Inelastic Neutron Scattering

Motions (mainly nuclear) and Spectrometers

Science and Technology Facilities Council

ISIS Neutron and Muon Source

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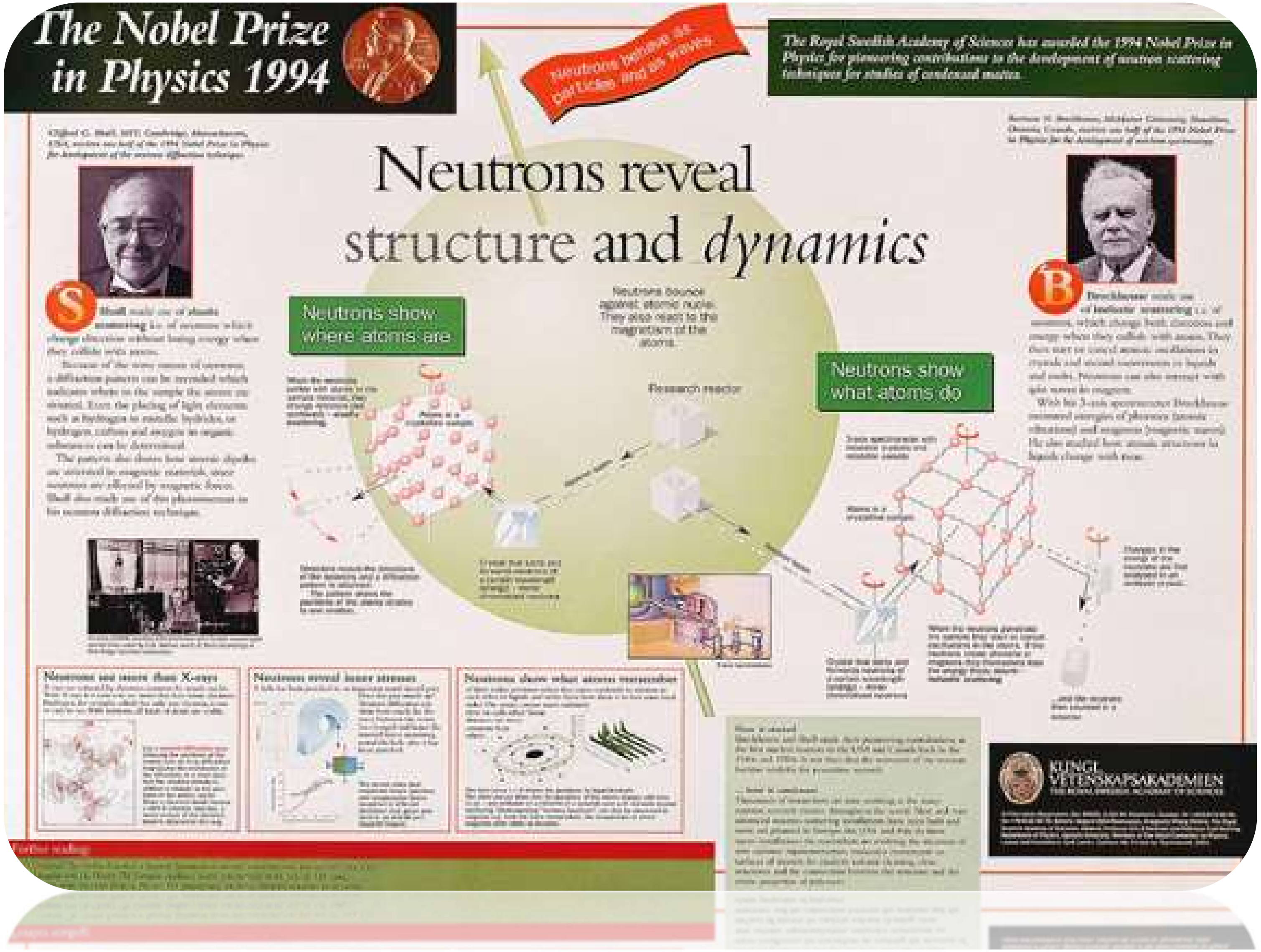
18th Oxford School on Neutron Scattering
11th September 2024



Neutrons tell us where atoms are and what atoms do

Take Home Messages

1. Neutron spectroscopy probes dynamics, whether it is vibrations in a crystal lattice, bond stretching modes, diffusion of species through pores or relaxation processes in polymers, magnetic excitations.

2. It can also, indirectly, inform on **structure**/binding

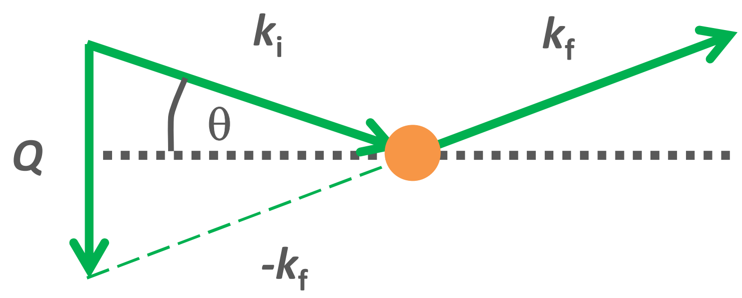
3. Variety of instrument types are available – get informed on which is best suited to your problem

4. Plethora of applications – ask or search the literature and talk to facility scientists

5. Offer unique info – complementary to other techniques, in particular atomic or molecular calculations.

Neutron Concepts (reminder)

Elastic and Inelastic Scattering



Elastic scattering:

$$|\boldsymbol{k}_i| = |\boldsymbol{k}_f| = \frac{2\pi}{\lambda}\ ; \quad \mathrm{Q} = 2|\boldsymbol{k}|\sin\boldsymbol{\theta} = \frac{4\pi}{\lambda}\sin\boldsymbol{\theta};$$

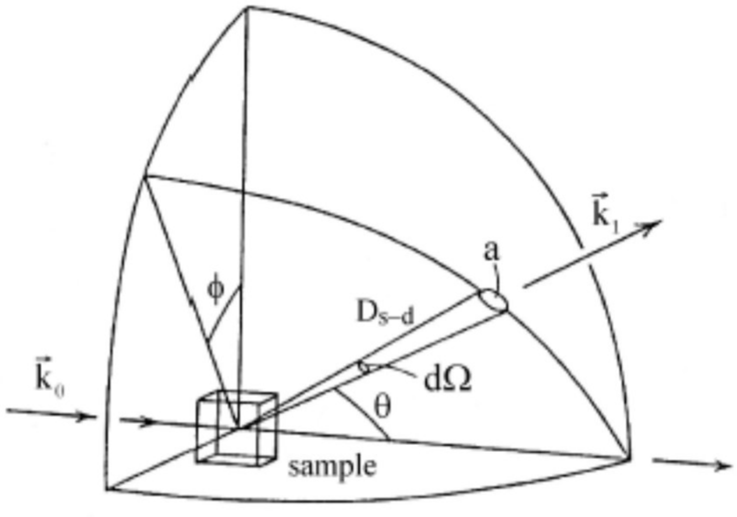
$$\boldsymbol{\Delta}E = \mathbf{0}$$

Inelastic scattering:

$$|\boldsymbol{k}_i| \neq |\boldsymbol{k}_f|;$$

$$\boldsymbol{\Delta E} = \hbar\boldsymbol{\omega} = E_i - E_f = \frac{\hbar}{2m}(k_i - k_f)$$

The Scattering Function, $S(\boldsymbol{Q},\omega)$



$$\frac{d^2\sigma}{d\Omega d\omega} \propto \frac{k_f}{k_i}\frac{\sigma}{4\pi}S(\boldsymbol{Q},\omega)$$

The **scattering function,** $S(\boldsymbol{Q},\omega)$ contains all the physics of the system (in space and time) and depends only on the system.

If your detector can analyse the energy of the neutrons, then the **double differential cross-section** can be defined as

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\begin{array}{c}number\ of\ particles\ scattered\ per\ s\ into\ a\ solid\ angle\\ d\Omega\ with\ final\ energies\ between\ E_f\ and(E_f+dE_f)\end{array}}{I_0 d\Omega dE_f}$$

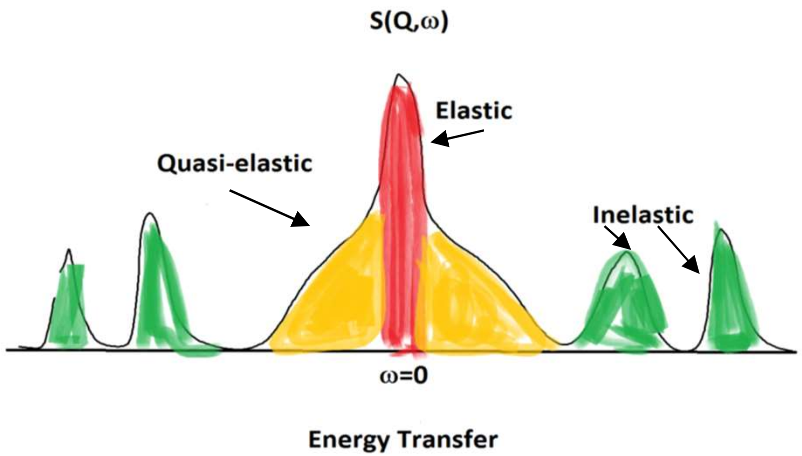
The Scattering Function, *S*(*Q*,ω)

$$\frac{d^2\sigma}{d\Omega d\omega} = \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{elastic}} + \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{inelastic}}$$

The Scattering Function, $S(Q,\omega)$

$$\frac{d^2\sigma}{d\Omega d\omega} = \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{elastic}} + \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{inelastic}} + \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{quasi-elastic}}$$

Elastic scattering – no energy exchange ħω=0. Ideally it is a δ function, in reality it is the resolution.



Quasi-elastic scattering (QENS)– a **small** energy exchange ħω≠0≈neV or few meV . Processes with a distribution of energies.

Inelastic scattering (INS) – energy exchange ħω≠0. Processes of discrete energy steps, quantised (vibrations or excitations).

Scattering Cross-section, σ

- σ is the total scattering x-section – probability that the neutron is scattered which depends on nucleus-neutron interactions as defined by the scattering length *b*. Complex number where the imaginary part is for the absorption.

$$\boxed{\sigma_{\mathrm{coh}} = 4\pi\bar{b}^2} \qquad \boxed{\sigma_{\mathrm{incoh}} = 4\pi(\overline{b^2} - \bar{b}^2)}$$

- Detector can only measure a finite angular range $\Delta\Omega$ and for an assembly of nuclei, the differential cross-section is

$$\frac{d\sigma}{d\Omega} = \sum_n \sum_m b_n b_m \exp(i\boldsymbol{Q}\cdot(\boldsymbol{r}_n - \boldsymbol{r}_m))$$

$$= \bar{b}^2 \sum_n \sum_{m\neq n} \exp(i\boldsymbol{Q}\cdot(\boldsymbol{r}_n - \boldsymbol{r}_m)) + \overline{b^2}$$

$$= \boxed{\bar{b}^2 \sum_n \sum_m \exp(i\boldsymbol{Q}\cdot(\boldsymbol{r}_n - \boldsymbol{r}_m))} + \boxed{(\overline{b^2} - \bar{b}^2)}$$

Coherent Scattering

Incoherent Scattering

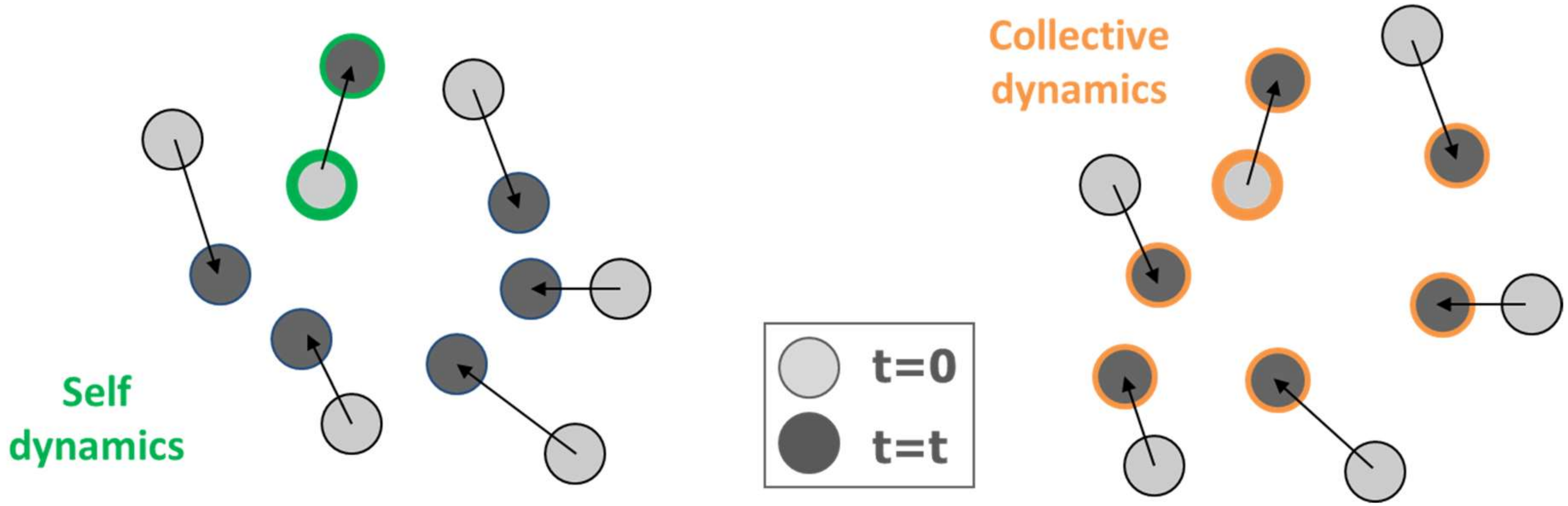
Scattering Function- Correlations

In the experiment we measure the total $S(\boldsymbol{Q},\omega)$ and that each term, coherent and incoherent is weighted by its respective cross-section σ

$$S(\boldsymbol{Q},\omega) = \boxed{S_{\text{inc}}(\boldsymbol{Q},\omega)} + \boxed{S_{\text{coh}}(\boldsymbol{Q},\omega)}$$

$$\boxed{S_{\text{inc}}(\boldsymbol{Q},\omega)} = \frac{1}{2\pi}\int_{-\infty}^{+\infty}\sum_{i}\langle \exp(-i\boldsymbol{Q}\cdot\boldsymbol{R}_i(0))\exp(-i\boldsymbol{Q}\cdot\boldsymbol{R}_i(t))\rangle \exp(-iwt)\,dt$$

$$\boxed{S_{\text{coh}}(\boldsymbol{Q},\omega)} = \frac{1}{2\pi}\int_{-\infty}^{+\infty}\sum_{i,j}\langle \exp(-i\boldsymbol{Q}\cdot\boldsymbol{R}_i(0))\exp(-i\boldsymbol{Q}\cdot\boldsymbol{R}_j(t))\rangle \exp(-iwt)\,dt$$



The Scattering Function, $S(Q,\omega)$

So, just like we can write

$$\frac{d^2\sigma}{d\Omega d\omega} = \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{elastic}} + \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{inelastic}} + \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{quasi-elastic}}$$

we could also write it in terms of:

$$\frac{d^2\sigma}{d\Omega d\omega} = \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{inc}} + \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\text{coh}}$$

$$S(\boldsymbol{Q},\omega) = S_{\text{inc}}(\boldsymbol{Q},\omega) + S_{\text{coh}}(\boldsymbol{Q},\omega)$$

Coherent inelastic scattering

Incoherent inelastic scattering

Incoherent quasi-elastic scattering

Coherent quasi-elastic scattering

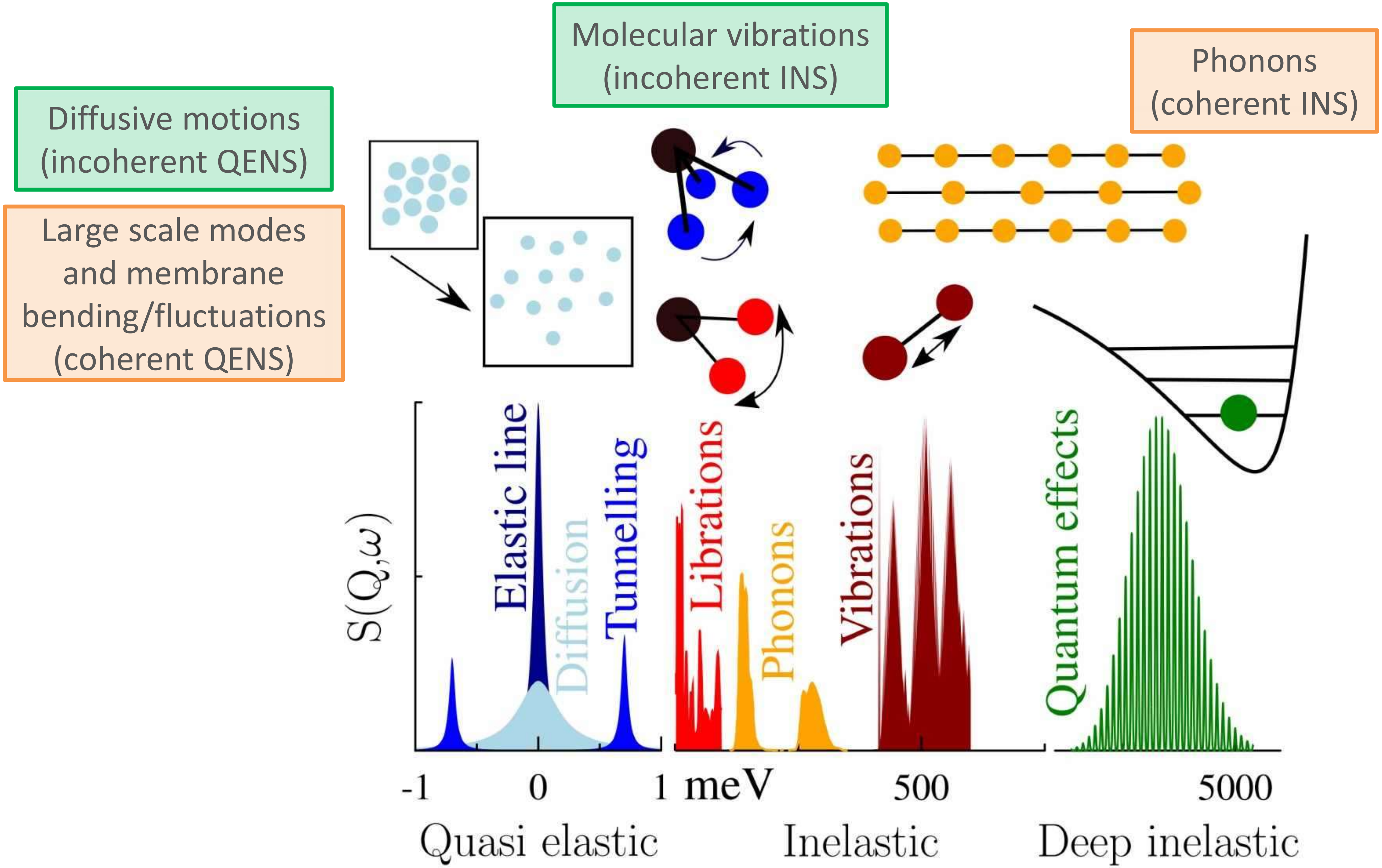
Coherent & Incoherent Scattering

- **Coherent** scattering
  - Describes **correlations between nuclei**
  - Peaks arise due to wave interference
  - Provides **structure** of the sample (elastic)
  - Describes the **collective dynamics** of nuclei (lattice vibrations, bending modes, cooperative relaxations…)

- **Incoherent** scattering
  - Contains no information about structure
  - For most elastic scattering measurements, it manifests itself as an inconvenient flat background but …
  - … it relates to the region of space accessible to the scatterers and yields information about the geometry of the motions.
  - Describes the **dynamics of individual particles**

Motions

Map of dynamical modes



1. Coherent Inelastic Scattering

Remember the double differential cross-section

$$\frac{d^2\sigma}{d\Omega d\omega} \propto \frac{k_f}{k_i}\frac{\sigma}{4\pi}S(\boldsymbol{Q},\omega)$$

For propagating excitations (e.g. lattice vibrations or spin waves), $\boxed{S(\boldsymbol{Q},\omega)_{coh}}$ has peaks at single frequencies $\hbar\omega_{ph}$ and the size of the peaks varies according to:

Lattice vibrations (phonons)

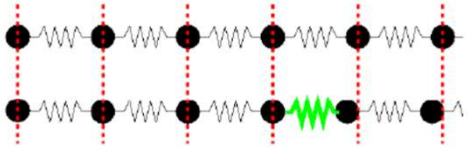
$$S(\boldsymbol{Q},\omega) \propto \exp[-2W(Q,T)] \times |G(\boldsymbol{Q})|^2 \times \left[n\left(\omega_{ph}\right)+1\right] \times 1/\omega_{ph} \times \boldsymbol{Q}^2$$

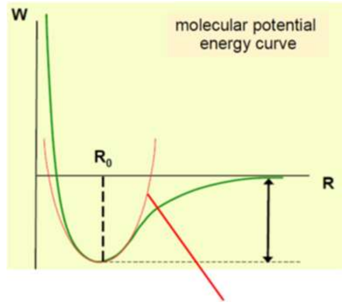
Spin waves (magnons)

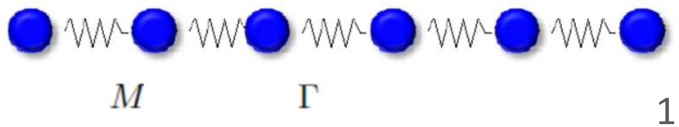
$$S(\boldsymbol{Q},\omega) \propto \exp[-2W(Q,T)] \times \left[n\left(\omega_{mag}\right)+1\right] \times 1/\omega_{mag} \times f^2(Q)$$

1. Coherent Inelastic Scattering

Atoms are considered to be oscillating in harmonic potentials. Dispersion relation gives angular frequency of phonon as function of momentum



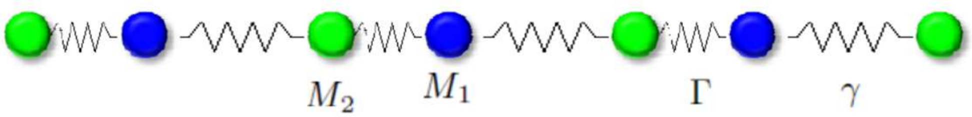


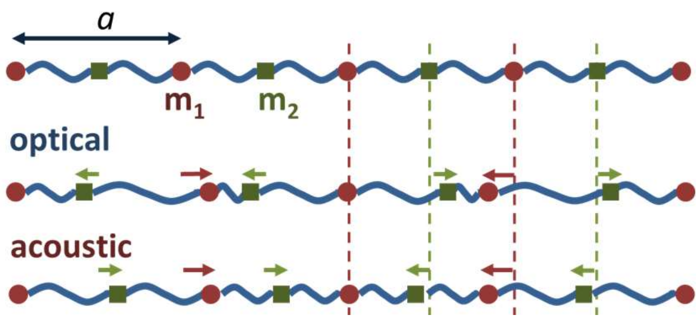


1D chain

$$\omega(k) = 2\sqrt{\frac{\Gamma}{M}}\,sin\frac{ka}{2}$$

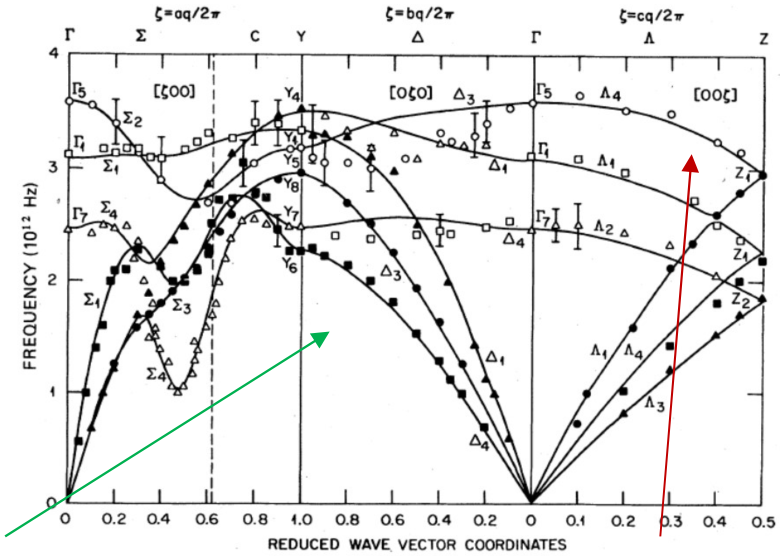
2D chain





Crummett *et al.* Phys Rev B (1979)

E.g. phonons in a-Uranium at T=300K



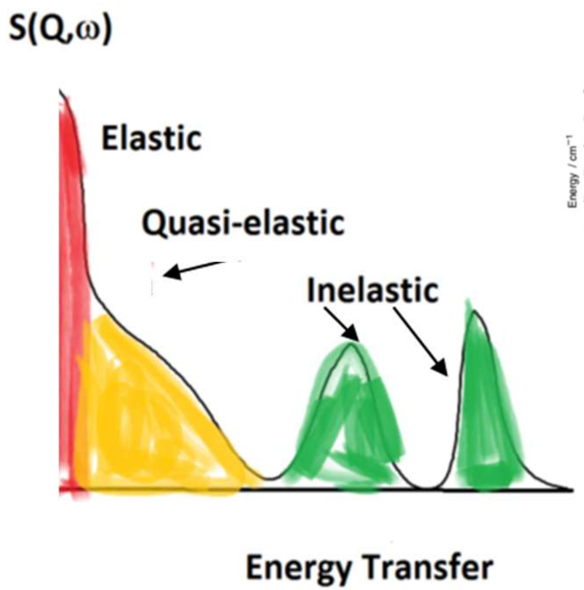
Acoustic phonon
In-phase vibrations

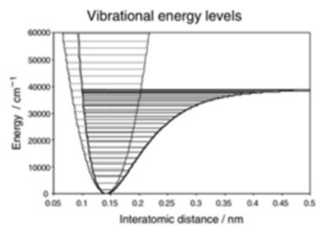
Optical phonon
Out of-phase vibrations

2. Incoherent Inelastic Scattering

If not a lattice but individual molecules, the incoherent inelastic intensity $S(\boldsymbol{Q},\omega_i)_{\text{incoh}}$ can be written for **vibrational modes**.

$$\boxed{S_{\text{inc}}(Q,n\omega_i)} \propto \frac{\left(Q^2U_i^2\right)^n}{n!}\exp(-Q^2U_{\text{total}}^2)$$





Vibrations are quantised thus appear as INS feature at given ħω≠0 values.

Incoherent scattering …

… describes **vibrational modes**

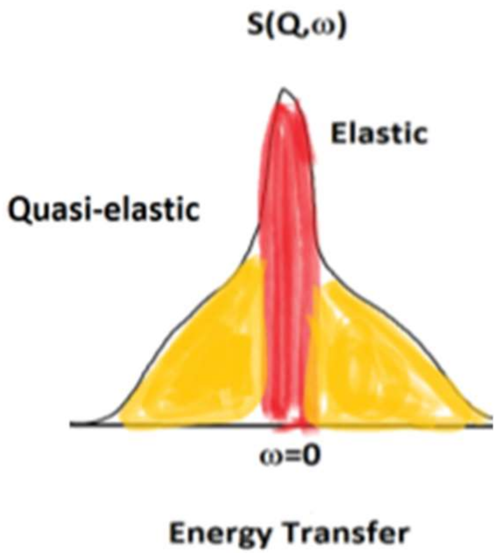
… stretching, rocking, bending, twisting…

… main association of vibrational spectroscopy technique (cm^{-1}) comparable to Raman or IR spectroscopy

… relies on large incoh x-section of H

… DWF minimised by measuring at low temperatures.

3. Incoherent Quasi-elastic scattering



We measure **the self correlation function**, ie. how particles move as a function of time. This corresponds to the incoherent signal which we can write as:

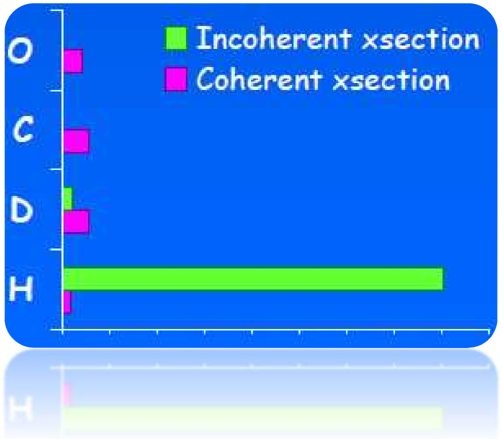
$$S_{\mathrm{inc}}(\boldsymbol{Q},\omega) = S_{\mathrm{vib}}(\boldsymbol{Q},\omega) \otimes S_{\mathrm{rot}}(\boldsymbol{Q},\omega) \otimes S_{\mathrm{trans}}(\boldsymbol{Q},\omega)$$

$$I_{\mathrm{self}}(\boldsymbol{Q},t) = I_{\mathrm{vib}}(\boldsymbol{Q},t) \times I_{\mathrm{rot}}(\boldsymbol{Q},t) \times I_{\mathrm{trans}}(\boldsymbol{Q},t)$$

It is a **convolution of components** which for simplicity are assumed **independent** motions. Note that in the time domain we multiply the terms (easier!)

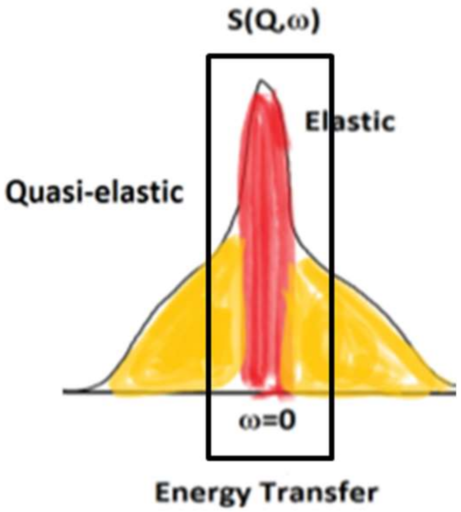
Can't forget the instrumental resolution:

$$S'(Q,\omega) = S(Q,\omega) \otimes R(Q,\omega)$$



QENS relies on the large incoherent xsection of hydrogen.

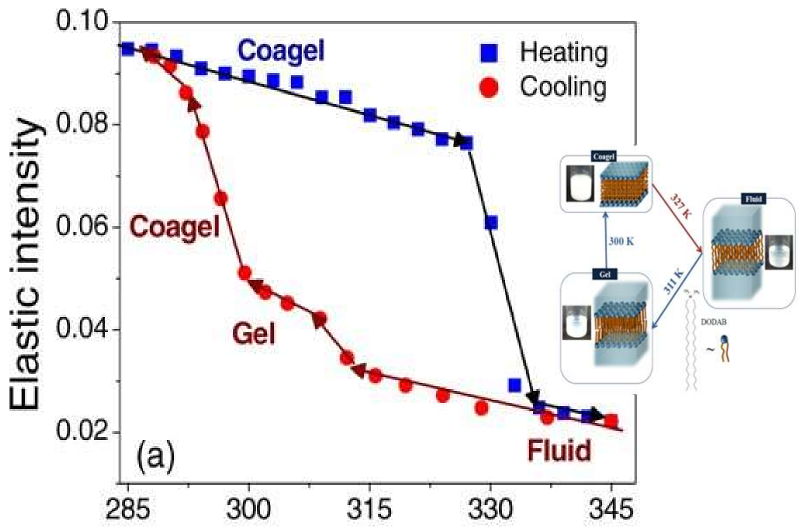
Fixed Energy Window Scans



Case of ***S(Q,*** $\omega \approx$ **0):** Elastic Fixed Window Scans

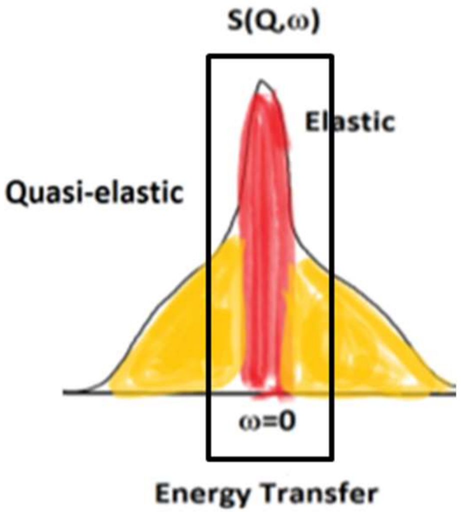
Measure the elastic intensity as a function of temperature (resembles a DSC scan).

