Inelastic neutron scattering

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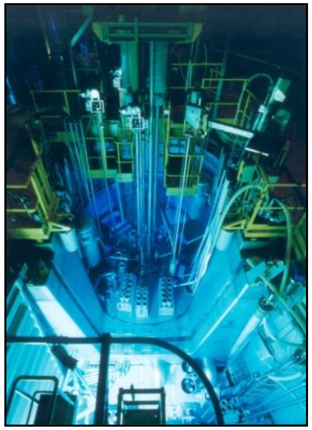
Why neutrons ?

Crystalline and magnetic structures

Elementary excitations in solids

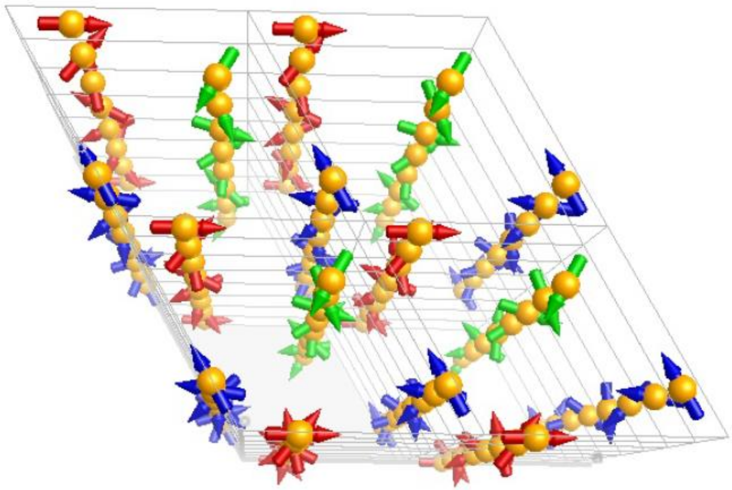


Large scale structures

Reflectometry (surface)

Quasi-elastic scattering

Diffraction



Crystalline and magnetic structures

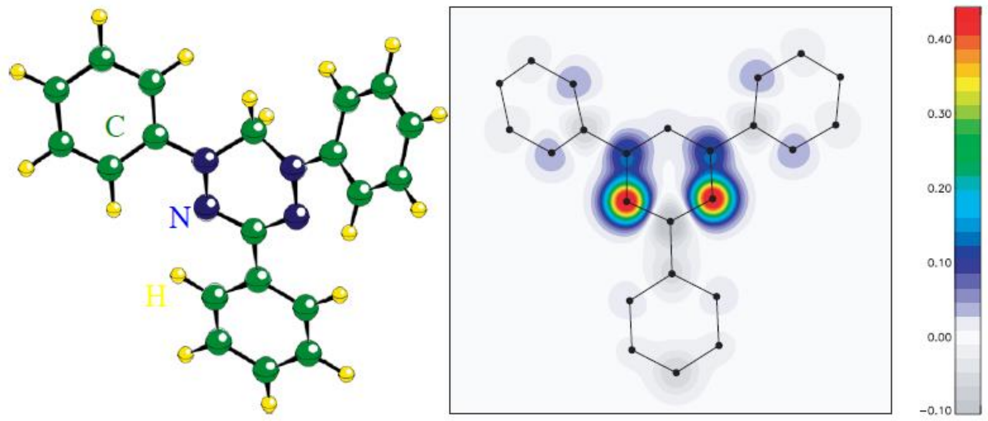


Fig. 4. View of the TPV molecule (left) and experimental magnetization distribution (right) as measured by polarized neutron diffraction.

Inelastic neutron scattering

The structure factor gives information about the structure:

$$F(Q) = \sum_{\ell} b_\ell \, e^{iQ\,r_\ell} e^{-W_\ell}$$

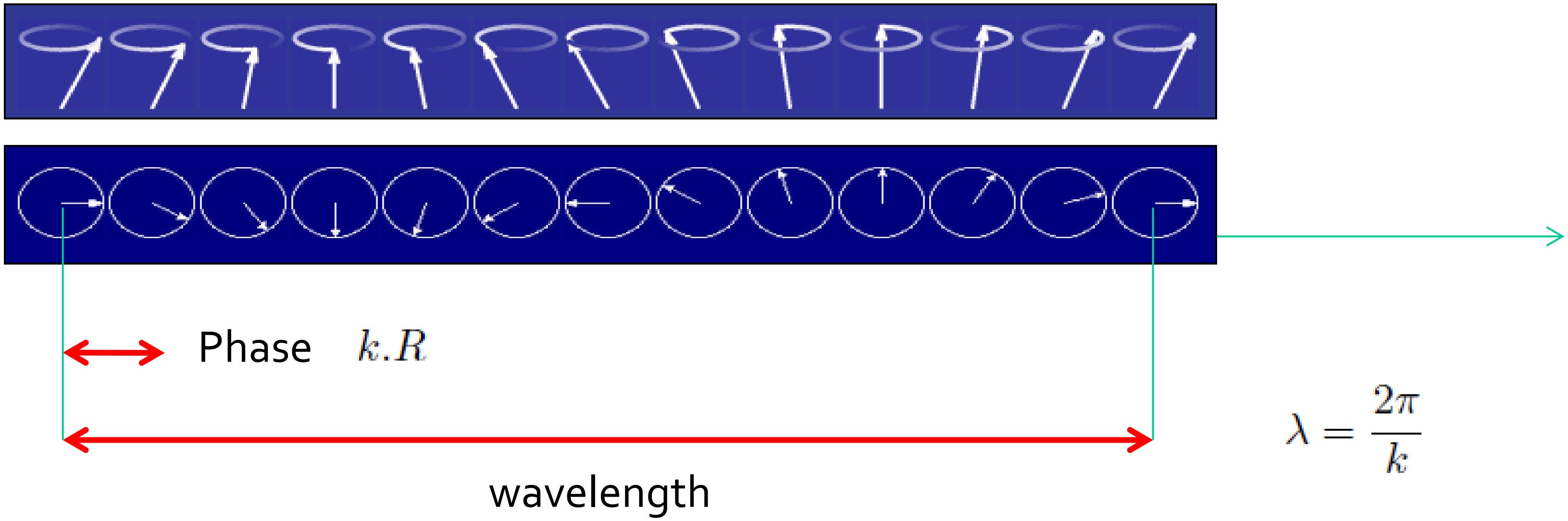
$$\vec{F}(Q) = \sum_{\ell} \vec{S}_{\perp,\ell} \, e^{iQ\,r_\ell} \, e^{-W_\ell}$$

What about the terms that stabilize this structure ?
What about the Hamiltonian of the system ?

Investigate the excited states ....

Example in magnetism

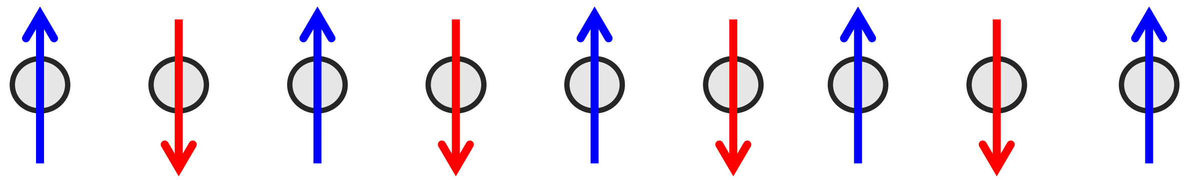
Classical elementary magnetic excitations are spin waves



$$\lambda = \frac{2\pi}{k}$$

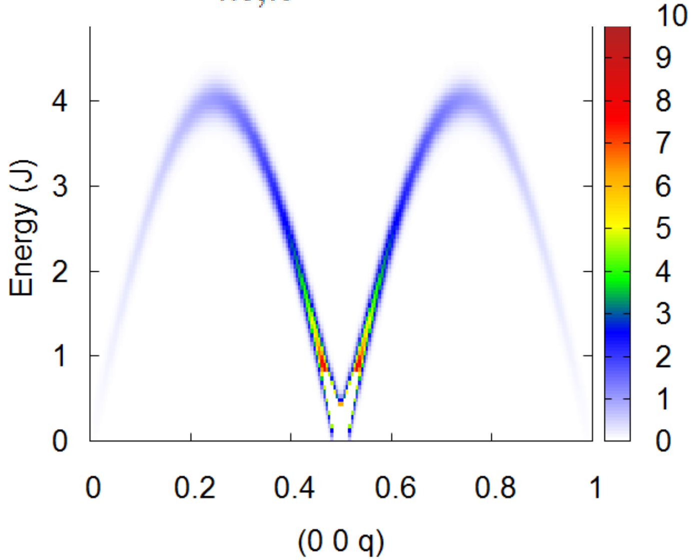
The dispersion relation connects the wavevector k and the frequency ω (energy)
Neutron directly measure ω(k)

Example in magnetism

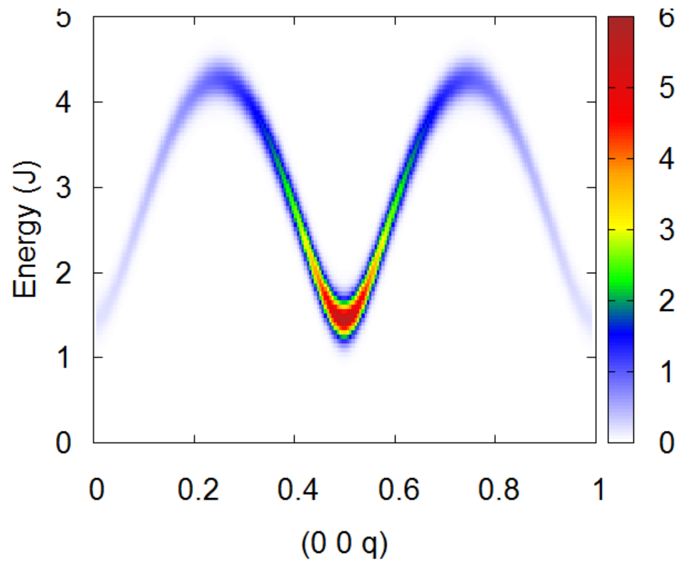


Same ground state configurations but different spectra (spin gap)

$$\mathcal{H} = \sum_{m,n} J_{m,n}\ \vec{S}_m.\vec{S}_n$$

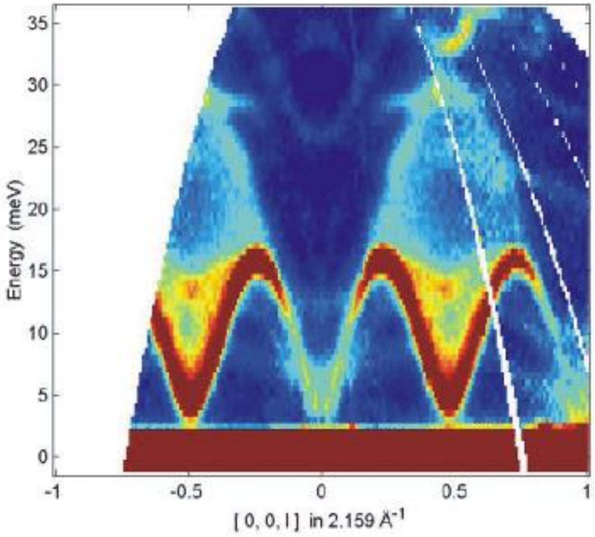


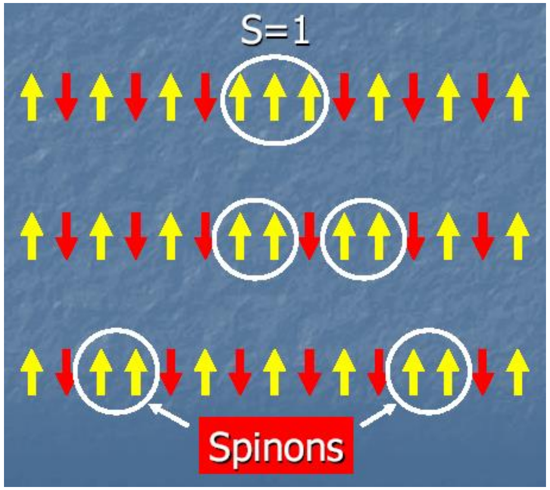
$$\mathcal{H} = \sum_{m,n} J_{m,n}\ \vec{S}_m.\vec{S}_n\ + \sum_{m} D\ (S_m^z)^2$$

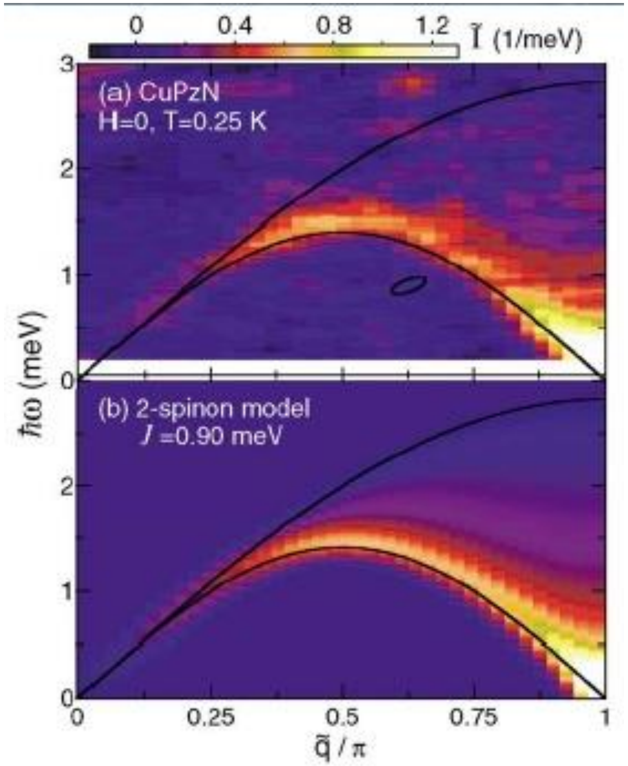


Example in magnetism

Unconventional magnetic excitations in 1D systems

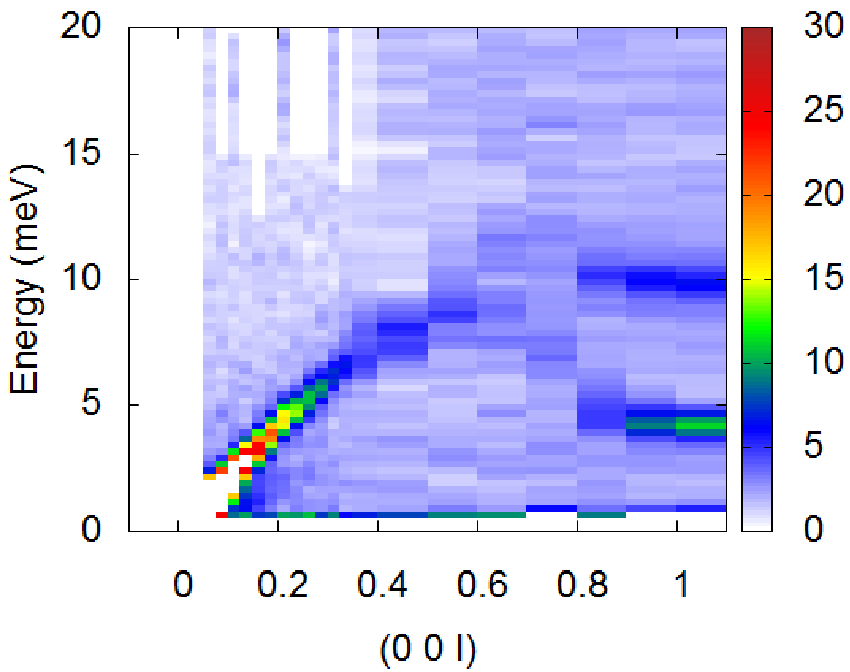




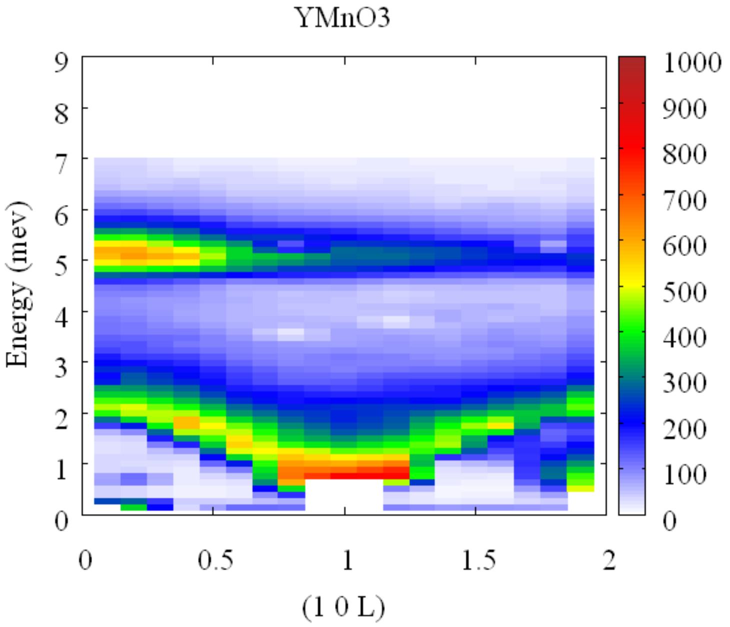


Lattice dynamics

Lattice dynamics in PbTe

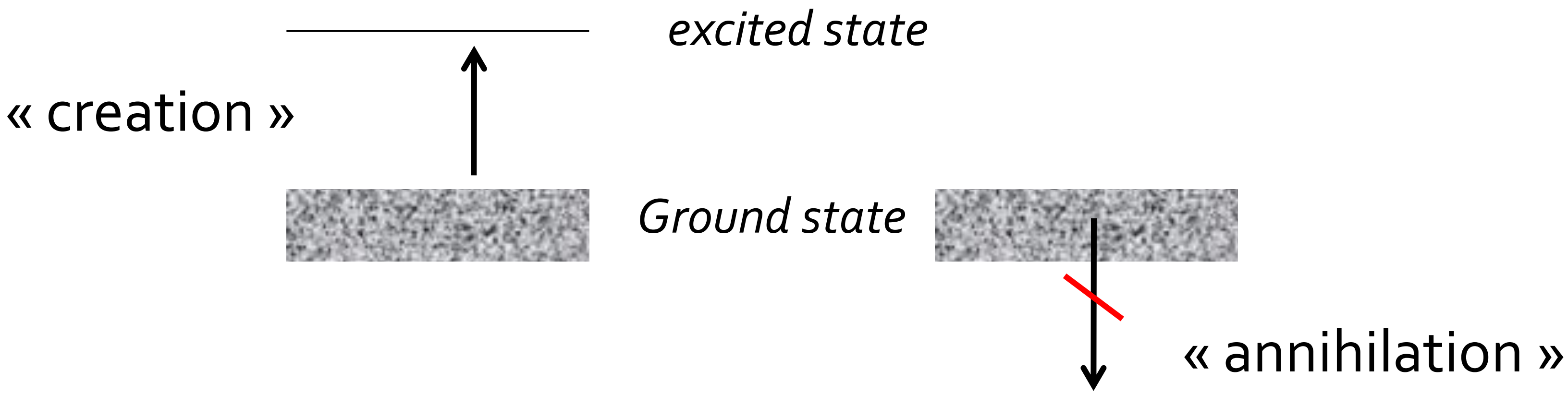


Spin dynamics in YMnO3



Detailed balance

At T=0 K: the system is in its ground state



Absence of symmetry between creation and annihilation processes

- No charge
- Massive particle

$$E = \frac{1}{2}mv^2$$

$$mv = \hbar k$$

$$E = \frac{\hbar^2 k^2}{2m}$$

- Spin ½

$$S = 1/2$$
$$S_z = \pm 1/2$$

- Interact with matter : nuclear & magnetic contributions
- Wavelength of thermal neutron : λ ~ a few Å $$\lambda = \frac{2\pi}{k}$$
- Energy of thermal neutrons : E ~ a few meV

Diffraction and spectroscopy are possible by virtue of both their Nuclear and Magnetic interaction with matter.

