPINS.

2. Make sure that the length scales **L** of the relevant motions lie within the range of the spectrometer. For example, consider the HFBS. ($\mathbf{Q} \sim 2\pi/\mathbf{L}$)

$$\mathbf{Q}_{min} = 0.25\ \text{Å}^{-1} \rightarrow \mathbf{L}_{max} \sim 25\ \text{Å}$$
$$\mathbf{Q}_{max} = 1.75\ \text{Å}^{-1} \rightarrow \mathbf{L}_{min} \sim 3.5\ \text{Å}$$

REMEMBER - $\mathbf{Q}_{min}$ and $\mathbf{Q}_{max}$ are <u>inversely</u> proportional to the incident neutron wavelength

More Rules of Thumb

Is your sample polycrystalline or amorphous?

Does ONLY the magnitude (not the direction) of $\mathbf{Q}$ matter?

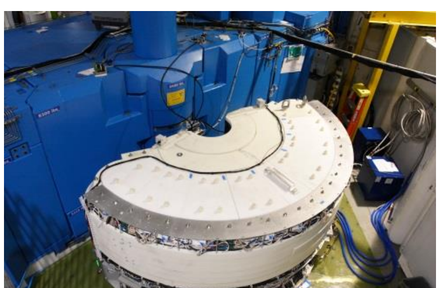
Is the expected $\mathbf{Q}$-dependence of the scattering weak?

This often means that you want to look at a large region of $\mathbf{Q}$-$\hbar\omega$ space, or that you can sum the data over a large region of $\mathbf{Q}$-$\hbar\omega$ space.

YES? Consider instruments with large analyzer areas.

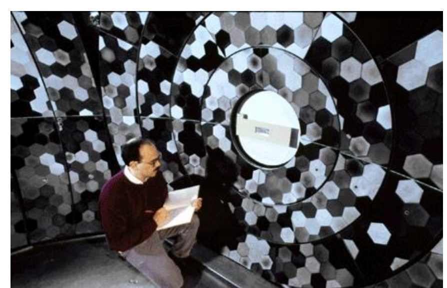
NO? Consider using BT7, SPINS, or NSE.

MACS



DCS

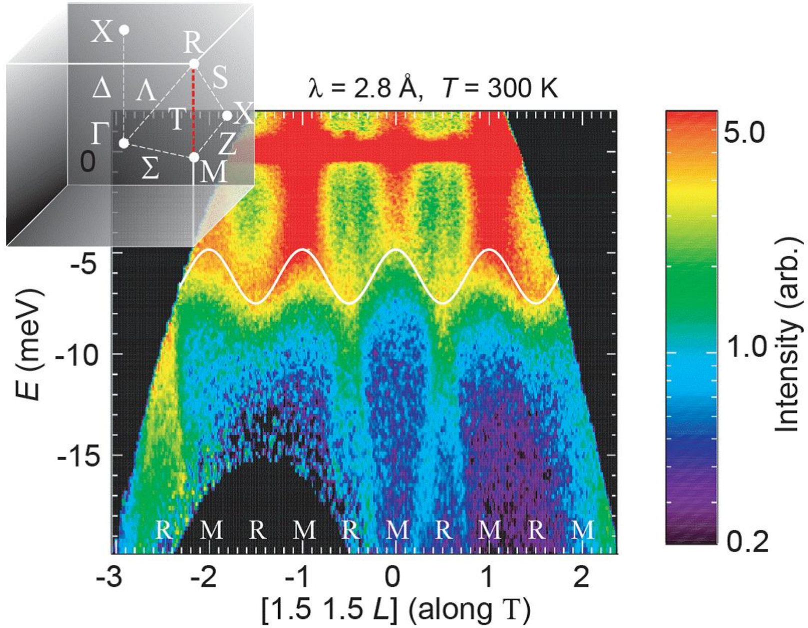
HFBS



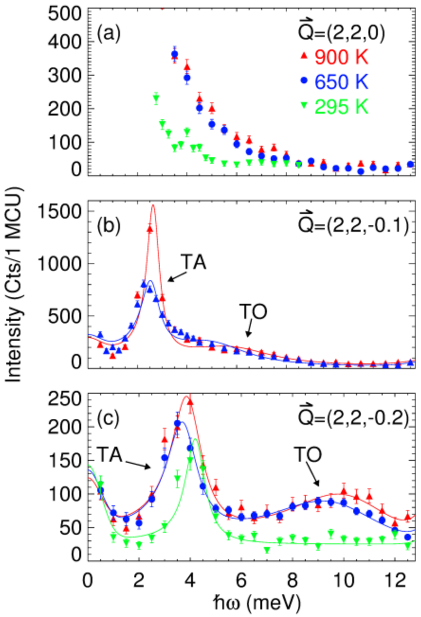
BT7

Example: DCS versus BT7

DCS Broad surveys in **Q-ω**



BT7 Limited regions in **Q-ω**



Rules of Thumb: (think carefully before violating)