Good for locating **transitions**, choosing at what temperatures to perform QENS measurements (**dynamics enter the spectrometer window),** and comparative studies (**parametric**).



Dubey, VGS et al, Scientific Reports (2018)

Fixed Energy Window Scans



Case of ***S(Q,*** $\omega \approx$ **0):** Elastic Fixed Window Scans

M … erature

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p … **spectrometer**
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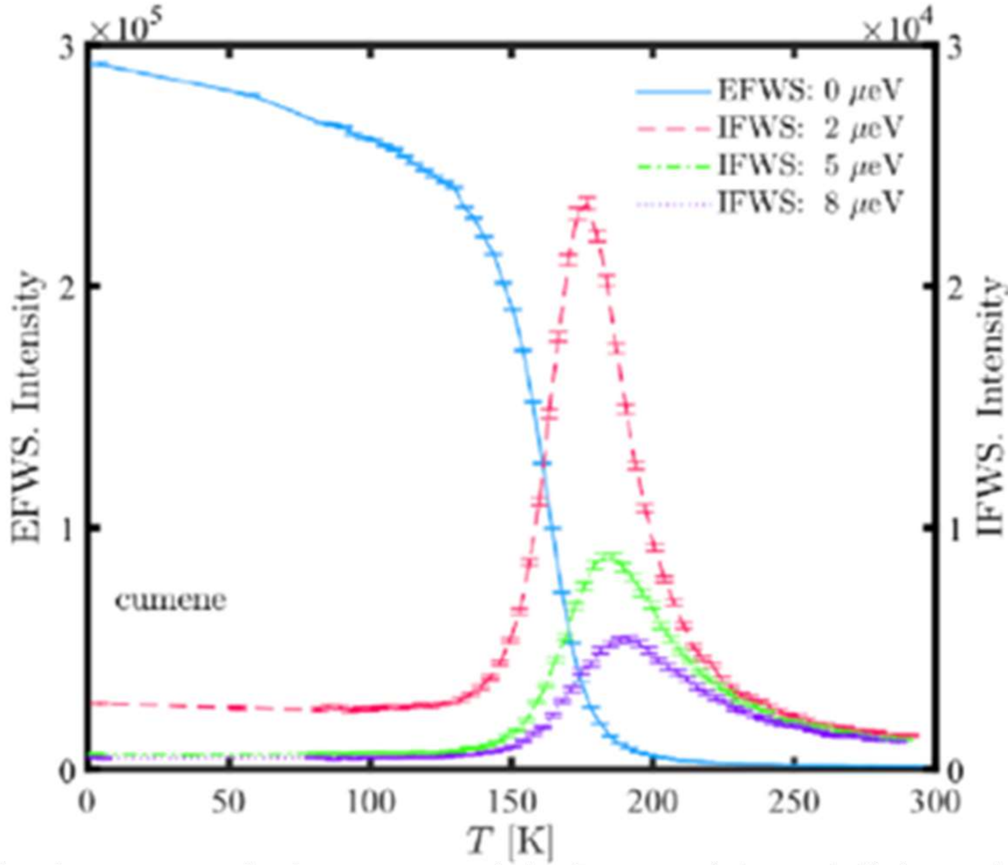
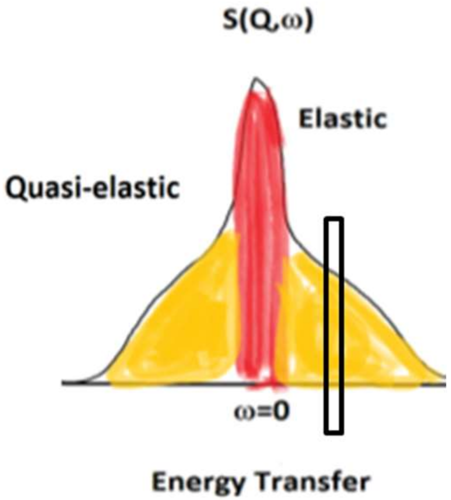


Scans



If tr … imescale,
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Peak shape and changes with Q provide additional information

Hansen et al, PRB, 2017

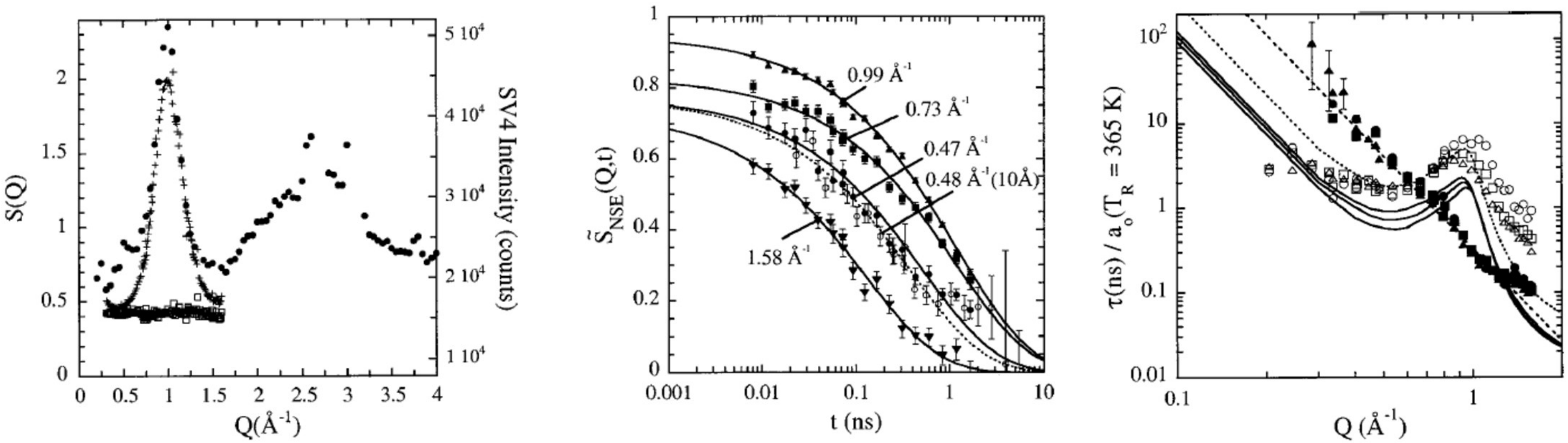
Different offsets correspond to different timescales

4. Coherent Quasi-elastic scattering

These are 'collective' dynamics, they come from **coherent part of the correlation function** and depend on the **structure factor**, ***S*(*Q*)**, ie. how atoms are distributed in space.

$$I_{\text{coll}}(Q,t) \approx I_{\text{self}}(Q,t)\left(\frac{Q}{\sqrt{S(Q)}},t\right)$$

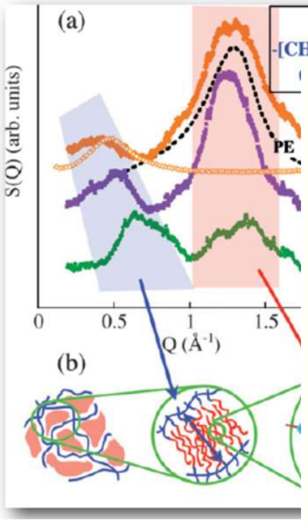
$$\tau_{\text{coll}}(Q,T) \approx a(T)\tau_{\text{self}}(Q,T)S(Q)^{1/\beta}$$

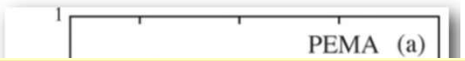


Molecular relaxations in polyisobutylene

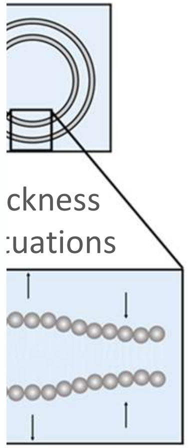
Farago et al, PRE, (2002)

2. Collective nuclear (or spin) dynamics





, surfactants)



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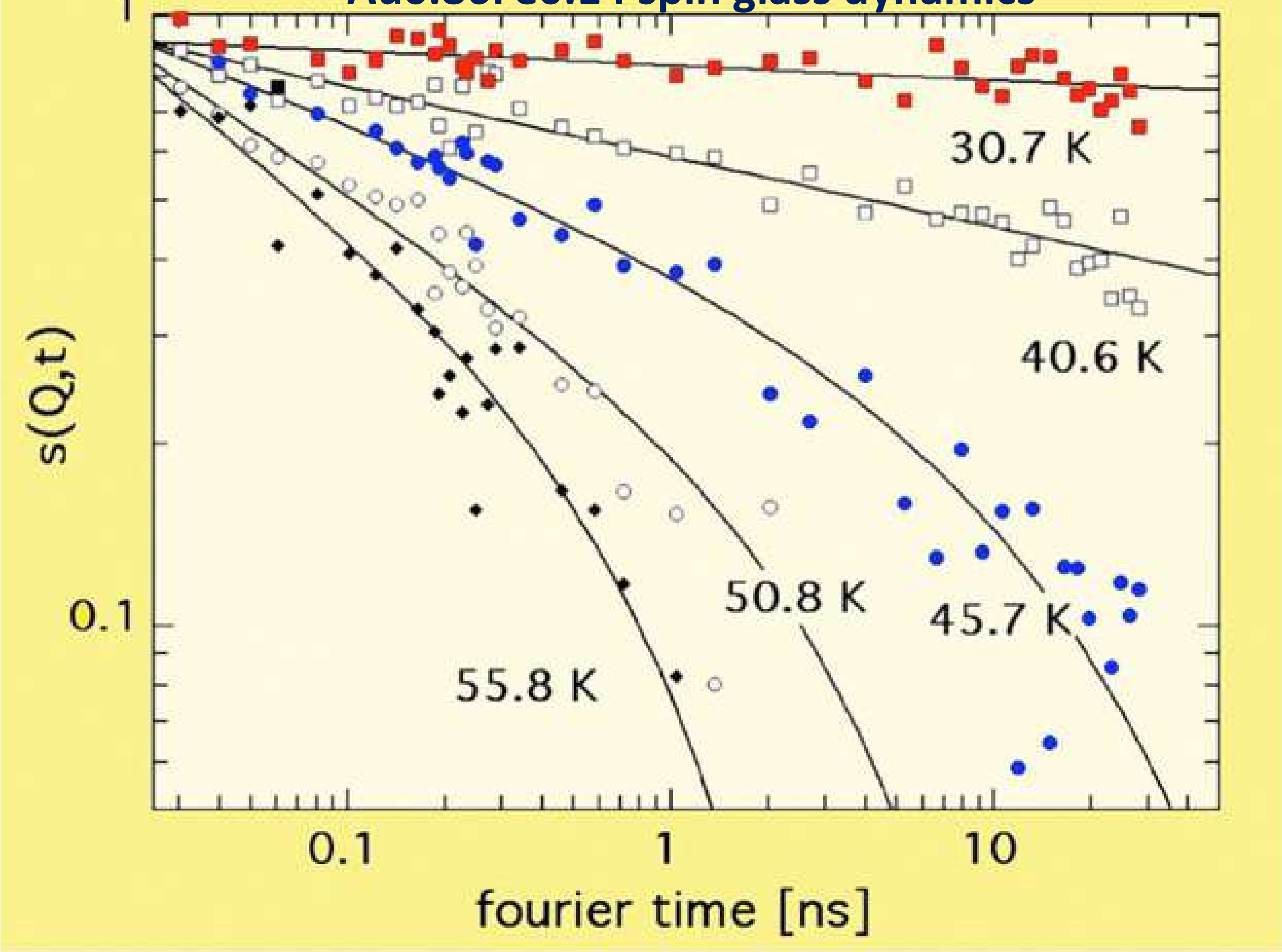
Arbe et al, Soft M

Au0.86Fe0.14 spin glass dynamics



Different Q
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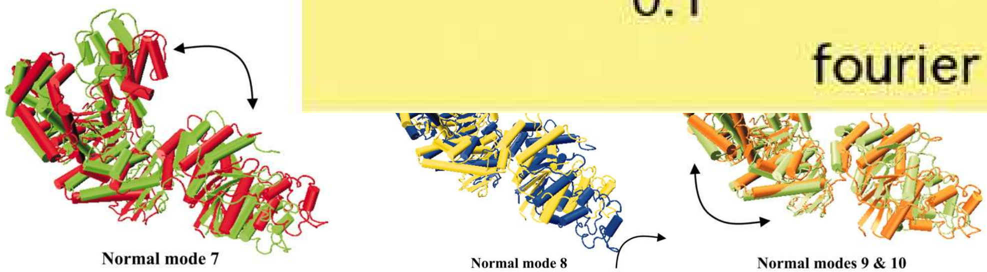
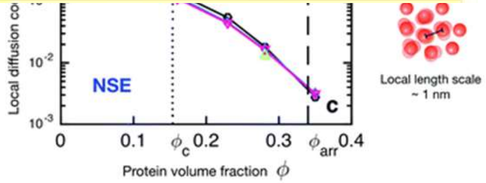
Assignment

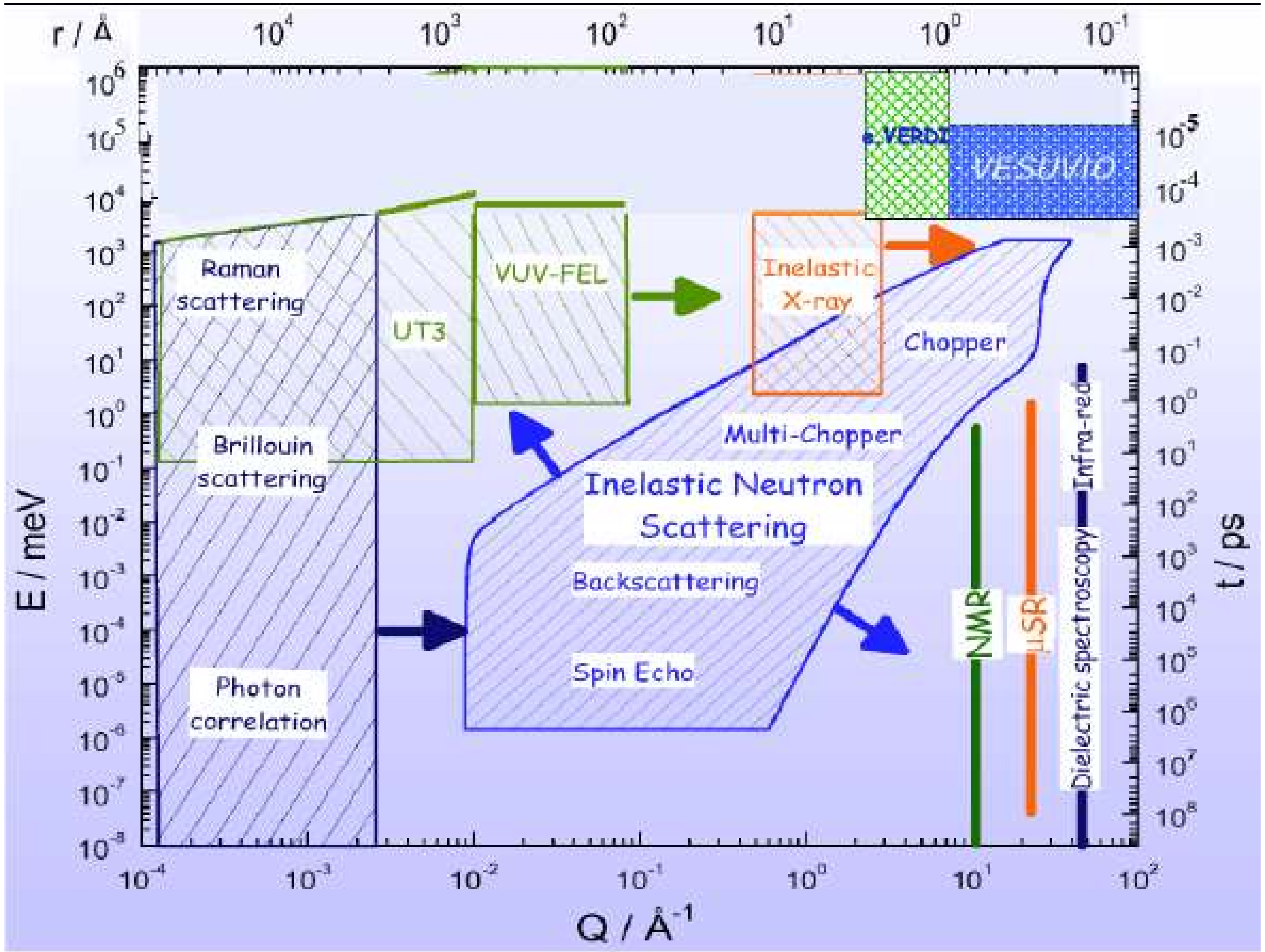




Bu et al, PNAS (2005)

Stradner and Schurtenberger, Soft Matter (2019)

Map of dynamical modes

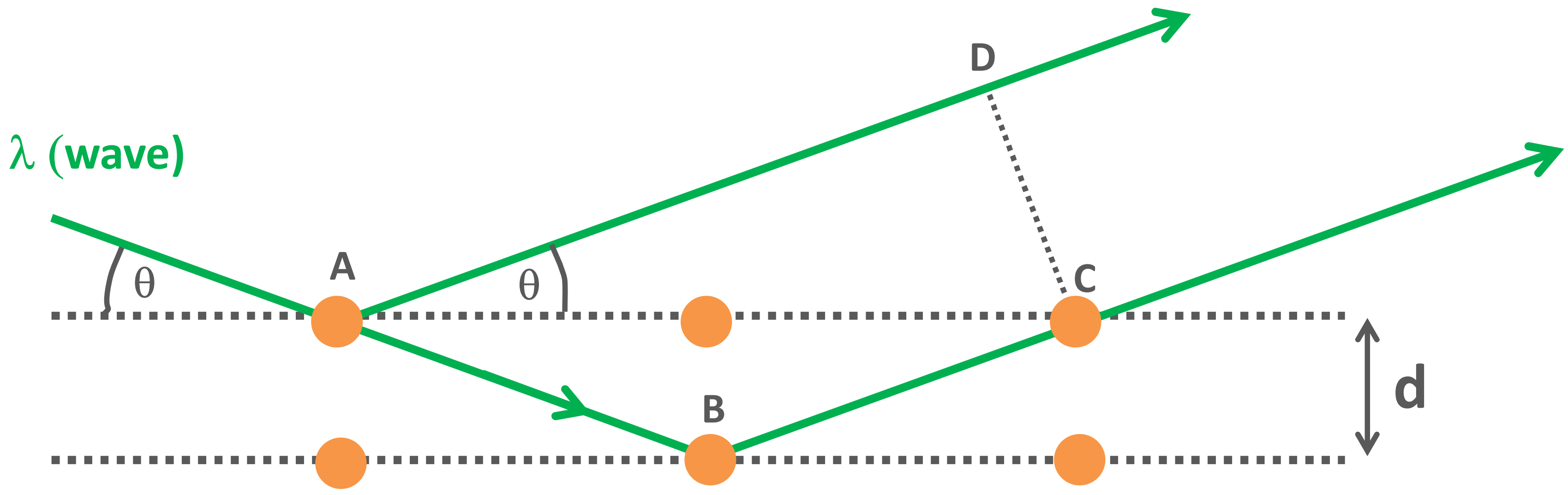


Inelastic Scattering Measurements (Instruments)

Need to measure k_i and k_f, how?

1) Bragg diffraction
2) Time-of-flight
3) Larmor precession

Bragg's Law



For **constructive interference, path difference** between the reflected wave and that traversing through, must equal $\boldsymbol{n\lambda}$

$$\text{Path difference} = (AB + BC) - (AD) = n\lambda$$

$$\sin\theta = \frac{d}{AB} = \frac{d}{BC} \qquad \tan\theta = \frac{2d}{AC} \qquad \cos\theta = \frac{AD}{AC}$$

$$n\lambda = \left(\frac{d}{\sin\theta} + \frac{d}{\sin\theta}\right) - \frac{2d}{\tan\theta}\cos\theta = \frac{2d}{\sin\theta}\left(1 - \cos^2\theta\right)$$

$$\boldsymbol{n\lambda = 2d\sin\theta}$$

Time-of-flight

De Broglie relations

wave and particle

$$p = \frac{h}{\lambda} \quad \text{and} \quad p = mv$$

$$\lambda[\text{Å}] = \frac{h}{mv} = \frac{3.956}{v\left[\frac{m}{ms}\right]}$$

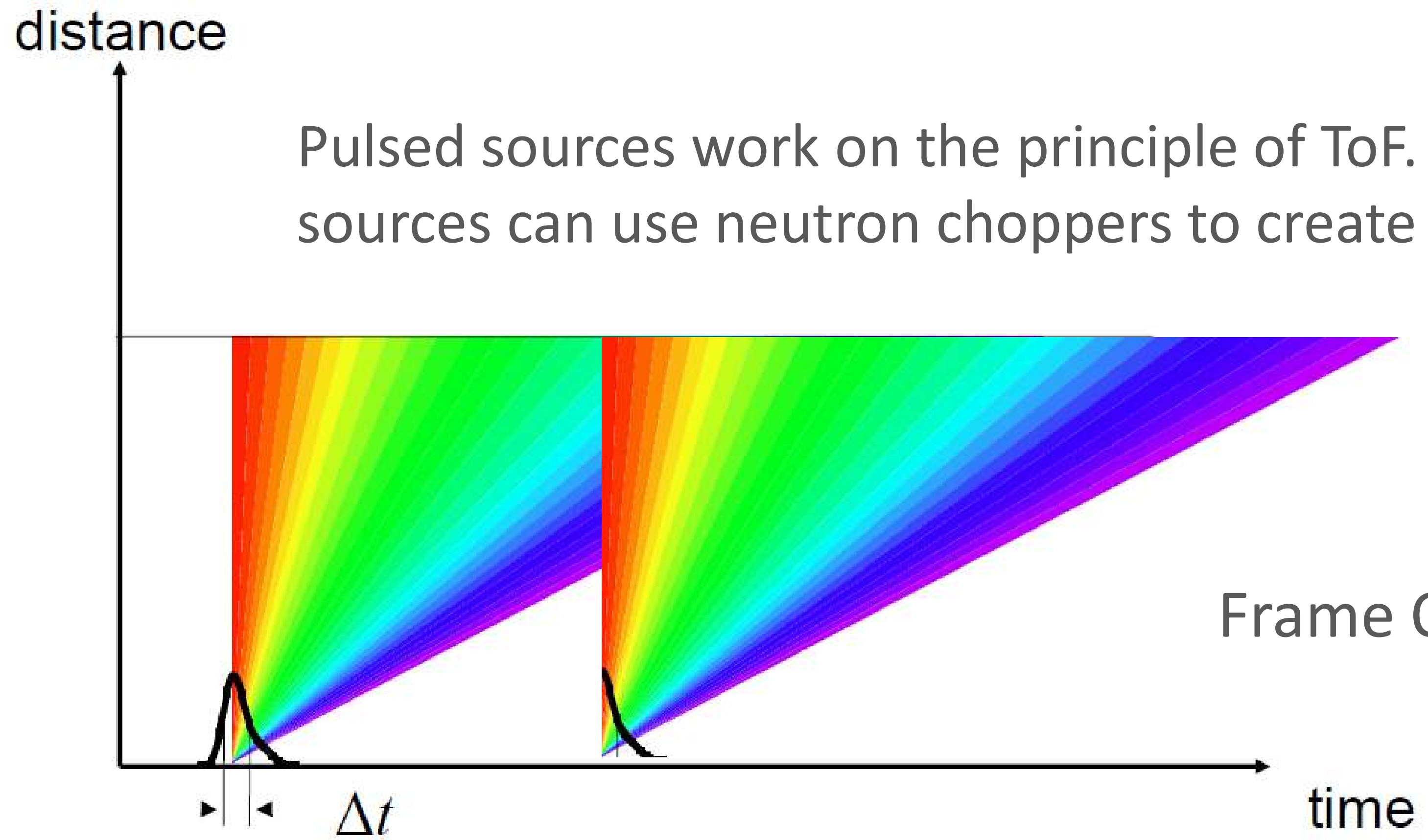
$$\hbar = \frac{h}{2\pi} = \frac{6.6\ \times 10^{-34}\text{J.s}}{2\pi}$$

$$m = 1.67 \times 10^{-27}\text{kg}$$

$$t[\text{ms}] = L[\text{m}] \times \lambda[\text{Å}]/3.956$$

$$t[\mu\text{s}] = L[\text{m}] \times \lambda[\text{Å}] \times 252.8$$

Pulsed sources work on the principle of ToF. Reactor sources can use neutron choppers to create a pulsed beam.



Basic needs for measuring neutron scattering probability

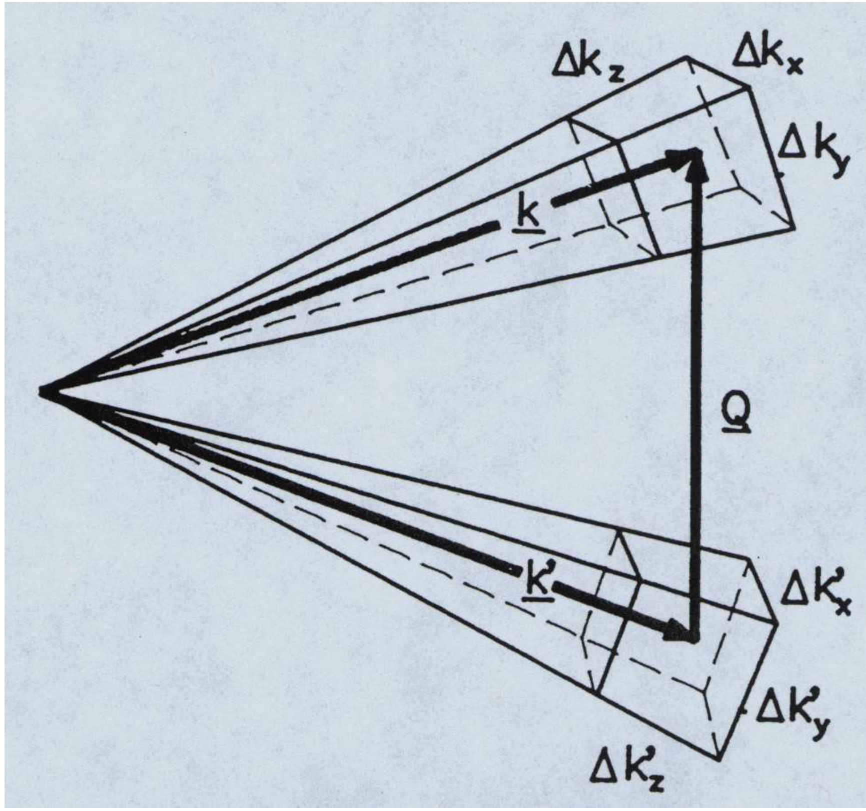
- **Source** of neutrons with wavelengths in the right range; need to transport neutrons to the experimental area.
- Need to determine the incident and neutron directions - **angular resolution**
- Need to determine the incident and scattered neutron wavelength (energy) - **wavelength (energy) resolution**
    - Bragg diffraction
    - Time of flight
- **Detectors** to measure counts at different scattering angles
- To cover desired range, need measurements at different wavelengths and/or many angles - **Q-range, E-range**
- Need to **filter** out unwanted neutrons
- Need to **polarise** beams

Instrumental Resolution

Uncertainties in the neutron wavelength and direction of travel => Q and E can only be defined with a certain precision

Total signal in a scattering experiment ∝ phase space volume

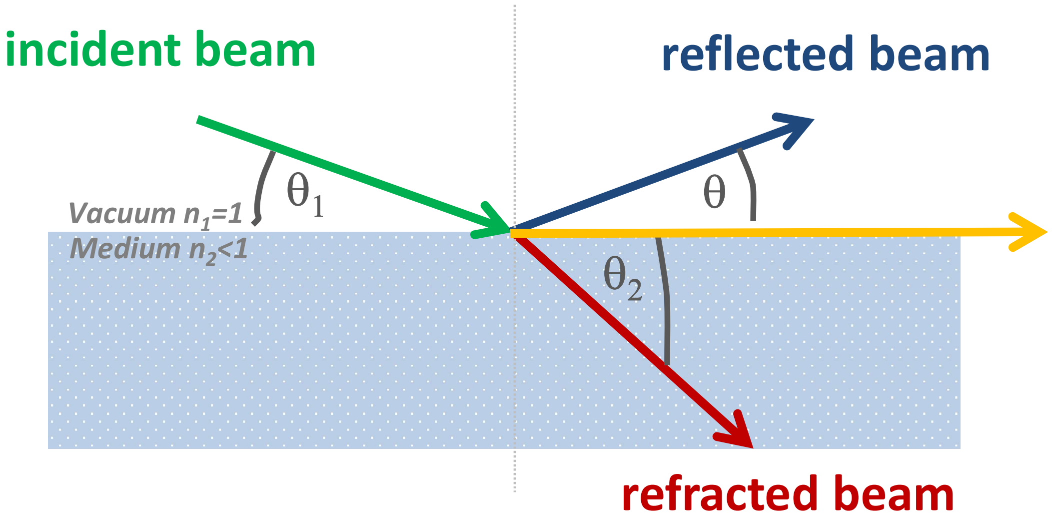
The better the resolution volume , the lower the count rate. It's always a trade-off between intensity and resolution.



Basic needs general (cont.)

- Large sample area, to achieve good count rates, since neutron sources are weak
- Need large sample-detector distance otherwise additional collimation is necessary for good angular resolution with large samples
- Need detectors all around sample to collect maximum number of scattered neutrons
- Need large amounts of shielding to minimise background in detectors (and personnel safety!). Also achieved with large source-sample distance
- *For ToF* – need long moderator-sample distance for good time resolution as dictated by pulse width plus long sample-detector distance.

Snell's Law



$$n_1 \cos\theta_1 = n_2 \cos\theta_2$$

$$n_2 = \frac{\cos\theta_1}{\cos\theta_2}$$

$$n_2 = \cos\theta_c \text{ if } \theta_2\text{=0}$$

Neutron guide tubes exploit that *n* for neutrons is < 1 in more materials, so total external reflection can occur at the boundary between material and vacuum.