- Bulk and not surface
- Light elements
- Magnetism, spins orientation

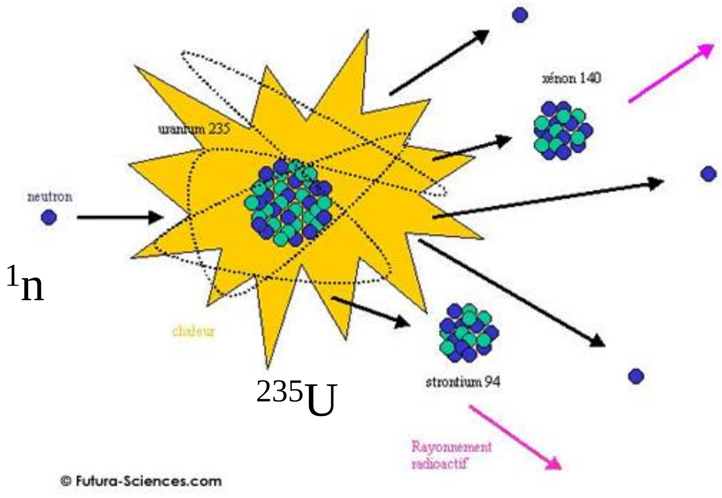
Neutron sources

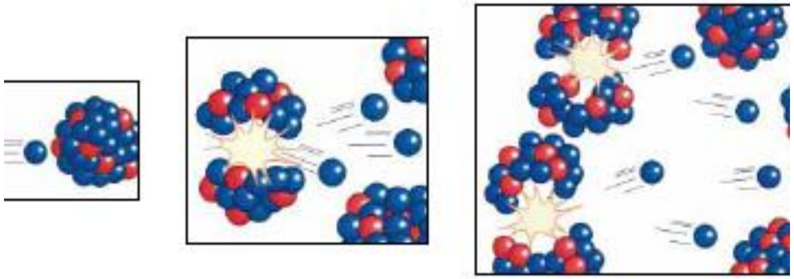
In Europe:
Reactors
ILL-Grenoble (France)
LLB-Saclay (France)
FRMII-Munich (Germany)
HMI-Berlin (Germany)
Spallation sources
ISIS-Didcot (UK)
PSI-Villigen (Switzerland)

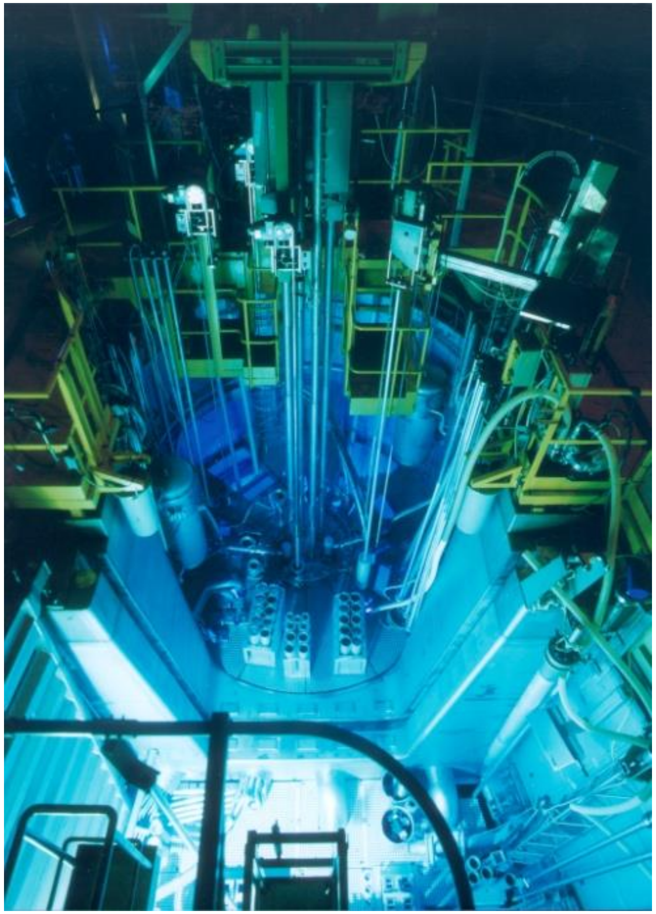
But also:

Dubna (Russia), JPARC (Japan)
SNS, DOE labs (USA), ANSTO (Australia)
Canada, India, …

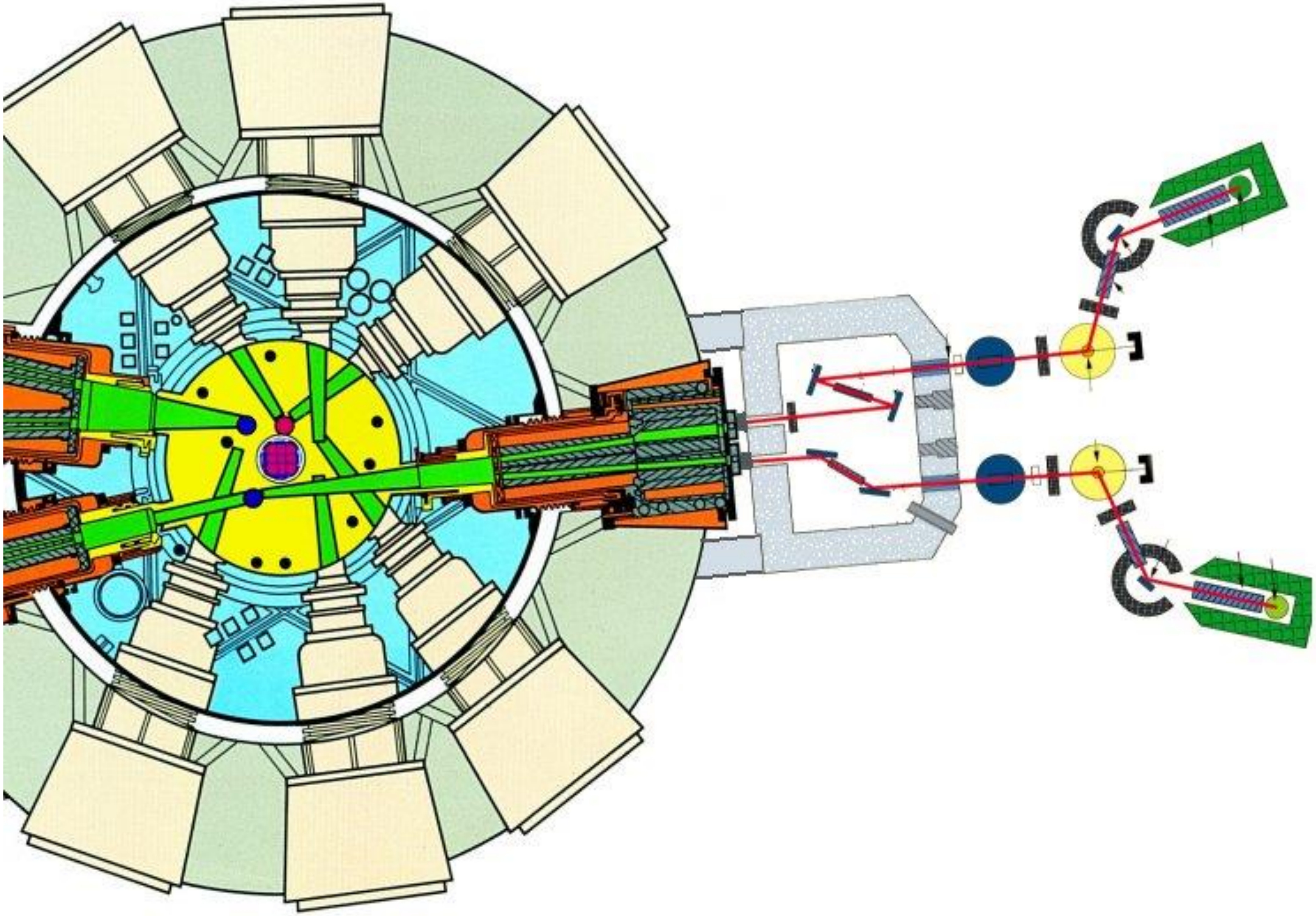
Neutron sources







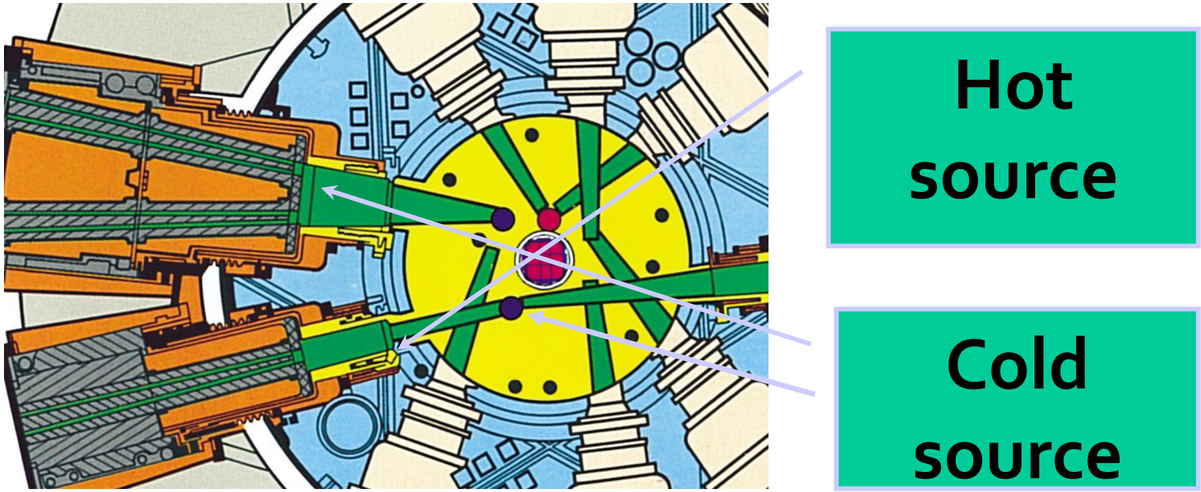
LLB



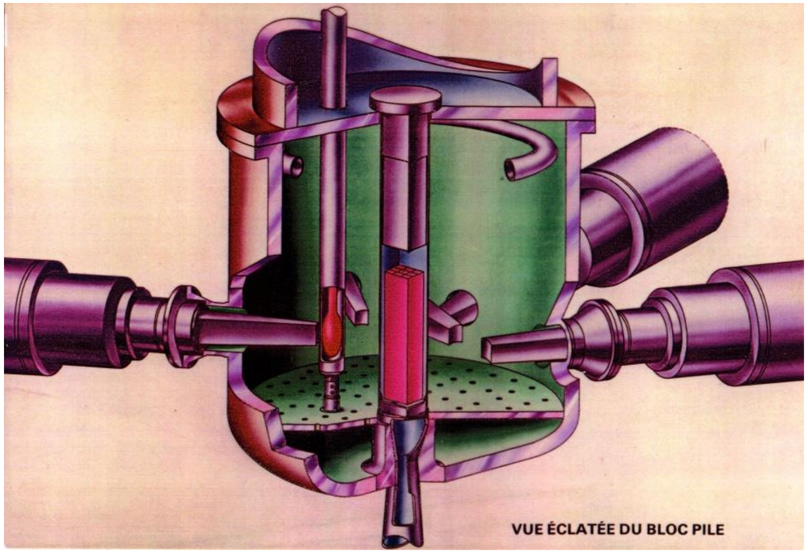
LLB

<u>9 beam tubes</u>

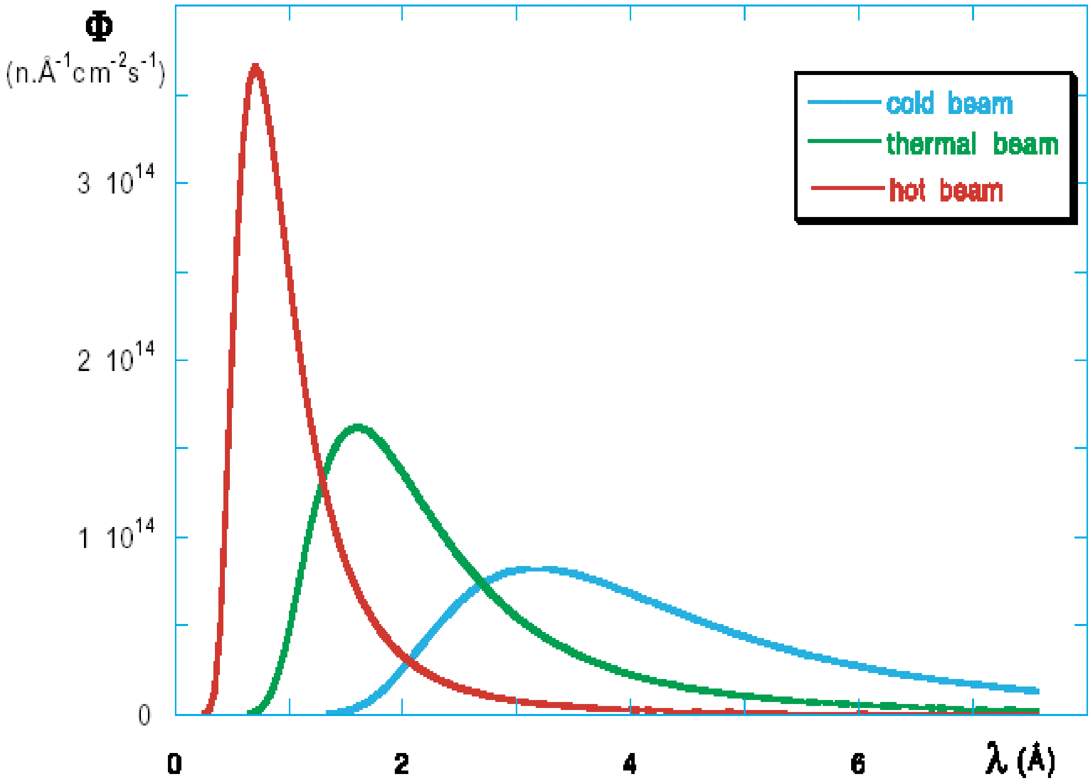
- 4 thermal
- 2 hot
- 3 cold



9 guides for cold neutrons



Hot, thermal and cold « sources »



Differential flux (per Å) given by local moderators

Hot Neutrons

Liquid and single crystal structures

Thermal Neutrons

Condensed matter

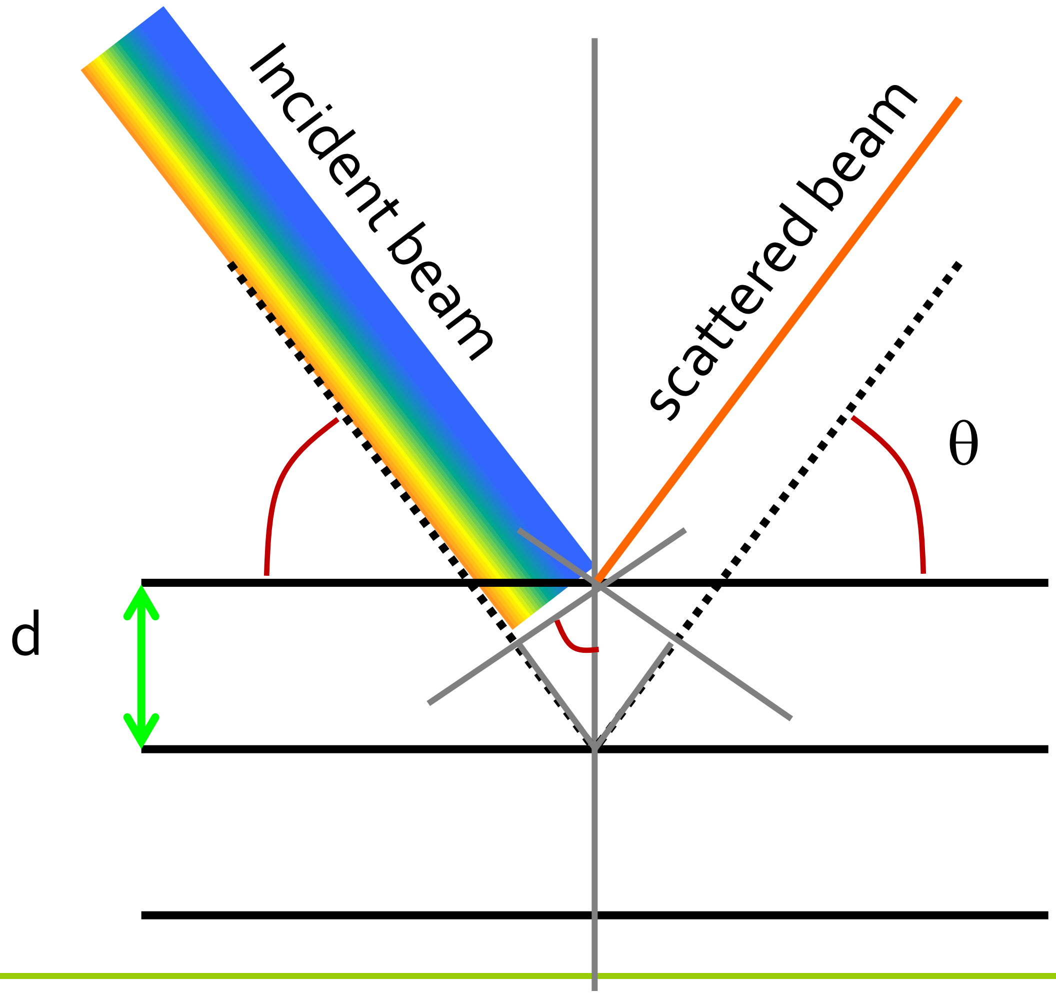
Cold neutrons

Condensed matter

Large scales structures

How to select an incident energy ?