DCS, MACS – systems requiring resolution < 400 μeV

BT7 – single crystals – resolution > 100 μeV

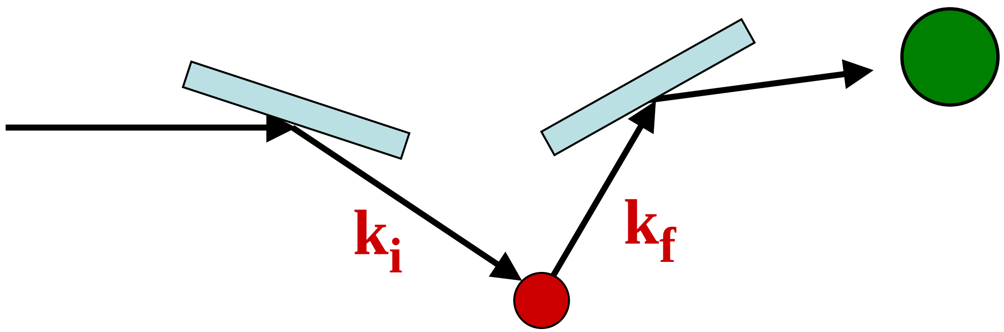
depends on collimation and monochrometer/analyzer

Things to Consider When Choosing BT7

Triple axis spectrometers are typically used when either -

(1) the *direction* of **Q** is important or

(2) the interesting region of **Q-ω** space is of *limited extent*.



One data point at a time …

Remember – **Intensity** ↓

Resolution ↑

Things to Consider When Choosing HFBS

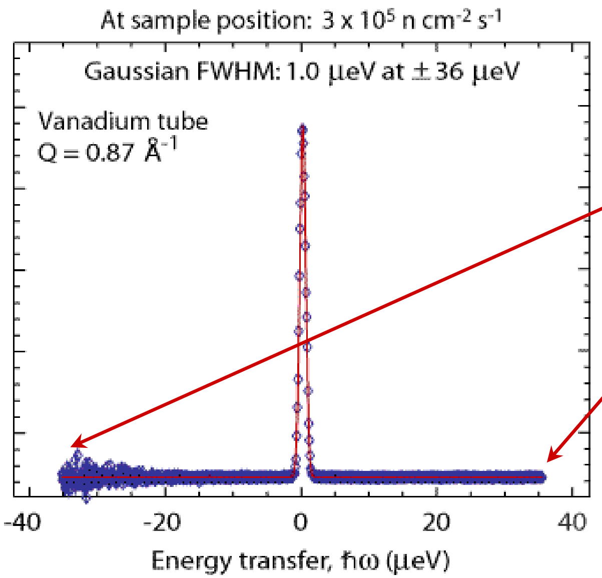
HFBS detects neutrons of E_f = 2.08 meV and uses monochrometer Doppler shifting of E_i to yield ΔE over a very narrow range (e.g., 75 μeV) but with high resolution

$\boxed{0.25\ \text{Å}^{-1} < \mathbf{Q} < 1.75\ \text{Å}^{-1}}$

Do the length scales of interest lie within this Q-range?

$\boxed{\delta\mathbf{Q} < 0.1 - 0.2\ \text{Å}^{-1}}$

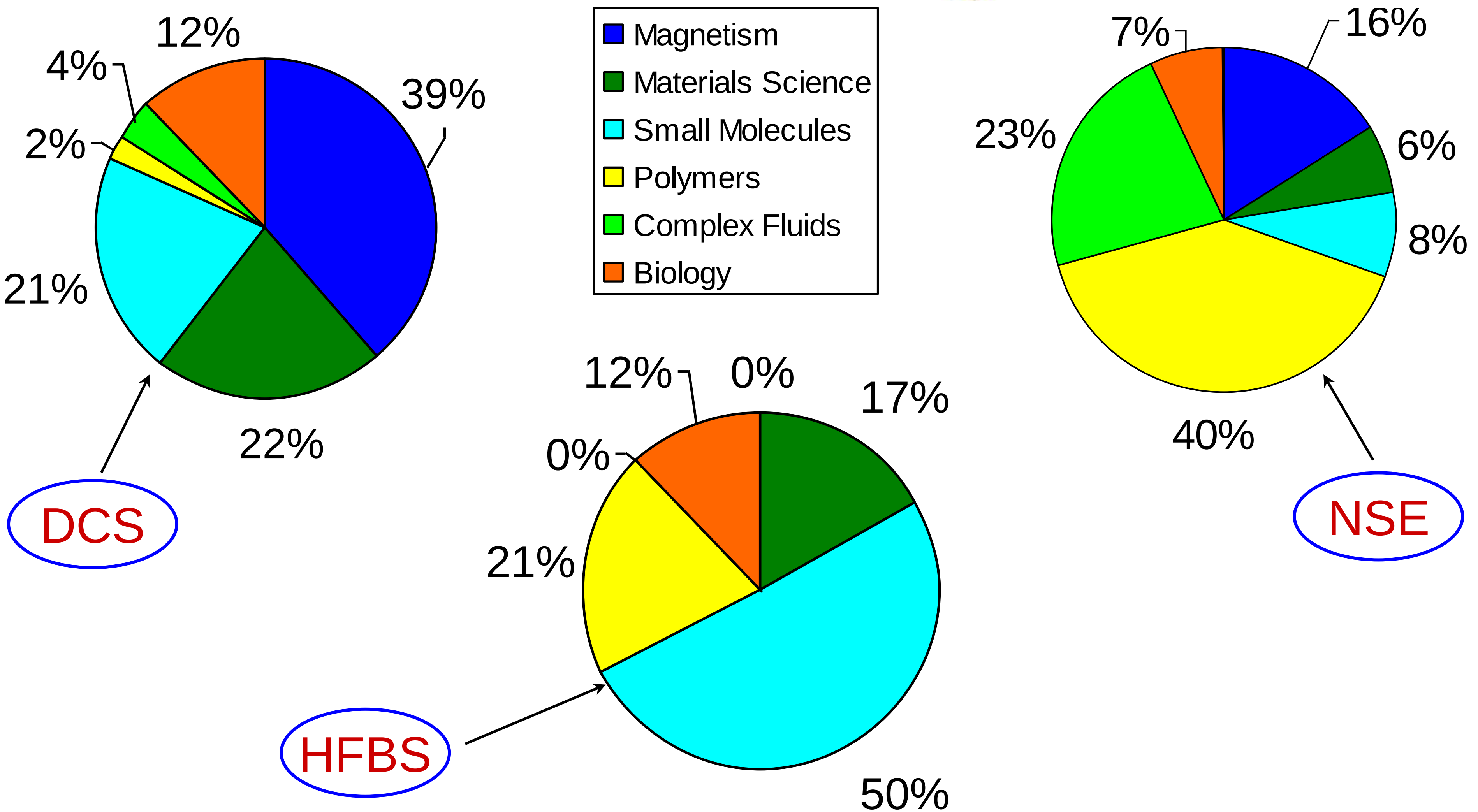
Can you live with such coarse Q-resolution?