θ_c is the critical angle of total reflection

N is number of atoms per cm3

b is bound coherent cross-section

$$n_2 = 1 - \lambda^2 \frac{Nb}{\pi}$$

$$\theta_c = \lambda\sqrt{Nb/\pi}$$

Neutron Guide Characteristics

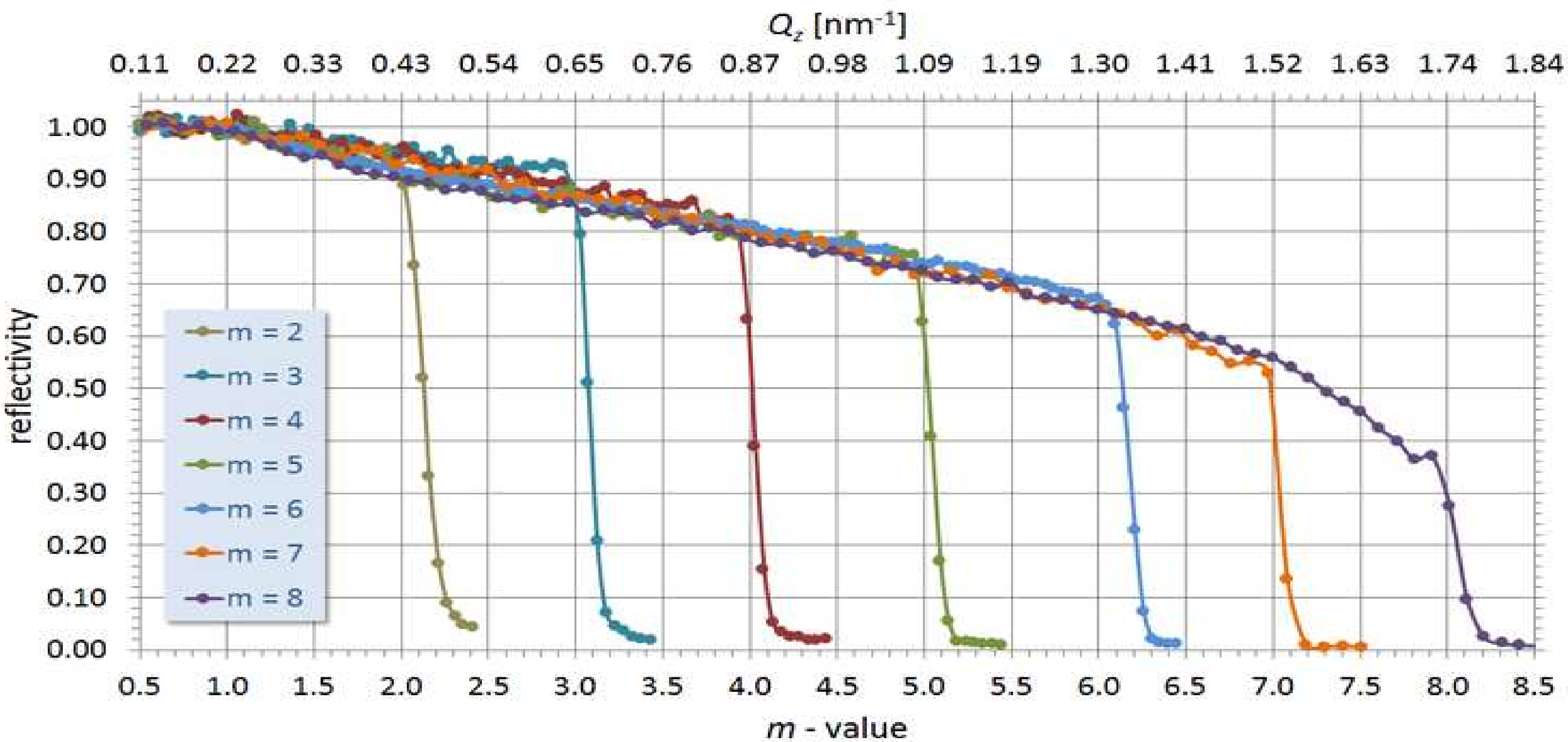
Natural Ni is most commonly used, with high θ_c of 1.7mrad @ λ=1Å. For Ni,

$$\theta_c(\mathrm{Ni}) = \lambda[\text{Å}] \times 0.1°;\ Q_c = 0.0218\text{Å}^{-1}$$

Reflectance requires extremely flat surfaces, so coatings on glass are typically used.

Better technology involves evaporating alternate layers of ^{58}Ni and Ti and of varying thicknesses.

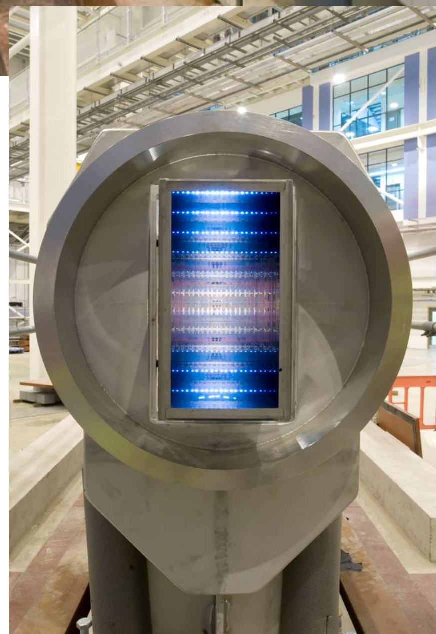
$$\theta_c(\mathrm{SM}) = m \times \lambda[\text{Å}] \times 0.1°$$



Courtesy of J. Stahn, PSI and Swiss Neutronics

Neutron Guides

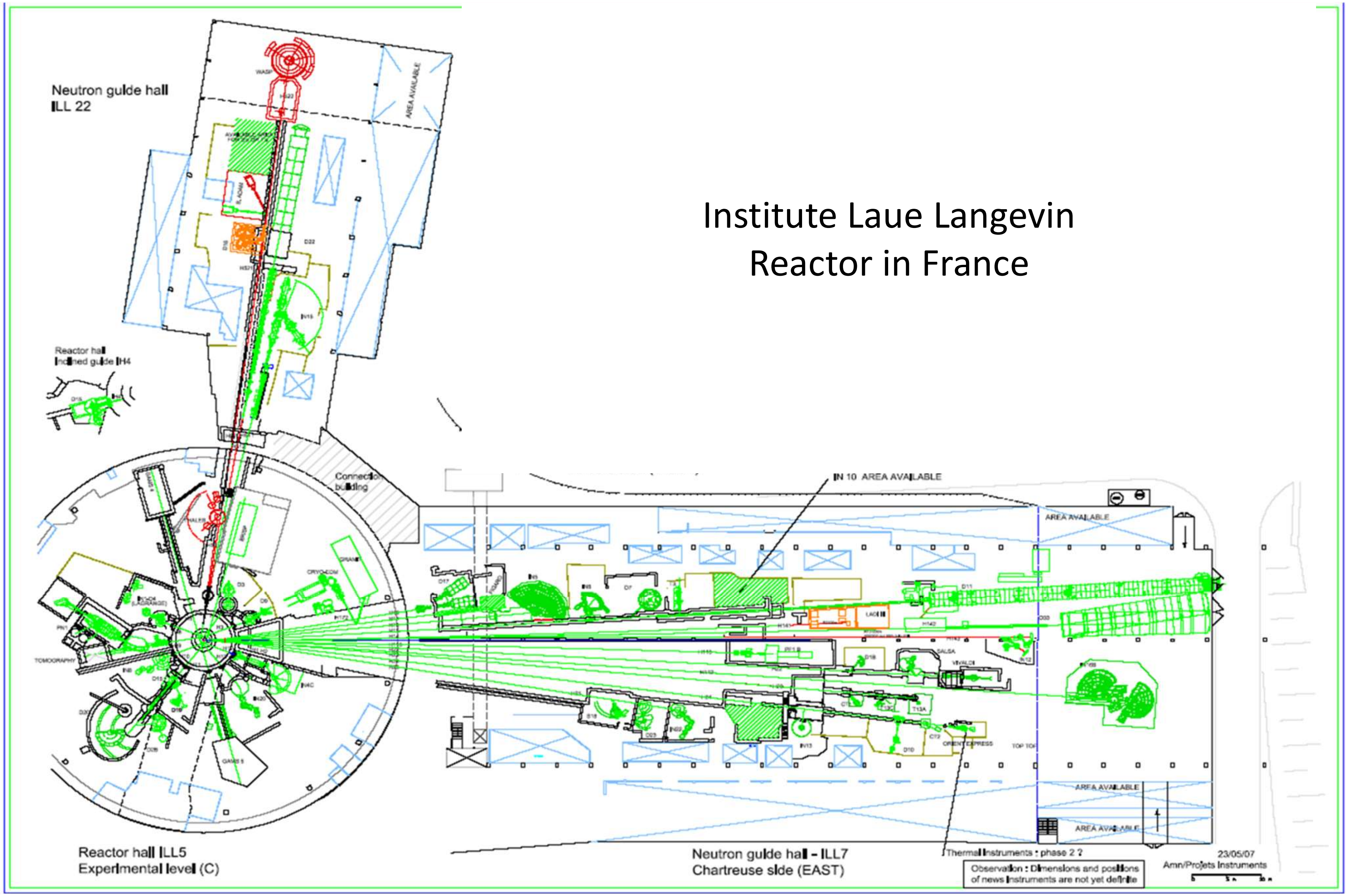
Swiss neutronics guides for NIST, USA



WISH guide on TS2 @ ISIS

Neutron transport up to ~ 100m in existing sources -> pushing to ~ 200m

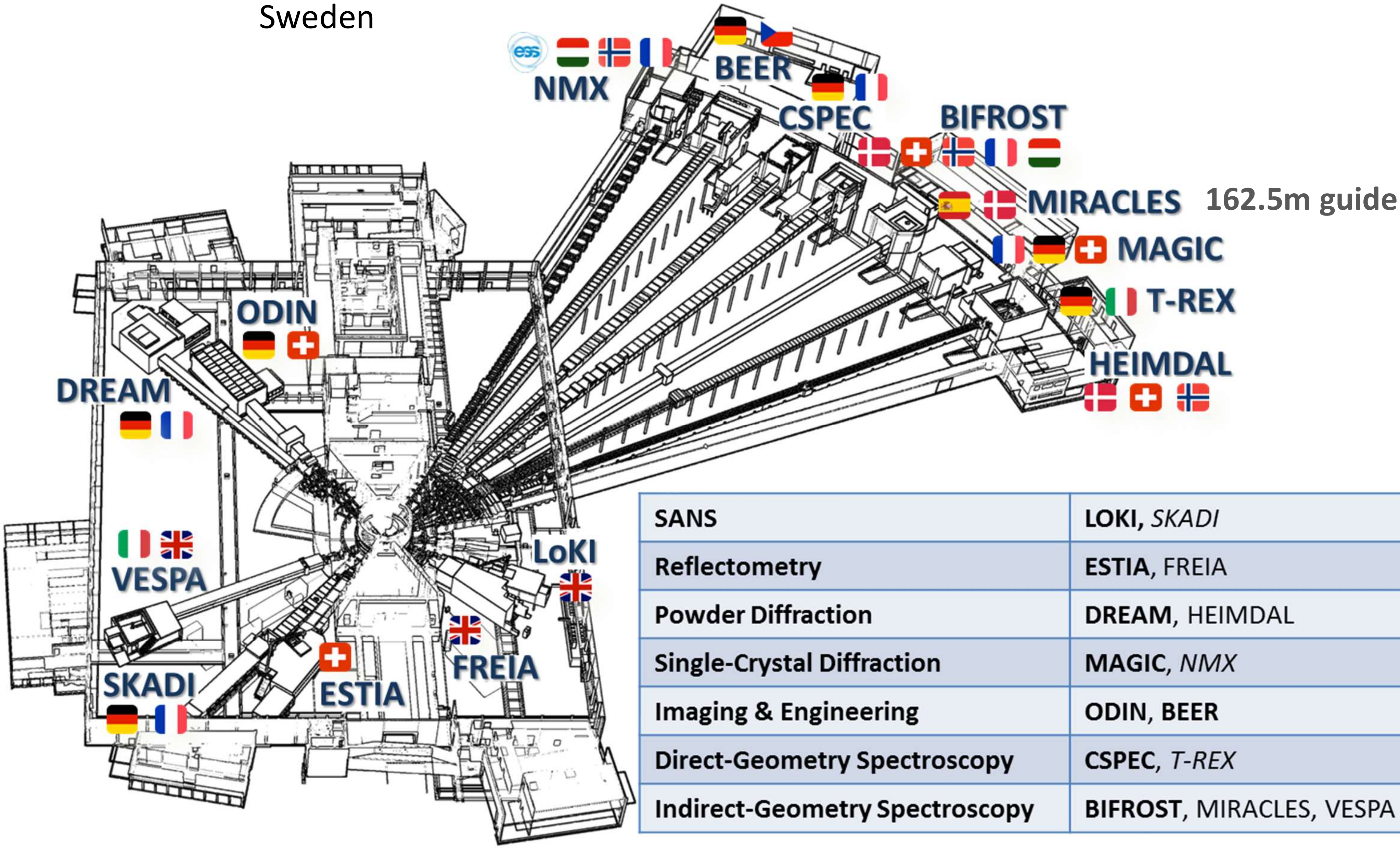
Distribution of neutrons by guides



Institute Laue Langevin
Reactor in France

Distribution of neutrons by guides

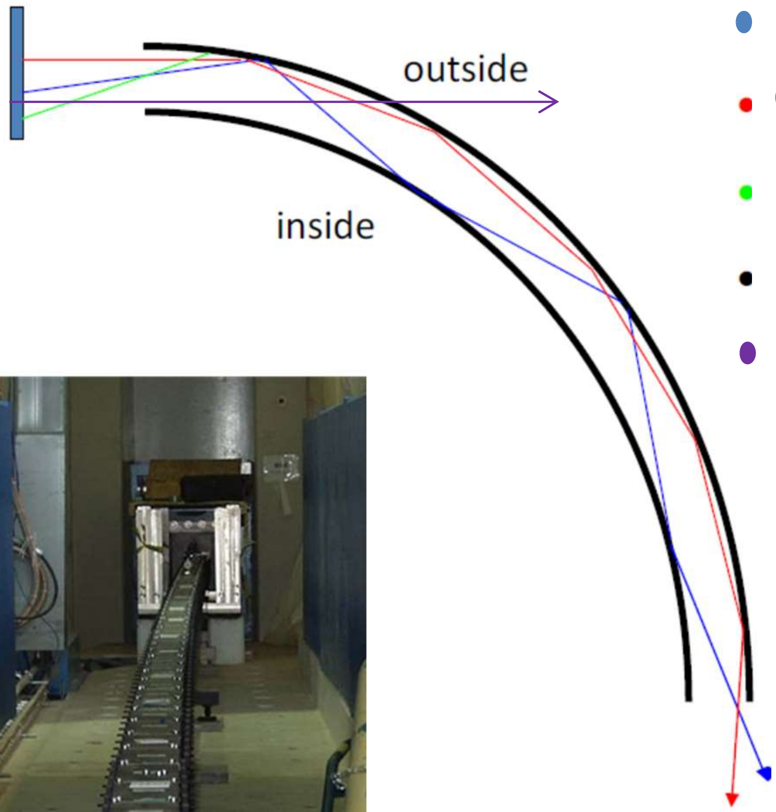
European Spallation Source
Sweden



SANS	**LOKI**, *SKADI*
Reflectometry	**ESTIA**, FREIA
Powder Diffraction	**DREAM**, HEIMDAL
Single-Crystal Diffraction	**MAGIC**, *NMX*
Imaging & Engineering	**ODIN**, **BEER**
Direct-Geometry Spectroscopy	**CSPEC**, *T-REX*
Indirect-Geometry Spectroscopy	**BIFROST**, MIRACLES, VESPA

Guide Types

Curved Guides

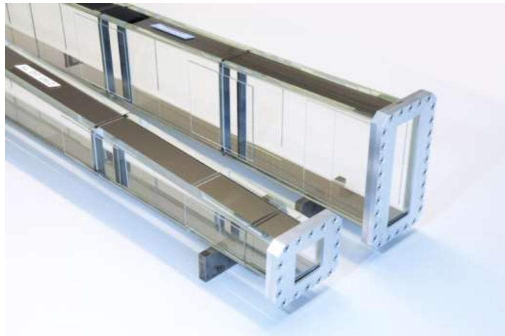


- Reflecting from both sides
- Garland reflections
- Exceed critical angle
- Fewer neutrons along inside face
- Fast neutrons and gamma rays

BASIS 82m
guide @ SNS

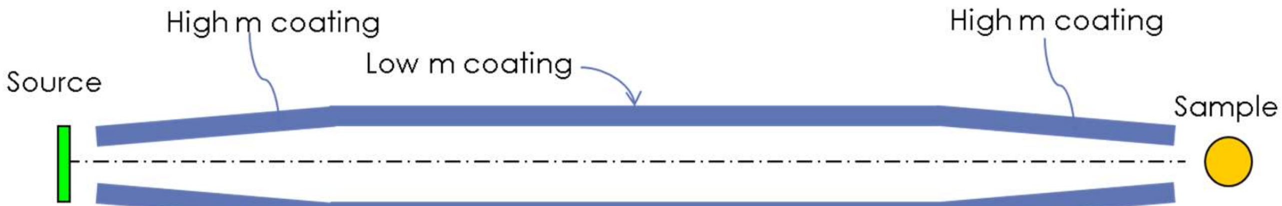
Straight Guide

TOSCA @ ISIS
beam line



D17 parabolic
focusing guide @ ILL

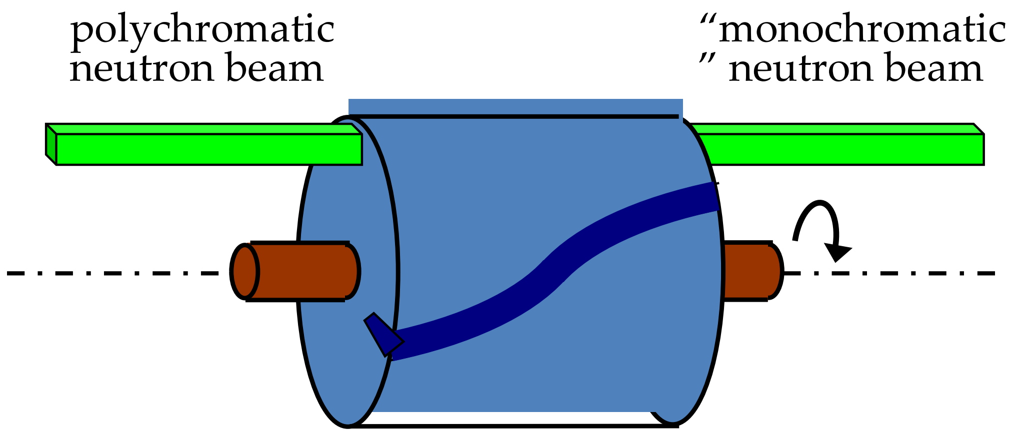
Ballistic Guide



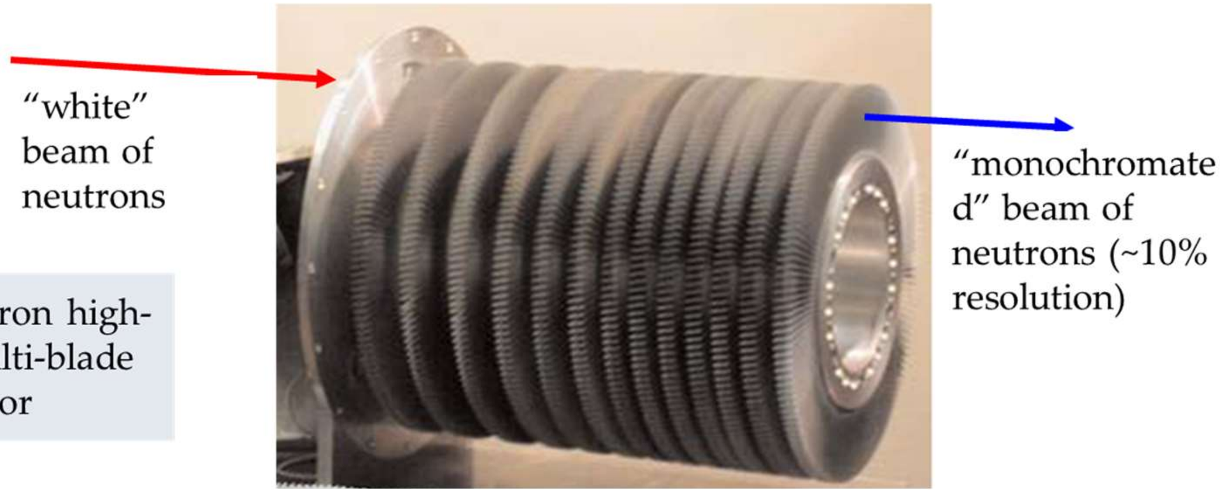
Energy/Wavelength Selection

Velocity Selector

If you can live with poor monochormatization ($\delta\lambda/\lambda \sim 10\%$) as it is commonly used in reactor small angle neutron scattering machines (SANS for measurements of large objects), you use a **velocity selector** (essentially a **rotating collimator**).



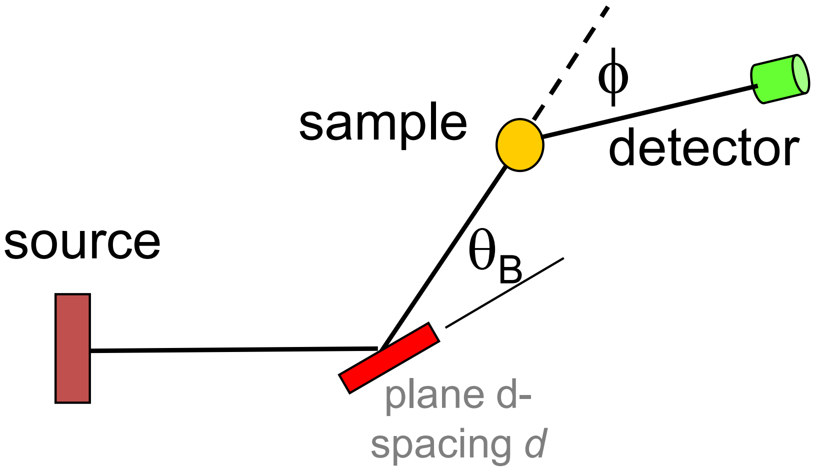
$$\frac{\Delta v}{v} = \frac{\Delta \lambda}{\lambda}$$

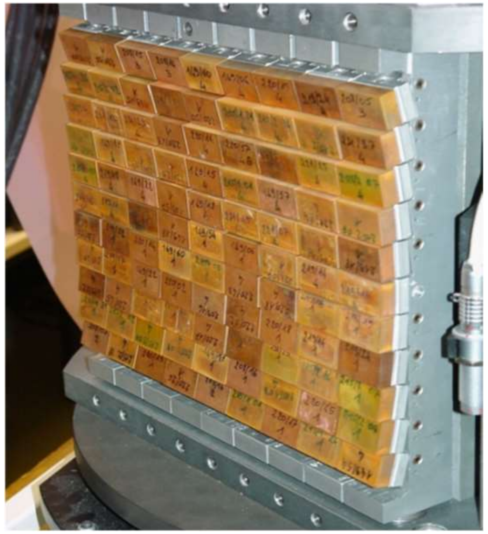


Lesker/Mirrotron high-resolution multi-blade velocity selector

Crystal Monochromators

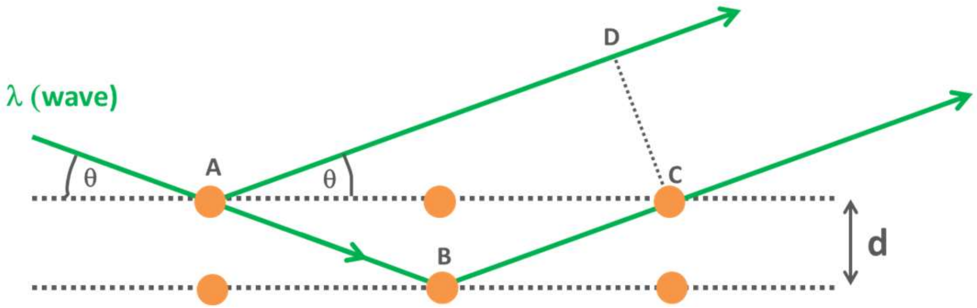
In a reactor, this is done by typically using **crystal monochromators** or ToF concept. The neutron wavelength (E_i) is selected through Bragg reflection. (E_i or E_f)





Copper 200

Silicon 111



	d-spacing
Graphite 002	3.355 Å
Copper 200	1.807 Å
Silicon 111	3.135 Å

Crystal Analyzers

Note that similar concept can be used to choose the final neutron wavelength (E_f) - analysers

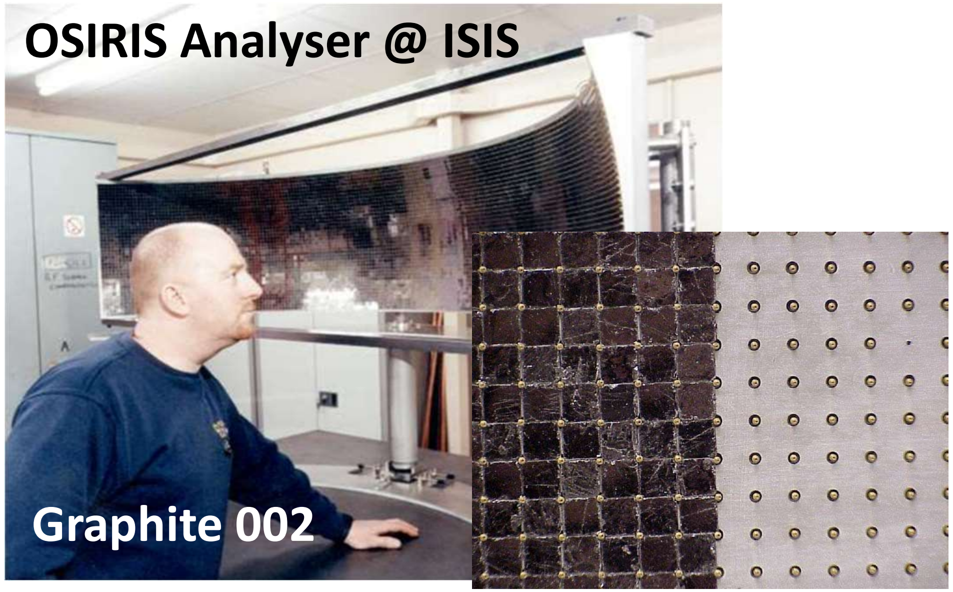
$$n\lambda = 2d\sin\theta$$

$$\frac{1}{2}\frac{\Delta E}{E} = \frac{\Delta\lambda}{\lambda} \sim \frac{\delta d}{d} + \cot(\theta)\delta\theta$$

Tight $\Delta\lambda/\lambda$ ~ 1%

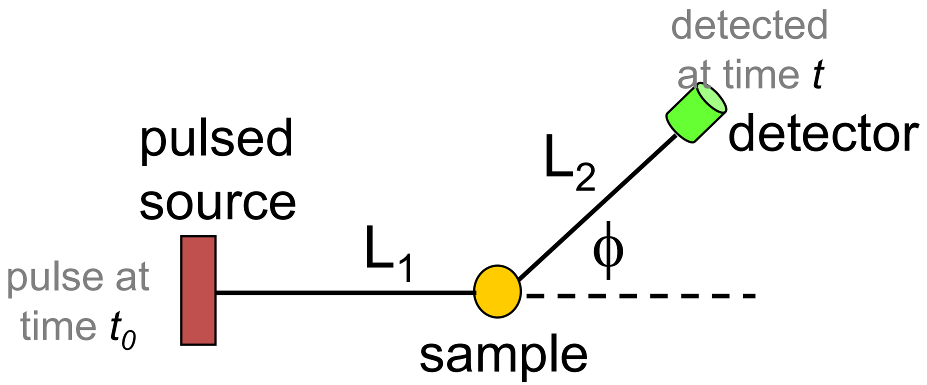
Poorer reflectivity

Higher order contamination



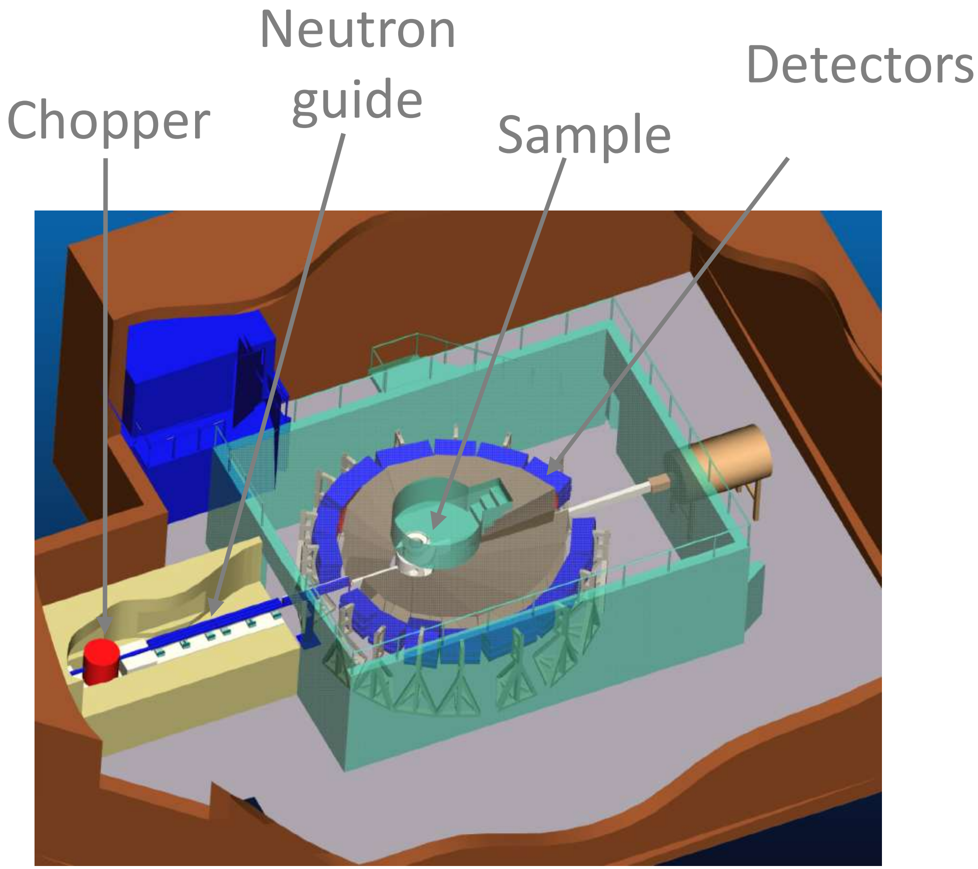
Pulses and Time-of-Flight

In a reactor, energy selection (E_i) can alternatively be achieved by creating pulses (using choppers) and using the pulsed source concept, **ToF concept**.



$$\lambda[\text{Å}] = \frac{t[\text{ms}]\times 3.956}{L[\text{m}]} = \frac{(t-t_0)\times 3.956}{(L_1+L_2)}$$

$$\delta\lambda \sim \delta t_0, \delta t, \delta L$$

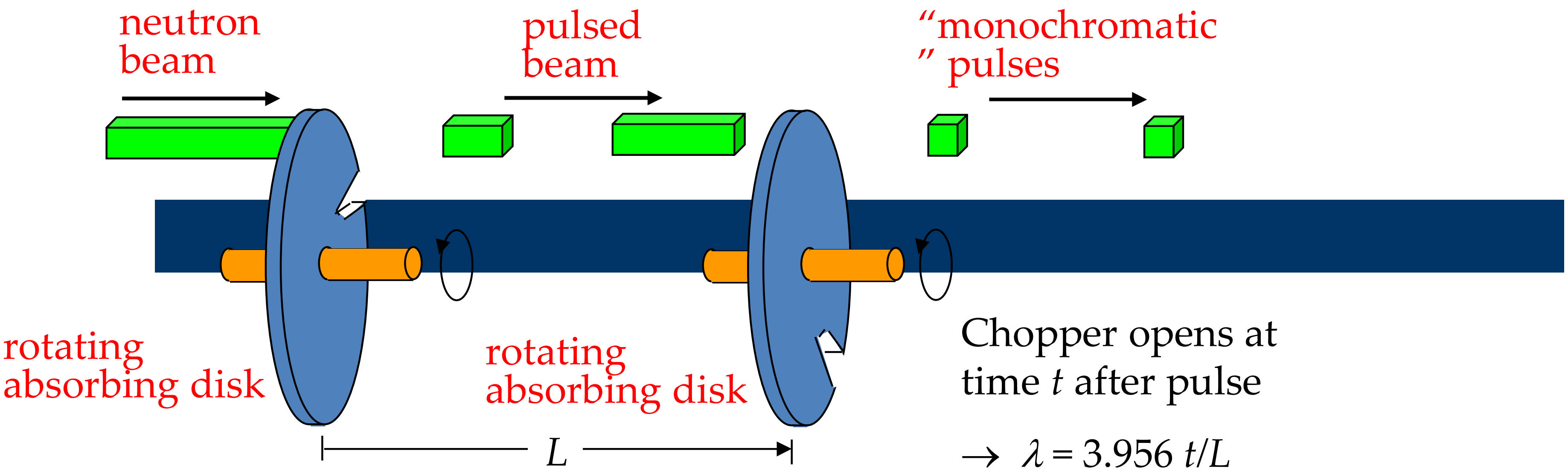


Powgen3 @ SNS

Disk Choppers

Allow a certain wavelength (or band) through, or multiple, i.e. as a **monochromator**.