The reactor provides a spectrum of neutrons with different E_i
But we need to select a unique incident energy



Phase difference
Bragg law

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

$$\lambda = \frac{2\pi}{k}$$

$$E = \frac{\hbar^2 k^2}{2m}$$

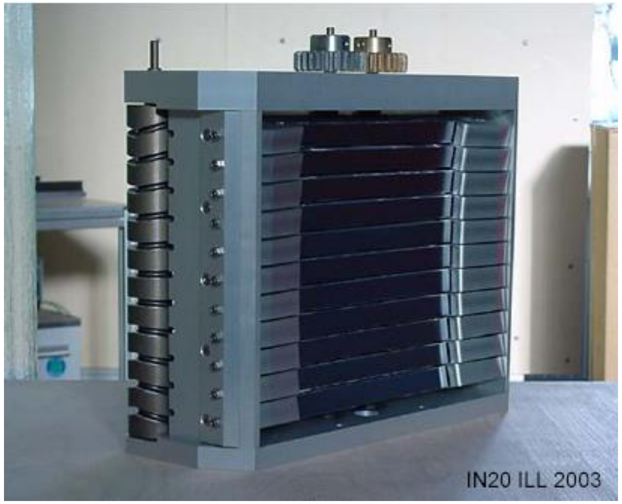
Monochromator

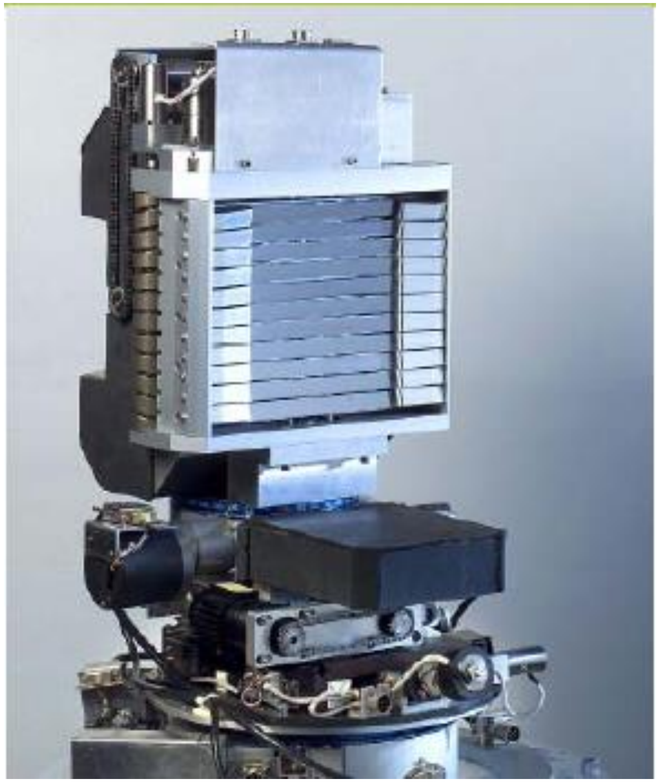
Graphite

Cu

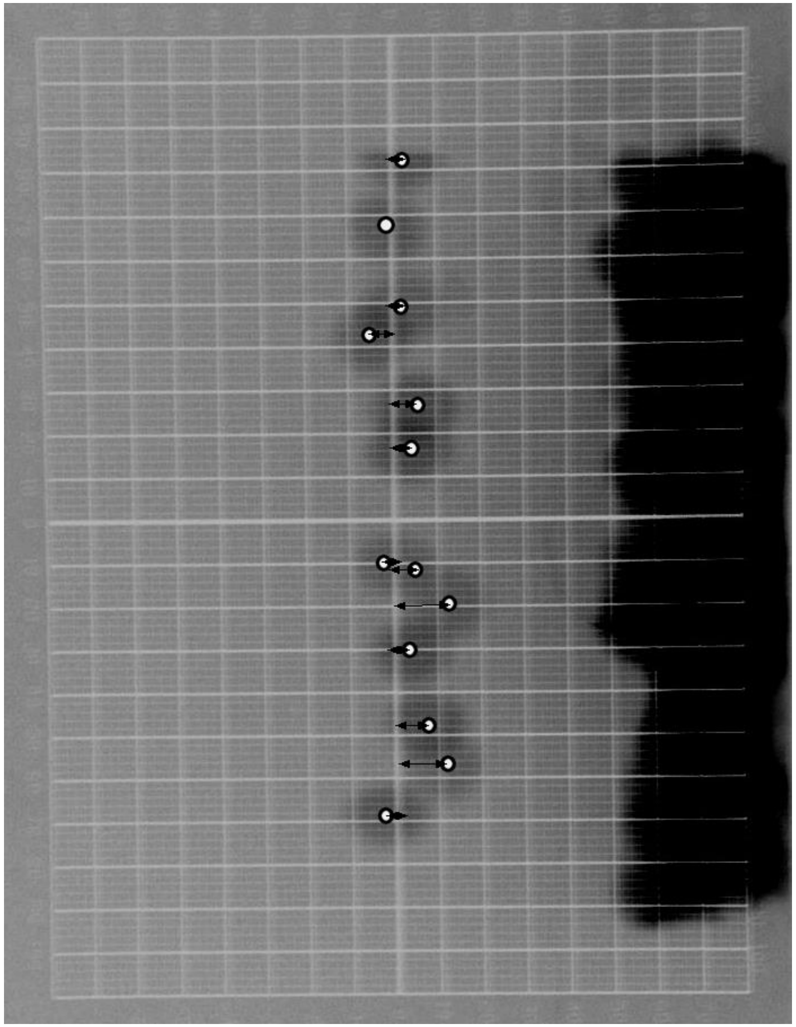
- Should not absorb
- High reflectivity
- d-spacing must be ok
- Large single crystals
- « Cheap » ...

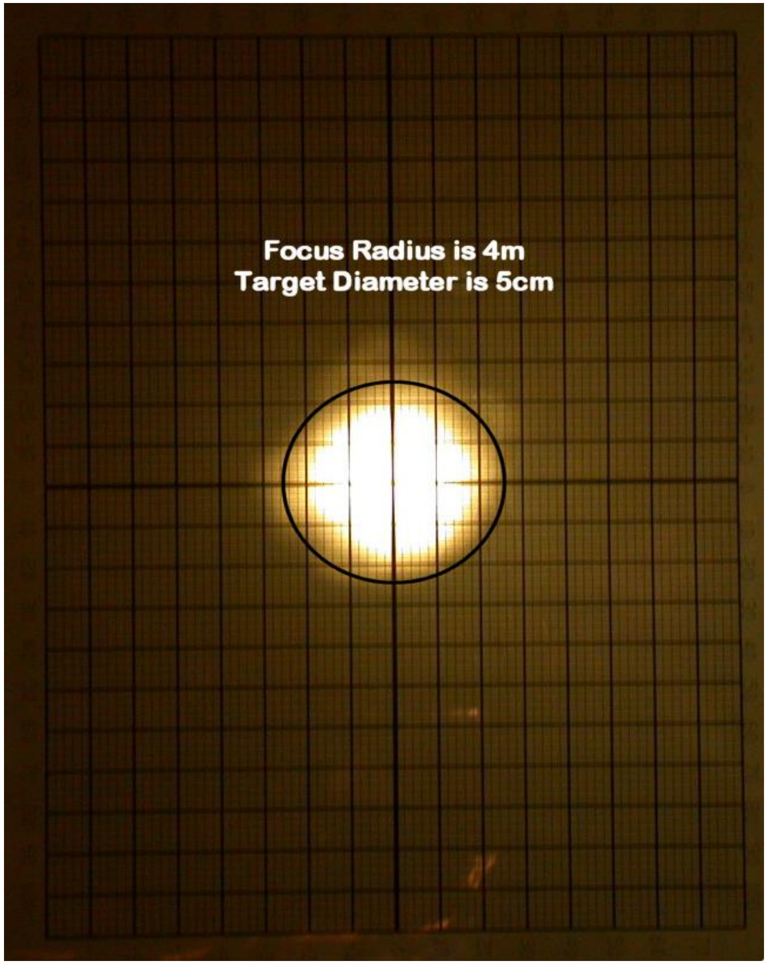
Monochromator





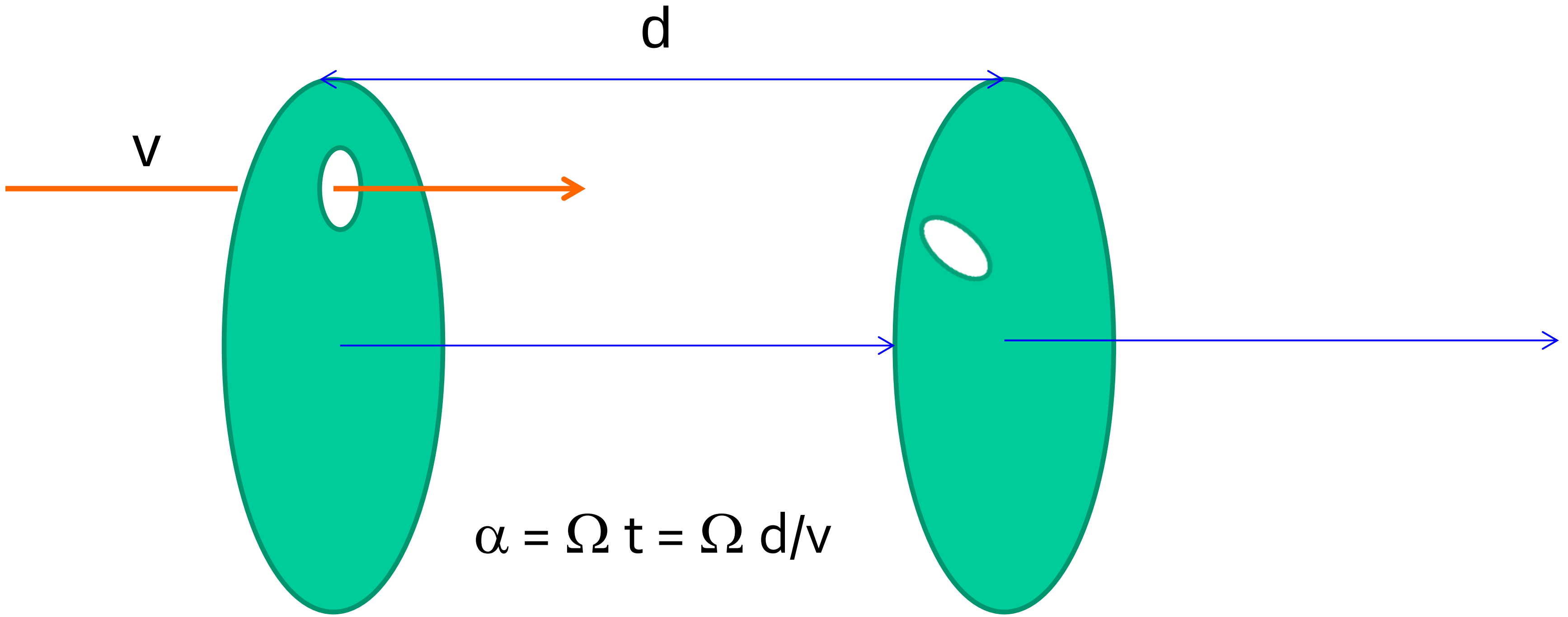
Monochromator





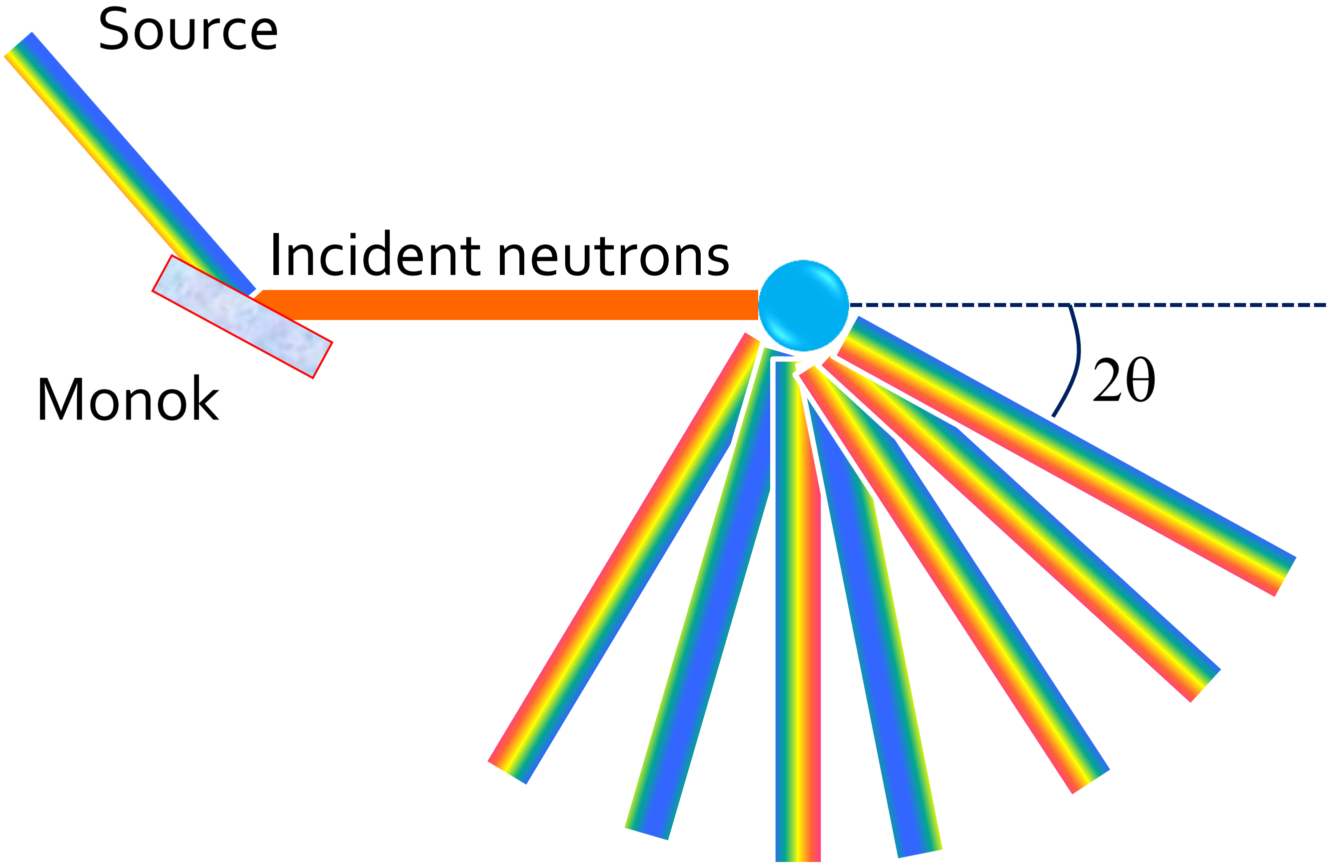
How to select an incident energy ?

Other possibilities : Disk Chopper



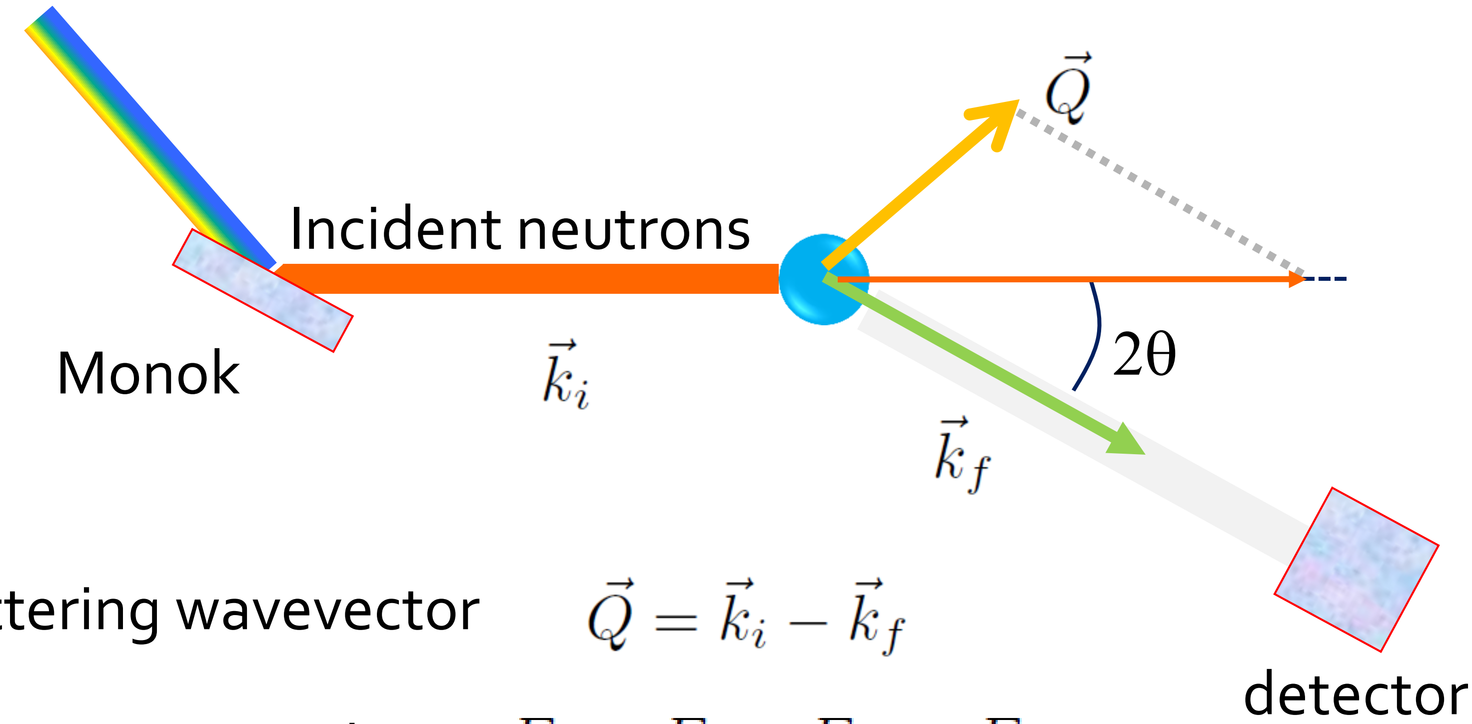
Produces a bunch of neutrons with a given incident energy

Neutron scattering



For a given θ, the neutrons gain or loose some energy depending on the scattering processs

Neutron scattering

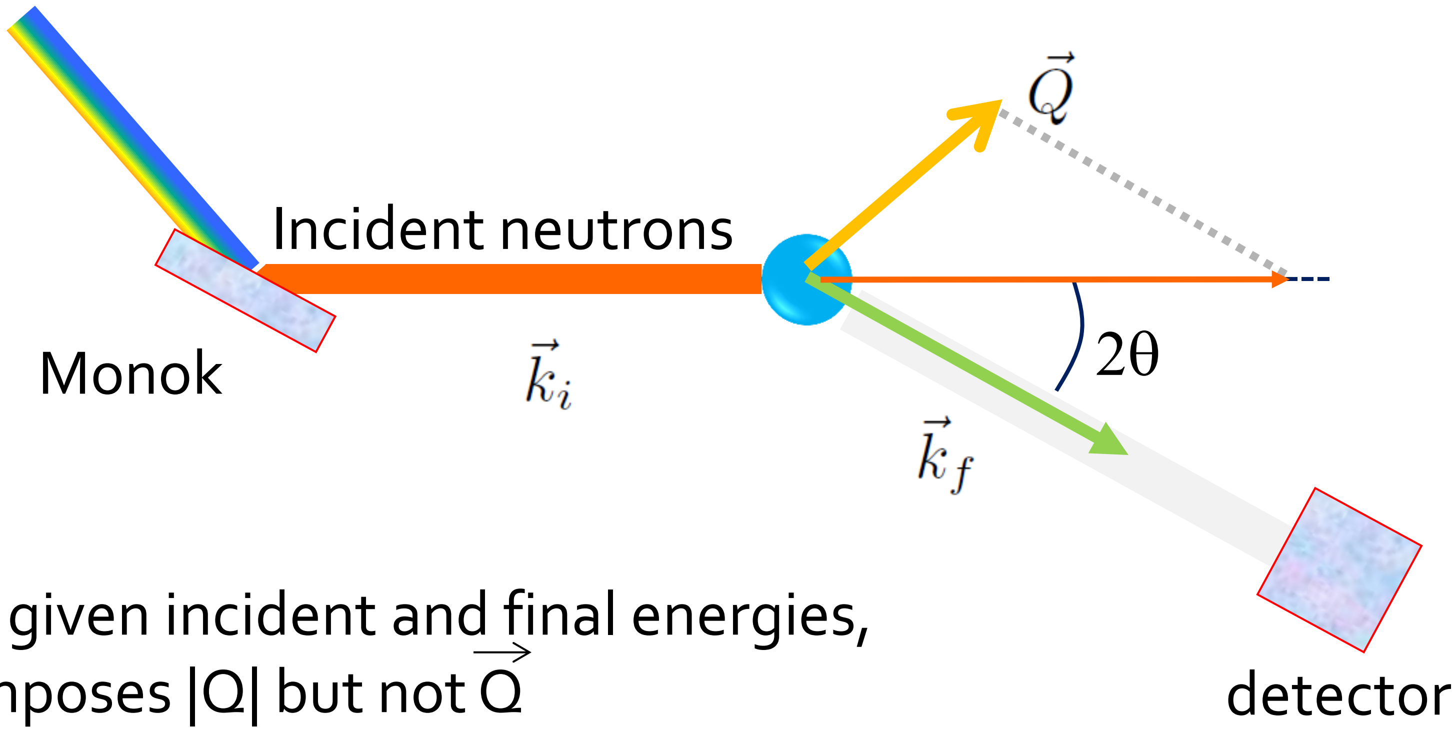


Scattering wavevector $\vec{Q} = \vec{k}_i - \vec{k}_f$

Energy conservation $E_\lambda + E_i = E_{\lambda'} + E_f$

Energy transfer $E = \hbar\omega = E_i - E_f = E_{\lambda'} - E_\lambda$

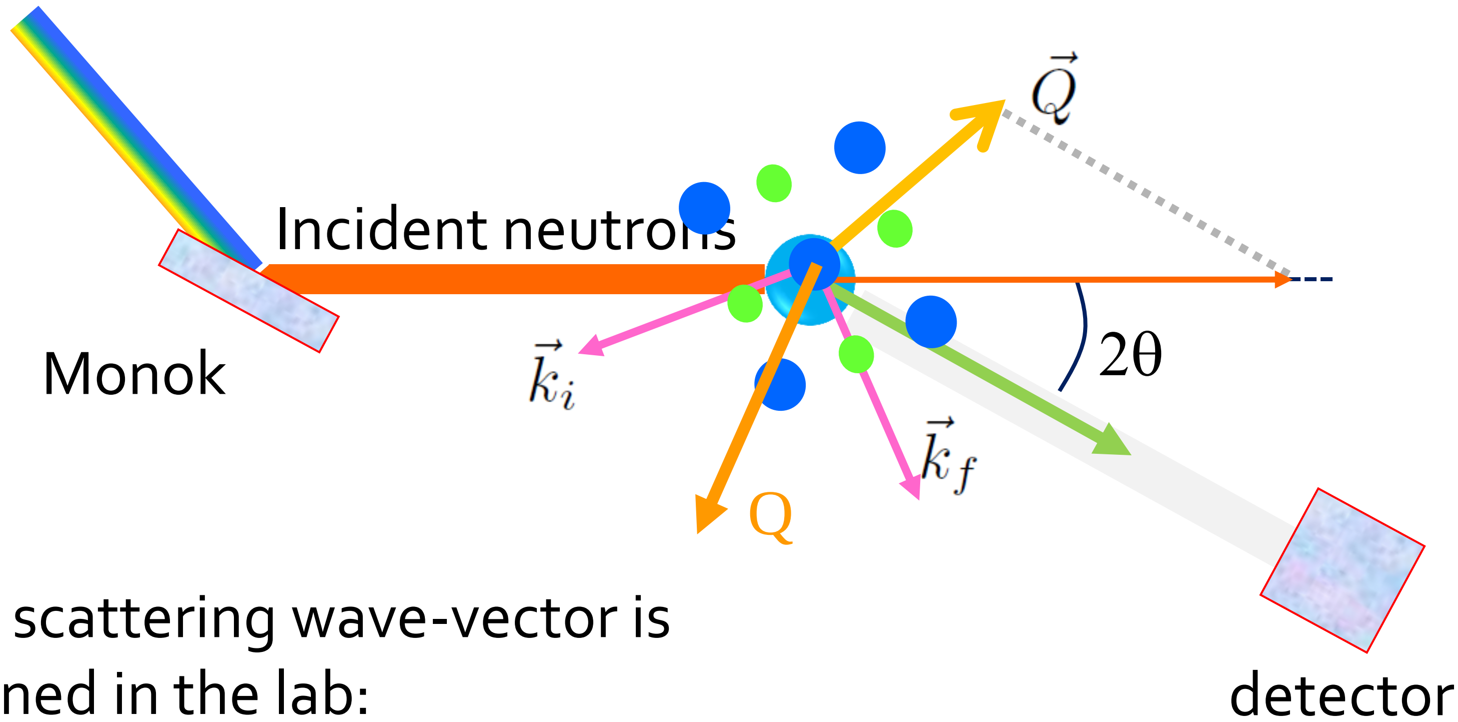
How to select |Q| ?