Do the features of interest lie within this hω-range?

Do you really require such good energy resolution δE ~ 1 μeV (or perhaps even better resolution)?

Who uses what instruments for their research



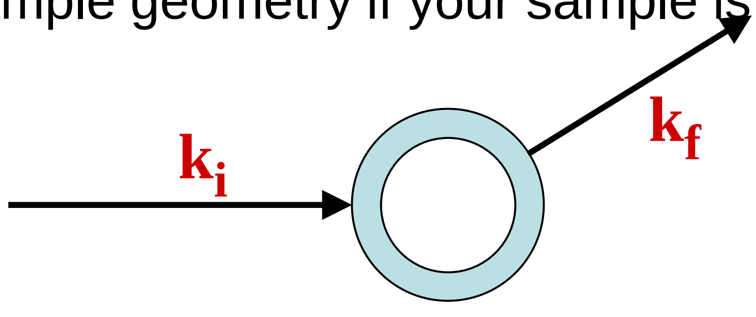
Sample "Design"

Single crystals yield the most information.

Increase the intensity by increasing the amount of sample.

If you have a powder, use a cylindrical container (rather than flat plate).

Annular may be the best sample geometry if your sample is absorbing.



Transmission of the beam should be ~70-90%.

$I/I_o = \exp(-n\sigma_A T)$

Almost all experiments of collective excitations involve coherent scattering
→ If sample contains H it should be deuterated (D).

General Sample "Design"

Know as much about your sample as possible!!
(Beamtime costs ~ $5000/day!!)

Other considerations:
What's the structure (in a general sense)?
Are there any phase transitions (or a glass transition)?
What isotopes are present?
Supplementary data from other measurements like …

Magnetization vs T

Muon spin relaxation

X-ray data

Specific heat vs T

Raman spectroscopy

General Sample "Design"

Try to avoid isotopes that are strongly absorbing.

^{6}Li ^{10}B ^{113}Cd ^{157}Gd

For a complete listing go to

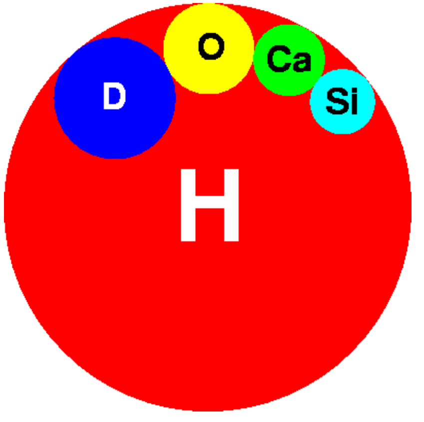
http://www.ncnr.nist.gov/resources/n-lengths

Sample Selective Deuteration

Does the sample contain H?

Remember: Neutrons LOVE H!!

Create a sample where -
the “interesting” portions are hydrogenated and
the “uninteresting” portions are deuterated.



How do I get time on an instrument?

Access to the neutron scattering instruments is totally merit-based. Open to all qualified users, but subject to an anonymous peer-review of proposals.

Calls for proposals are issued about twice/yr.

Next deadline for new proposals: Not scheduled, likely fall 2022

Further information on submitting proposals :

http://www.ncnr.nist.gov/programs/CHRNS/CHRNS_prop.html

Enjoy the Science With Neutrons!

Questions