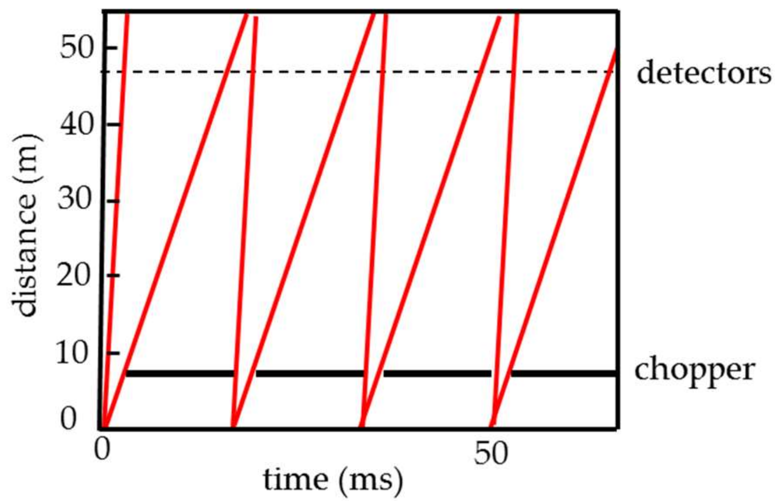
Alternatively, removes unwanted frames in pulsed sources, as a **bandwidth limiter.**



Chopper opens at time t after pulse

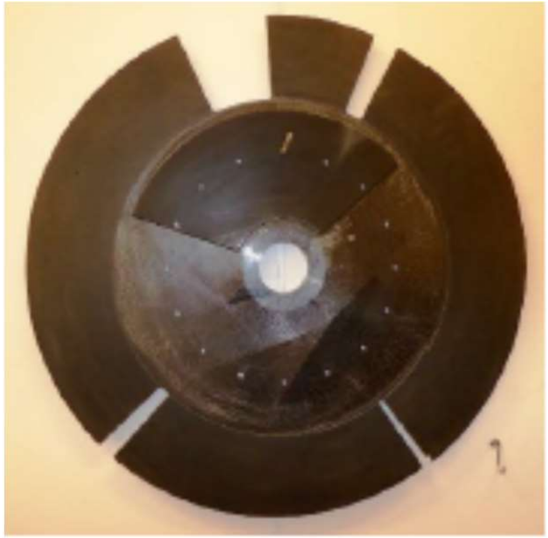
$\rightarrow \lambda = 3.956\ t/L$

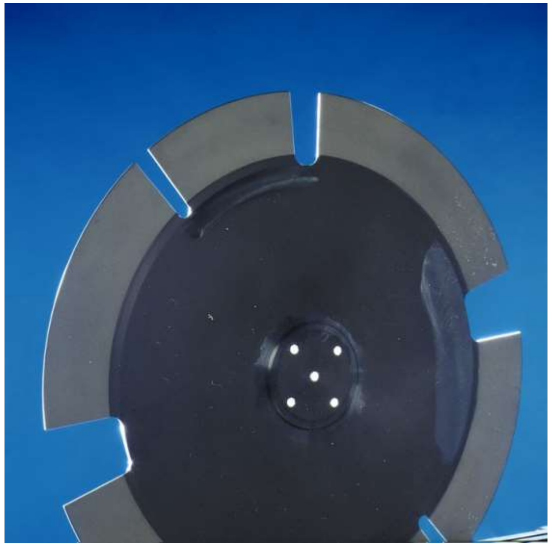
If the downstream chopper has a wider opening and/or the chopper runs slower, then it lets through a broad but limited band. This is used to prevent frame overlap.

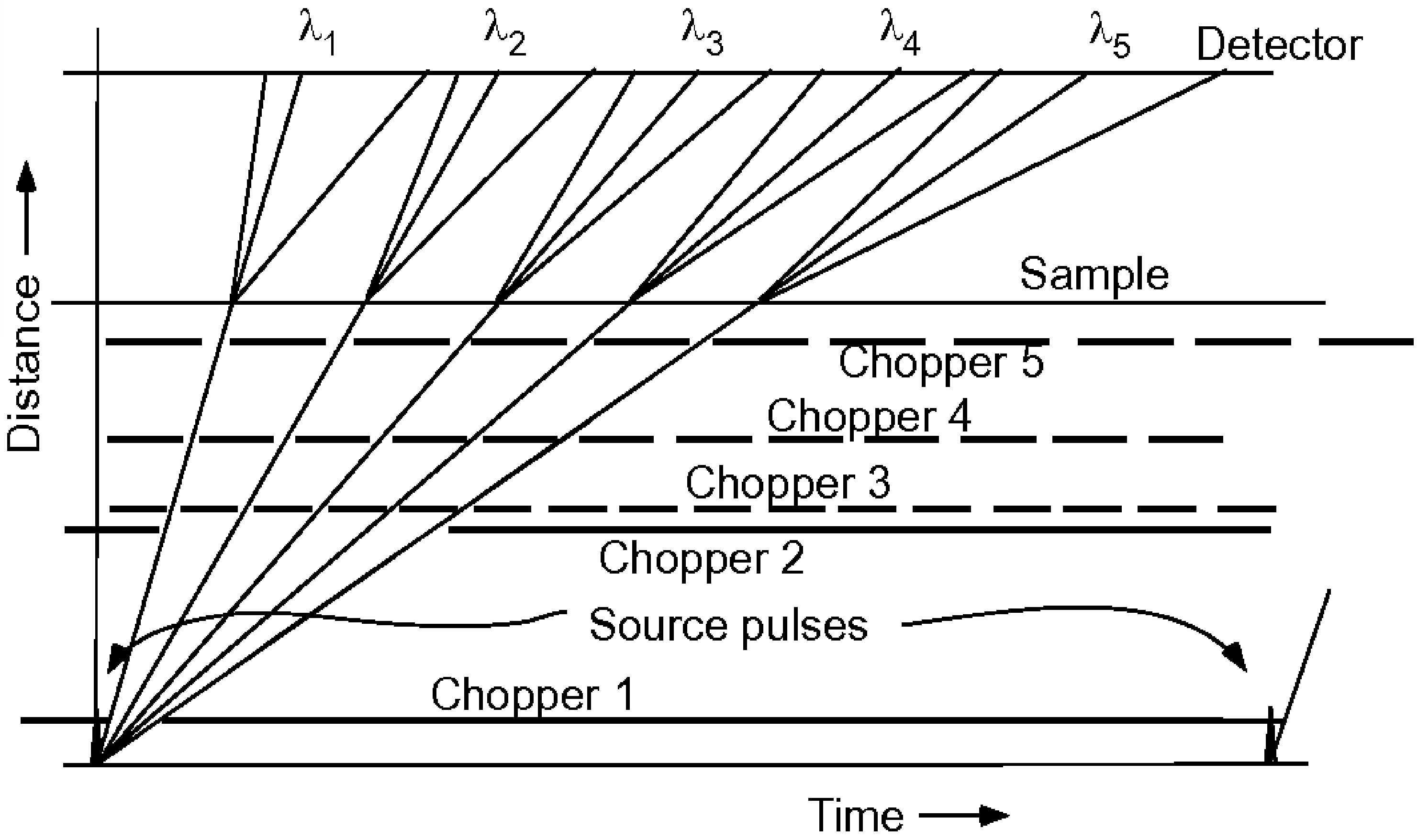


Disk Choppers

Design enables possibility of selecting a variety of wavelengths (trade band width, resolution and intensity). Also cascade allows for multiple wavelengths in one pulse (**Repetition Rate Multiplication)**

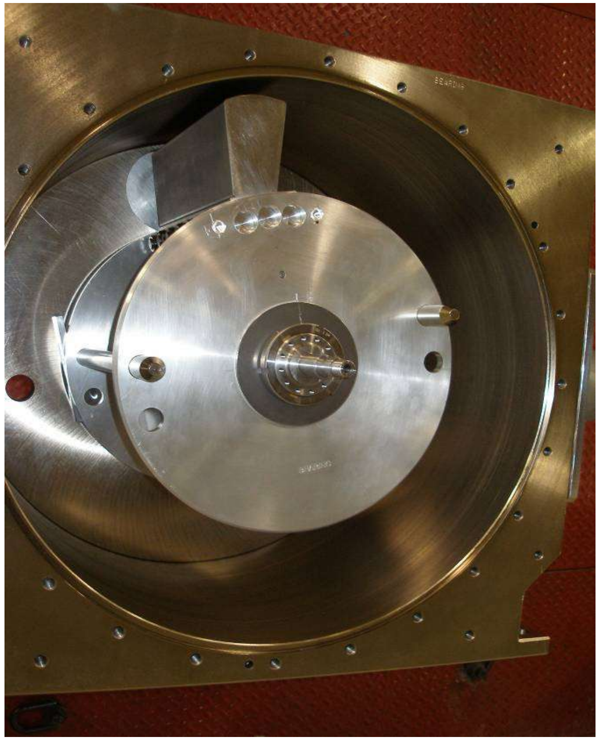




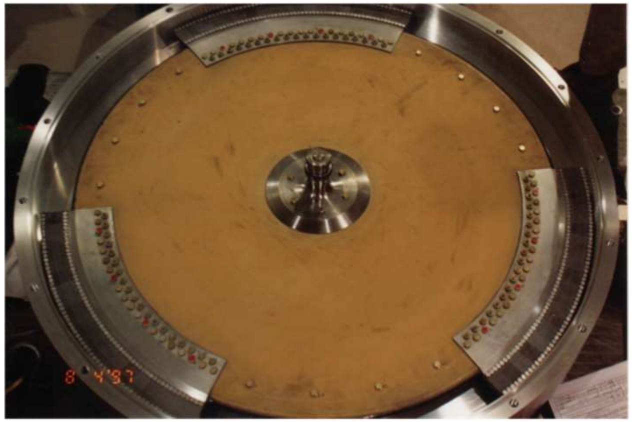


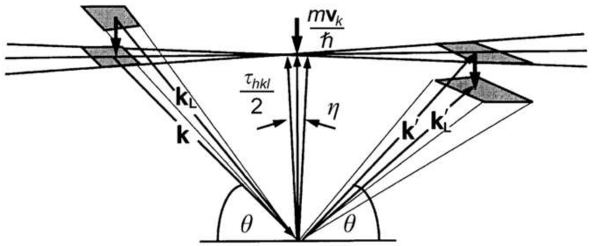
Other Types of Neutron Choppers

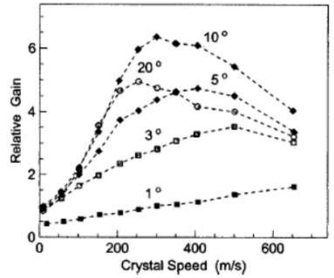
Horizontal axis T0 chopper



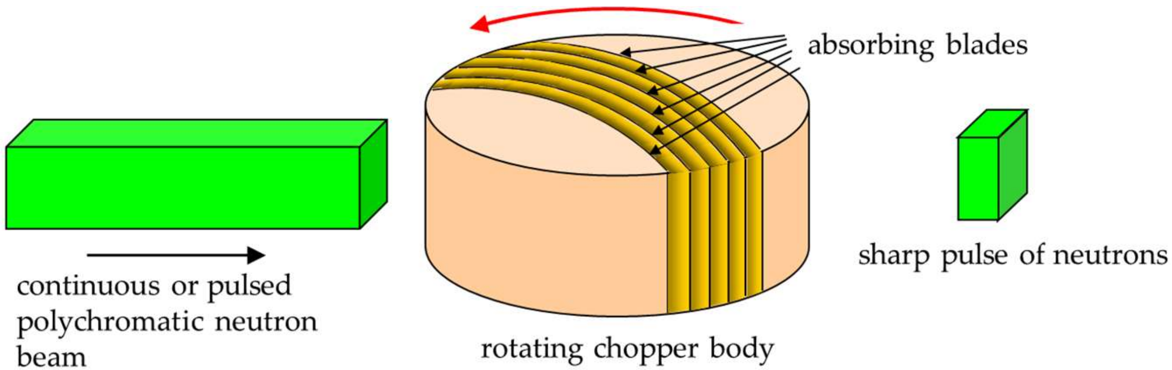
Phase Space Transform chopper using moving crystals







Fermi chopper

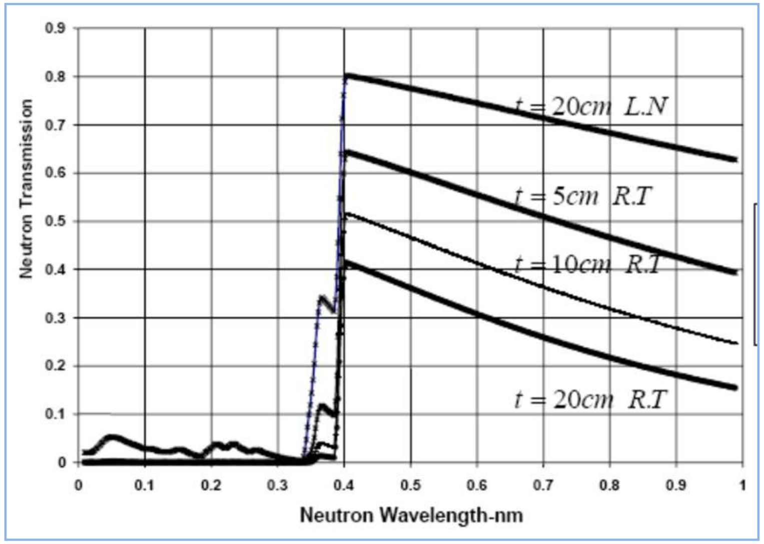


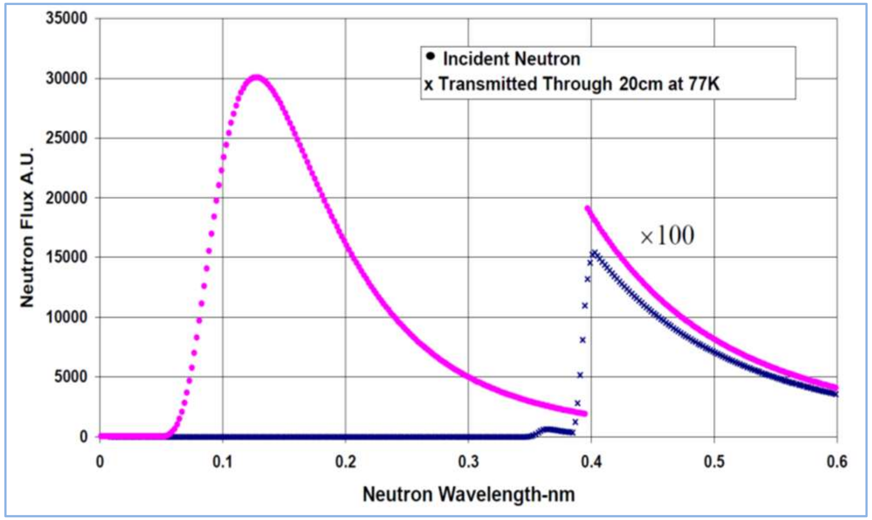
600 Hz rotor

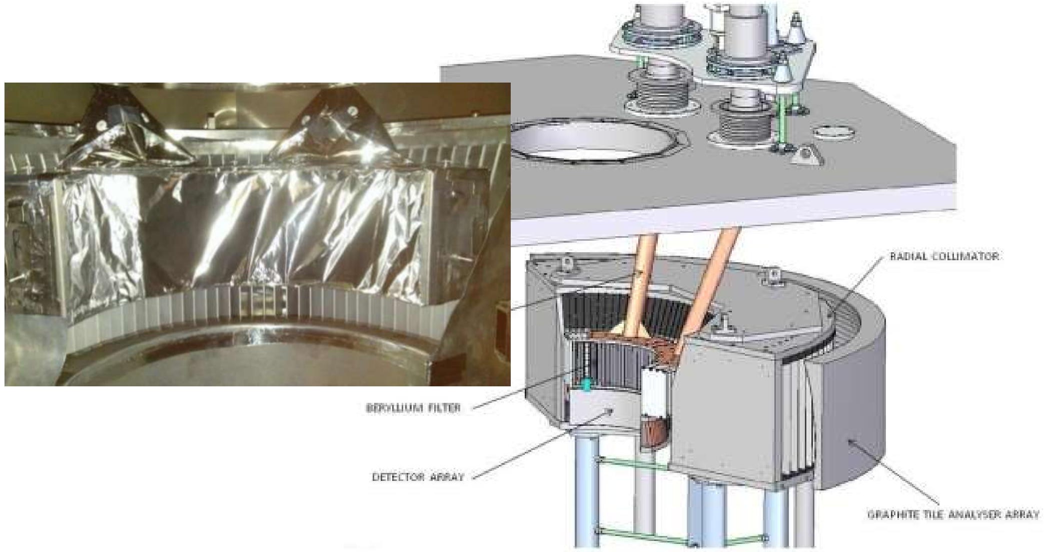
Filters – mainly Beryllium

Cooled Beryllium is used to remove higher order contamination from crystal monochromators (eg. PG002)

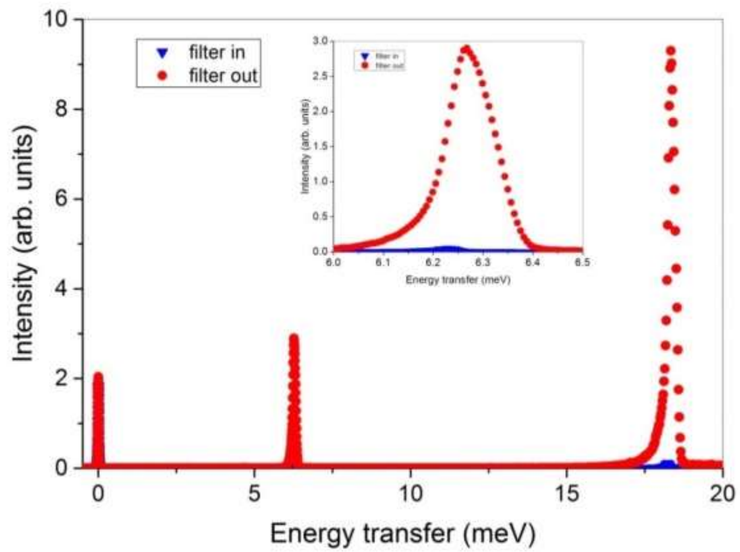
polycrystalline Be filter







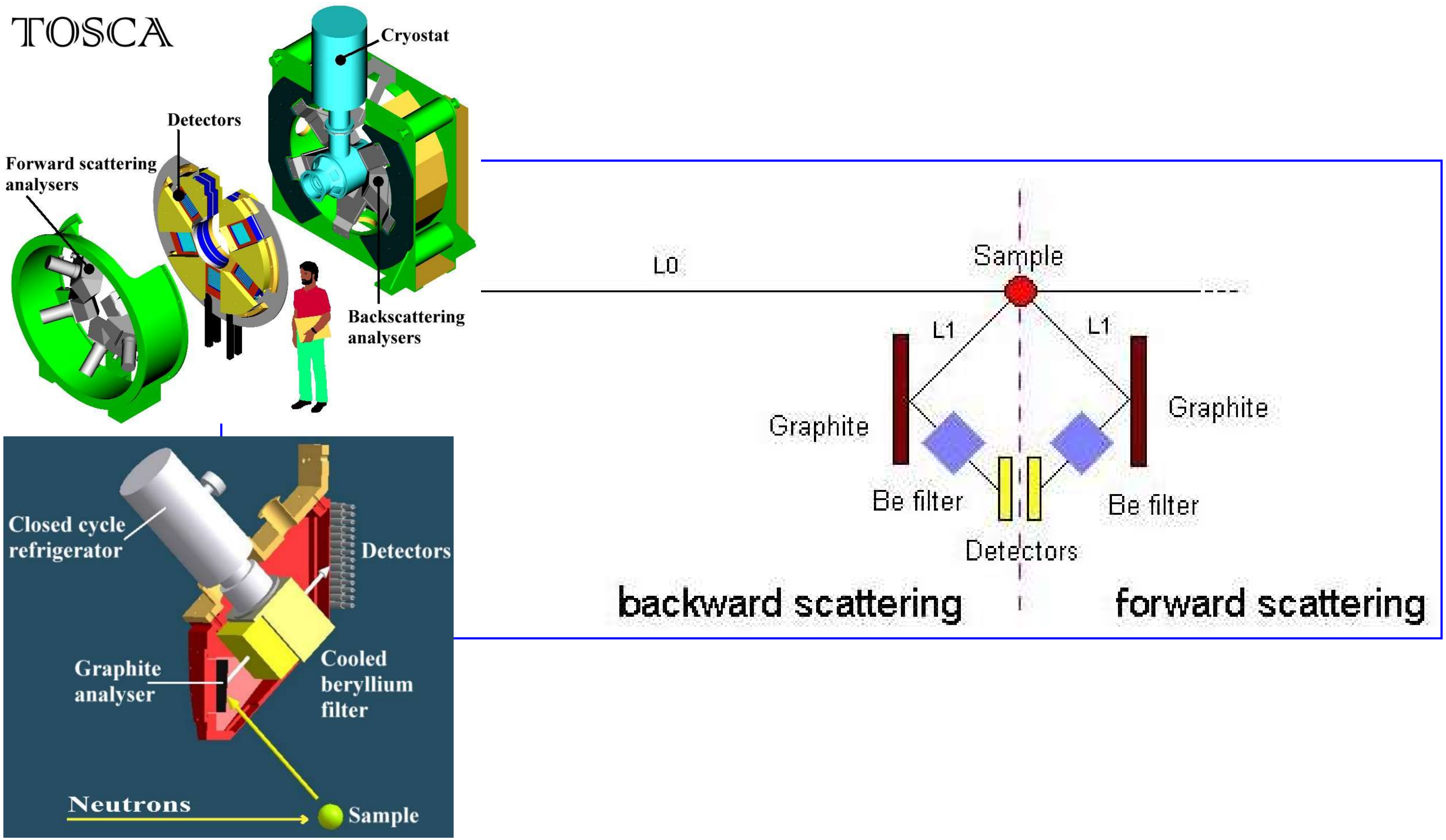
OSIRIS @ ISIS



M. Wahba, Egypt J. Sol. **25**, 215-227 (2002)

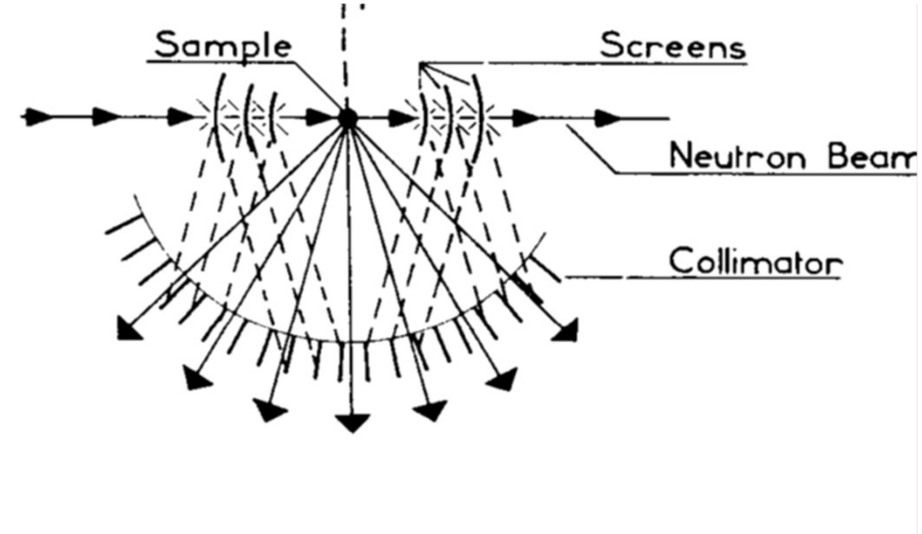
Filters – mainly Beryllium

Cooled Beryllium also is a big part of some inelastic neutron spectrometers such as TOSCA @ ISIS



Radial Collimators

- Large angular coverage
- Collimation of 0.2° to several degrees
- Blades of polymer or glass coated with absorbing material (Gd_2O_3, ^{10}B)
- Can be used to defined beam direction and angular resolution



Neutron Detection

^{3}He tubes

n + ^{3}He -> ^{3}H + ^{1}H + 0.764MeV

>1 mm resolution

High efficiency

Low gamma-sensitivity

Position sensitive

3He supply problem $$$c

Scintillators

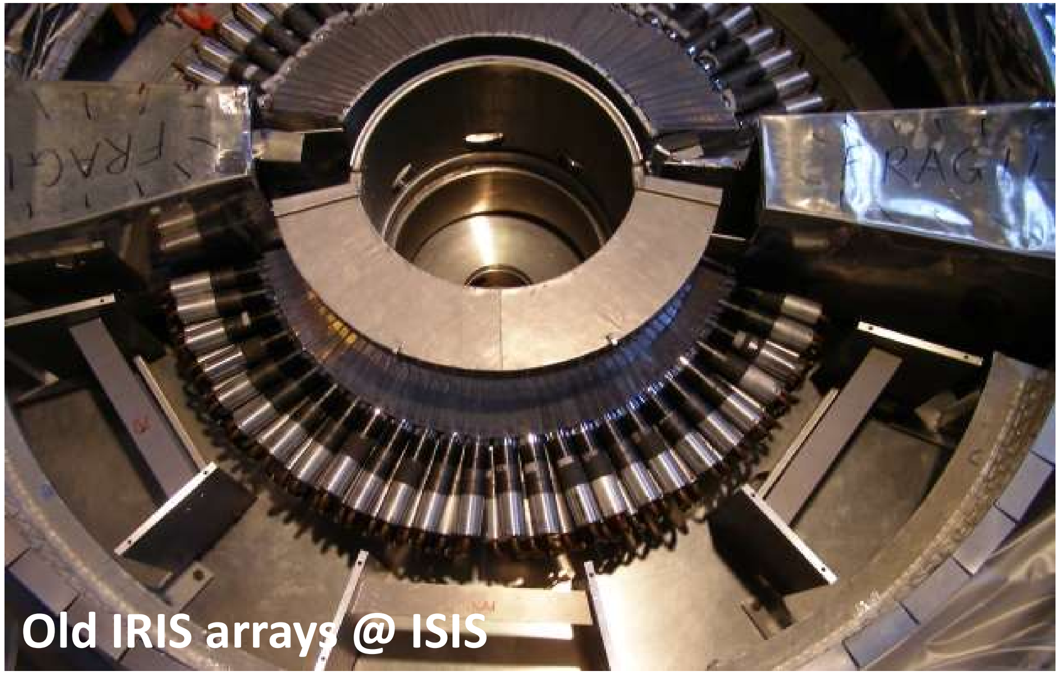
n + ^{6}Li -> ^{4}H + ^{3}H + 4.79MeV

<1 mm resolution

Medium efficiency

Some gamma-sensitivity

Sensitive to magnetic fields


Old IRIS arrays @ ISIS

Neutron Detection

^{10}B multigrids

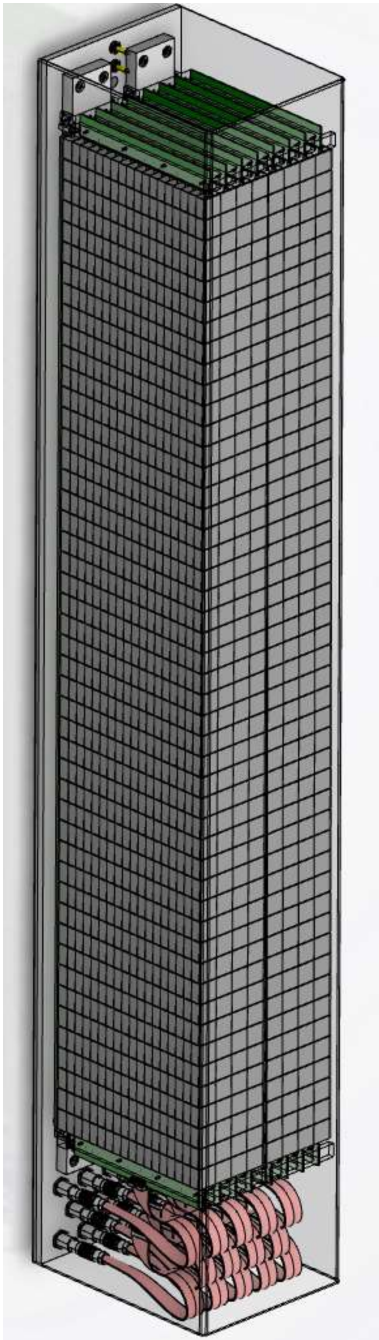
$n + {}^{10}B \rightarrow {}^{7}Li + 4^{1}He + 0.48MeV$

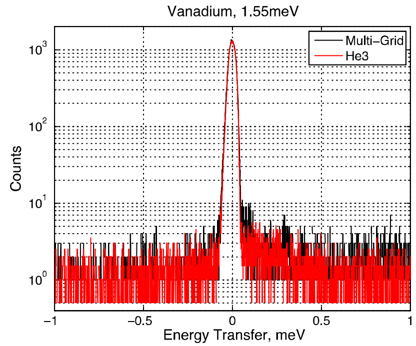
New technology under development

Basis for ESS?