For given incident and final energies,
θ imposes |Q| but not $\overrightarrow{Q}$

$$\vec{Q} = \vec{k}_i - \vec{k}_f$$
$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta$$

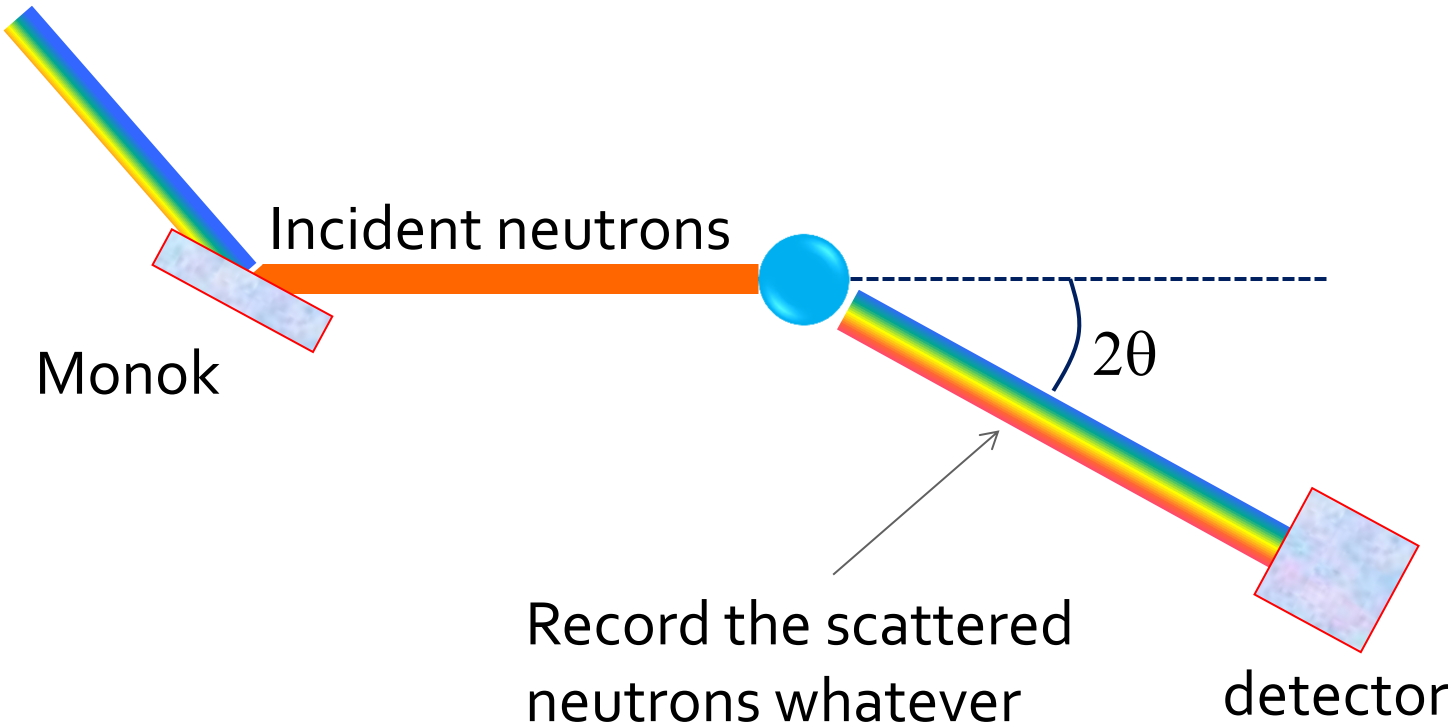
How to select $\vec{Q}$?



The scattering wave-vector is defined in the lab:

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

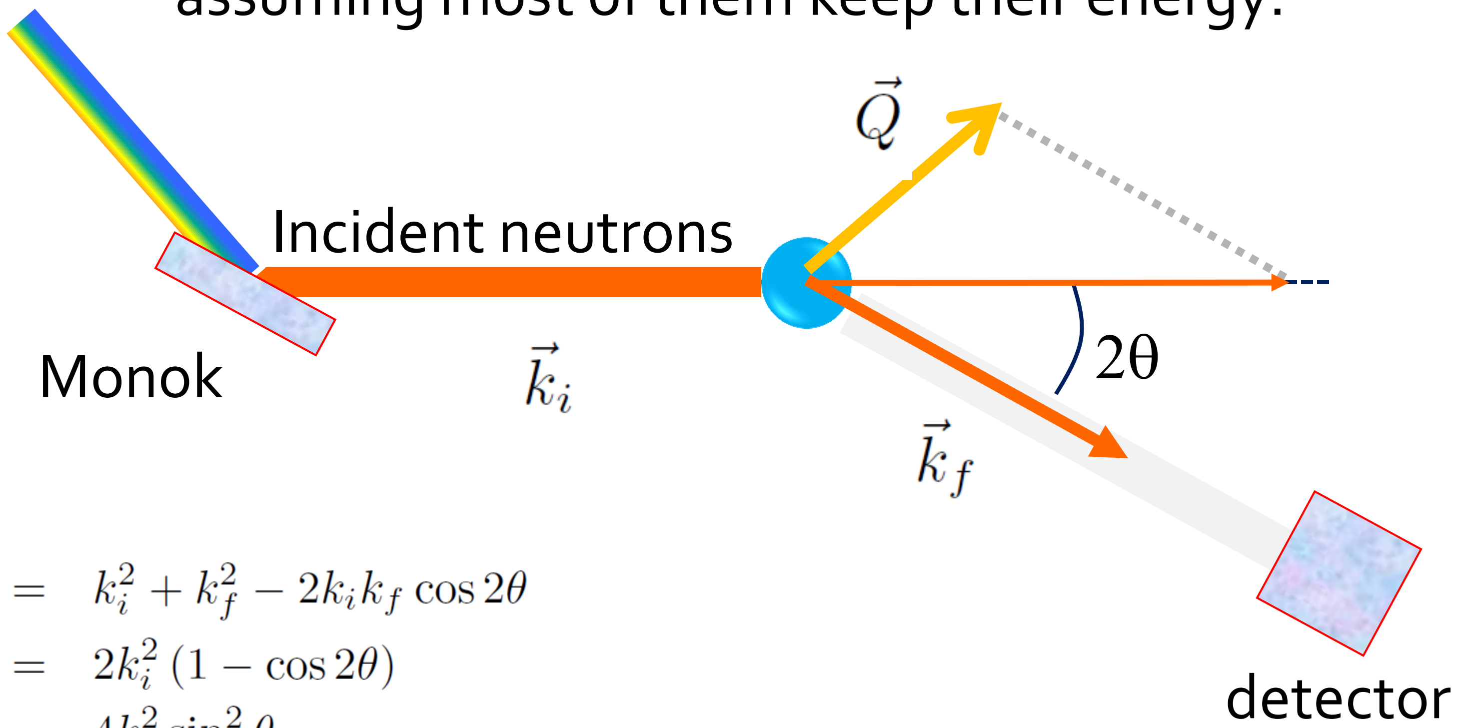
Diffraction



Record the scattered neutrons whatever their final energy

Diffraction

assuming most of them keep their energy:

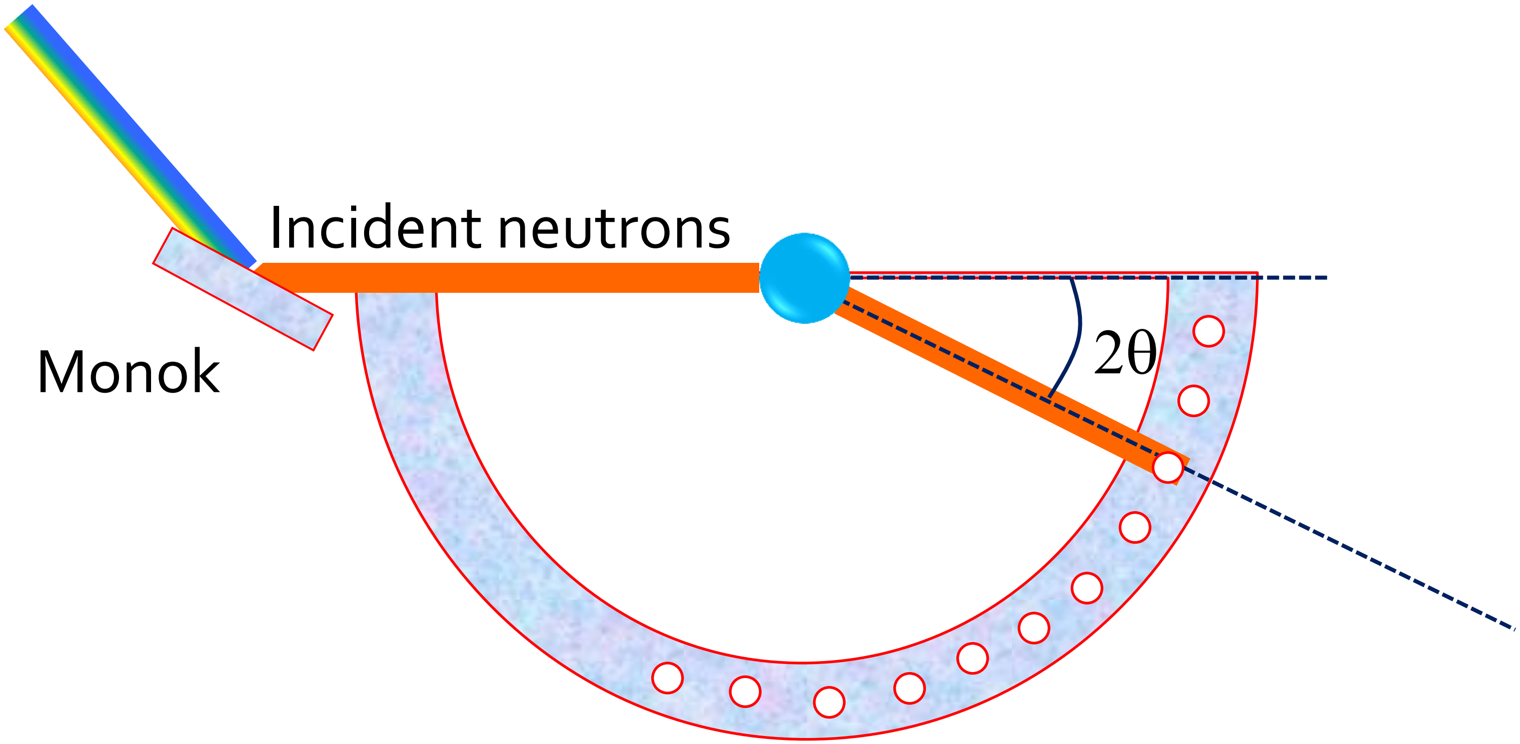


$$
\begin{aligned}
Q^2 &= k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta \\
&= 2k_i^2 (1 - \cos 2\theta) \\
&= 4k_i^2 \sin^2 \theta \\
Q &= 2k_i \sin \theta \\
\frac{2\pi}{d} &= 2\frac{2\pi}{\lambda} \sin \theta
\end{aligned}
$$

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

Bragg law

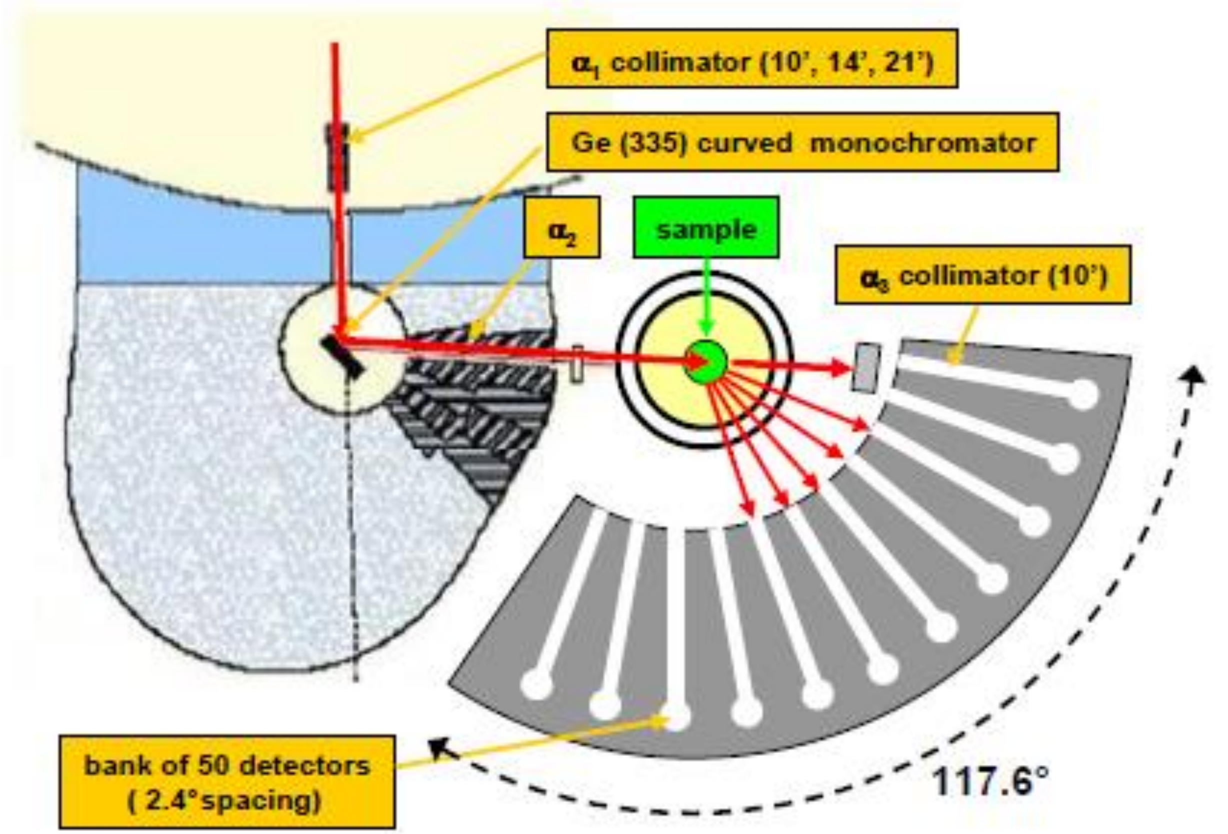
Diffraction



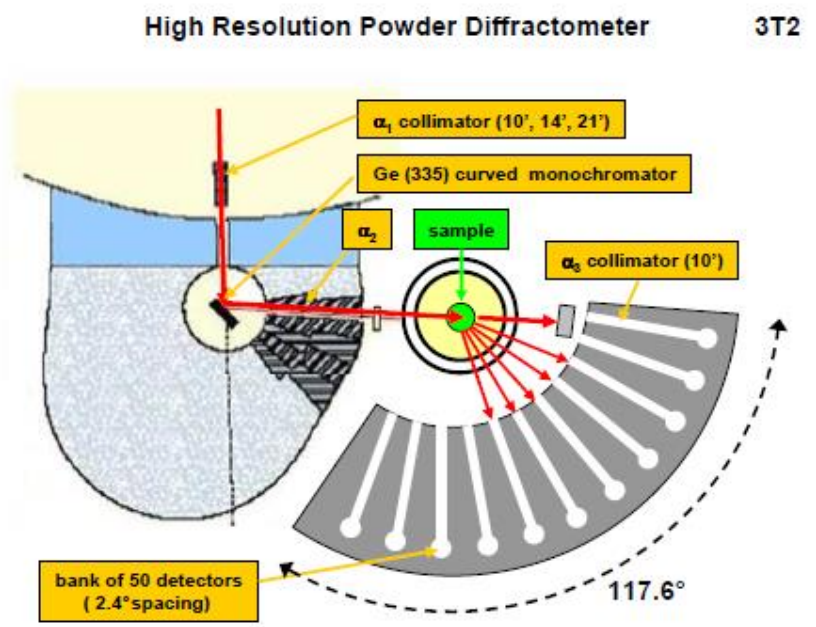
We still have to rotate the sample even if the experiment looks more efficient with a detector bank