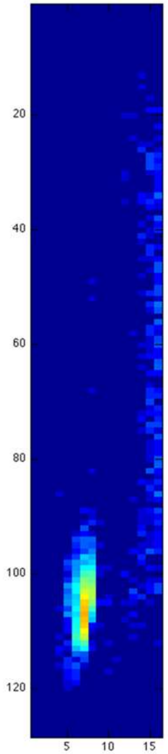
B layer thickness limited to ~1μm

Low $$ for large area

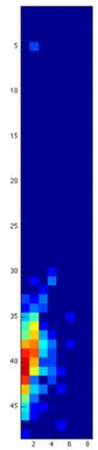




^{3}He



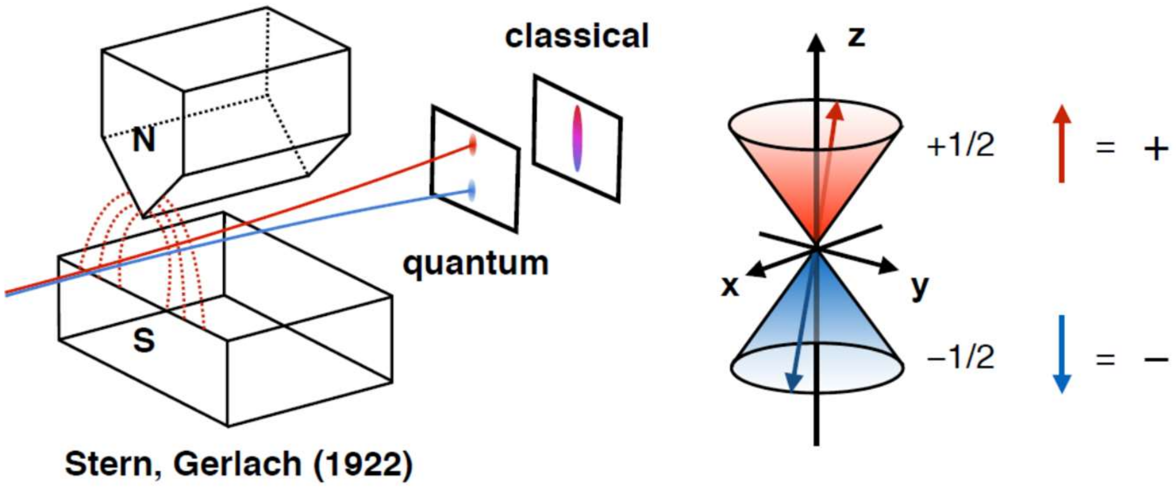
MG

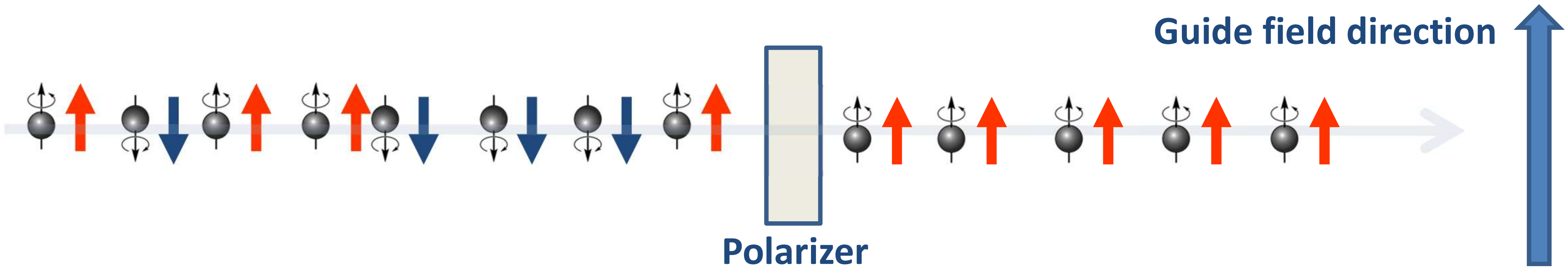


Prototype tested on CNCS @ SNS

Neutron polarisation

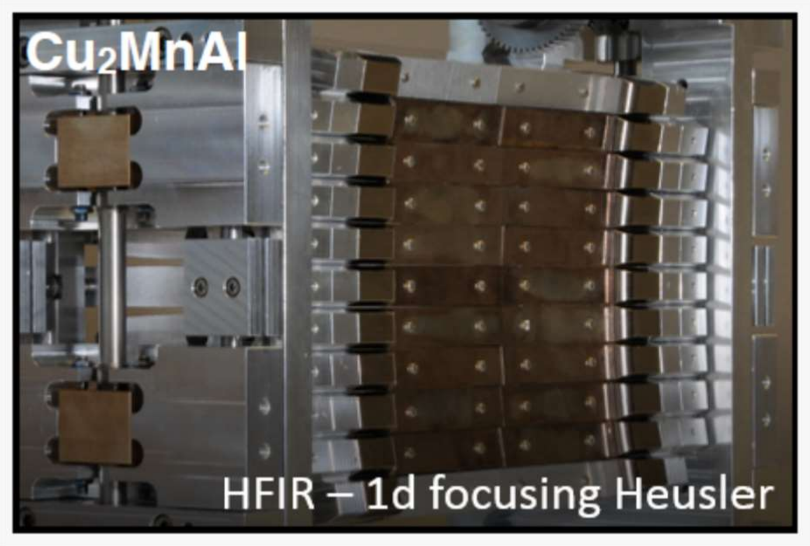
Remember that neutrons posses an inherent **magnetic moment** related to their **spin-angular momentum S=1/2**. In a magnetic field, beam polarisation is a vector pointing in the direction of the field.





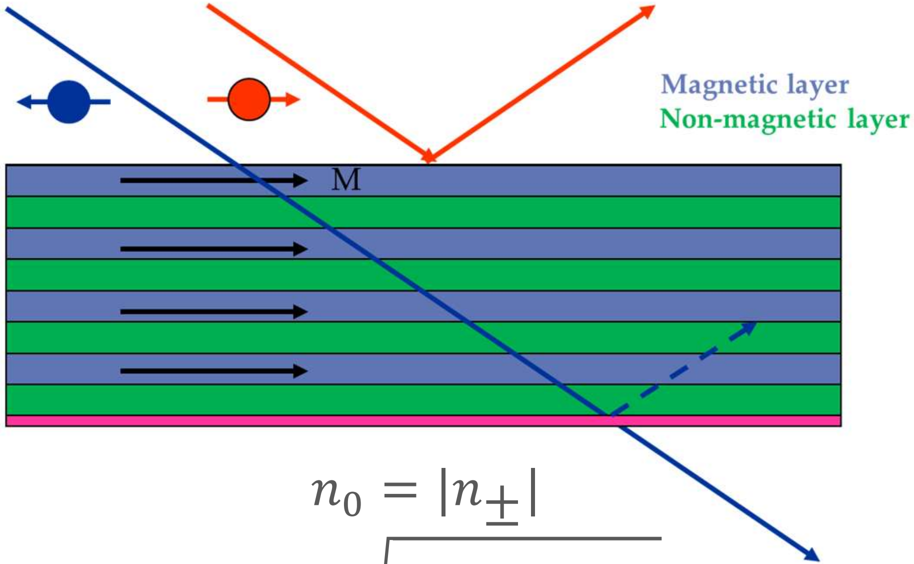
Neutron polarisers

Heussler crystal



$$b_{\mathrm{coh}} = b_{\mathrm{mag}}$$

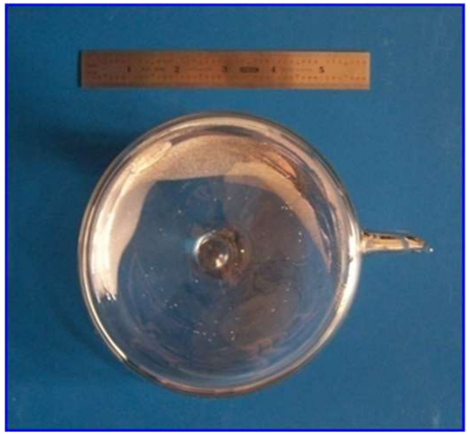
Polarising supermirrors



$$n_0 = |n_{\pm}|$$

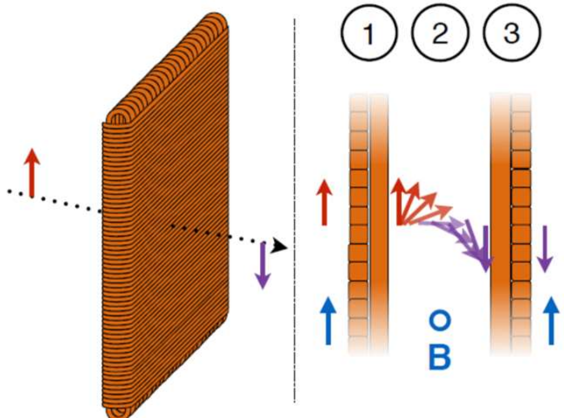
$$n_{\pm} \propto \sqrt{\rho_{\mathrm{coh}} \pm \rho_{\mathrm{mag}}}$$

^{3}He spin filters

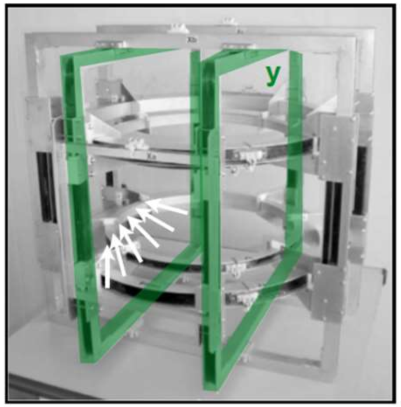


$$\sigma_{\mathrm{a}}(\uparrow\downarrow) = 5931\mathrm{b}; \sigma_{\mathrm{a}}(\uparrow\uparrow) \sim 0\mathrm{b}$$

Mezei Flippers



XYZ field rotators

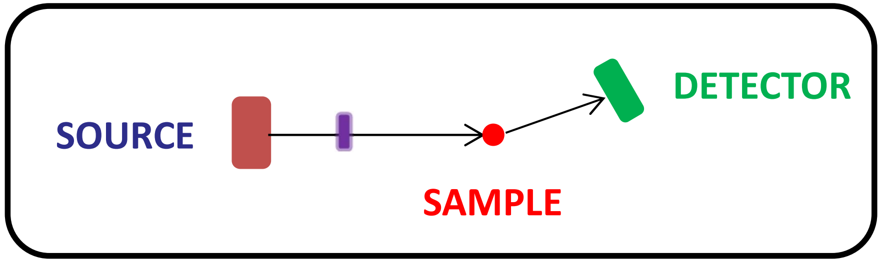


Neutron Spectrometers

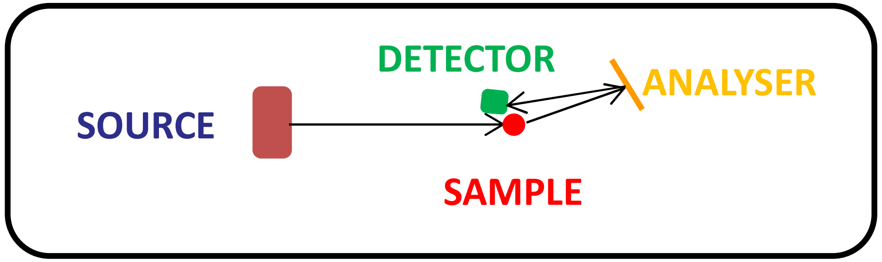
Neutron Spectrometers

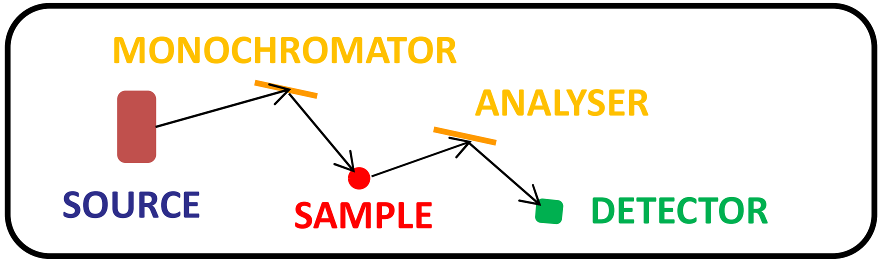
Frequency domain S(Q,ω)

Direct geometry



Indirect geometry or Backscattering*

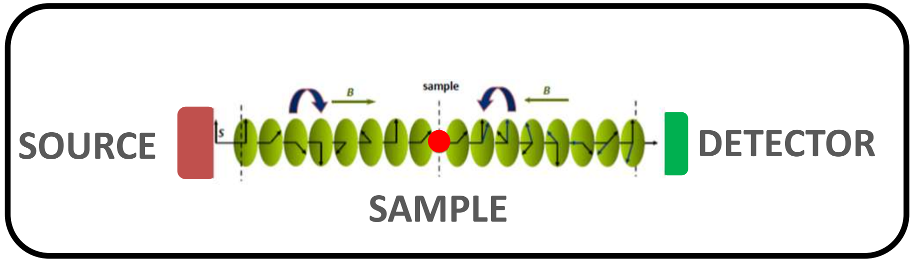




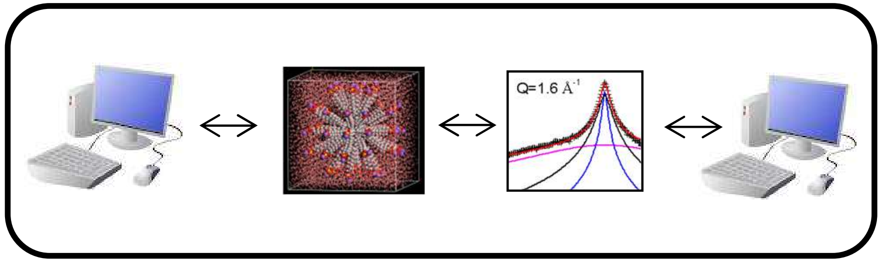
Triple-Axis

Time domain I(Q,t)

Neutron Spin Echo



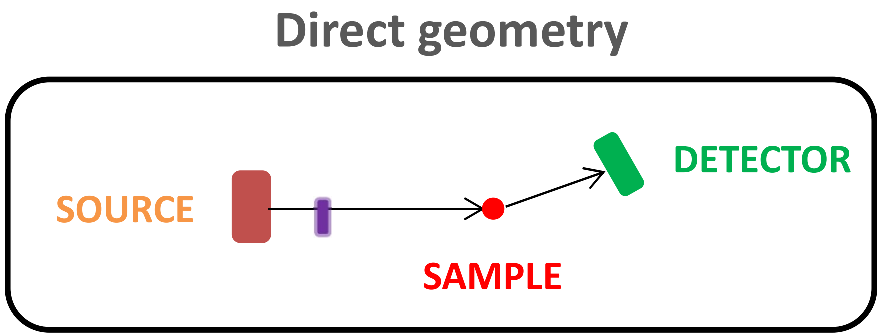
Molecular Dynamics Simulations



** This is essentially a special case of TAS*

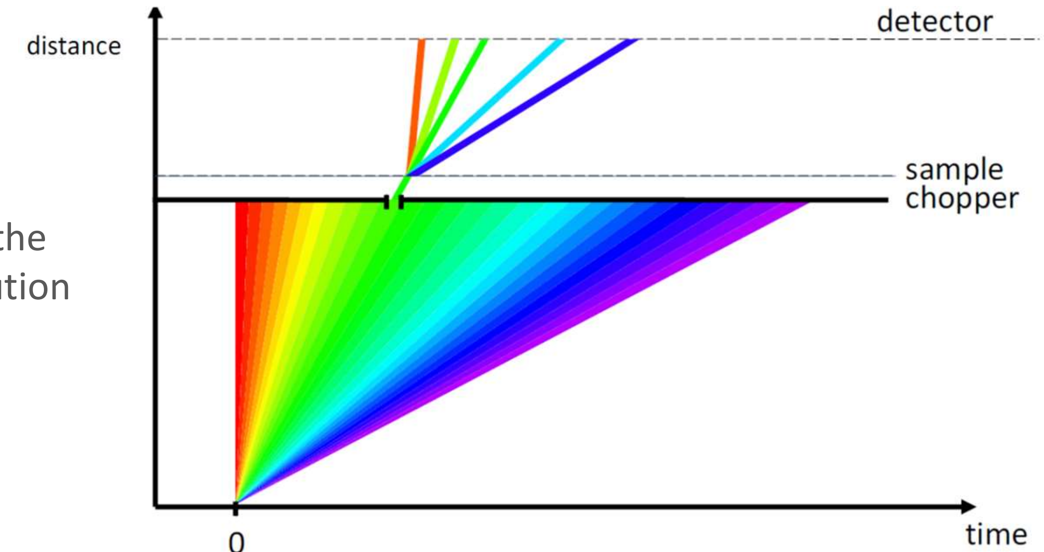
Direct Geometry
(or Chopper spectrometers)

Direct Geometry



- Send neutrons of known **fixed** $\boldsymbol{E_i}$ **(**$\boldsymbol{v_i}$**)** – neutron can loose only as much energy as it has but can gain any.
- Source-sample and sample-detector distances known (order of m)
- Time at which neutron is sent, known, t_0
- Time at which neutron is detected tells us $E_{f;}$ thus we know ΔE

ΔE=0 as given by the instrumental resolution



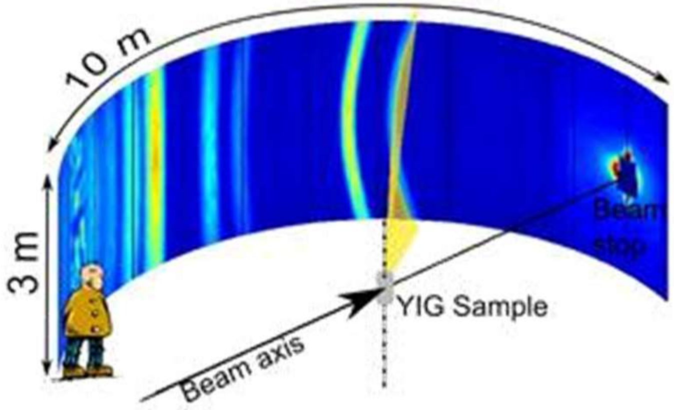
or Chopper Spectrometers

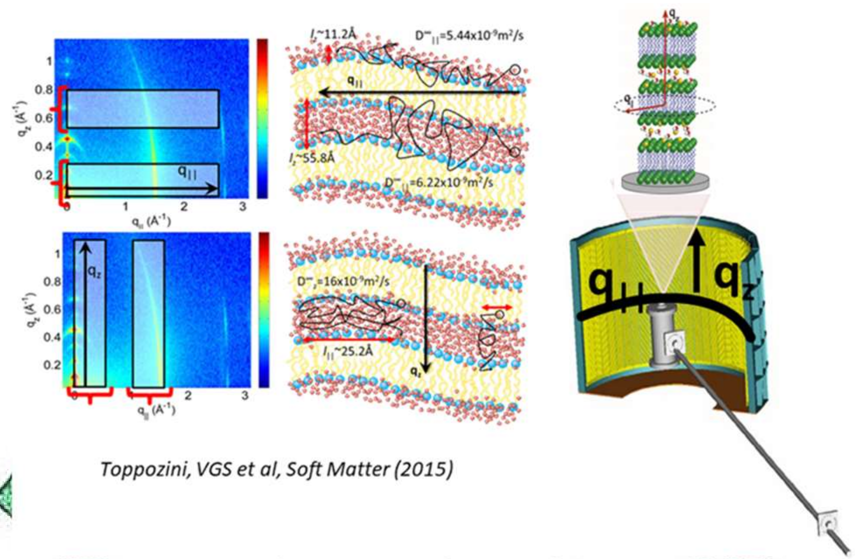
Monochromatisation	
6 counter-rotating choppers	
Chopper velocity Vo (revolution per minute)	2000 rpm to 17000 rpm
Incident wavelength λ_o	1.8 Å to 20 Å

Sample	
elastic energy resolution at 5.0 Å, 8500rpm	~ 100 µeV
max neutron energy loss	$\hbar\,\omega_{max}$ 0.6×E_o $E_{min} \approx 0.4\times E_o$ (λ_{max} = 1.5λ_o)
max energy gain for the neutrons	$E_{max} \sim \infty$ . . .
max momentum transfer Qmax	11.48/λo Å^{-1}
horizontal divergence (no collim.)	~ 0.1×2×λ_o [°]
vertical divergence (no collim.)	~ 0.1×3×λ_o [°]
Collimation before sample	- / 30' / 60'
beam size at the sample	~ 15×50 mm^2
flux at the sample at 5.0 Å	6.83×10^5 [n/cm^2/s]

Detectors	
detector shape	cylindrical
detector surface [m^2]	30.0
fight path (radius of reading) [m]	4.0
effective detection height [m]	3.0
scattering angular range [°]	-12° to 135°
vertical angular range [°]e	±20.55°
solid angle covered	1.8 sr (~0.6π)
min. & max. momentum transfer [Å^{-1}]	0.2/λ [Å] - 11.8/λ [Å]
spatial resolution [cm^2]	2.6 x 2.6
angular resolution [° / rad / ']	0.37° / 6.10^{-3} rad / 22
detector type	position sensitive ^{3}He counters
gas mixture	4.75 bar ^{3}He +1.25 bar CF_4
detector efficiency	~ 80% for Eo = 3.27 meV (5 Å)
dead time / max. count rate (per tube)	≤ 10 µs / ≥ 100 kHz
material	stainless steel
number of detectors units (32 tubes/unit)	12
tubes diameters [inches/cm]	1' / 2.54

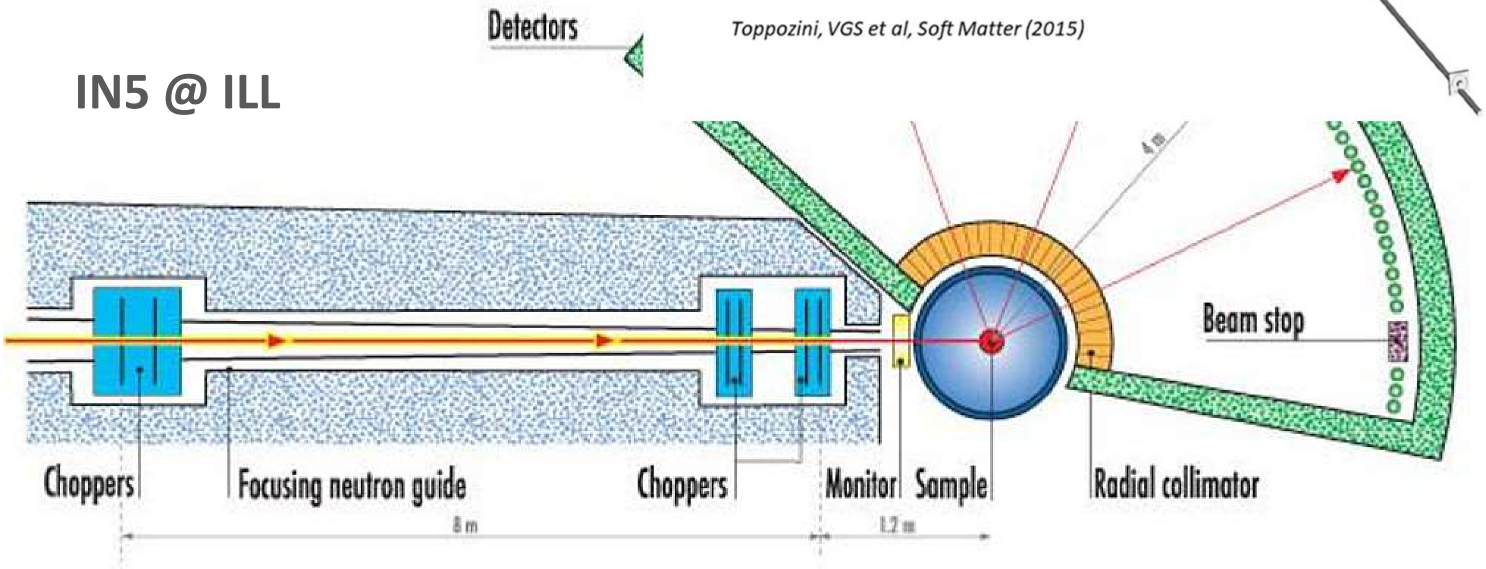
Typical layout – consist of a number of choppers, large detector arrays of ^{3}He tubes of varying heights, position sensitive, typically covering -15 to 160deg and have a radial collimator. Tank is evacuated or Ar filled. Incident energies between 1meV and 1eV.





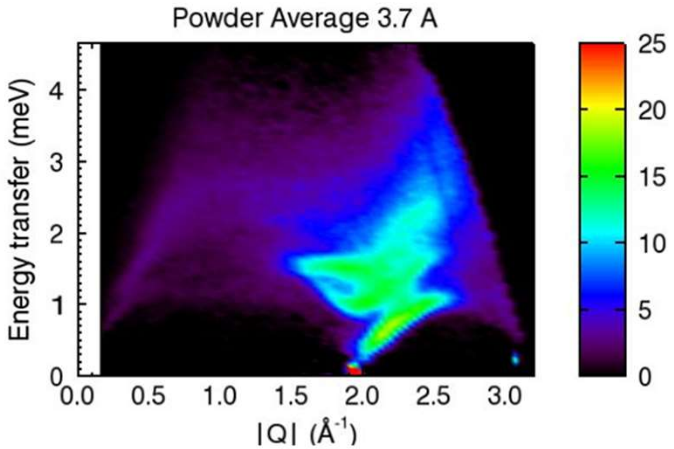
Toppozini, VGS et al, Soft Matter (2015)

IN5 @ ILL

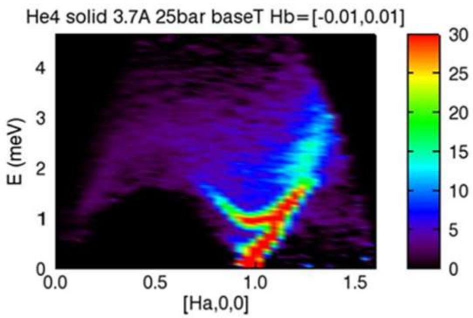


Chopper Spectrometers

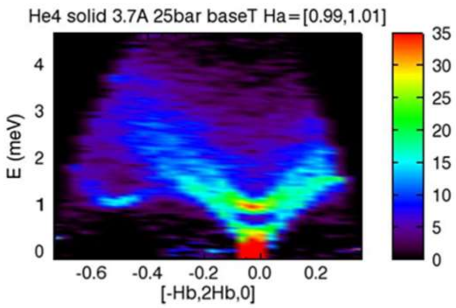
Solid Helium



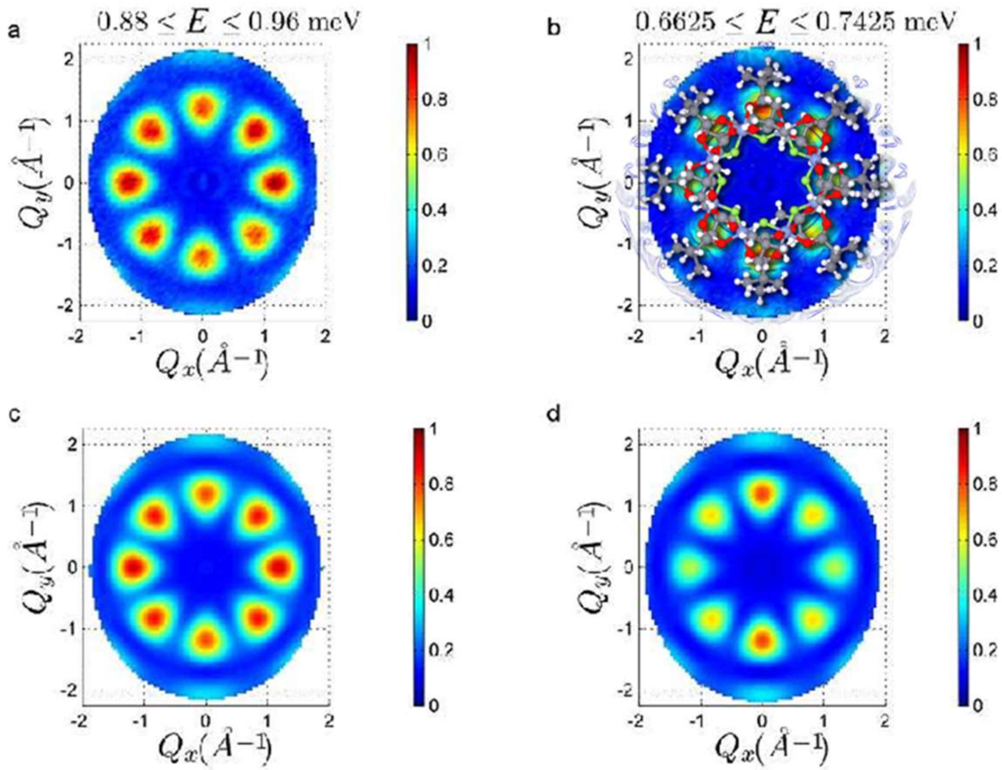
Longitudinal



Transverse



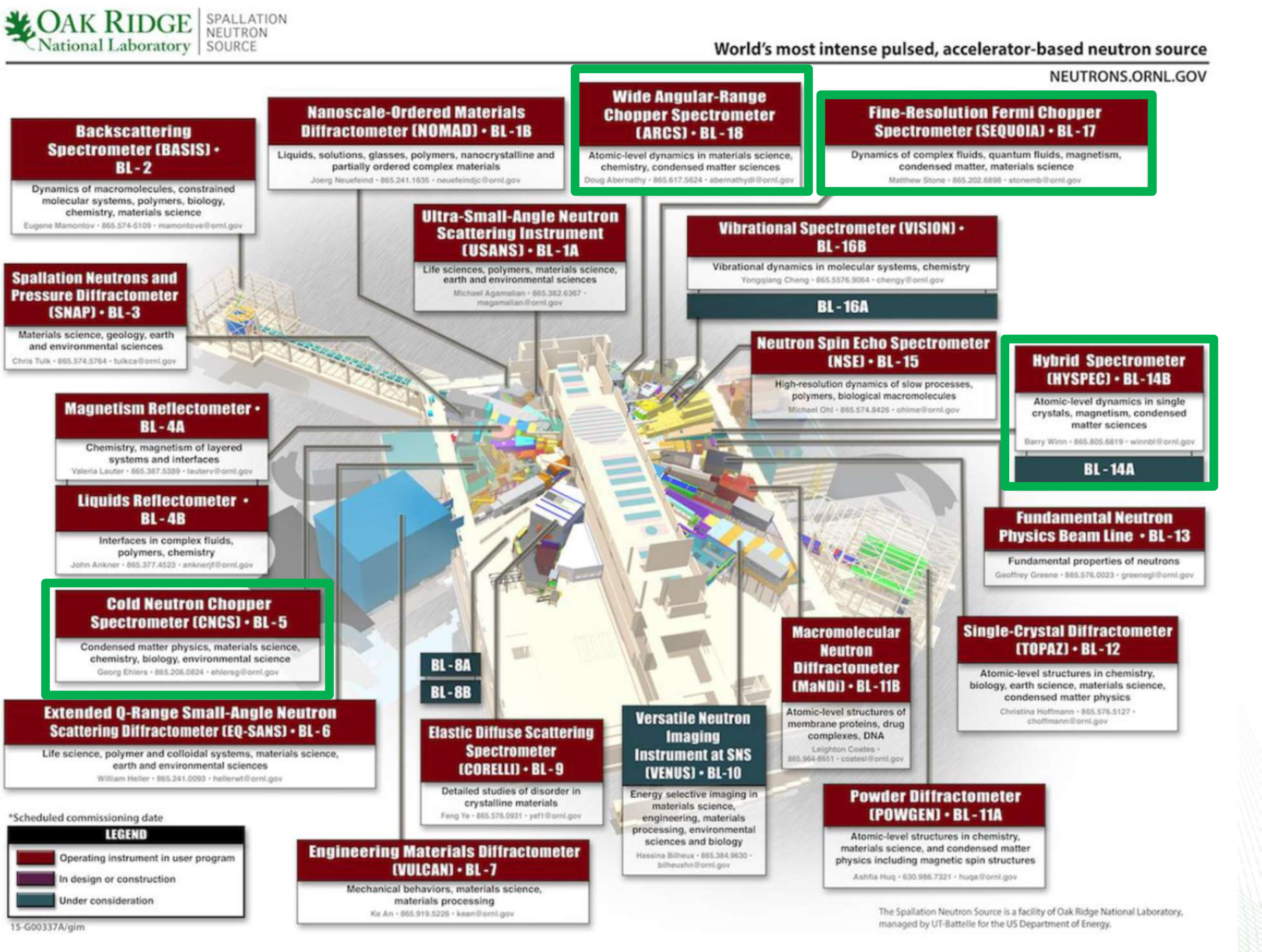
Spin dynamics in molecular nanomagnets



Baker *et al.*, Nature Physics 8, 906-911 (2012)

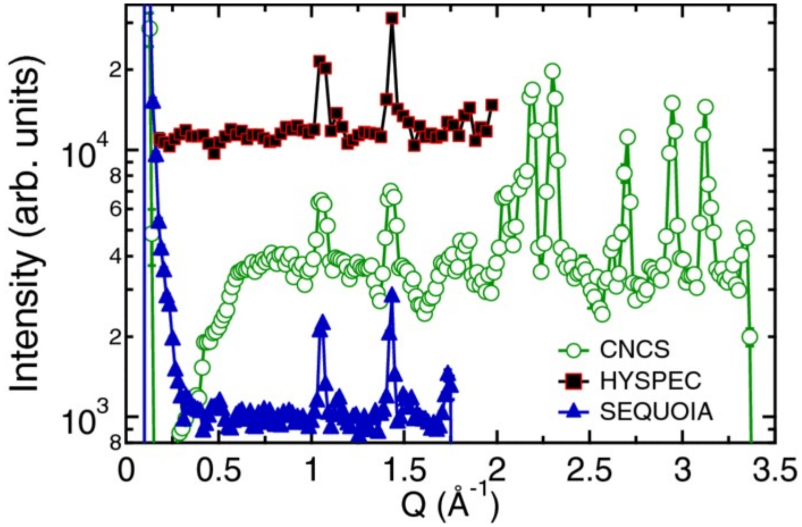
Can be found at reactors or spallation sources

Time-of-flight Spectrometers

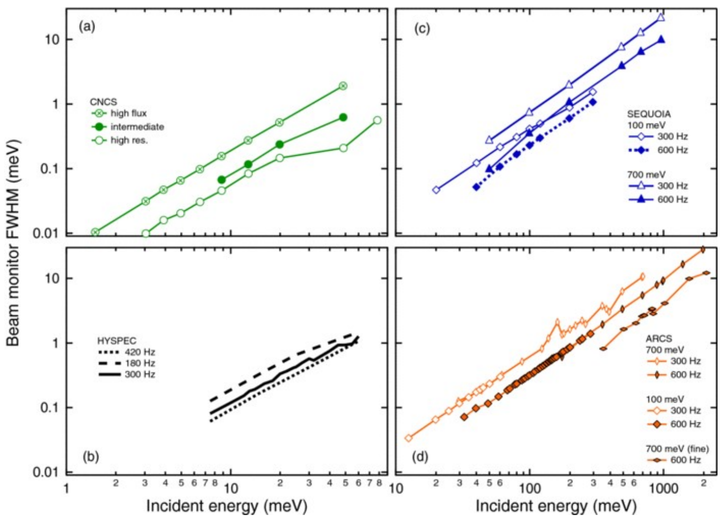


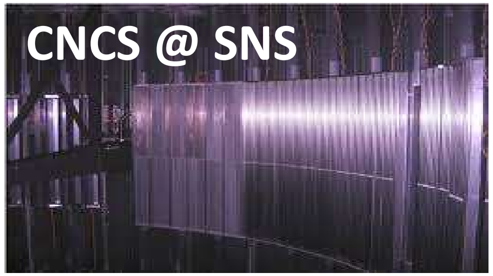
Time-of-flight Spectrometers

Intensity as a function of incident E

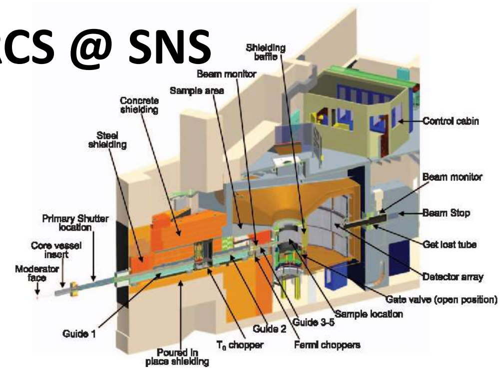


Resolution as a function of incident E

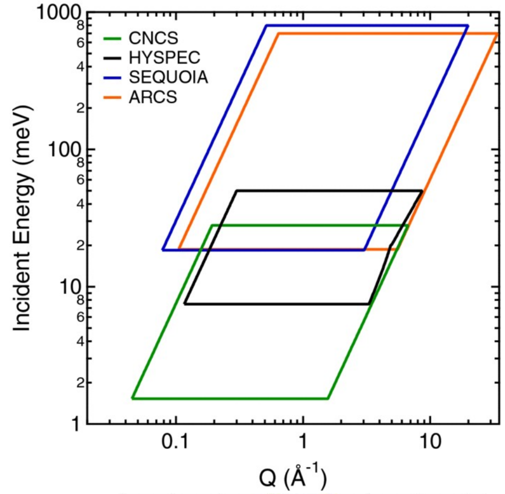




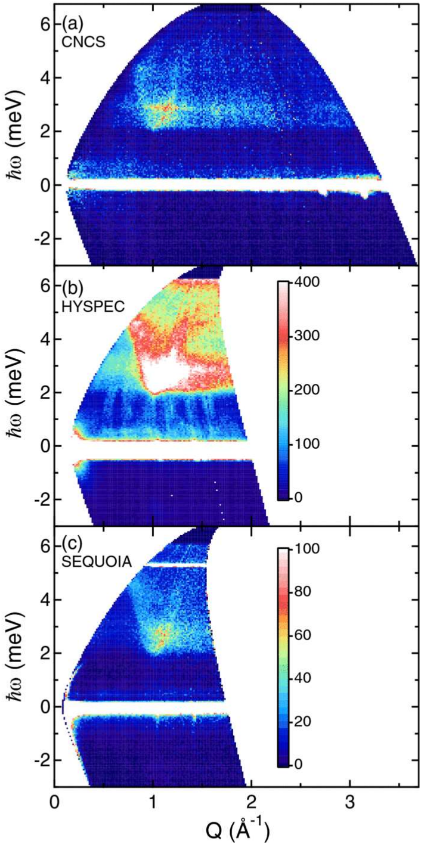
ARCS @ SNS



Q-space as a function of incident E



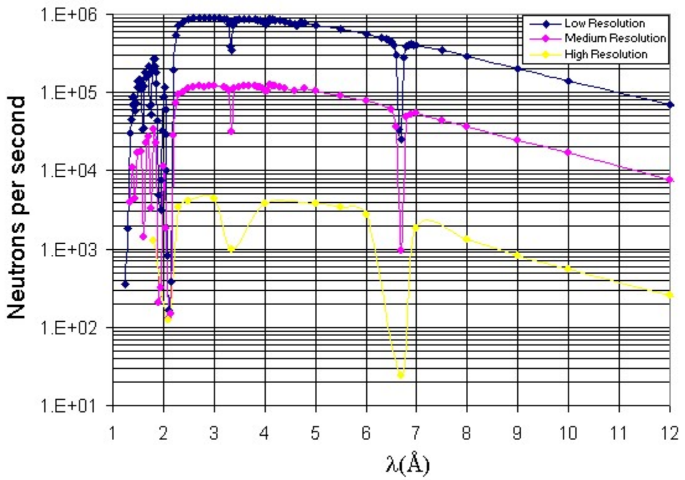
Energy window as a function of elastic Q



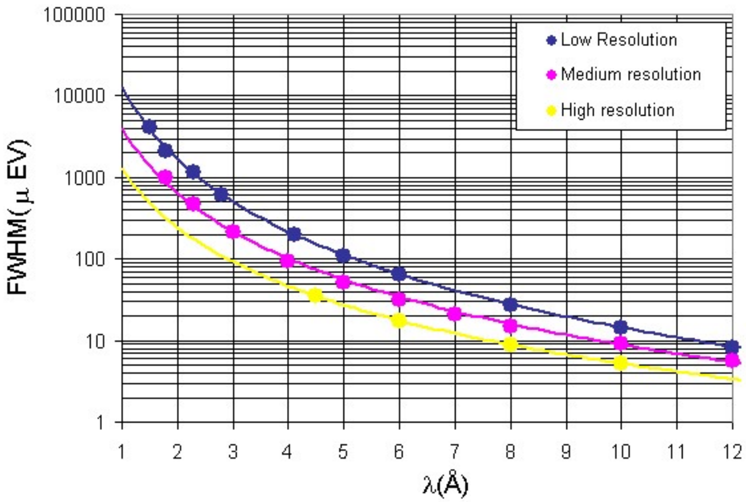
Chopper Spectrometers

DCS @ NIST

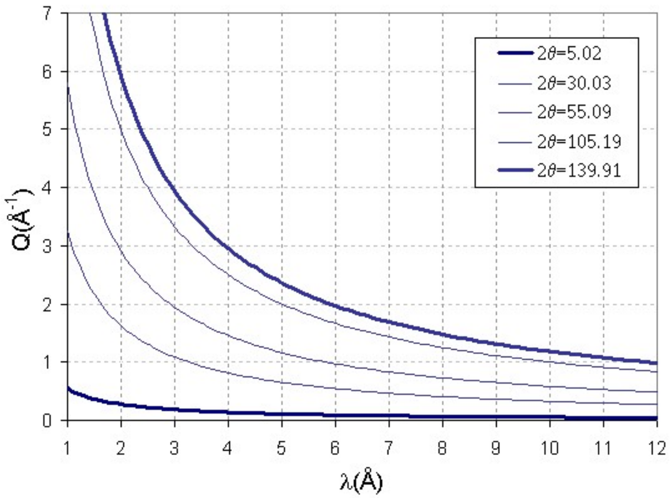
Intensity as a function of λ and ΔE



Energy resolution ΔE as a function of λ



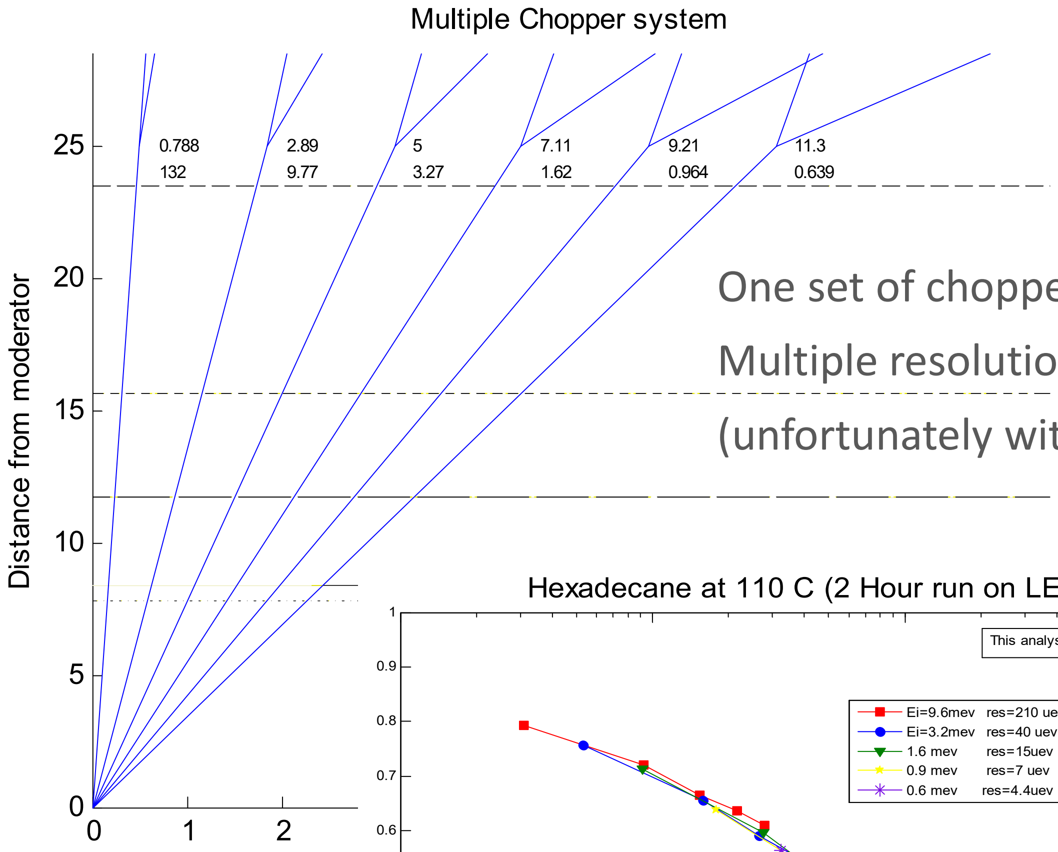
Elastic scattering vector Q, as a function of λ



- Medium energy resolution (10's ueV to meV's)
- Large DE and Q range
- **Flexible** in choosing Q-E space
- Picosecond dynamics

Chopper Spectrometers

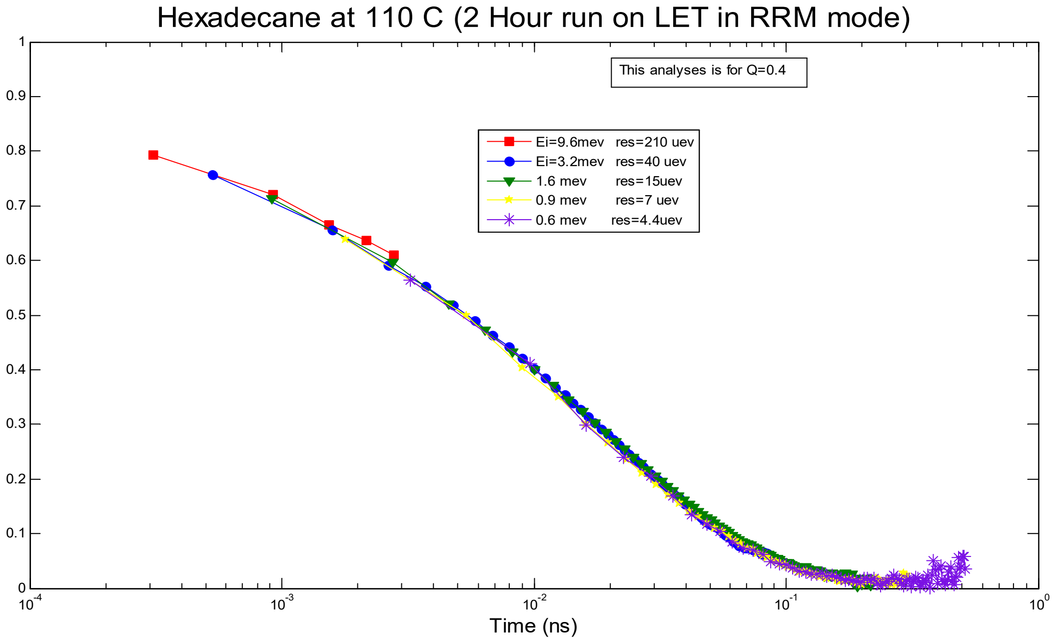
Clever chopper cascades have enable Repetition Rate Multiplication



One set of chopper settings give multiple E_i's

Multiple resolutions + energy ranges at once

(unfortunately with varying Q)



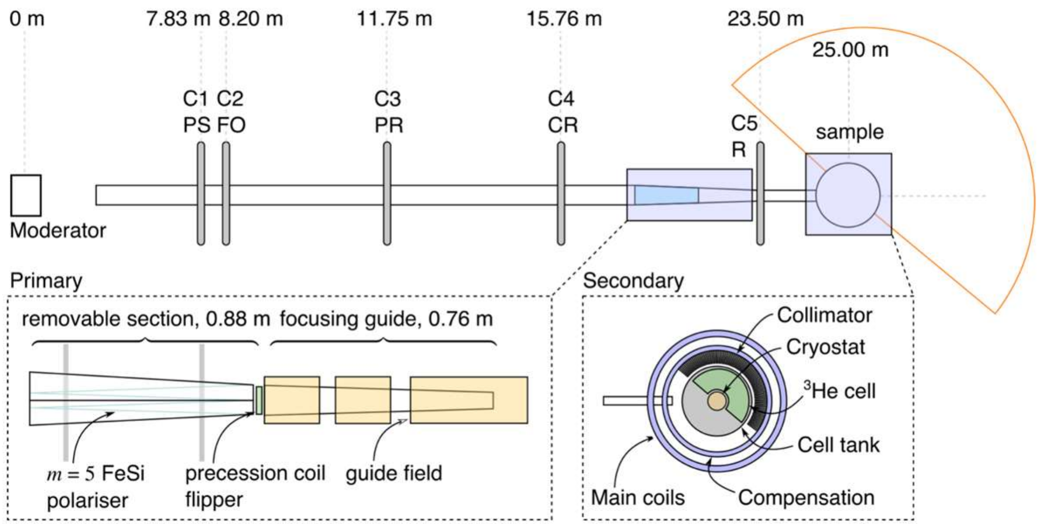
LET @ ISIS

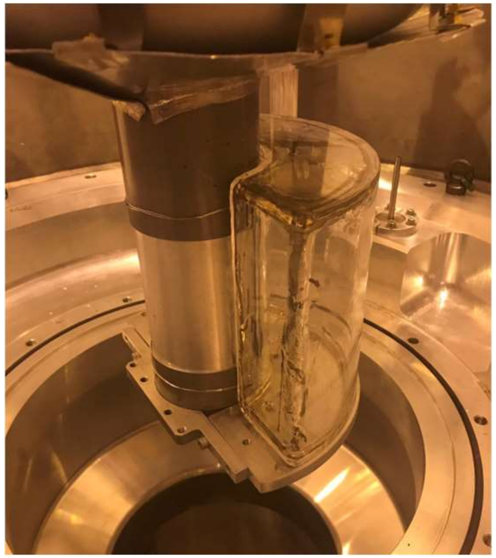
Closes gap in S(Q,w) space

Polarisation Analysis

Since neutrons allow for polarised beams, PA can be used to separate signals:

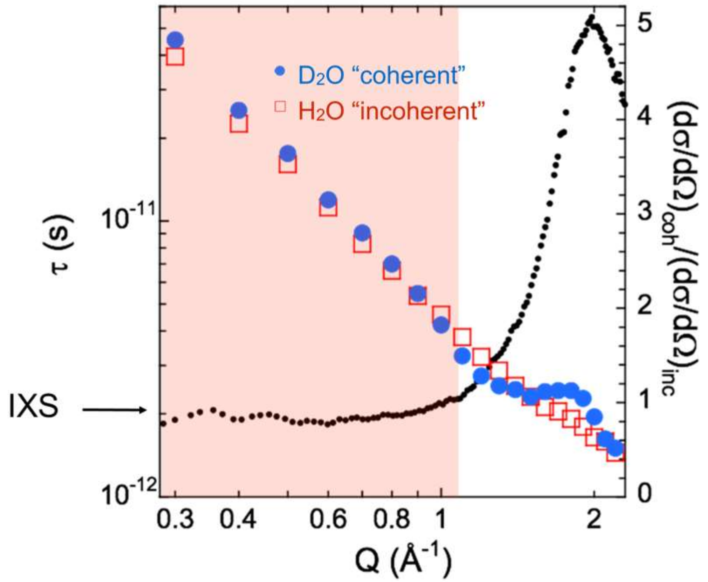
1. Nuclear from magnetic inelastic scattering
2. Where deuteration is difficult in multi-component systems
3. When there is structural peak contamination (non-H)
4. Where coherent and incoherent scattering are difficult to distinguish (e.g. Na, ^{7}Li, D …)
5. To prove assumptions!





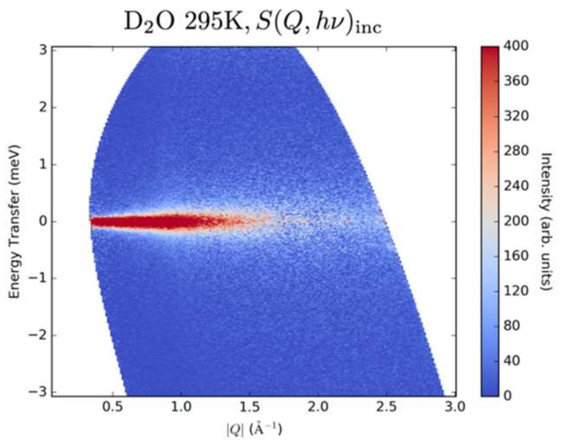
Polarisation Analysis Example

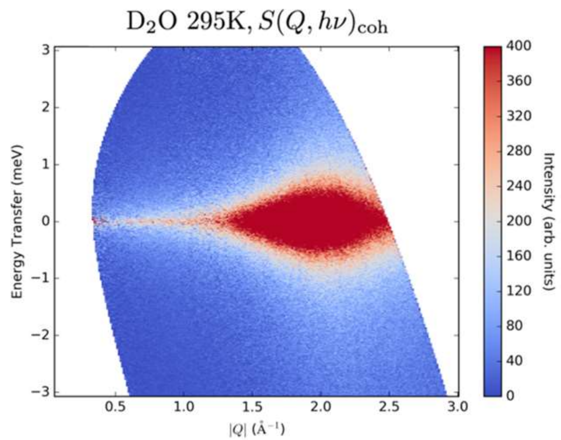
Dynamics of D2O at the mesoscale measured on IN5 @ ILL (unpolarized)

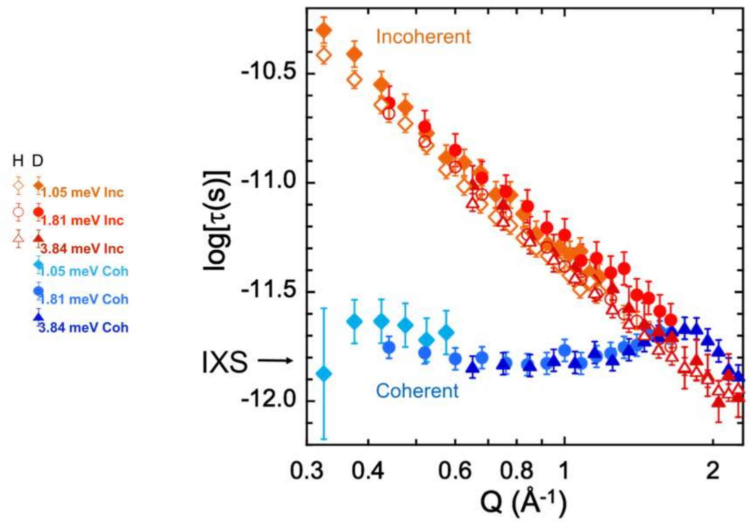


Ratio of coherent/incoherent from D7@ ILL (diffraction mode)

1st (proper) PLET experiment on QENS

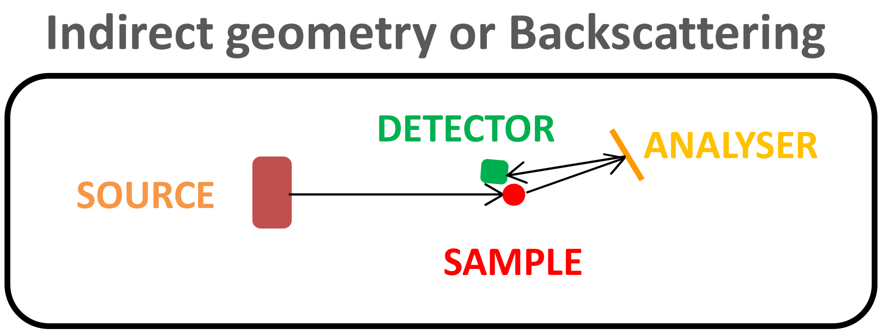






Indirect Geometry

Indirect Spectrometers or Backscattering



- Send neutrons of a known band of wavelengths or E_i (v_i)s (defines your energy window)
- In reactor source, use a Doppler drive; in a spallation source, use choppers
- Analyser crytals reflect back only a **fixed E_f** (Bragg's Law)
- Times & distances known, so detected neutron gives us ΔE
- Higher energy resolution ΔE at a flux penalty

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

$$\frac{1}{2}\frac{\Delta E}{E} = \frac{\Delta\lambda}{\lambda} \sim \frac{\delta d}{d} + \cot(\theta)\delta\theta$$

in backscattering $\theta \to \pi/2$

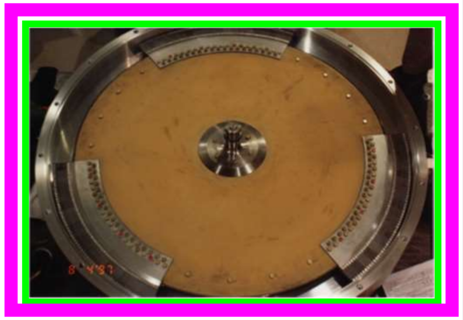
$$\cot(\theta) = \frac{\cos\theta}{\sin\theta} \to 0$$

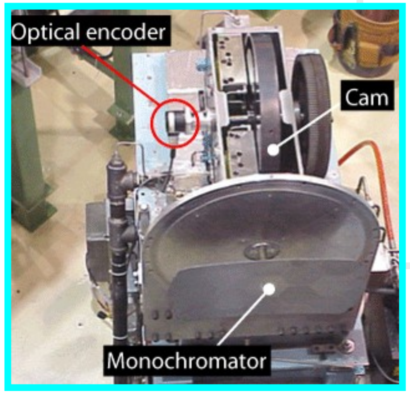
Backscattering @ Reactors

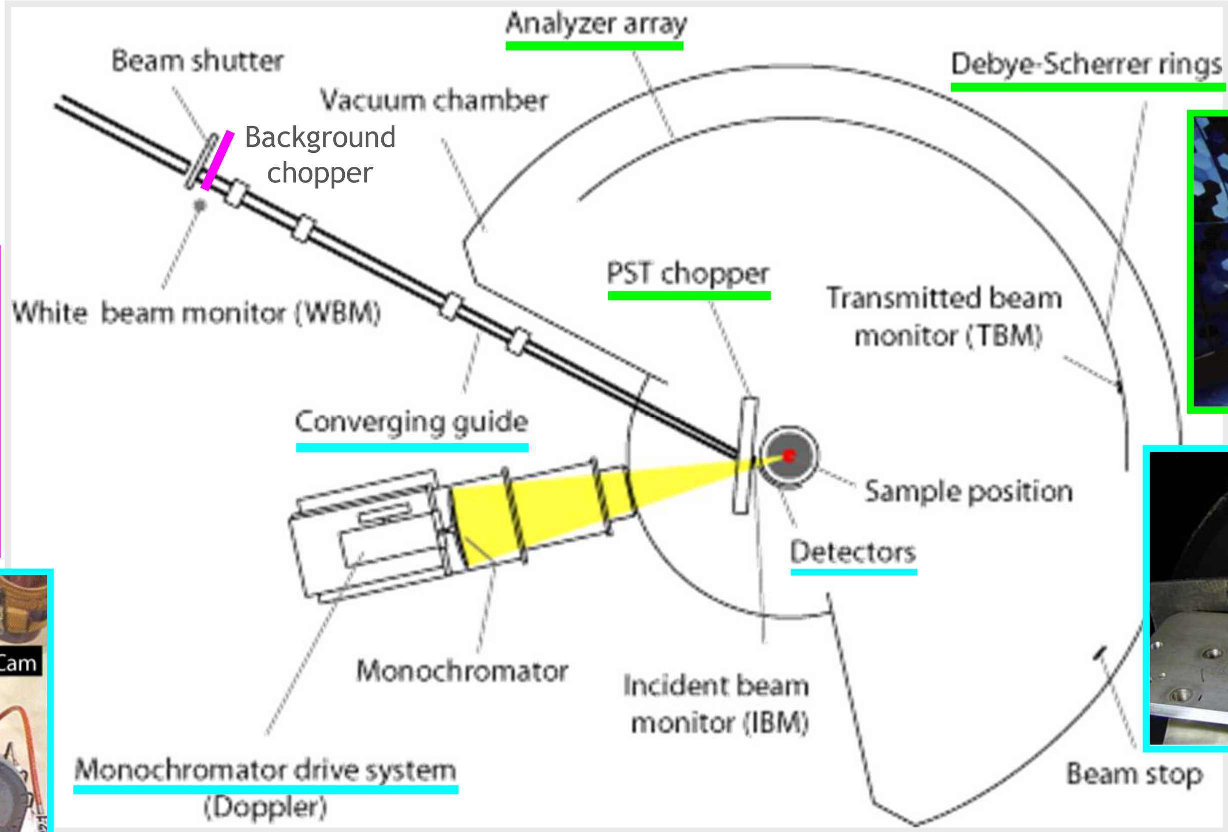
HFBS @ NIST

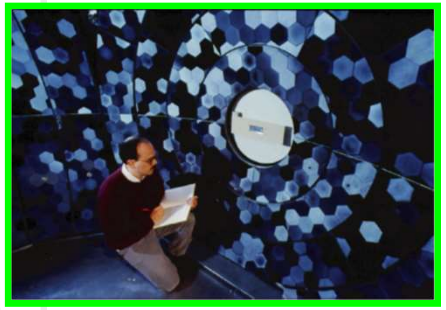
Typical layout – consist of a Doppler monochromator, large analyser bank of **Si111 crystals** (final energy is 2.08meV), small array of 3He detectors, typically covering 15 to 130deg. Tank is evacuated.

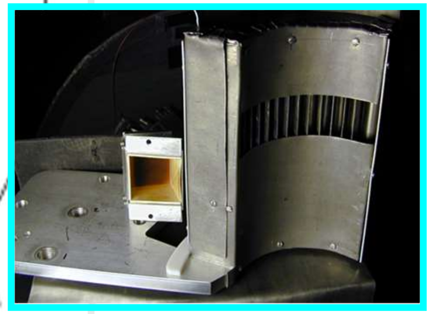
- 20MW reactor
- 41m long
- filter + NVS





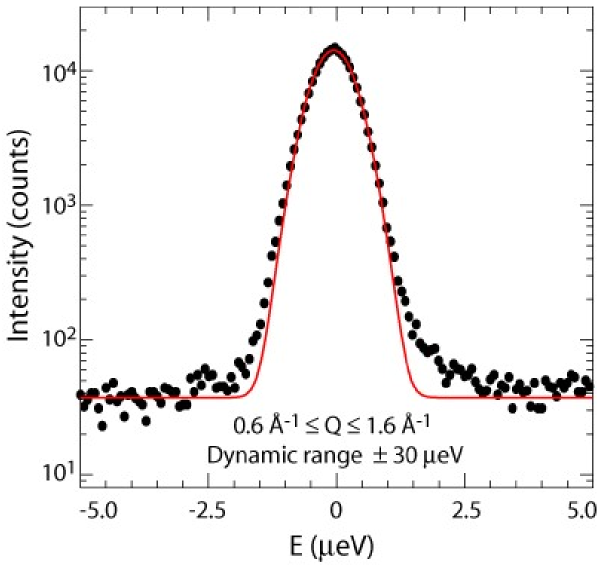


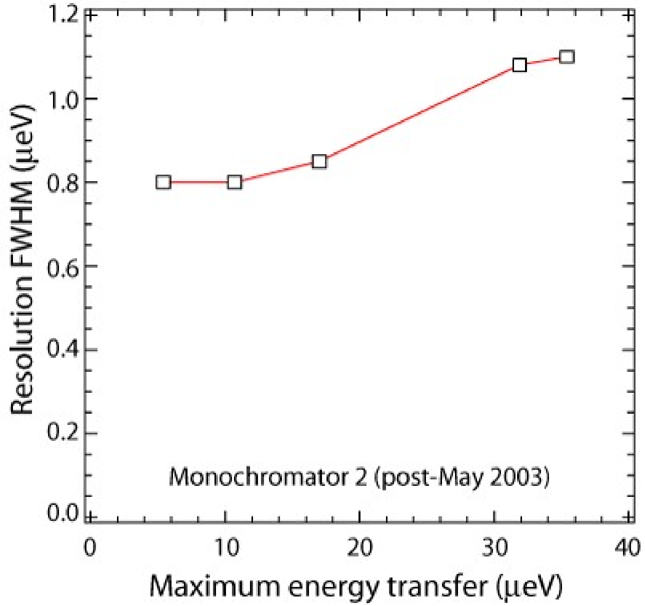


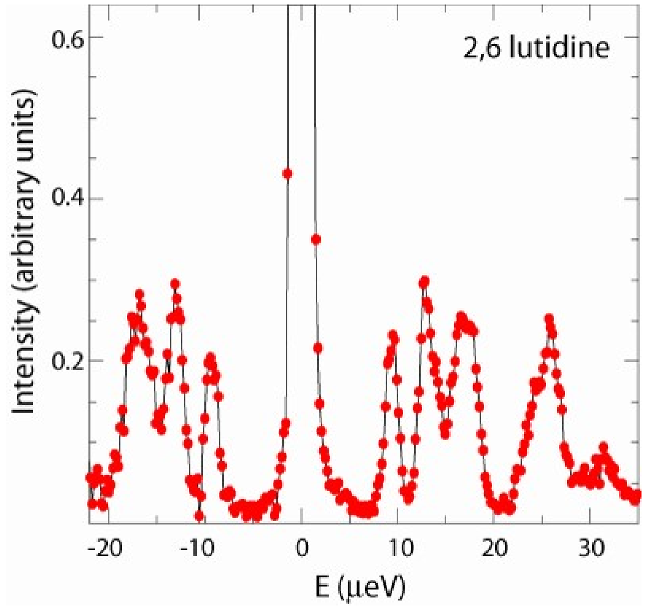


λ_0 = 6.271 Å and E_0 = 2.08 meV

Backscattering @ Reactors

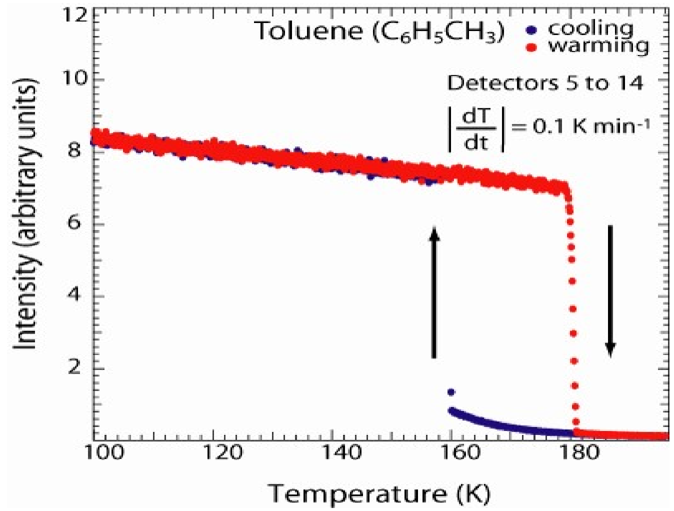






Energy Range	Detectors: 1-4 (μeV)	Detectors: 5-16 (μeV)
11 μeV	1.07	**0.79**
17 μeV	1.13	**0.83**
36 μeV	1.60	**1.04**

Detectors 1-4 are slightly out of backscattering geometry and have a degraded resolution. Detectors 5-16 are in backscattering geometry and have almost linear resolution averaged in the above table.