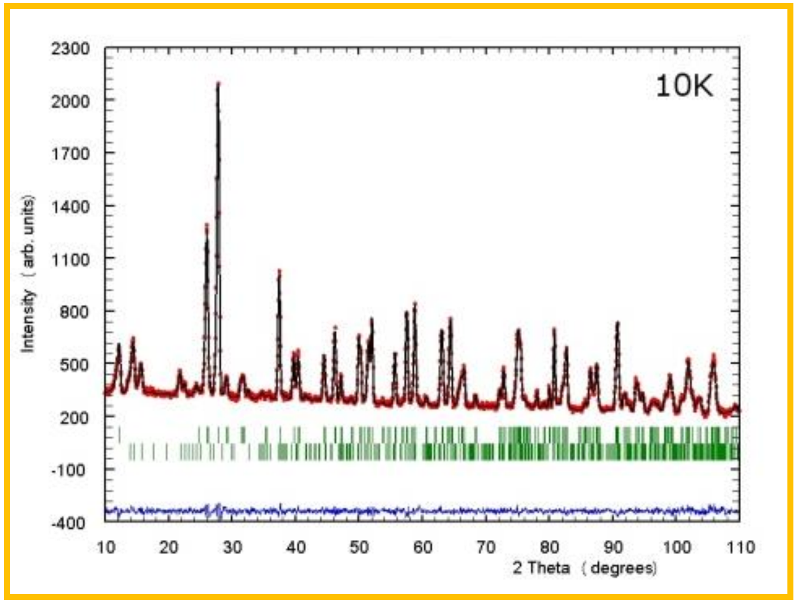
Diffraction

High Resolution Powder Diffractometer 3T2

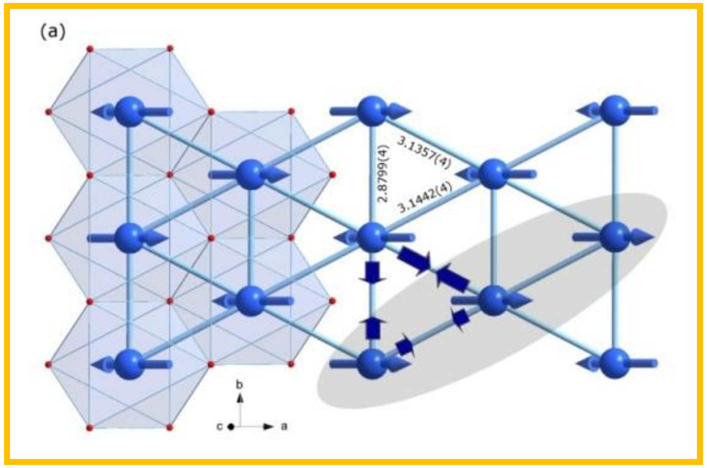


Diffraction



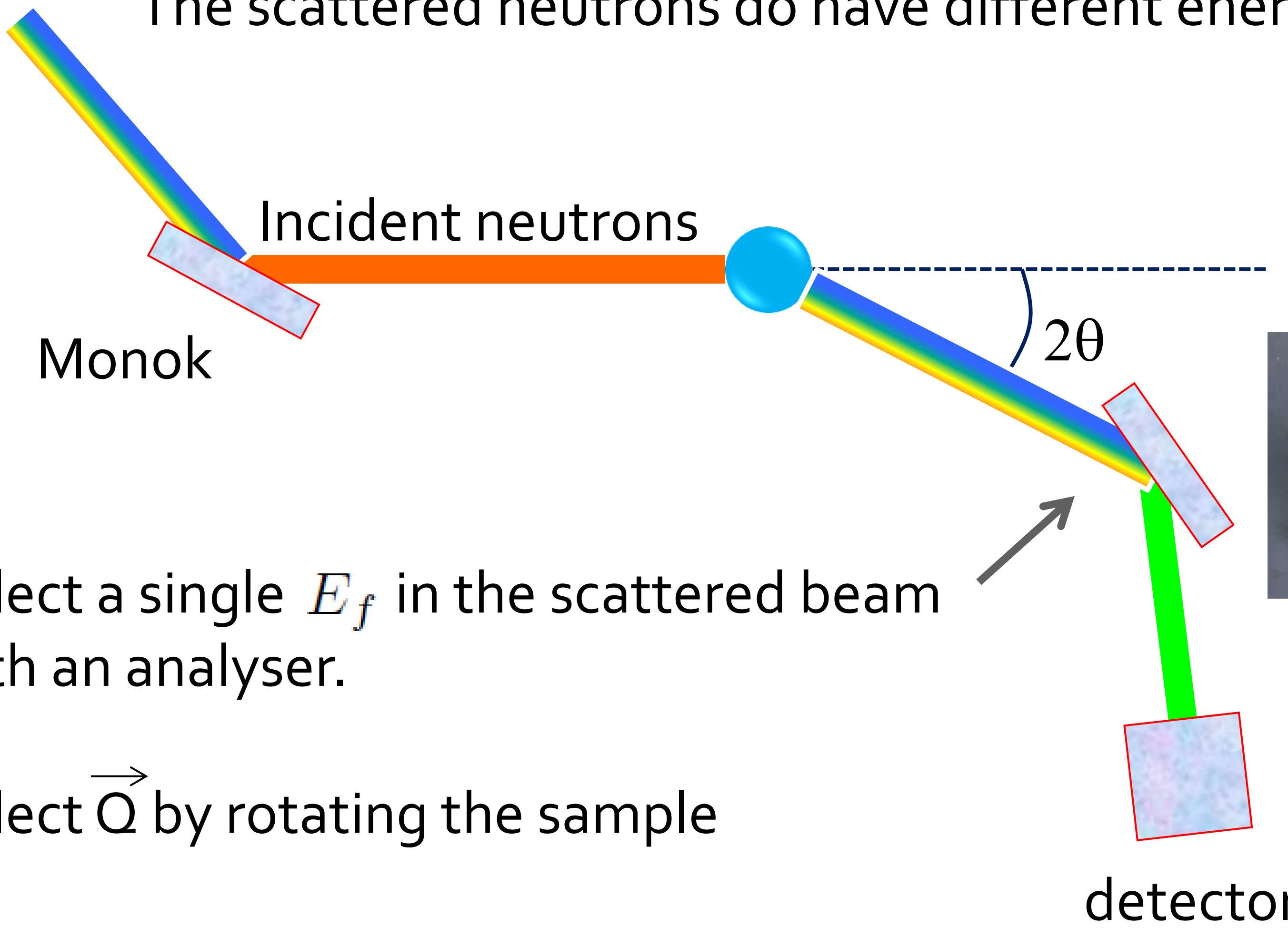


Unit cell and space group



Inelastic scattering

The scattered neutrons do have different energies

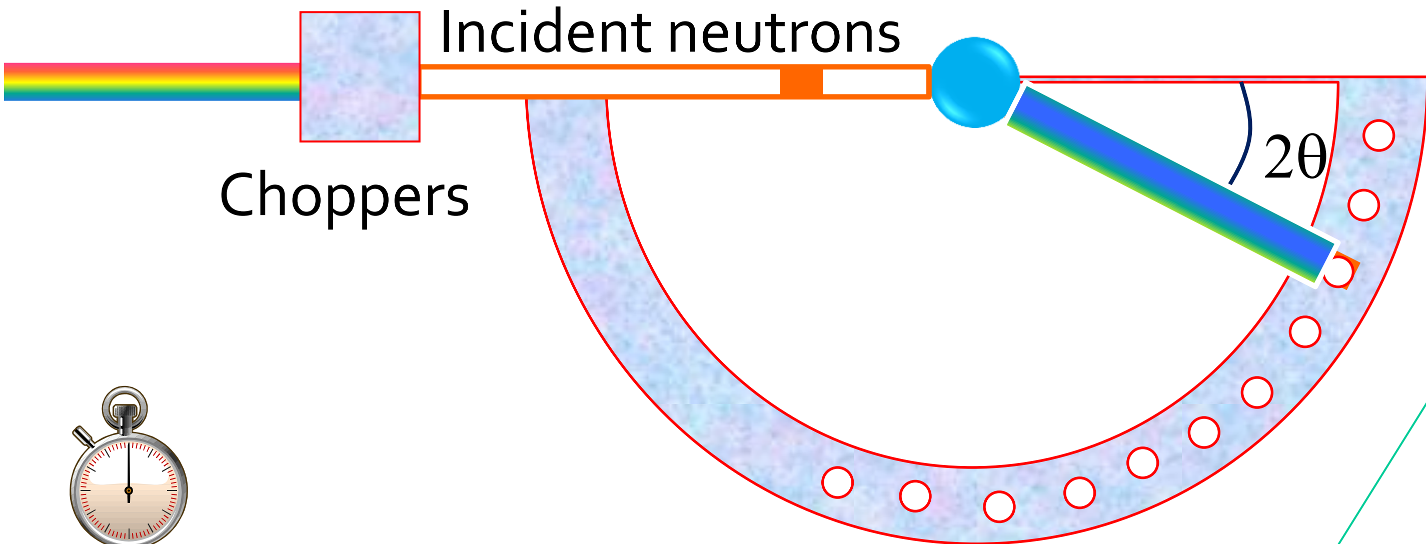


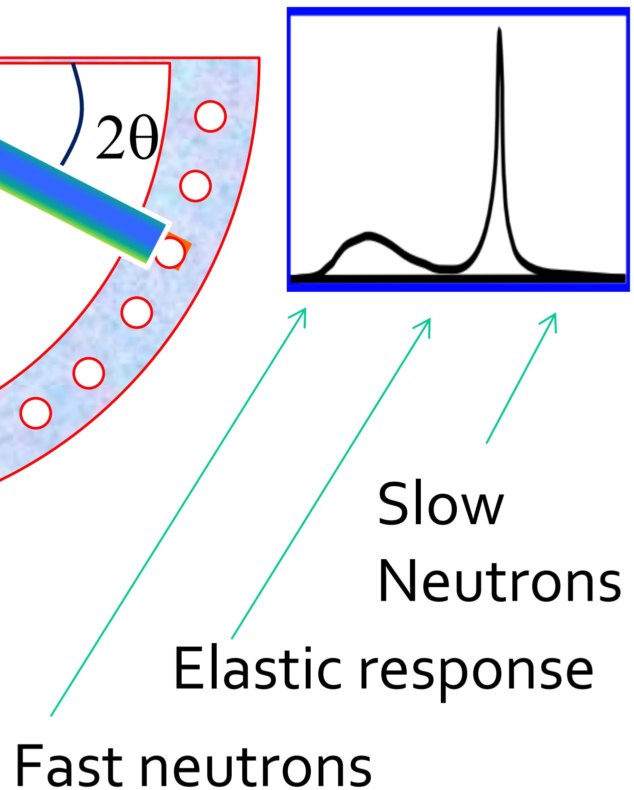
Select a single E_f in the scattered beam with an analyser.

Select $\overrightarrow{Q}$ by rotating the sample

Inelastic scattering

The scattered neutrons do have different energies

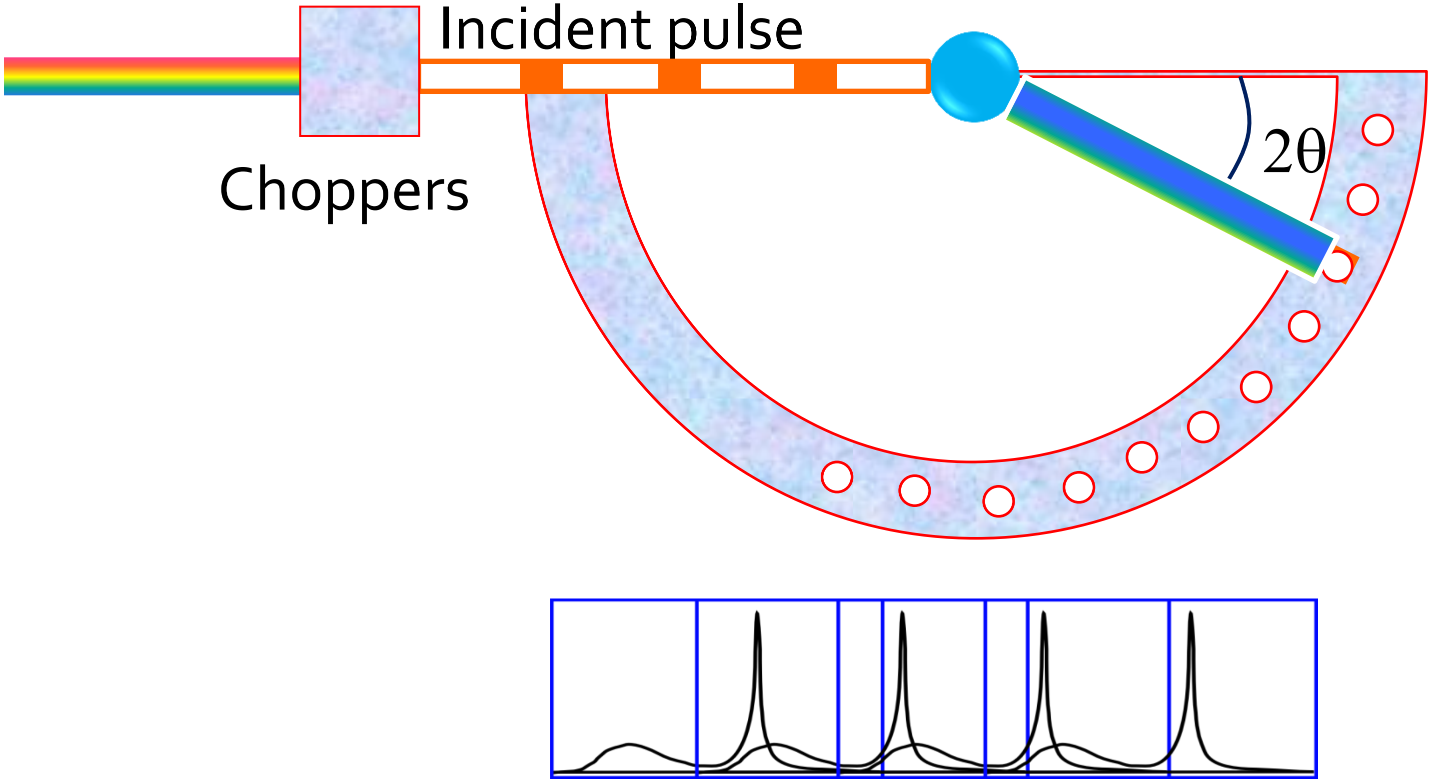




Select a single E_f in the scattered beam
via the « time of flight » $t = \dfrac{d}{\sqrt{2E_f/m_n}}$

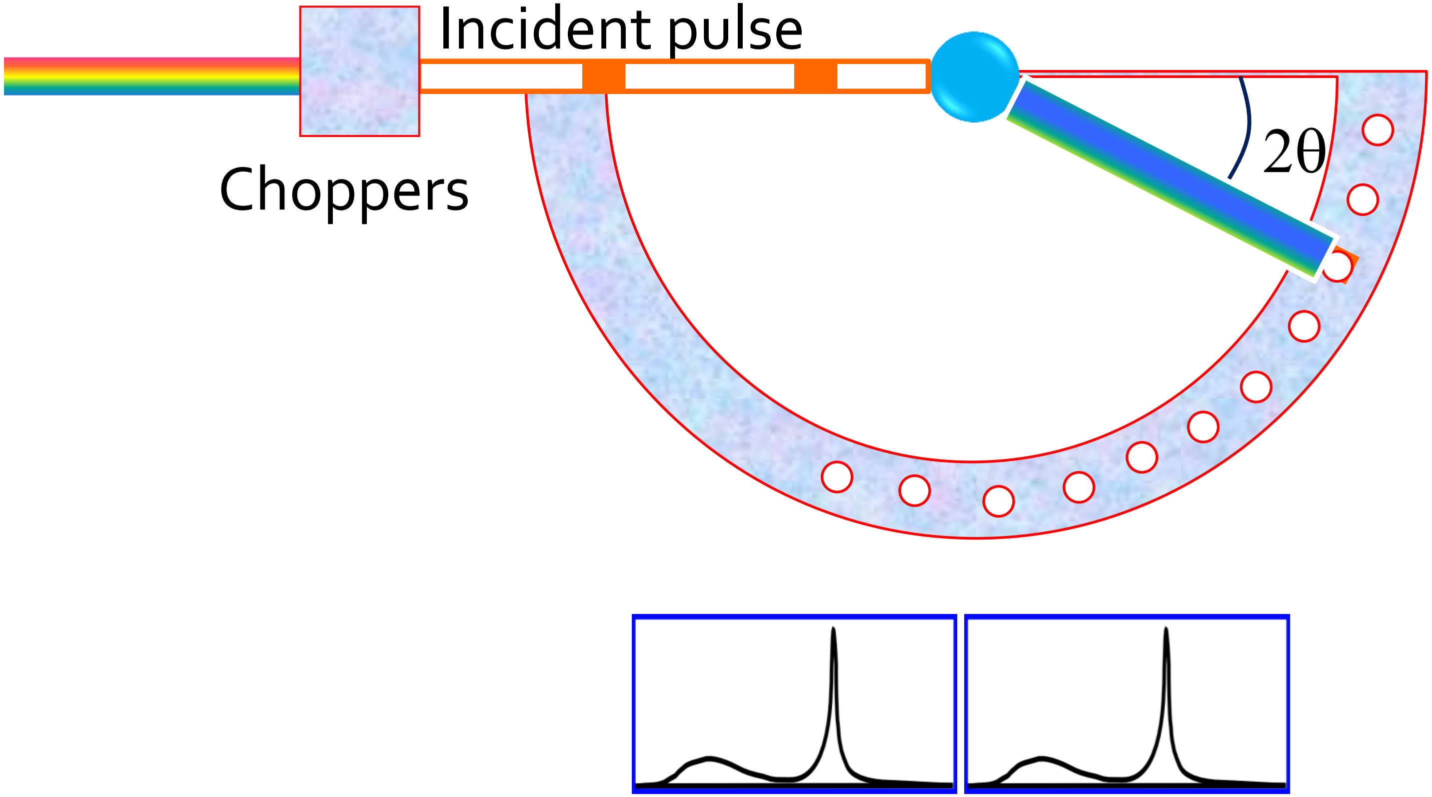
Inelastic scattering

Chop the beam



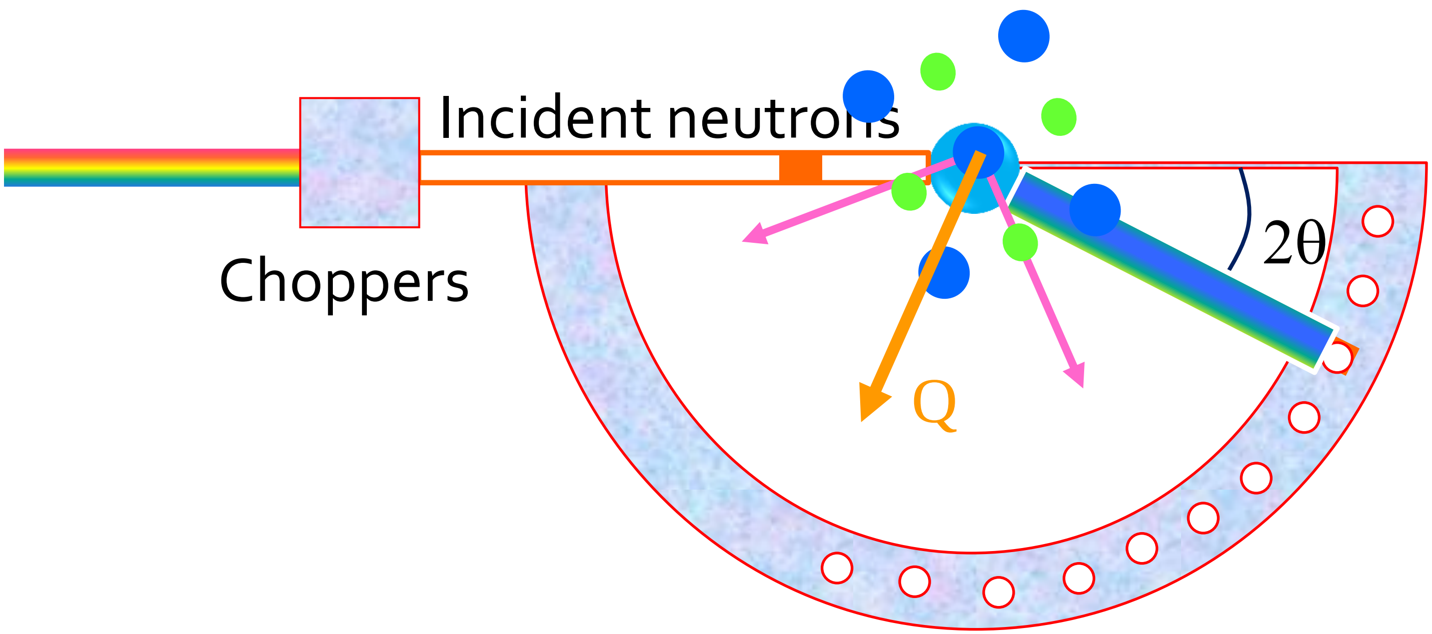
Inelastic scattering

Chop the beam



Inelastic scattering

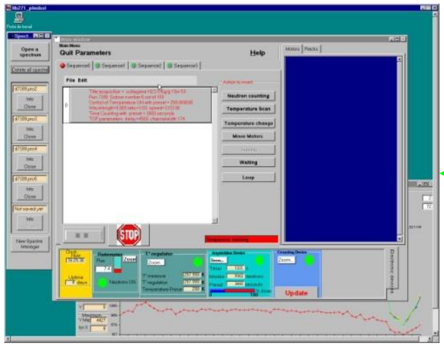
The scattered neutrons do have different energies