- High energy resolution (~1 ueV)
- Limited ΔE and Q range
- **Limited flexibility** in choosing Q-E space
- Picosecond and nanosecond dynamics
- Multiple modes (EFWS, QENS)

Backscattering @ Reactors

Changing the analyser crystal enables choice of energy resolution, Q-range and energy window. As a result of the crystal reflectivity, choice of scattered beam intensity.

modes	Si 111		Si 311	BATS	*GaAs 200*
	standard E-resolution	high E-resolution	access to high Q with high E-resolution	wide E-transfer range	*best E-resolution (in commissioning)*
analysers	Si 111 'strained' *6 new full height large angle analysers (LAA). SANS: old IN16 analysers with half height*	Si 111 'polished' *3 old IN16 type LAA and SANS (half height ±19°)*	Si 311 'strained' *6 old IN16 type LAA; no SANS (half height ±19°)*	Si 111 or Si 311 *commissioned with Si 111 only*	*GaAs 200* *~ 0.3 m2 prototype analyser*
monochromator	Si 111 'strained'	Si 111 'polished'	Si 311 'strained'	- none - *short 'white' pulses*	*GaAs 200*
final wavelength (Å)	6.271	6.271	3.275	$\Delta\lambda/\lambda$ ~ 12%	*5.654*
final energy (meV)	2.08	2.08	7.63	2.08 or 7.63	*2.56*
Q-range (Å-1)	0.1 - 1.8	0.1 - 1.8	0.7 - 3.5	~ 0.2 - 1.8	
energy resoution (µeV)	~0.75	~0.30	~ 2.0	~ 1.2 to 8	*~ 0.075*
energy transfer (µeV)	± 31	± 31	± 59	for Si 111 k_f centered: ± 200 k_f off-set: -850…1000	*< ± 5*
flux at the sample (n/cm2/s) @ 58 MW	HF: $6.0 \cdot 10^5$ LB: $8.7 \cdot 10^4$	~ factor 8 lower than standard	HF: $1.2 \cdot 10^5$	~ $1.2 \cdot 10^6$ (with 8° slit and ~ 3.5µeV resolution)	*estimated a factor 50 - 100 lower than Si 111*

IN16B @ ILL

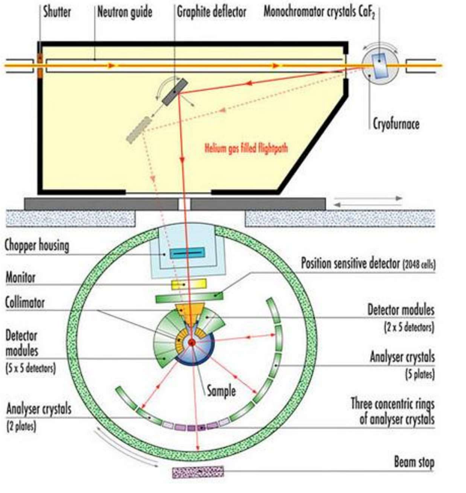
Monochromator	
$CaF_2(422)$	
temperature range	$-196 < T_M/°C < 250$
energy range	$-125 < \Delta E/\mu eV < 150$
angular range	$81° < \theta_M < 89°$
incident energy ($T_M \geq 25°C$)	16.45 meV
incident wavelength ($T_M \geq 25°C$)	2.23 Å
energy resolution	8 µeV

Sample	
sample size	3.5 x 3.5 cm²
flux at sample	2×10^4 n cm^{-2} s^{-1}

Analyser	
$CaF_2(422)$	
Q-range	$0.2 < Q/Å^{-1} < 4.9$
Q-resolution	$\Delta Q/Å^{-1} < 0.1$

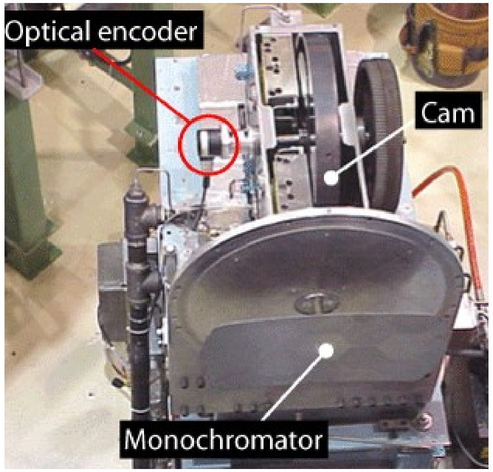
Detectors	
monitors	2
35 ^{3}He detectors	$1 < Q/Å^{-1} < 4.9$
^{3}He PSD detector	$0.2 < Q/Å^{-1} < 0.8$
background per detector	0.5 - 4 cpm

IN13 @ ILL



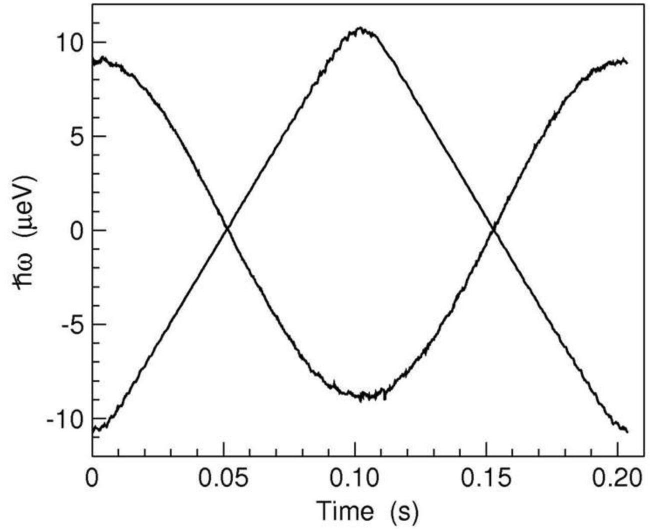
Scanning the incident energy

Initial wavelength band can be selected in two ways at a reactor source backscattering spectrometer:



Doppler shifting the energy

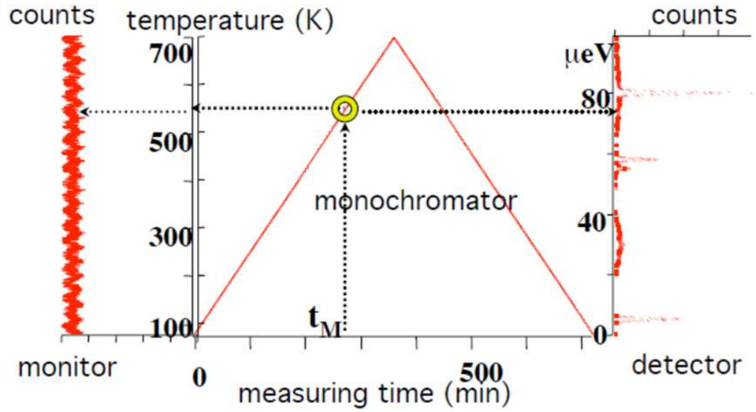
(Eg. IN16B, HFBS)



$$d(T) = d_0(1 + \alpha T + \cdots)$$

$$\frac{\delta E}{E_i} = \frac{\delta d(T)}{d_0}$$

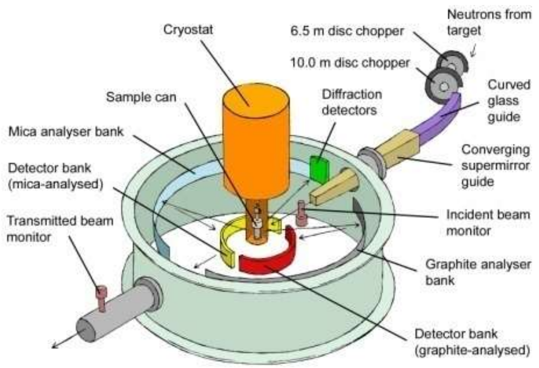
Change the monochromator temperature which changes the crystal d-spacing (eg. IN13)



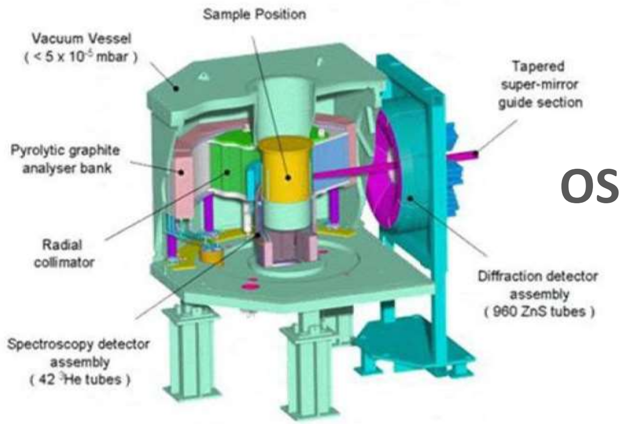
Backscattering @ Spallation Sources

Combination of time-of-flight technique plus crystal analyser backscattering.

Typical layout – consist of choppers, analyser bank of **PG002** (final energy is 1.845meV) or **Si111 crystals** (final energy is 2.08meV), ^{3}He (or scintillator) detectors, typically covering 15 to 150deg. Tank is evacuated. For PG need cooling and Be filter.



IRIS @ ISIS



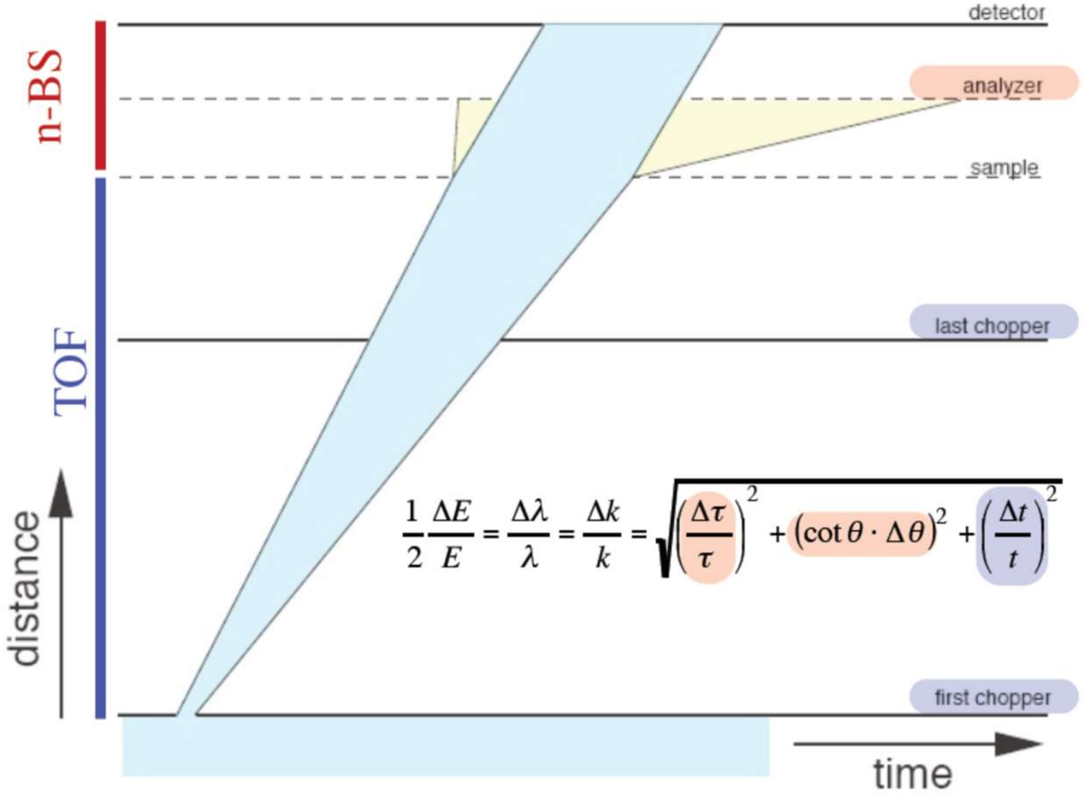
OSIRIS @ ISIS

BASIS @ SNS

DNA @ SNS

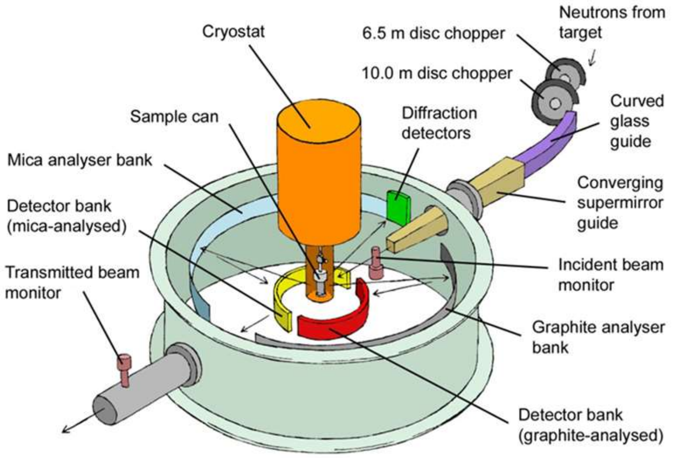
Backscattering @ Spallation Sources

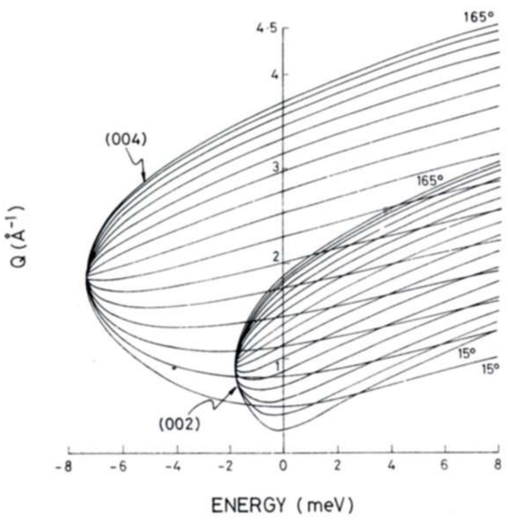
Combination of time-of-flight technique plus crystal analyser backscattering.
Bandwith set by choppers.



- Poorer resolution than reactor backscattering (near backscattering) plus other contributions to the resolution – need long L1, but still higher than direct.
- Larger ΔE but similar Q range
- Limited flexibility in Q-E space
- Picosecond-nanosecond
- Again, be flexible with choice of crystal

Backscattering @ Spallation Sources





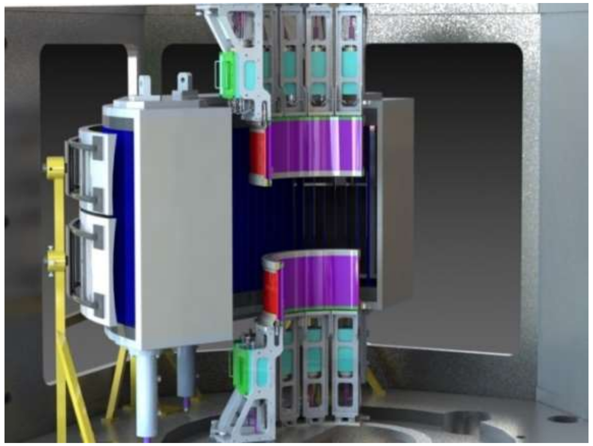
	PG 002	PG 004	Mica 002	Mica 004	Mica 006
Analysing Energy (meV)	1.84	7.38	0.207	0.826	1.86
Dynamic Range (meV)	-0.4 to +0.4	-3.5 to +4.0	-0.02 to +0.02	-0.15 to +0.15	-0.4 to +0.4
Resolution (μeV)	17.5	54.5	1.0	4.5	11.0
Q-range ($Å^{-1}$)	0.42 to 1.85	0.84 to 3.70	0.13 to 0.62	0.26 to 1.24	0.40 to 1.87

In addition, running the choppers at for eg. 16Hz we are able to open the window to -1 to 30meV

7 ^{3}He diffraction detectors at $2\theta \approx 170°$

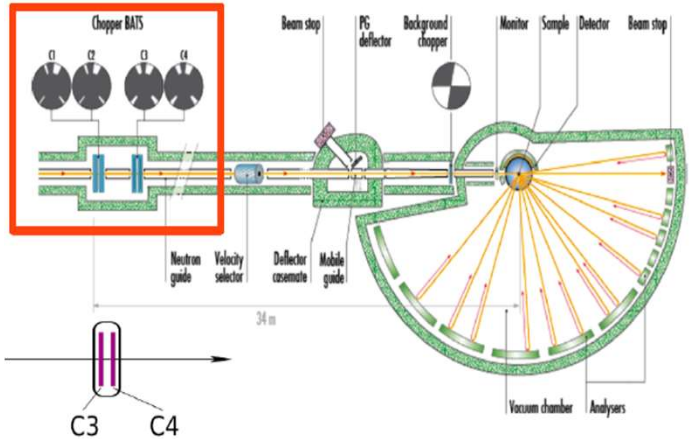
Reactor meets Spallation

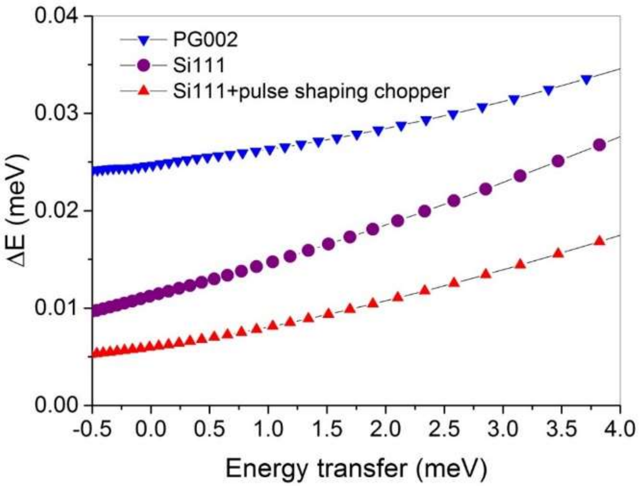
Seeing a trend towards widening the accessible time scale of backscattering instruments. Towards Si111 spallation sources and innovative use of choppers on IN16B to create pulses and wider energy widnow.

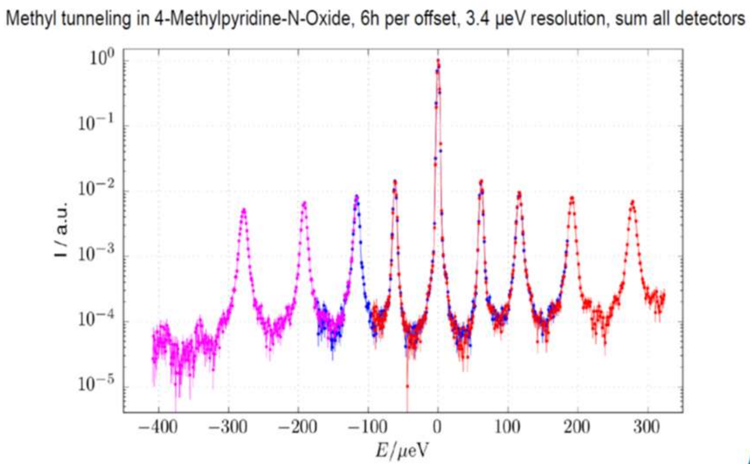


OSIRIS upgrade
5-5000ueV

BATS option on IN16B

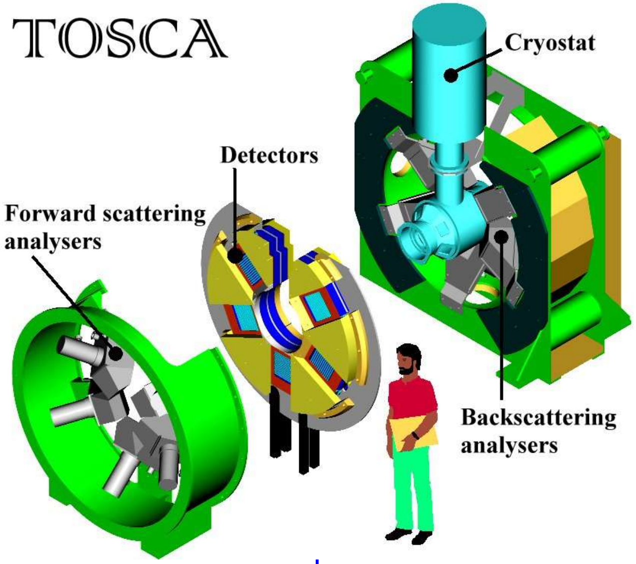


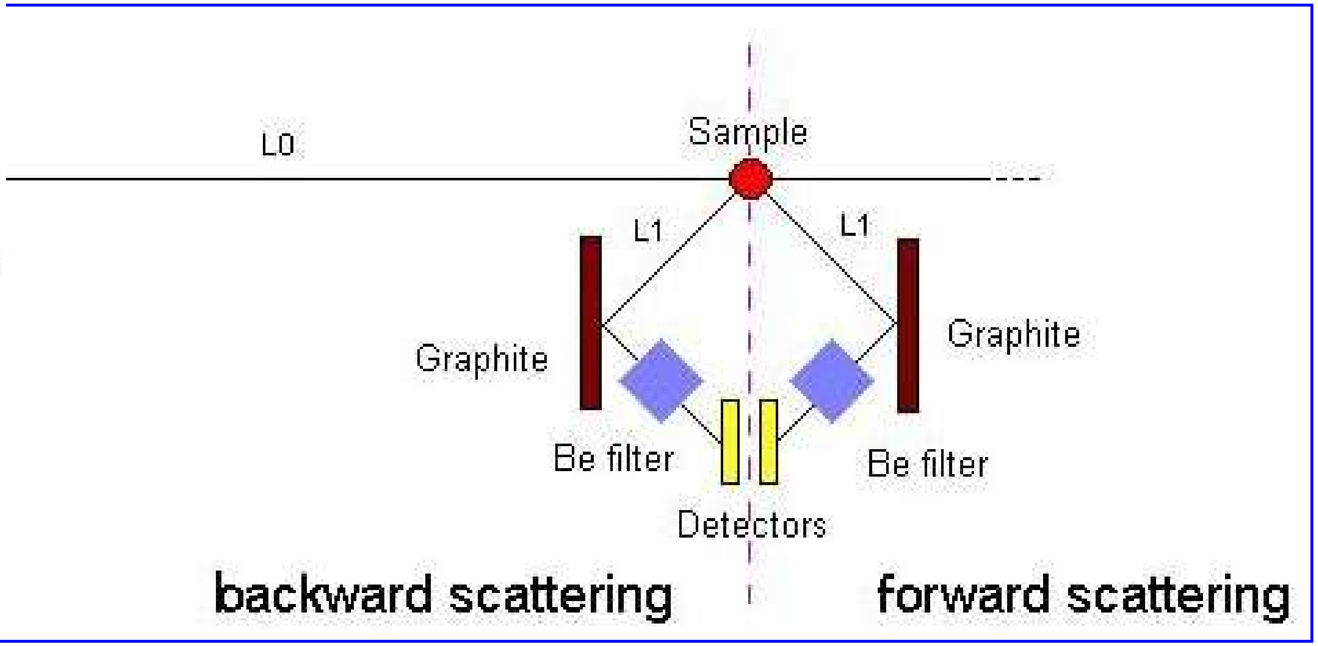


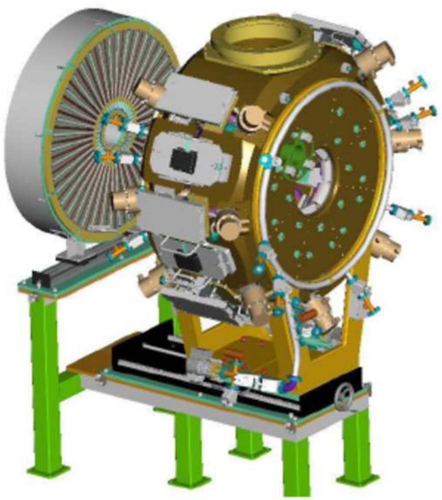


Inverted Geometry- Vibrational Spectrometers

A white beam is sent to the sample and a small portion that are scattered at 45deg or 145deg impinge on the analyser crystals. Those satisfying the PG 002 Bragg condition on the analyser are selected.





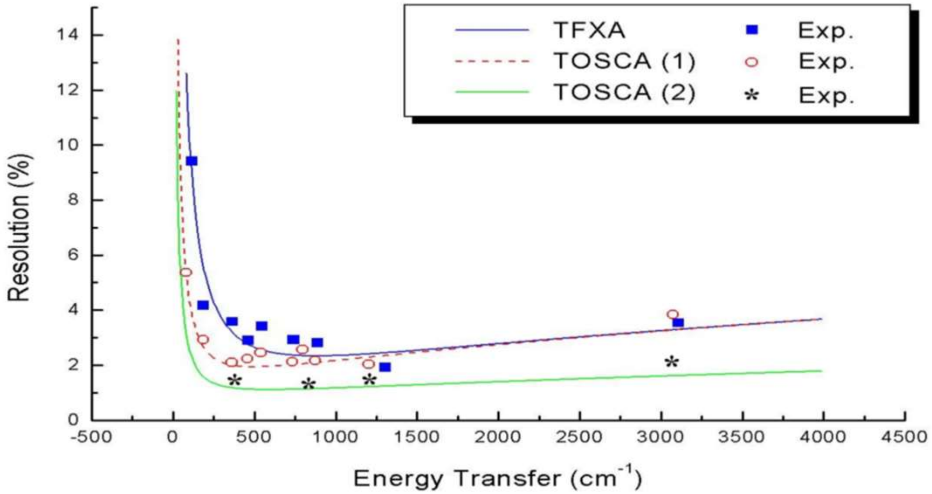


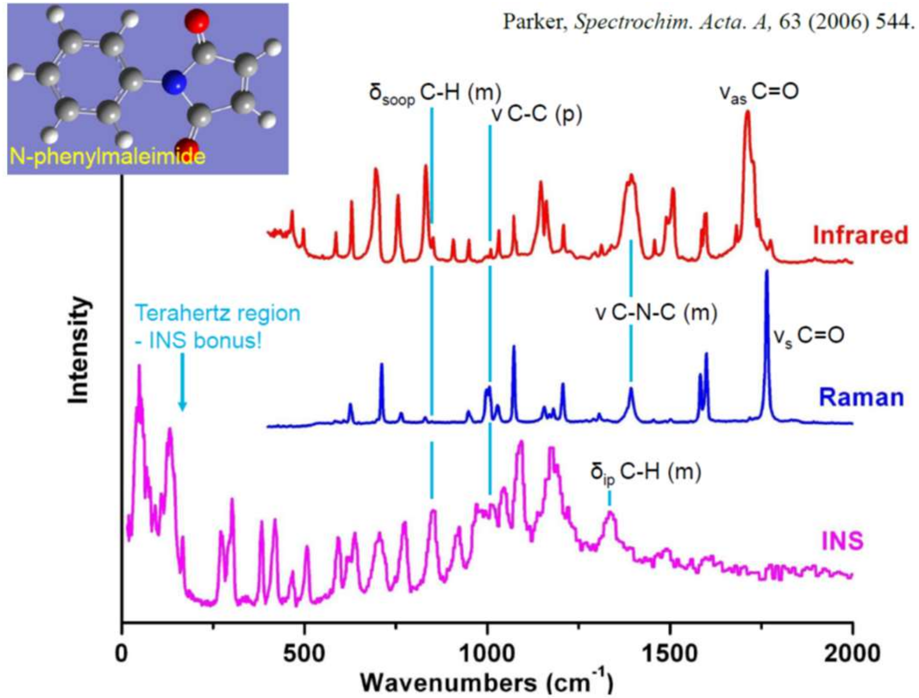
VISION @ SNS

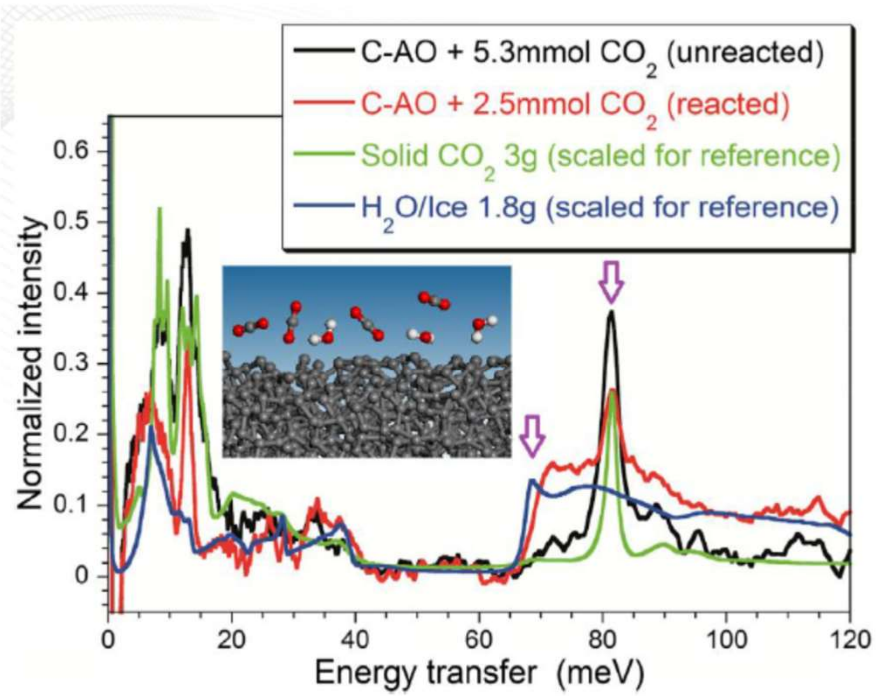
$$t = t_i + t_f = \frac{L_0}{v_0} + \frac{L_1}{v_1}$$

$$v = \sqrt{2E/m}$$

Inverted Geometry- Vibrational Spectrometers







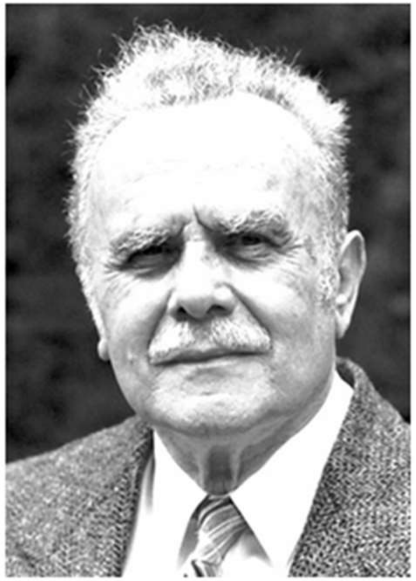
- Resolution is almost constant with energy transfer.
- Very broad energy transfer range (0-120meV)
- Used to be limited by sample mass, but great achievements lately (mg)

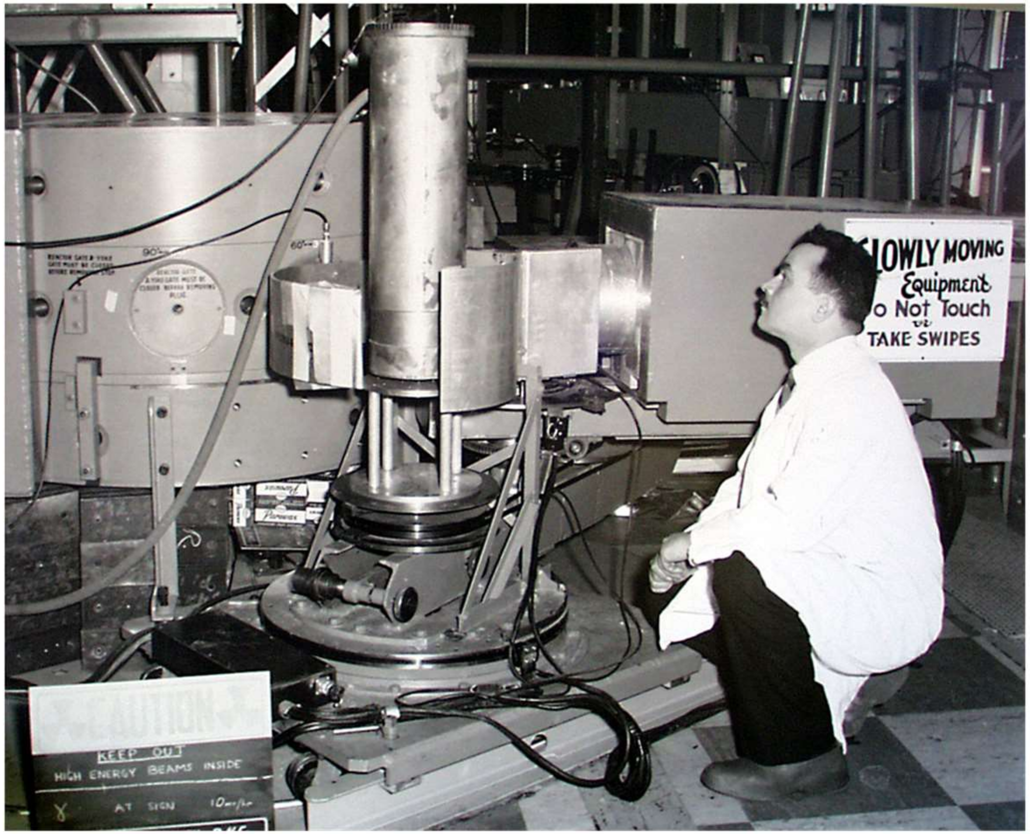
Triple-Axis Spectrometers

Triple-axis spectrometers @ reactors (e.g. ILL)

The Nobel Prize in Physics 1994
Bertram N. Brockhouse, Clifford G. Shull

Bertram N. Brockhouse - Facts





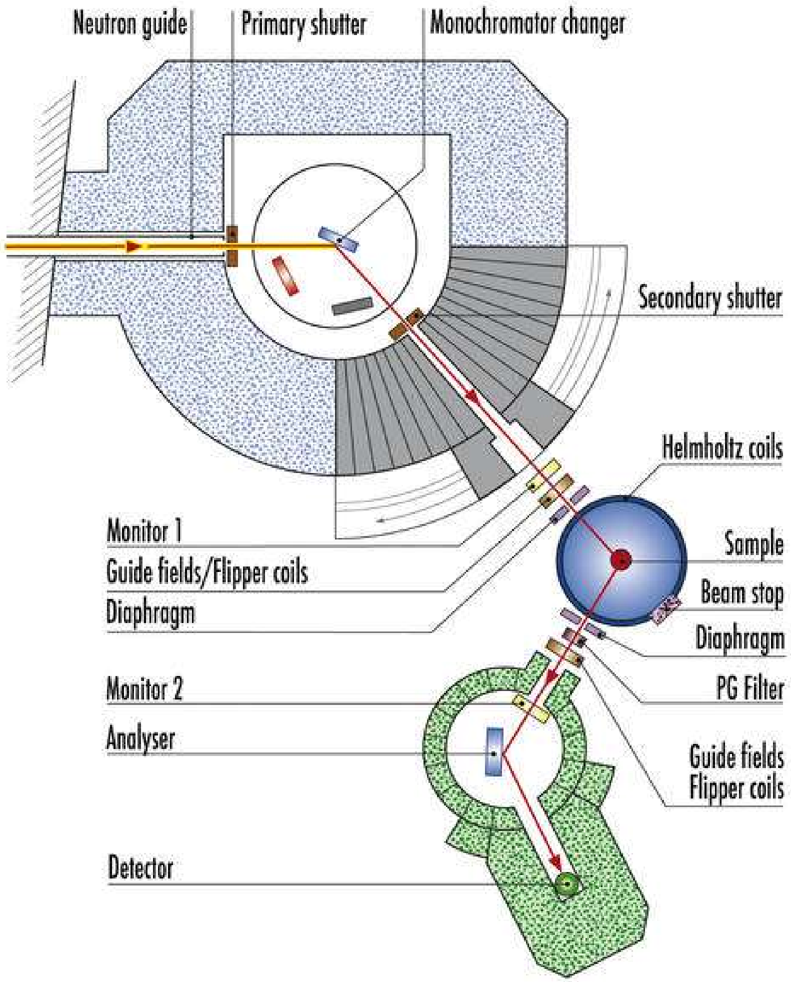
Source: Atomic Energy of Canada Limited, Chalk River, Ontario (CC BY-NC-ND 2.0)

Bertram N. Brockhouse. *Nobelprize.org.* Nobel Media AB 2013.

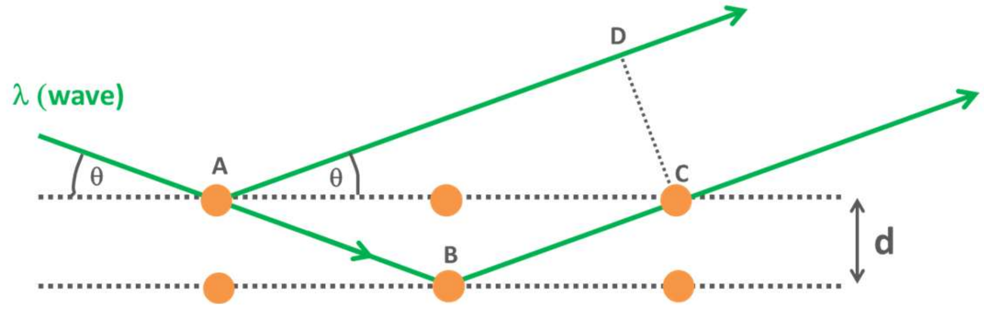
Triple-Axis Spectrometers

Using Bragg diffraction -> using crystals as monochromators and analysers.

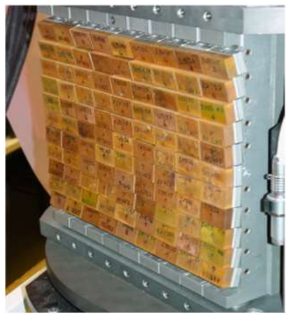
Selecting a specific neutron wavelength through Bragg reflection (either E_i or E_f)



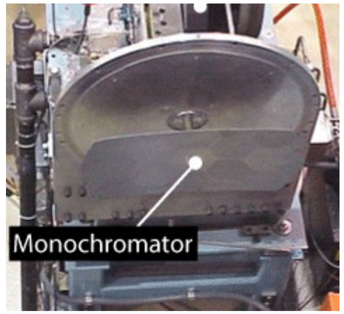
Bragg's Law $n\lambda = 2d\sin\theta$



Graphite 002
d = 3.335 Å



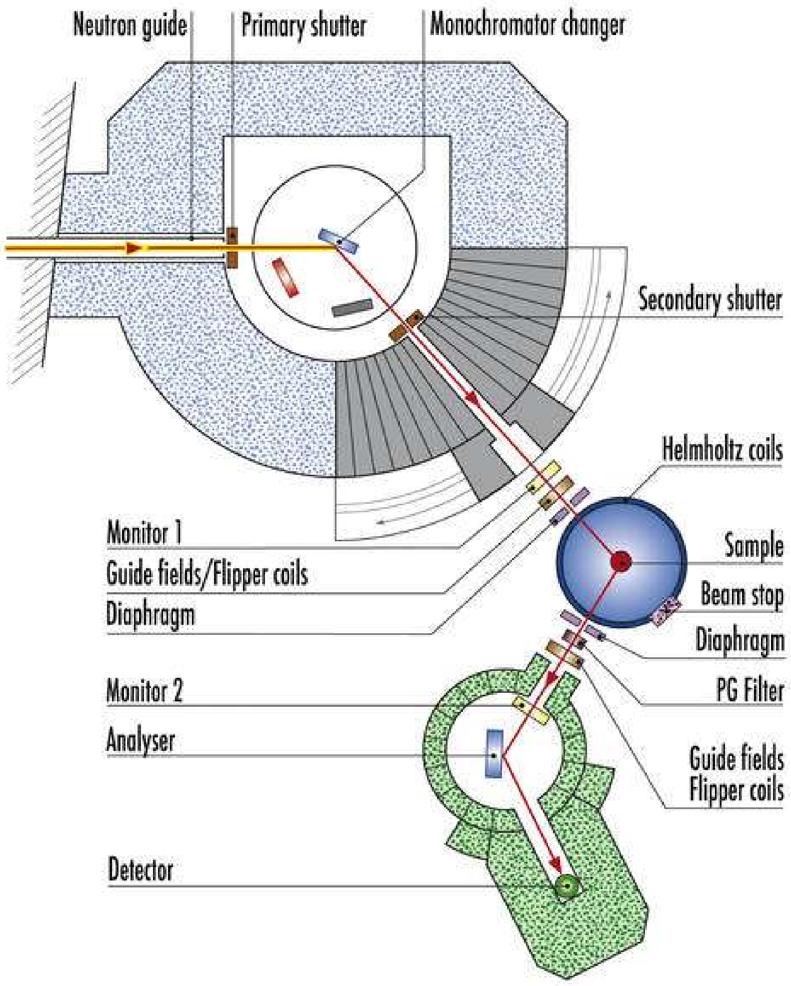
Copper 200
d = 1.807 Å



Si 111
d = 3.135 Å

Triple-Axis Spectrometers

Triple-axis spectrometers @ reactors (e.g. ILL)



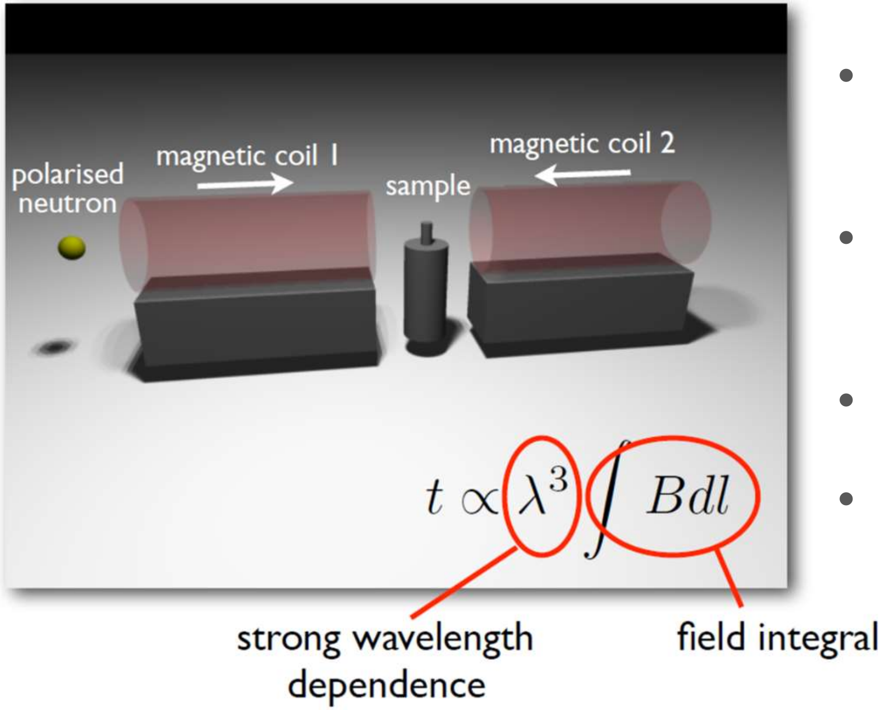
IN22 @ ILL

- Good for single-crystal excitations
- Very flexible
- Can focus all intensity on a single point in reciprocal space
- Can scan either constant ***Q*** or constant *E* depending on excitation
- Can use multiplexing and polarisation analysis
- Can be slow
- Crystals can give rise to higher-order effects and spurions
- Measurement along high-symmetry directions
- ‘Coarse’ E resolution

Neutron Spin-Echo

Neutron Spin-Echo

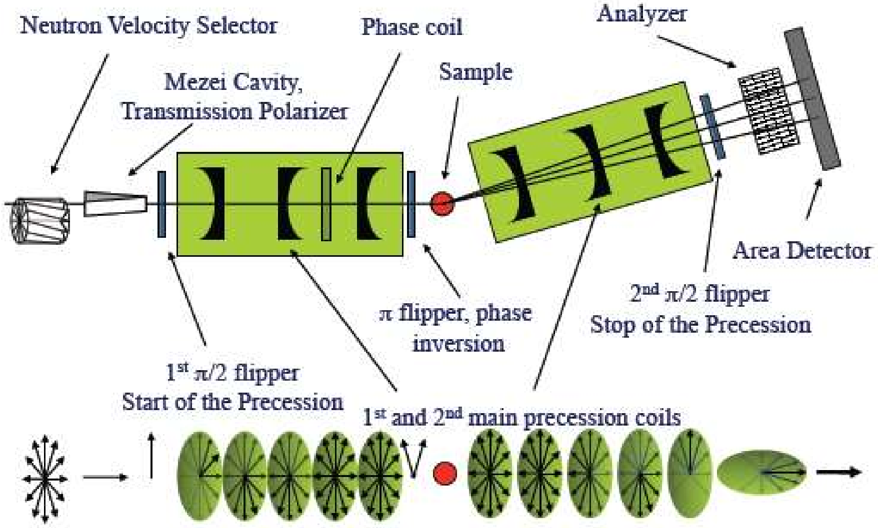
- NSE uses the neutron's **spin polarisation** to encode the difference in energies between incident and scattered beams.
- Neutron perform **Larmor precessions** in two antiparallel magnetic fields, before and after the sample, resulting in polarization of the neutrons.
- Precession angles are equal and opposite and the difference is analysed at the detector. Small energy transfers lead to a change in the precession angle and thus a decrease in measured polarization.



- NSE is the neutron spectroscopy with highest energy resolution
- Time covered is 1ps < t < 1us (equivalent to neV resolution)
- Momentum transfer range is $0.01 < Q < 4\text{Ang}^{-1}$
- Unlike other neutron spectrometers, measures in I(Q,t), real time.

Neutron Spin-Echo

- Velocity selector determines mean velocity with a spread of 10-20%
- Beam goes through polarizer with all spins aligned to velocity direction
- First $\pi/2$ flipper changes the spin to be perpendicular to magnetic filed of the coils and precession starts
- Neutron precesses and faster neutrons will spend less time than slow neutrons and their final precession angle will be smaller
- π flipper rotates the spin direction of 180deg around the z-axis and y component changes sign (B_{in} and B_{out} are parallel).
- Neutron interacts with the sample and exchange momentum and energy, changing direction and velocity.
- Apply equivalent field, those exchange will not regain initial polarisation. Second $\pi/2$ flipper stops precession.



NG5 @ NIST

precession angle

Field integral

$$\varphi = \gamma_L \frac{\int \vec{B} \cdot d\vec{l}}{v}$$

neutron gyromagnetic ratio

neutron velocity

Neutron Spin-Echo

In neutron experiment we measure the **polarisation P** over all precession angles:

$$P_x = \langle \cos\varphi \rangle = \langle \cos(\varphi_{in} - \varphi_{out}) \rangle$$

Precession angles in and out are given as before and after scattering from the sample, related to their respective velocities:

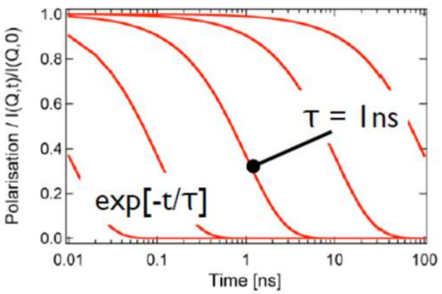
$$P_x = \langle \cos[\gamma_L(\frac{\int \vec{B}_{in}\cdot d\vec{l}}{v_{in}} - \frac{\int \vec{B}_{out}\cdot d\vec{l}}{v_{out}})] \rangle$$

To first order the angle is proportional to the energy transfer at the sample with the proportionally constant being the **spin echo time *t***:

$$\varphi = t\omega$$

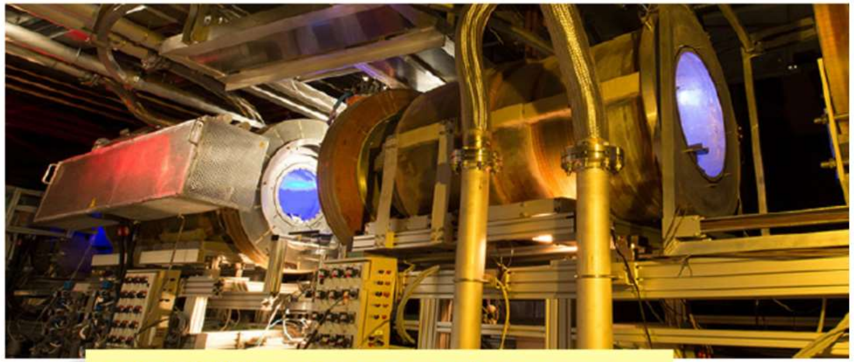
The polarisation is also related to the scattering function and if QENS signal is a Lorentzian, the the Fourier transform is an exponential decay:

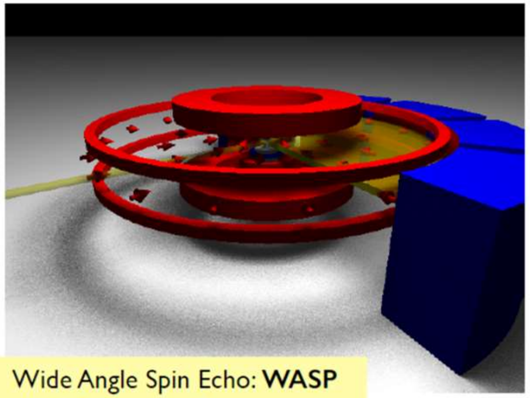
$$P_x(Q,t) = \frac{\int S(Q,\omega)\cos(\omega t)d\omega}{\int S(Q,\omega)d\omega} \qquad P_x(Q,t) = \frac{\int [\Gamma^2 + \omega^2]^{-1}\cos(\omega t)d\omega}{\int [\Gamma^2+\omega^2]^{-1}d\omega} = e^{-\Gamma t} = e^{-t/\tau}$$



$$t = \frac{\varphi}{\omega} = \frac{\hbar}{m}\frac{\gamma_L \int \vec{B}\cdot d\vec{l}}{\bar{v}^3} = \frac{m^2\gamma_L \int \vec{B}\cdot d\vec{l}}{2\pi h^2}\lambda^3$$

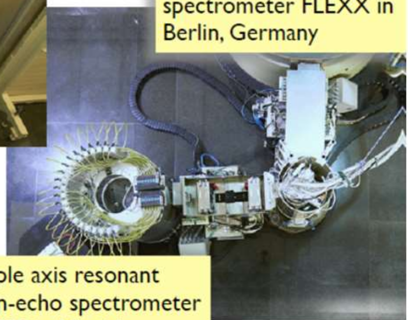
Neutron Spin-Echo Spectrometers


Today's IN15: measures up to 1 μs


Wide Angle Spin Echo: **WASP** (ILL, Millennium Programme)

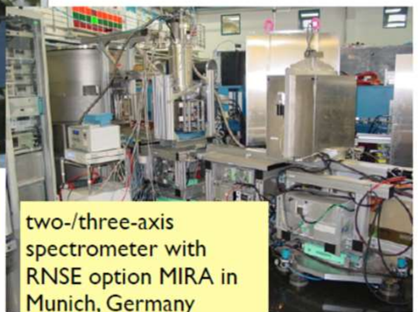

High resolution NSE at NIST, USA


Resonant NSE RESEDA in Munich, Germany


RNSE option on TAS spectrometer FLEXX in Berlin, Germany

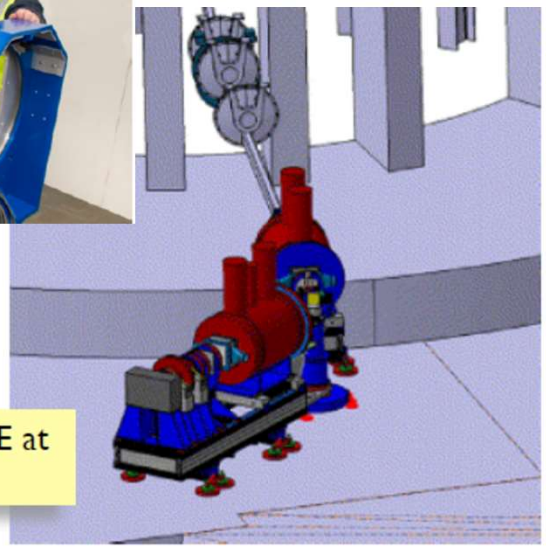
Triple axis resonant spin-echo spectrometer TRISP in Munich, Germany


High resolution spectrometer J-NSE in Munich, Germany


two-/three-axis spectrometer with RNSE option MIRA in Munich, Germany

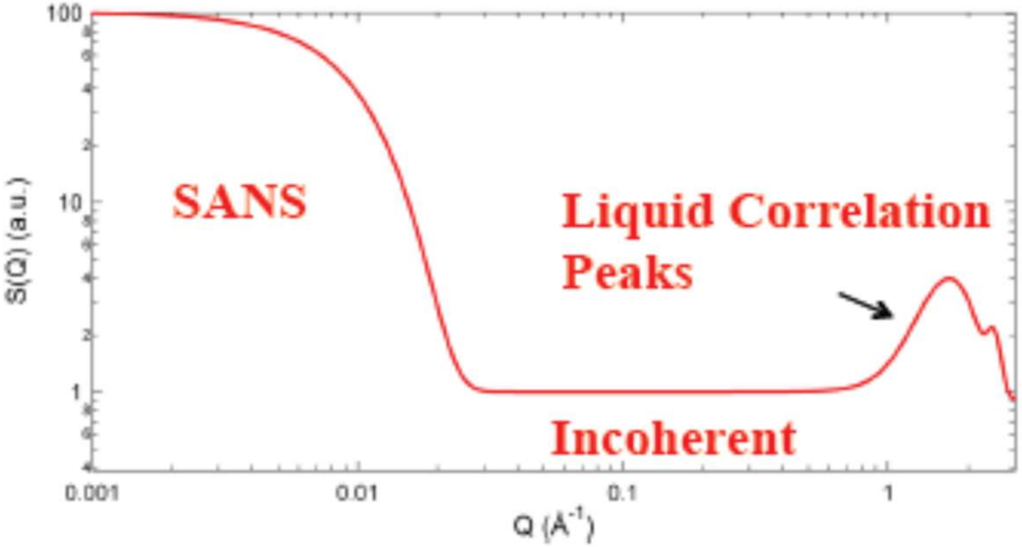

Spin-echo SANS with optional NSE OFFSEC at ISIS, UK

Traditionally @ reactor sources


High resolution NSE at SNS, USA

Neutron Spin-Echo

The best signal in NSE is at the peaks and is very complementary to the SANS structural information.

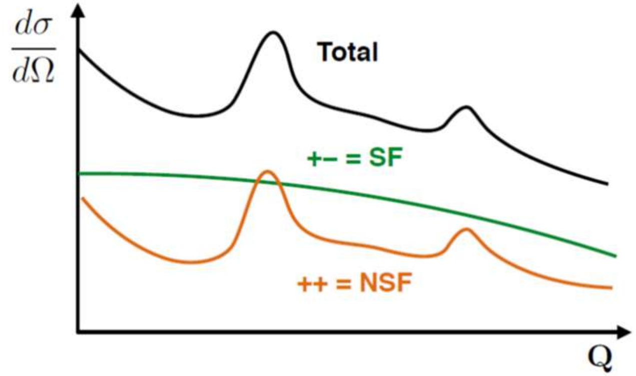


$$I_{\mathrm{NSE}}(Q,t) = \frac{I_{\mathrm{coh}}(Q,t) - \frac{1}{3}I_{\mathrm{inc}}(Q,t)}{I_{\mathrm{coh}}(Q,0) - \frac{1}{3}I_{\mathrm{inc}}(Q,0)}$$

$$\boxed{b_{coh} = \overline{\mathbf{b}}; \quad b_{inc} = \sqrt{\overline{\mathbf{b}^2} - \overline{\mathbf{b}}^2}}$$

Nuclear coh.
Isotope inc.

Spin inc.
$\sqrt{B^2 I(I+1)}$



NSF: $\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{++} = \left(\frac{d\sigma}{d\Omega}\right)_{--}} = \left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{coh+II}} + \frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}$

SF: $\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{+-} = \left(\frac{d\sigma}{d\Omega}\right)_{-+}} = \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}$

$$\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{++}} = \boxed{\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{coh+II}}} + \boxed{\frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}}$$

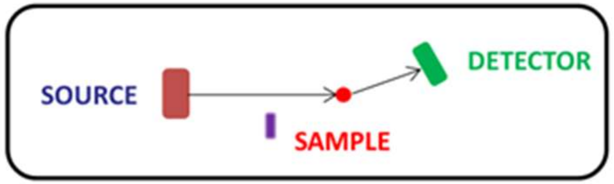
$$\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{+-}} = \boxed{\frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}}$$

Neutron Spectrometers – Which one?

Neutron Spectrometers

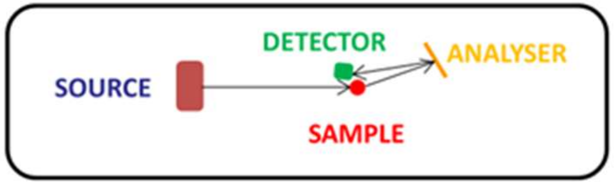
Frequency domain S(Q,ω)

Direct geometry



- Lower energy resolution
- Larger DE and Q range
- Flexible in choosing Q-E space
- Good Q resolution
- Picosecond
- Self-correlation function (H)
- IN5, LET, CNCS, MAPS, MERLIN
- For INS better for higher energy features and more flexible tuning.
- Polarisation analysis available

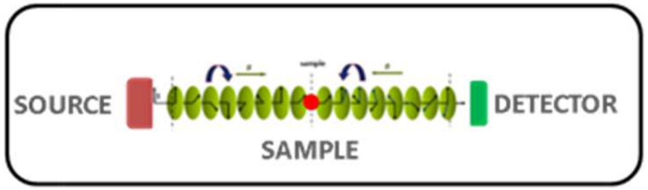
Indirect geometry or Backscattering



- Higher energy resolution
- Smaller DE and Q range
- Fixed Q-E space
- Picosecond-nanosecond
- Self-correlation function (H)
- Coarse Q resolution
- DNA, IRIS, IN16B, HFBS, VISION, TOSCA
- For INS excellent resolution and sensitivity below ~ 2000cm^{-1}
- Polarisation analysis under development

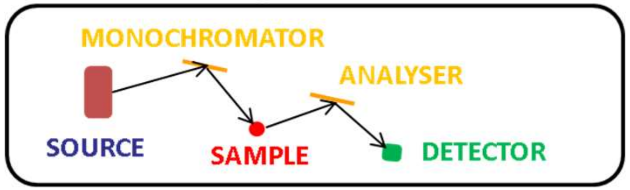
Time domain I(Q,t)

Neutron Spin Echo



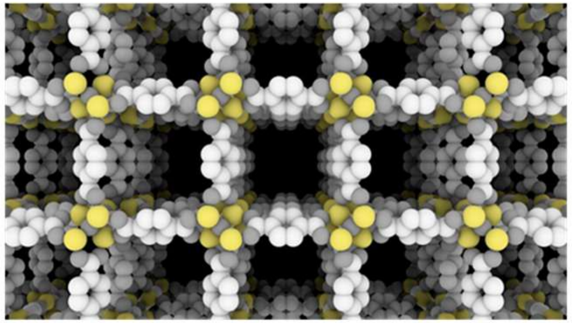
- Highest energy resolution
- Large dynamic and Q range
- Nanosecond dynamics
- Collective dynamics (H/D) normally
- Q resolution not very high generally
- Signal is often weak
- IN15, NIST-NSE
- Polarisation analysis in-built

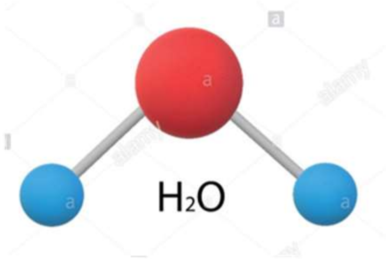
Triple-Axis

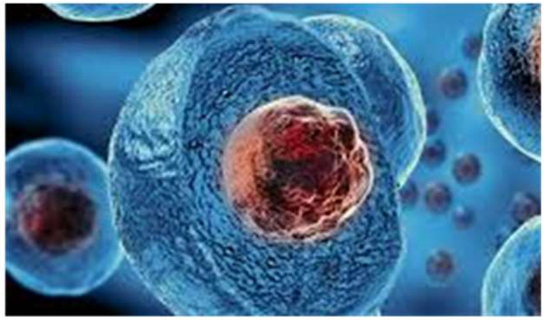


- High flux but point by point (compare with DG)
- Cold and hot neutrons, mainly high energies
- Used for collective excitations and magnetic excitations
- Polarisation analysis available
- Coarser resolution and control over Qresolution

Neutron Spectrometers – Which one?







Totally depends on the problem!

You need to know a certain amount about your system before embarking in neutron spectroscopy

Talk to the facility scientists!

Further Reading & Credits

- Neutron Scattering: A Primer by Roger Pynn (1990): http://library.lanl.gov/cgi-bin/getfile?19-01.pdf and http://la-science.lanl.gov/cat_materials.shtml#neutron
- Experimental Neutron Scattering: B. T. M. Willis and C. J. Carlile, OUP (2013); http://global.oup.com/academic/product/experimental-neutron-scattering-9780199673773
- J. S. Gardner, Ehlers, G. , Faraone, A. and Garcia Sakai, V. (2020), High-Resolution Neutron Spectroscopy using Backscattering and Neutron Spin-Echo Spectrometers in Soft and Hard Condensed Matter, Nature Reviews Physics.
- F. Mezei, C. Pappas, T. Gutberlet (Eds.): “Neutron Spin-Echo Spectroscopy (2nd workshop)”, Lecture Notes in Physics 601, Springer, Heidelberg, (2003)
- … and many others

These lectures have been made using material from many people including Ken Andersen, Ian Anderson, Timmy Ramirez-Cuesta, Stewart Parker, Goran Nilsen, Peter Fouquet, Ross Stewart, Bernhard Frick, Dan Neumann.