Select $\overrightarrow{Q}$ by rotating the sample

$$\vec{Q} = \vec{k}_i - \vec{k}_f$$
$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta$$

An instrument consists in …

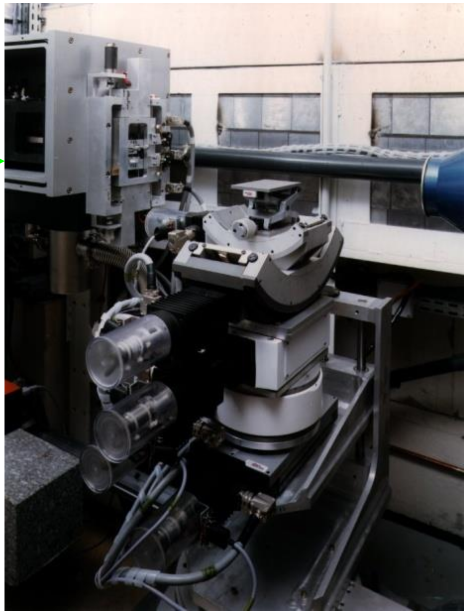


Control Software

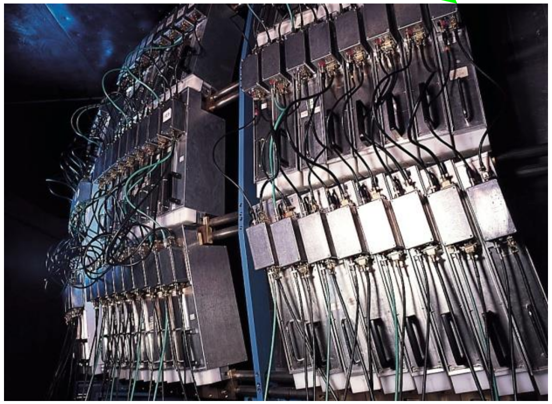
Data storage

Data treatment

Electronics

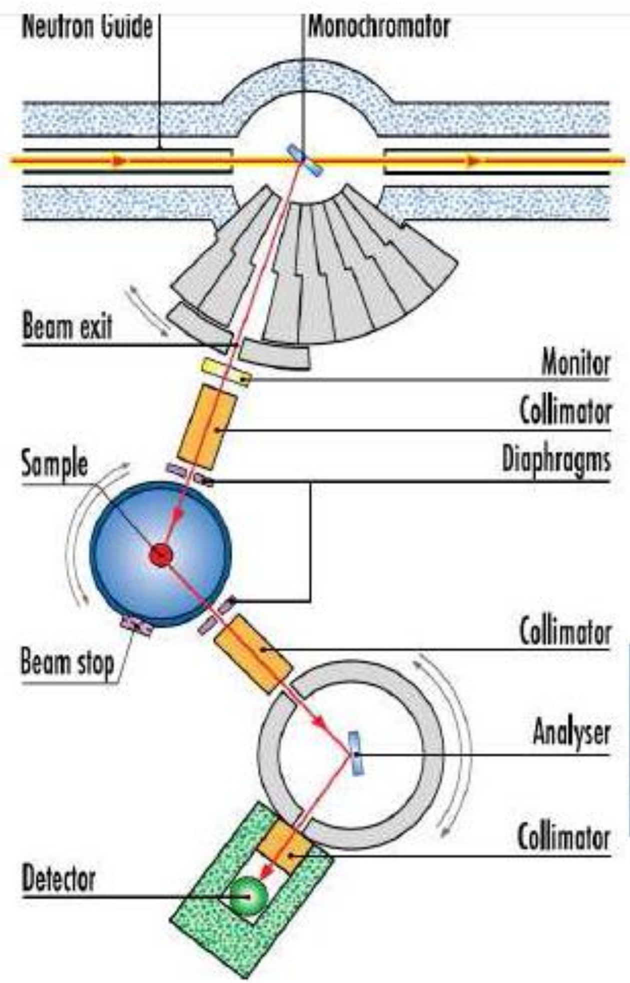


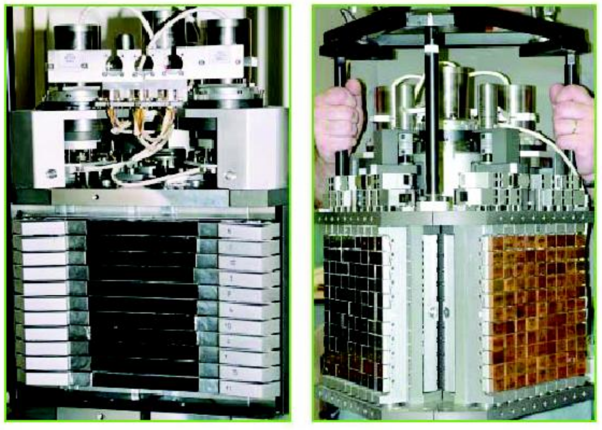
Positionning



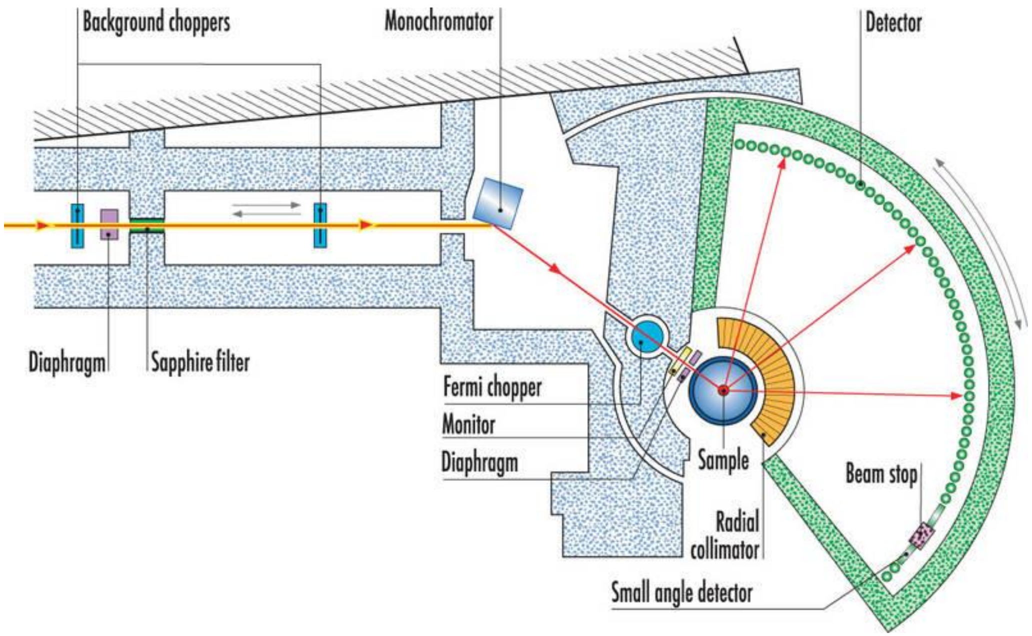
Detector

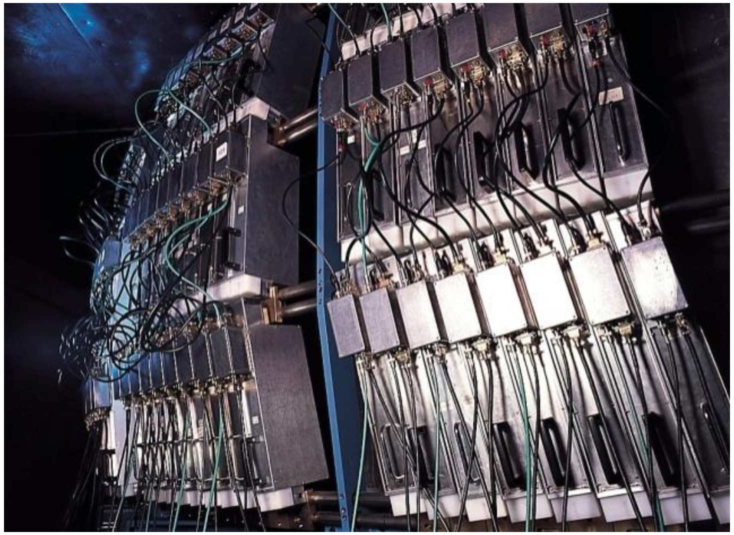
Triple axis


Neutron Guide
Monochromator
Beam exit
Monitor
Collimator
Diaphragms
Sample
Collimator
Beam stop
Analyser
Collimator
Detector



Time of flight

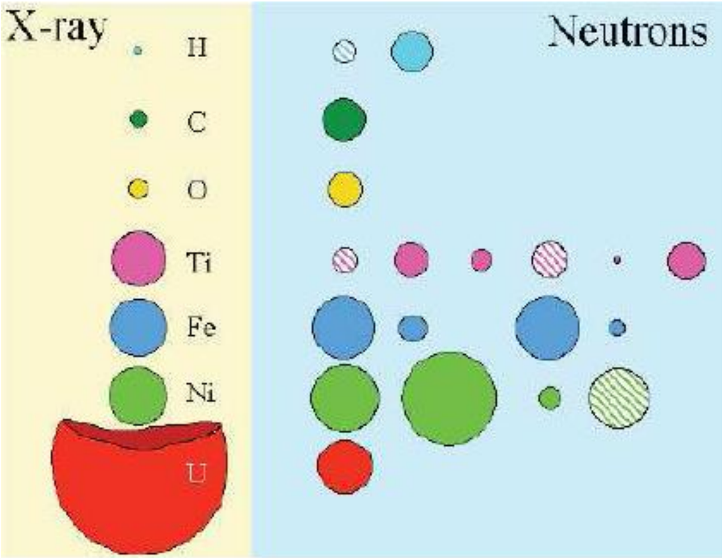




Cross section, nuclear interaction

Nuclear interaction with nuclei

$$\hat{V}_n(\vec{r}) = \frac{2\pi\hbar^2}{M} b\delta(\vec{r} - \vec{R}).$$



b= scattering length

- *positive or negative*
- *depends on the isotope*

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E'} = \frac{k_f}{k_i} \sum_{i,j} b_i \, b_j \int_{-\infty}^{+\infty} dt \, \langle e^{iQ \, R_i} \, e^{-iQ \, R_j(t)} \rangle \, e^{-i\omega t}$$

Cross section, nuclear interaction

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\sum_{i,j} b_i\, b_j \int_{-\infty}^{+\infty} dt\, \langle e^{iQ\ R_i}\ e^{-iQ\ R_j(t)}\rangle\ e^{-i\omega t}$$

Frozen lattice

$$R_i(t) = R^o_m + r_\ell$$

Nothing moves

Unit cell

Atom in the cell

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\,\delta(\omega) \sum_{m,n,\ell,\ell'} b_\ell\, b_{\ell'}\ e^{iQ\ (R^o_m + r_\ell - R^o_n - r_{\ell'})}$$

$$= \delta(\omega) \sum_{m,n} e^{iQ\ (R^o_m - R^o_n)} \sum_{\ell,\ell'} b_\ell\, b_{\ell'}\ e^{iQ\ (r_\ell - r_{\ell'})}$$

$$= \delta(\omega) \sum_{m,n} e^{iQ\ (R^o_m - R^o_n)}\ |F(Q)|^2$$

« simple » definition of the structure factor

$$F(Q) = \sum_\ell b_\ell\ e^{iQ\ r_\ell}$$

Cross section, nuclear interaction

Vibrating lattice

$$R_i(t) = R_m^o + r_\ell + u_{m,\ell}(t)$$

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i} \sum_{m,n} e^{iQ\,(R_m^o - R_n^o)} \sum_{\ell,\ell'} b_\ell\, b_{\ell'}\, e^{iQ\,(r_\ell - r_{\ell'})}$$

$$\int_{-\infty}^{+\infty} dt\, \langle e^{iQ\,u_{m,\ell}}\, e^{-iQ\,u_{n,\ell'}(t)} \rangle\, e^{-i\omega t}$$

Harmonic approximation

$$\langle e^{iQ\,u_{m,\ell}}\, e^{-iQ\,u_{n,\ell'}(t)} \rangle = e^{-W_\ell - W_{\ell'}}\, e^{\langle Q\,u_{m,\ell}\; Q\,u_{n,\ell'}(t)\rangle}$$

$$W_\ell = \frac{1}{2} \langle\, [Q\,u_{m,\ell}]^2\, \rangle$$

Debye-Waller factor

Cross section, nuclear interaction

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\sum_{m,n} e^{iQ\,(R^o_m - R^o_n)} \sum_{\ell,\ell'} b_\ell\, b_{\ell'}\, e^{iQ\,(r_\ell - r_{\ell'})}\, e^{-W_\ell - W_{\ell'}} \int_{-\infty}^{+\infty} dt\, e^{\langle Q\, u_{m,\ell}\; Q\, u_{n,\ell'}(t)\rangle}\, e^{-i\omega t}$$

Series expansion

$$e^{\langle Q\, u_{m,\ell}\; Q\, u_{n,\ell'}(t)\rangle} \approx 1 + \langle Q\, u_{m,\ell}\; Q\, u_{n,\ell'}(t)\rangle\; + \;...$$

Elastic term

$$F(Q) = \sum_\ell b_\ell\; e^{iQ\; r_\ell} e^{-W_\ell}$$

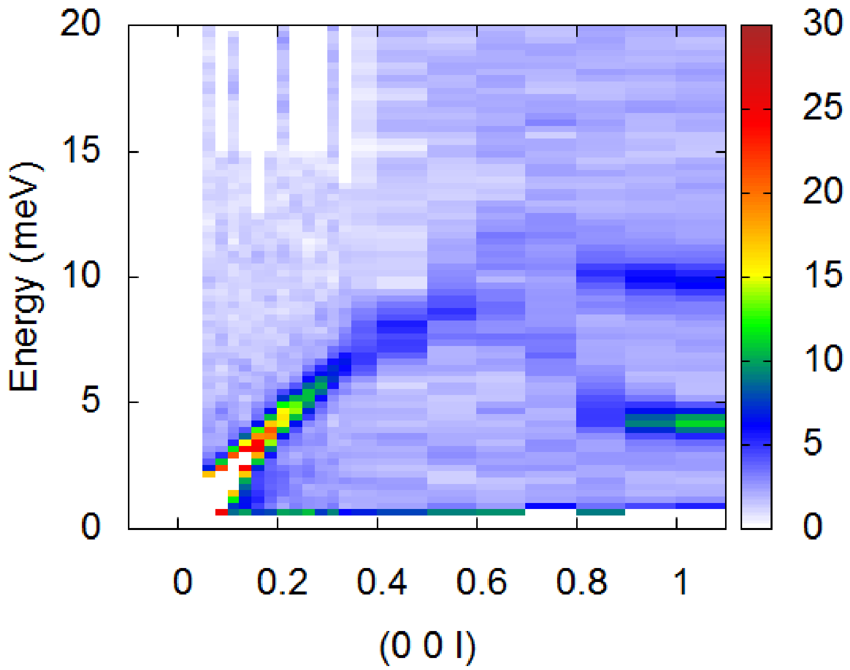
Inelastic term : phonons

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \sum_s A_s\; [(1+n(\omega_{Q,s}))\;\delta(\omega-\omega_{Q,s})\; + \; n(\omega_{Q,s})\;\delta(\omega+\omega_{Q,s})]$$

$$F_s(Q) = \sum_\ell b_\ell\; e^{iQ\; r_\ell}\; e^{-W_\ell}\; \frac{1}{\sqrt{M_\ell \omega_{q,s}}}\; \left(\vec{Q}.\vec{e}_{q,\ell}\right)$$

Cross section, nuclear interaction

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E'} = \sum_s A_s \left[(1 + n(\omega_{Q,s}))\ \delta(\omega - \omega_{Q,s}) + n(\omega_{Q,s})\ \delta(\omega + \omega_{Q,s}) \right]$$



Intensity ≠ o (Bkg) if

$k_i - k_f = Q$

$E_i - E_f = \omega(Q)$

Cross section, magnetic interaction

Dipolar field created by the spin (and orbital motion) of unpaired electrons

$$E_{ne} = -\mu_n . B_e$$

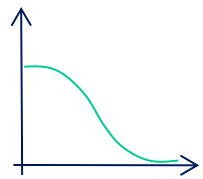
$$B_e(R) = \frac{\mu_o}{4\pi}\left(\mathrm{rot}(\frac{\mu_e \times R}{R^3}) - e\ v_e \times \frac{R}{R^3}\right)$$

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\,(\gamma r_o)^2 \sum_{i,j} \int_{-\infty}^{+\infty} dt\ \langle \vec{S}_{\perp,i}\ \vec{S}_{\perp,j}(t)\ e^{iQ\ R_i}\ e^{-iQ\ R_j(t)} \rangle\ e^{-i\omega t}$$

Cross section, magnetic interaction

Unit cell position

Form factor of unpaired electrons in a given orbital (tabulated)



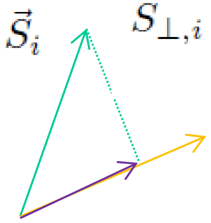
Atomic positions within the unit cell

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}(\gamma r_o)^2 \sum_{m,n} e^{iQ\,(R_m - R_n)} \sum_{i,j} f_i(Q)\; f_j^*(Q)\; e^{iQ\,(r_i - r_j)}\; e^{-W_i - W_j}$$

$$\int_{-\infty}^{+\infty} dt\; \langle \vec{S}_{\perp,m,i}\; \vec{S}_{\perp,n,j}(t)\; \rangle\; e^{-i\omega t}$$

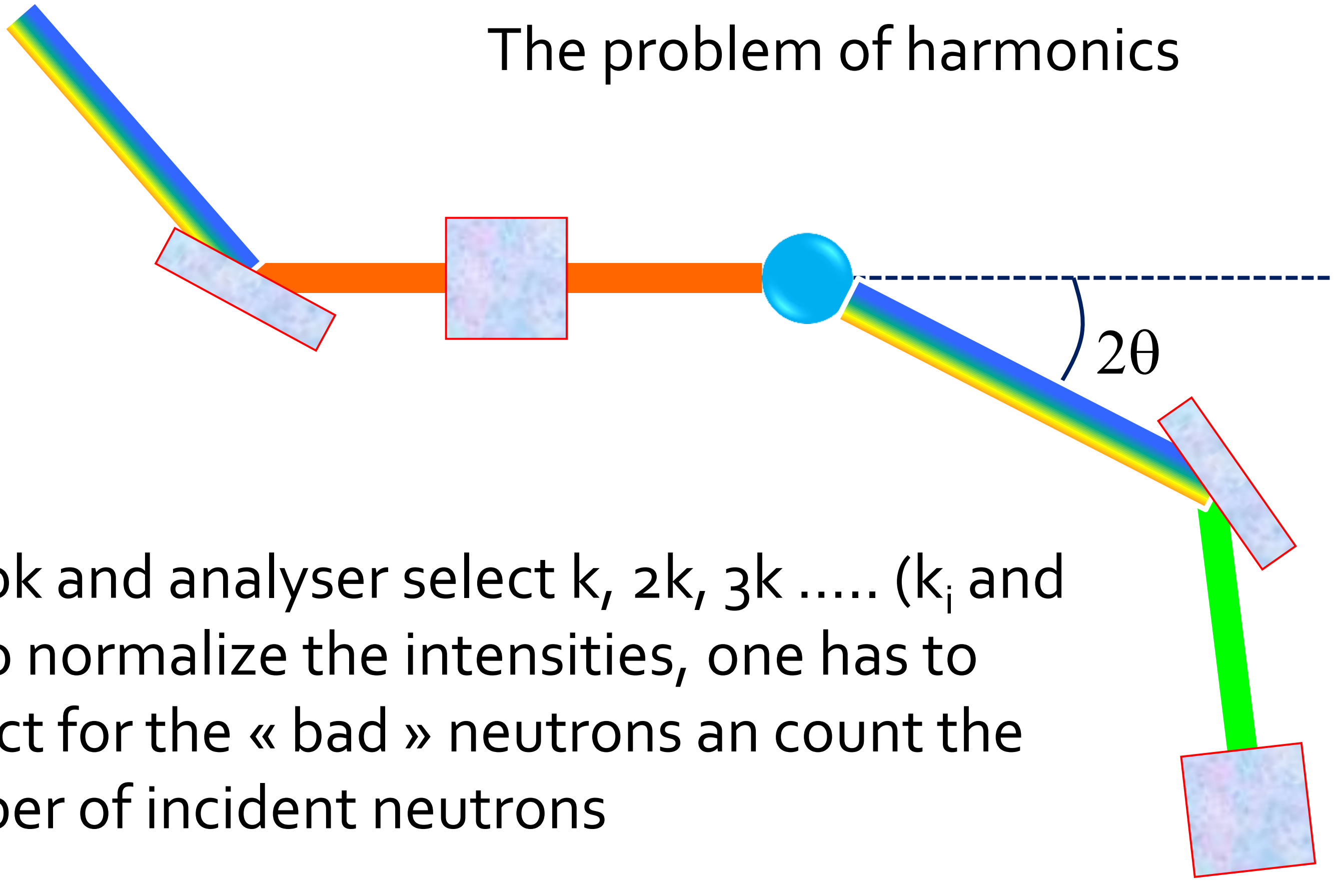
Debye-waller factor (thermal motion of the ions)

- Spin-spin correlation function
- spin components perp to Q (dipolar interaction)
- ω(k) for spin waves in close analogy with the case of phonons



$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \sum_s A_s\; [(1 + n(\omega_{Q,s}))\;\delta(\omega - \omega_{Q,s}) \;+\; n(\omega_{Q,s})\;\delta(\omega + \omega_{Q,s})]$$

Triple axis in practice

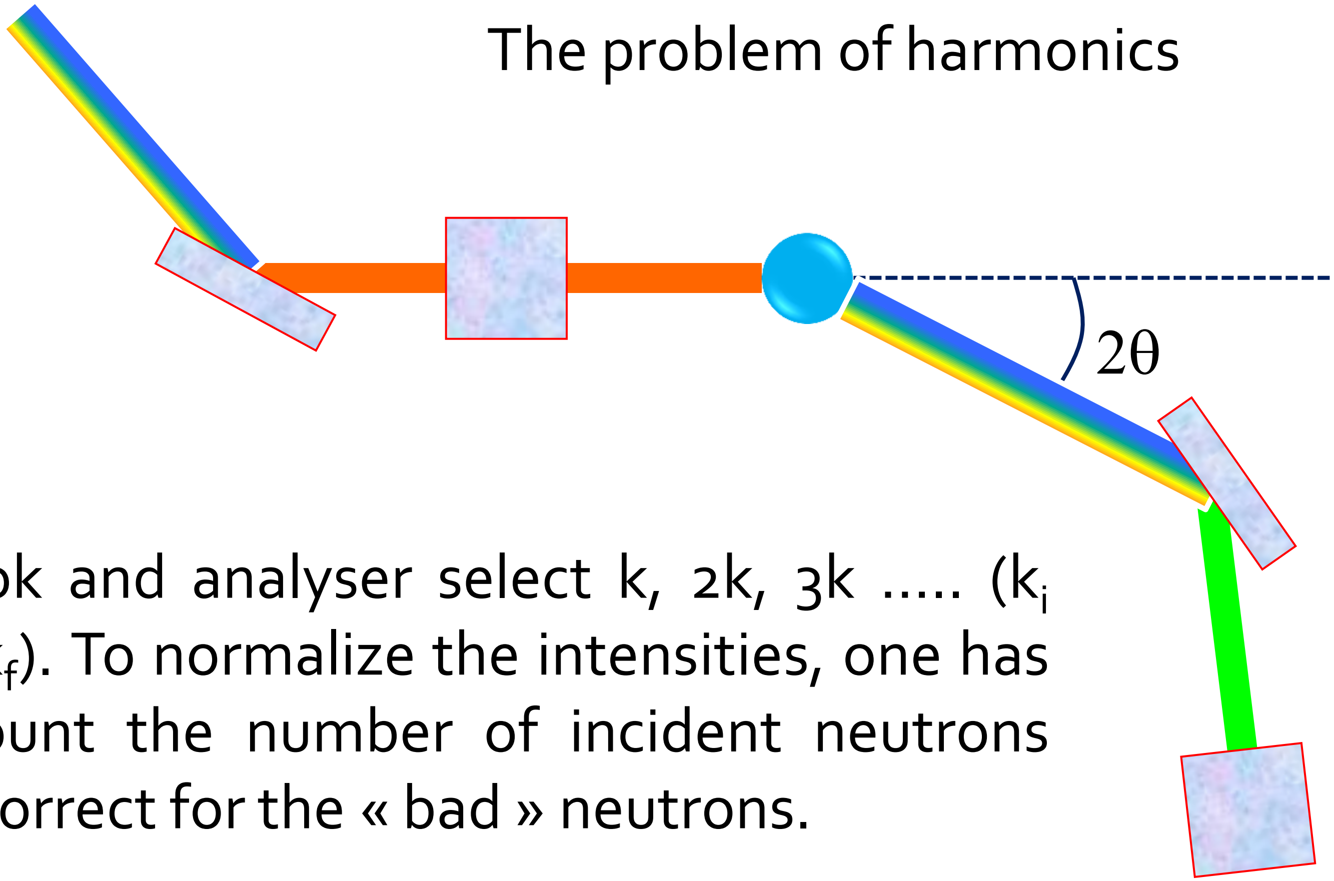


The problem of harmonics

Monok and analyser select k, 2k, 3k ..... (k_i and k_f). To normalize the intensities, one has to correct for the « bad » neutrons an count the number of incident neutrons

"monitor" : low sensibility detector

Triple axis in practice

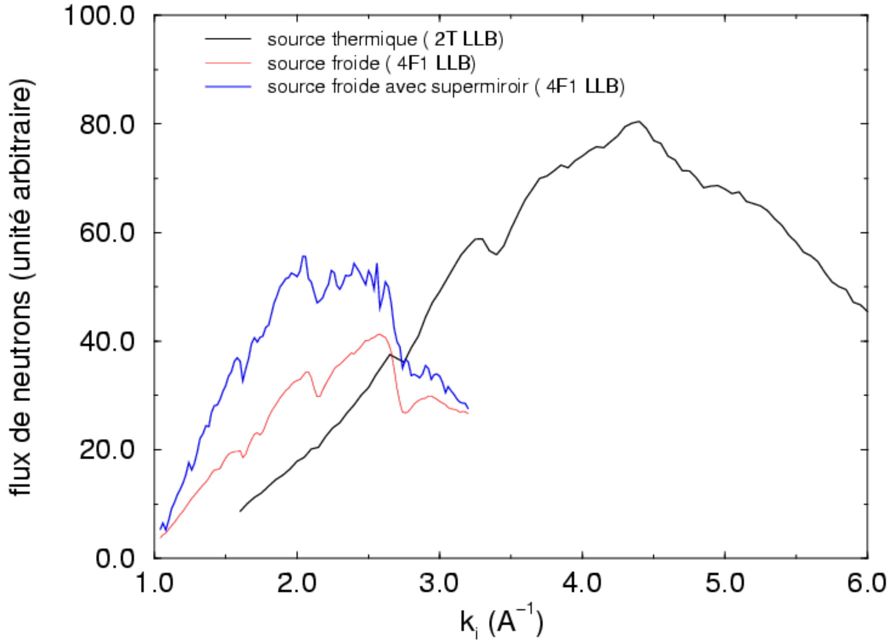


The problem of harmonics

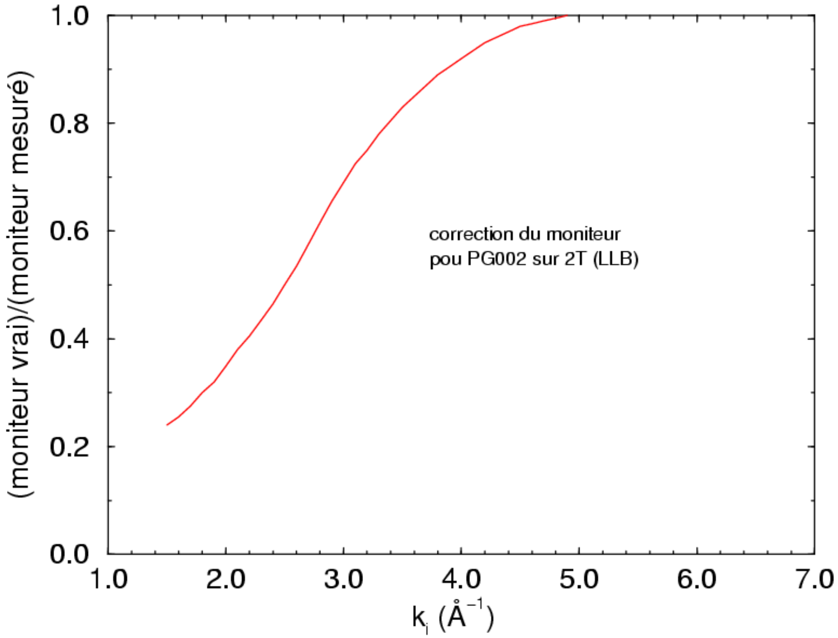
Monok and analyser select k, 2k, 3k ..... (k_i and k_f). To normalize the intensities, one has to count the number of incident neutrons and correct for the « bad » neutrons.

"monitor" : low sensibility detector

Flux

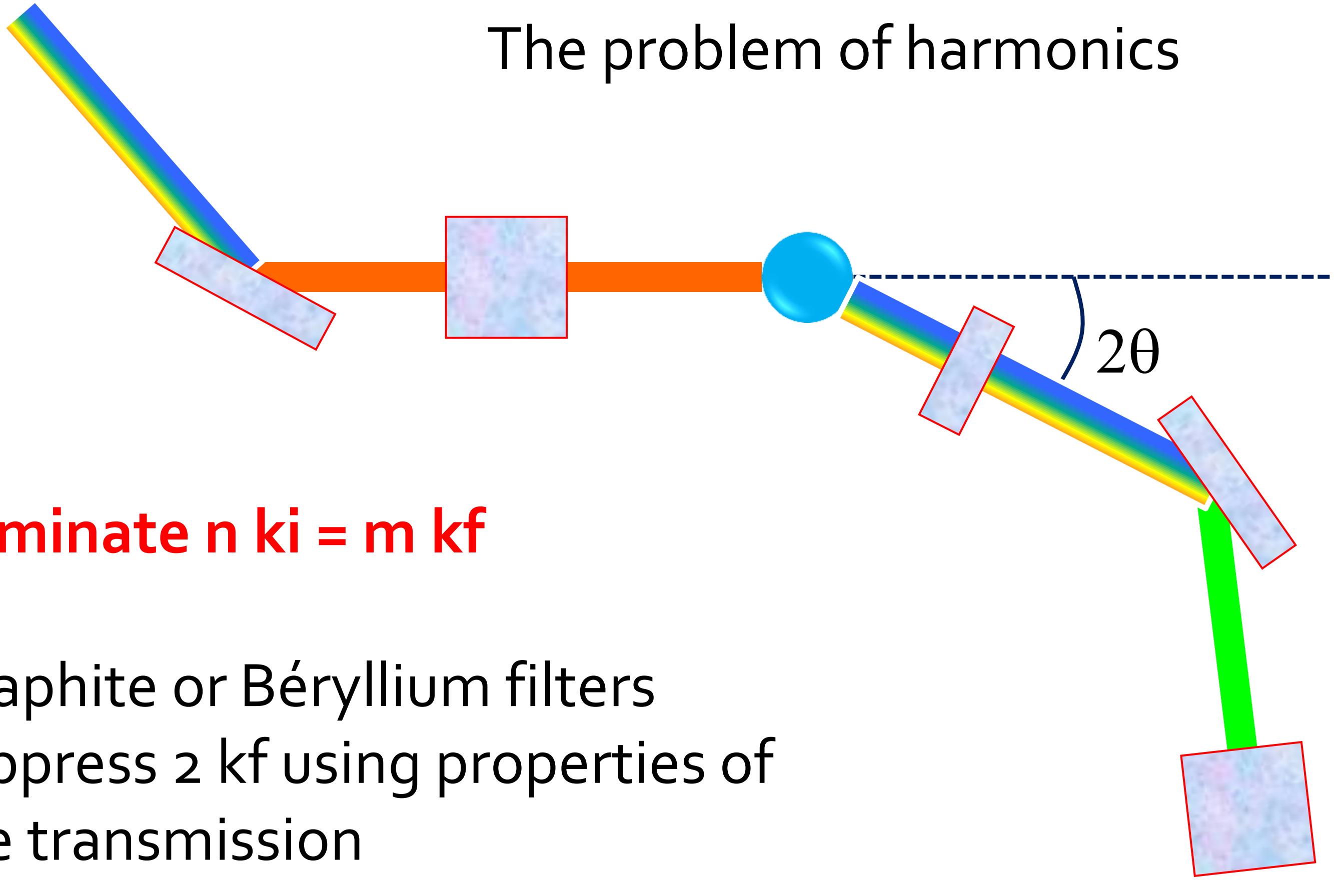


Monitor correction



Filters

The problem of harmonics



Eliminate n ki = m kf

Graphite or Béryllium filters suppress 2 kf using properties of the transmission

3kf is not suppressed !!

Magic values

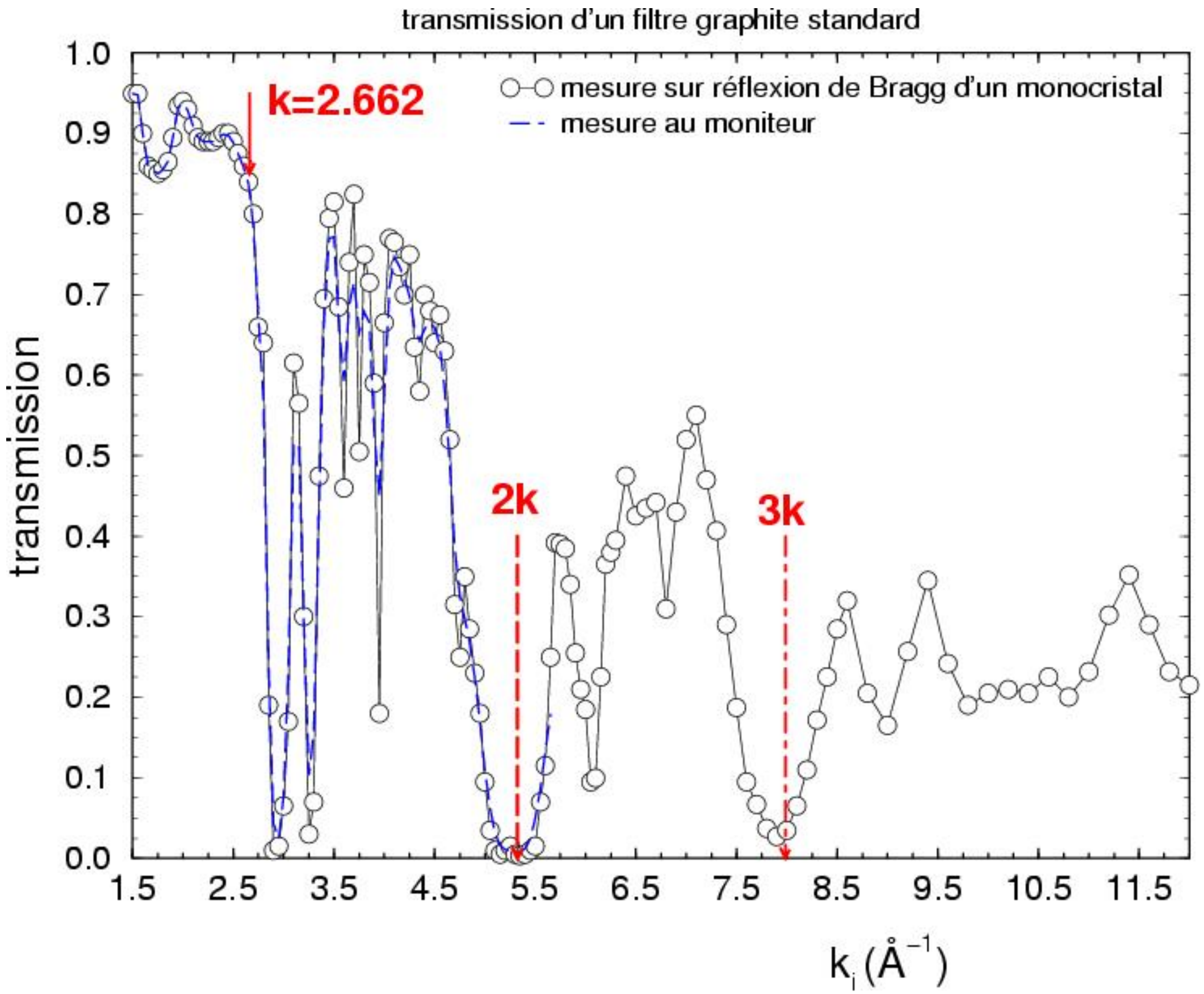
k=2.662 Ǻ$^{-1}$

k=1.97 Ǻ$^{-1}$

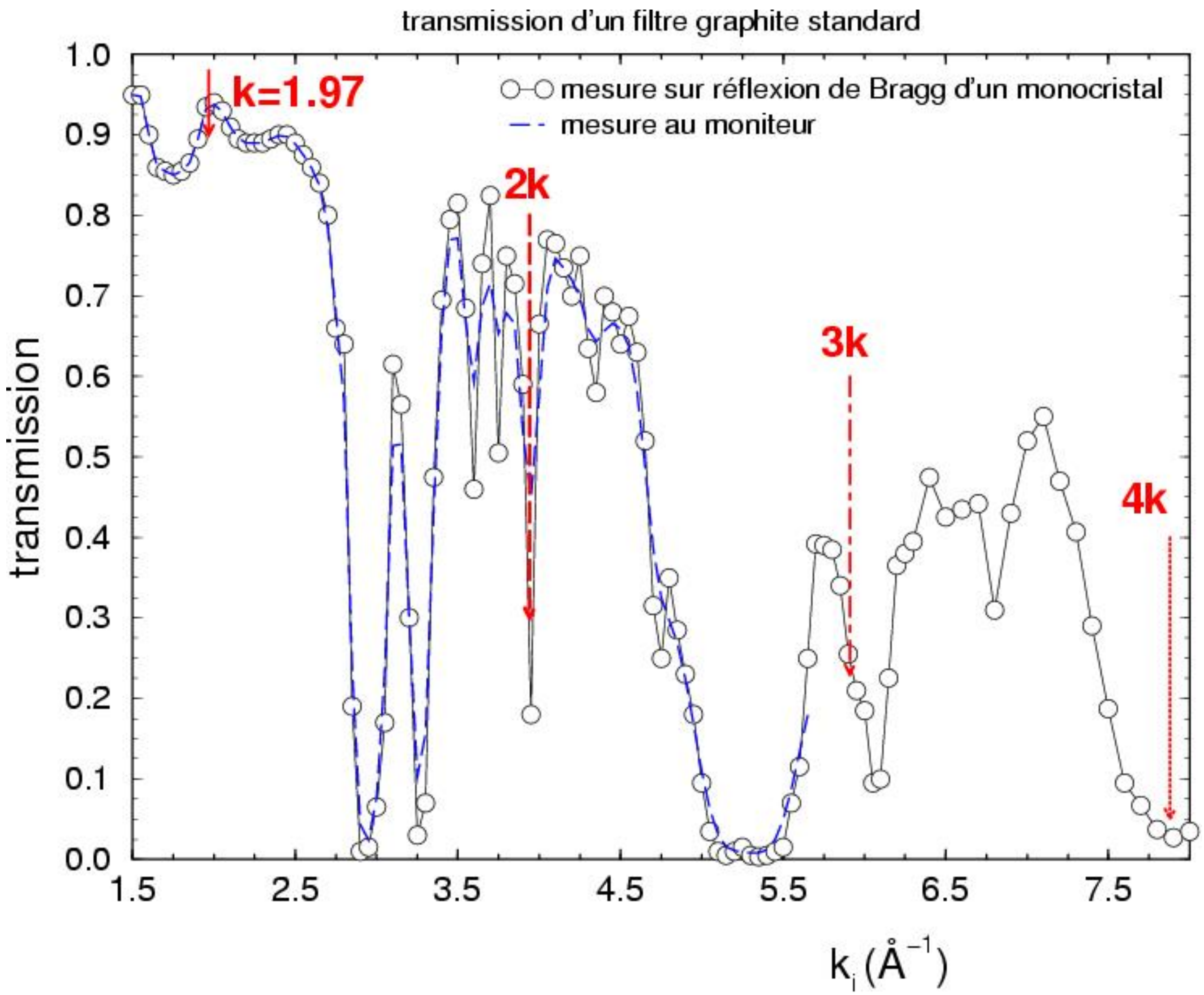
k=1.64 Ǻ$^{-1}$

k=3.85 Ǻ$^{-1}$

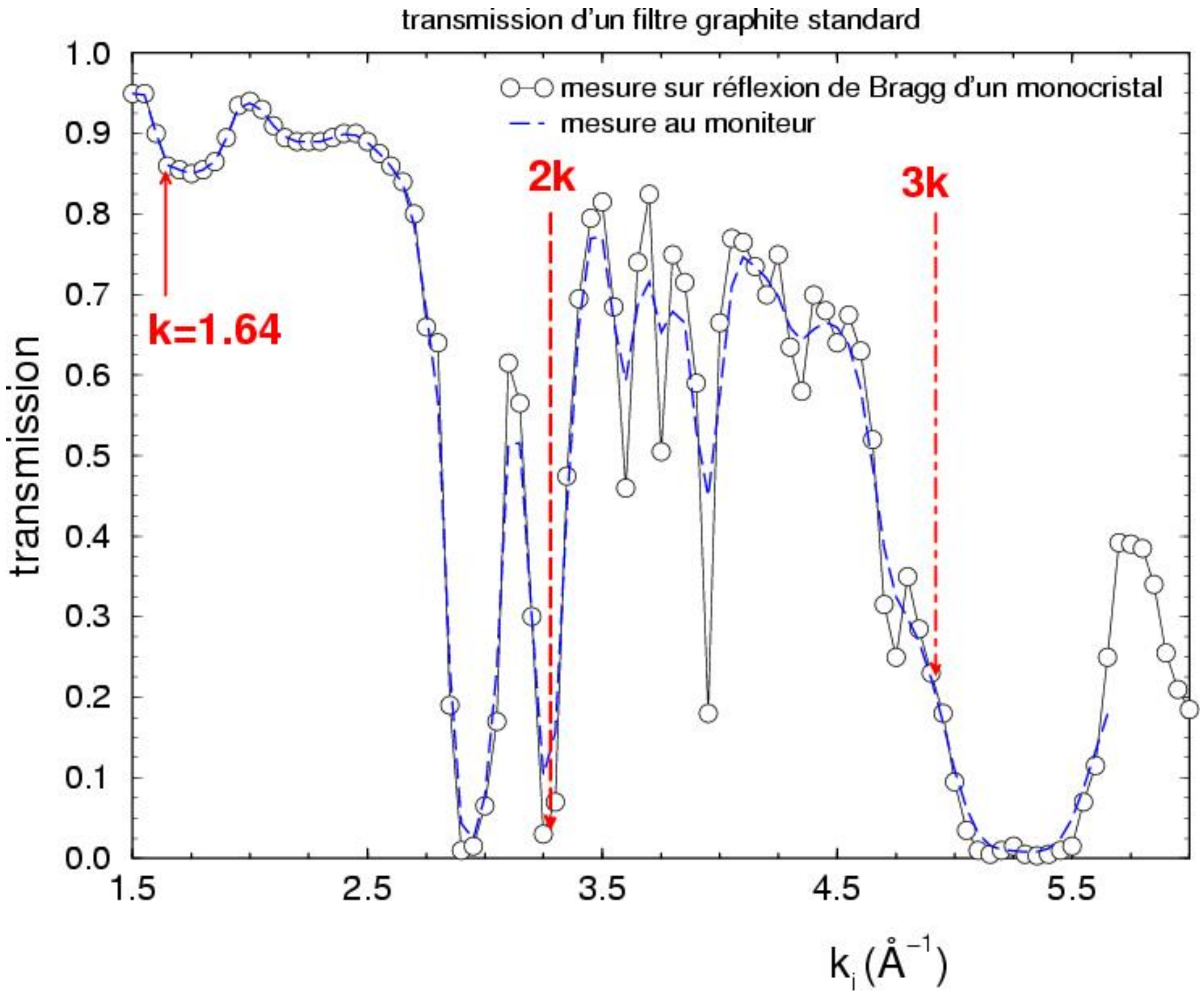
k=4.10 Ǻ$^{-1}$



k=1.97 Ǻ$^{-1}$

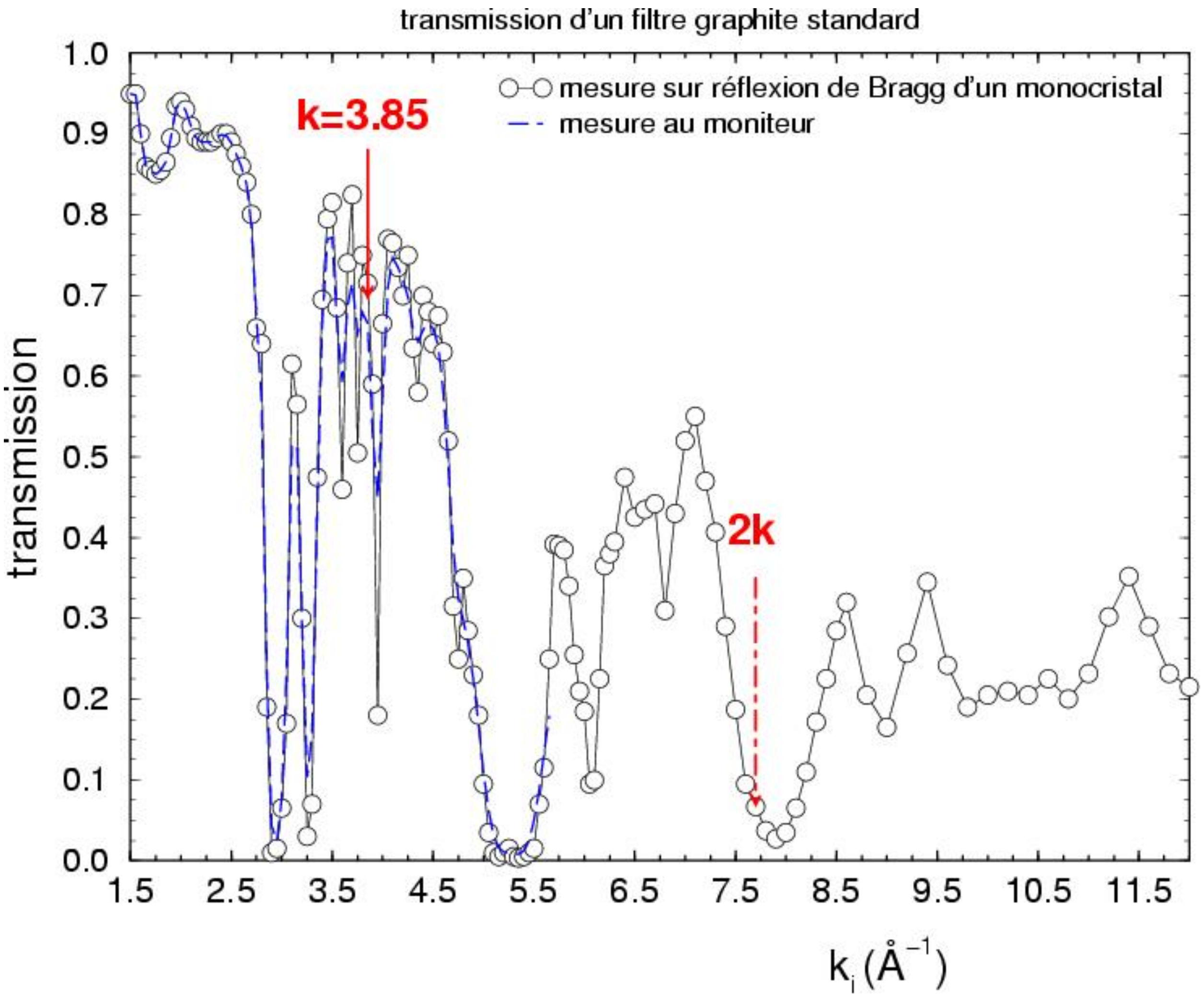

transmission d'un filtre graphite standard
mesure sur réflexion de Bragg d'un monocristal
mesure au moniteur
k=1.97
2k
3k
4k
transmission
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
1.5
2.5
3.5
4.5
5.5
6.5
7.5
k_i (Å^-1)

$k=1.64\ \text{Å}^{-1}$



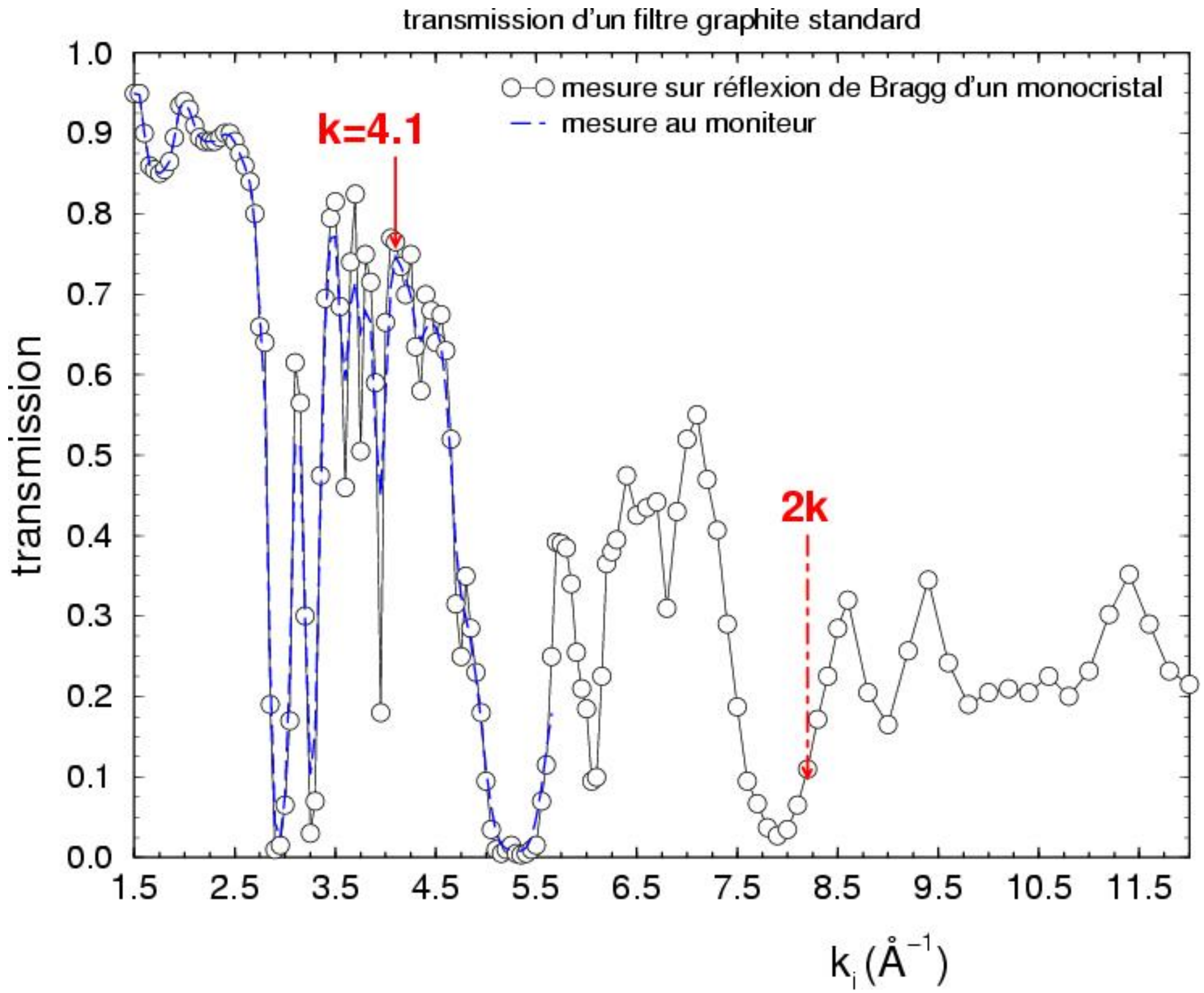
transmission d'un filtre graphite standard
mesure sur réflexion de Bragg d'un monocristal
mesure au moniteur
2k
3k
k=1.64
transmission
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
1.5
2.5
3.5
4.5
5.5
k_i (Å^-1)

k=3.85 Ǻ$^{-1}$



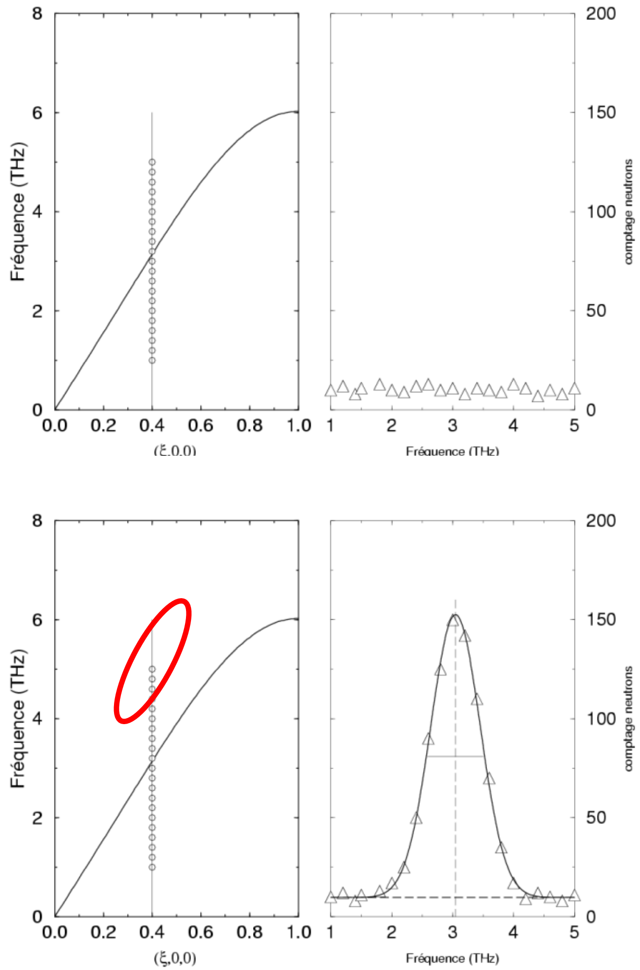
transmission d'un filtre graphite standard
mesure sur réflexion de Bragg d'un monocristal
mesure au moniteur
k=3.85
2k
transmission
k_i (Å^-1)
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
1.5
2.5
3.5
4.5
5.5
6.5
7.5
8.5
9.5
10.5
11.5

k=4.10 Ǻ$^{-1}$



transmission d'un filtre graphite standard
mesure sur réflexion de Bragg d'un monocristal
mesure au moniteur
k=4.1
2k
transmission
k_i (Å^{-1})
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
1.5
2.5
3.5
4.5
5.5
6.5
7.5
8.5
9.5
10.5
11.5

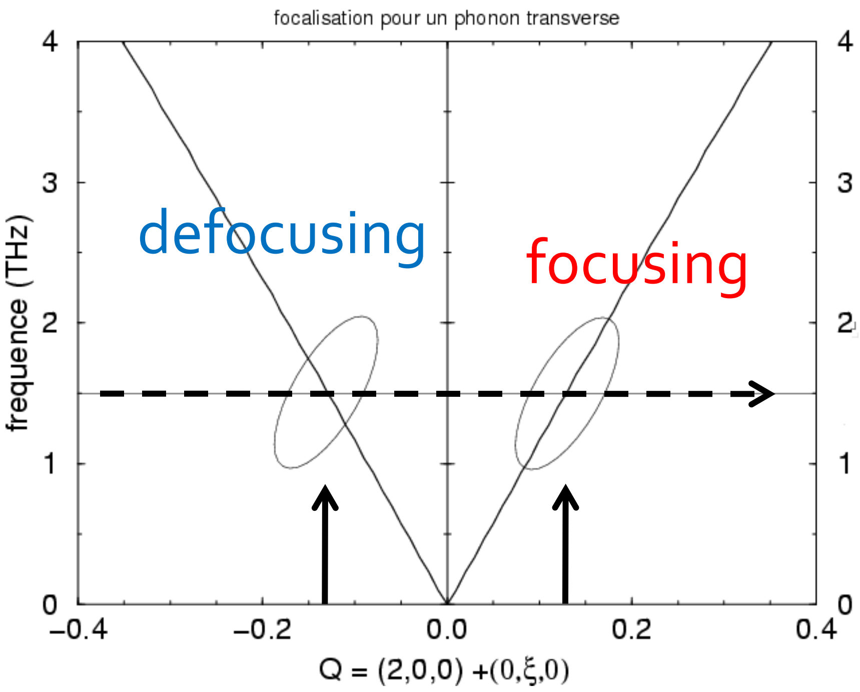
Resolution

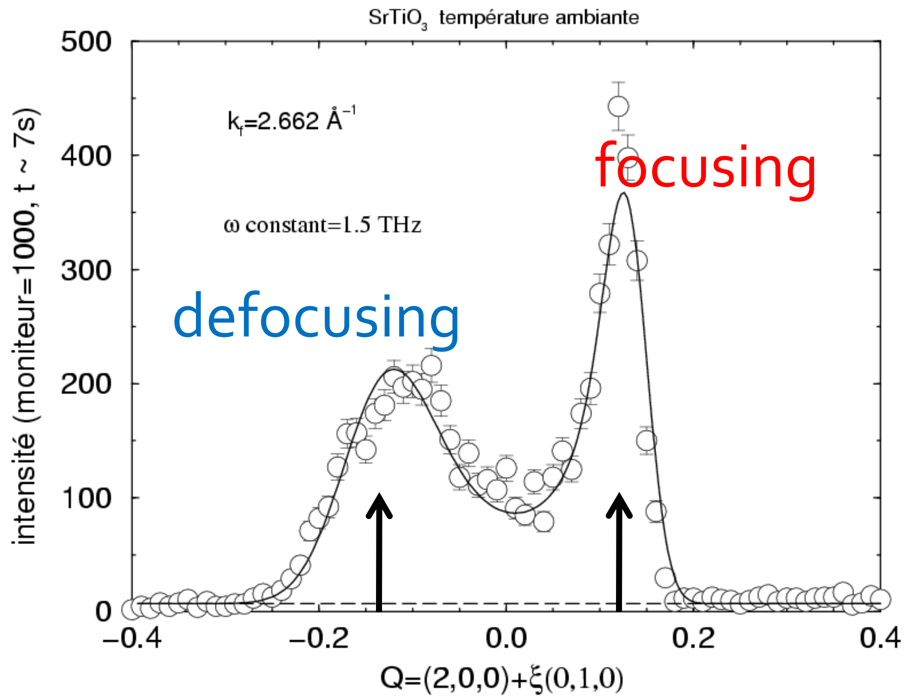


$$S(\vec{Q},\omega) \otimes R(\vec{Q},\omega)$$

The neutron spot in phase-space is an ellipsoid

Focusing effect on two modes at –q and +q





Examples

Spin dynamics in Manganites

$LaSrMnO_3$

(M. Hennion and F. Moussa)

Examples

Example 2

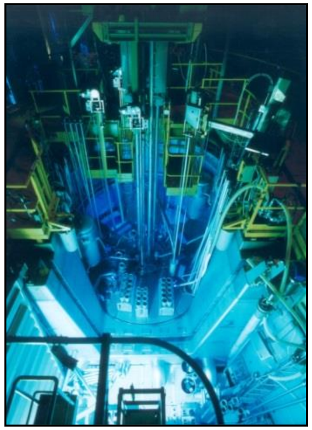
Spin dynamics in Dy thin film

(K. Dumesnil, C. Dufour, IJL Nancy France)

Summary

Crystalline and magnetic structures

Elementary excitations in solids



Large scale structures

Quasi-elastic scattering

Reflectometry (surface)

Inelastic scattering : dispersion of elementary excitations : phonons, spin waves can be measured in (Q,ω) space. With the help of a model, it becomes possible to determine the structure and measure physical parameteres as k, J, D ...