INTRODUCTION TO NEUTRON SCATTERING

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References

https://www.neutron-sciences.org/

Articles from
Eric Ressouche, Mechthilde Enderle, Stéphane Raymond
And many others …

Explore matter

Different states of matter :
liquid, solid, gas, plasmas,
Liquid crystals, smectics, nematics,
macro-molecules,
Magnetic phases, superconductors, Kondo,
spin liquids, Luttinger liquids…

Correlations ?
How move the atoms ?
How move the magnetic moments ?
Are these movements correlated ?

Phase Transitions
spontaneous broken symmetry,
quantum phase transition, topological phase transition …

Explore matter

1895 : RX discovery (Röntgen)

1913, Bragg (son and father) use XR to determine the structure of NaCl and of many other salts (Nobel prize 1915)

Diffraction with massive particles ?

YES if $\lambda \approx d$

☺ : λ of thermal neutrons ≈ few Å

Explore matter with neutrons

Diffraction with massive particles ?

Complementary to Xrays because the interaction between neutrons and matter is different from the interactions between light and matter :

+ isotopes
+ magnetism
+ volume

AND :
E ≈ typical energy of excitations (meV): **spectroscopy in reciprocal space**

Explore matter with neutrons

More generally, neutron scattering allows to :

Measure correlations between atomic positions
in space and time

Measure correlations between magnetic moment
orientations
in space and time

Where are the atoms ?

What do they do ?

A neutron is …

- Not charged
- Has a mass

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

$$\begin{aligned} S &= 1/2 \\ S_z &= \pm 1/2 \end{aligned}$$

- Carries a spin ½
- Also a « wave of matter »

A neutron is …

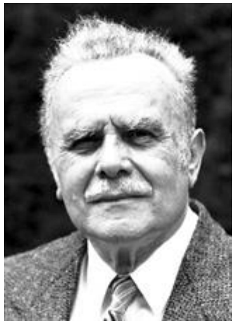
1911 : nucleus (Rutherford)

1913 : « planets model » (Bohr)

1932 : neutron discovery (J. Chadwick)

1940-1950 : construction of reactors in the US and in Canada
(Chalk River's NRX reactor, ORNL's graphite reactor)
E. Wollan et C. Shull : diffraction

1956: construction of the first 3-axis spectrometer at Chalk-River by B. Brockhouse.



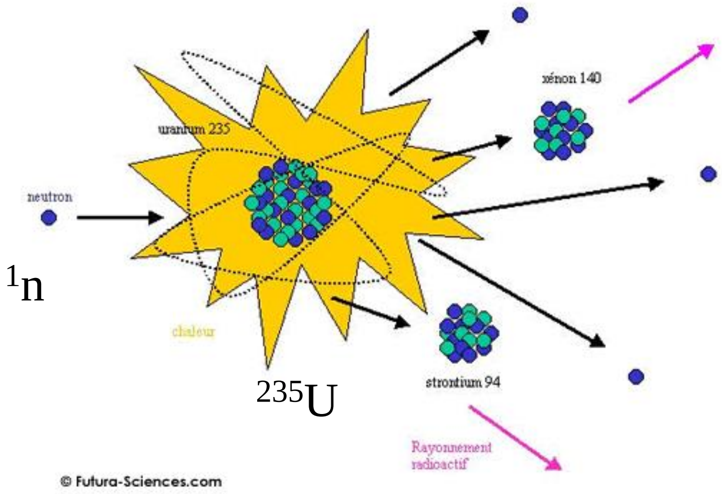
Bertram N. Brockhouse

Clifford G. Shull

Producing neutrons

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

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


uranium 235
xénon 140
neutron
^1n
chaleur
^{235}U
strontium 94
Rayonnement radioactif
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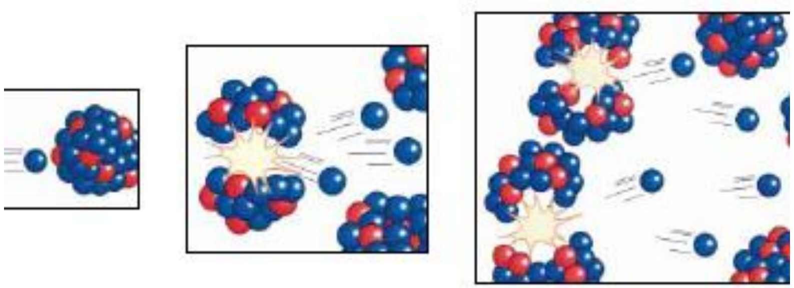
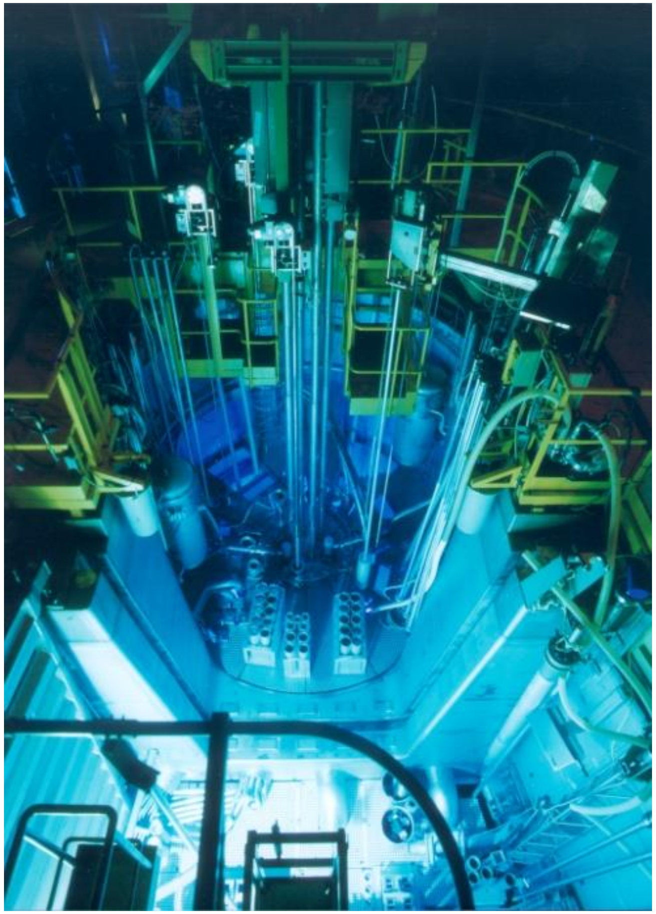
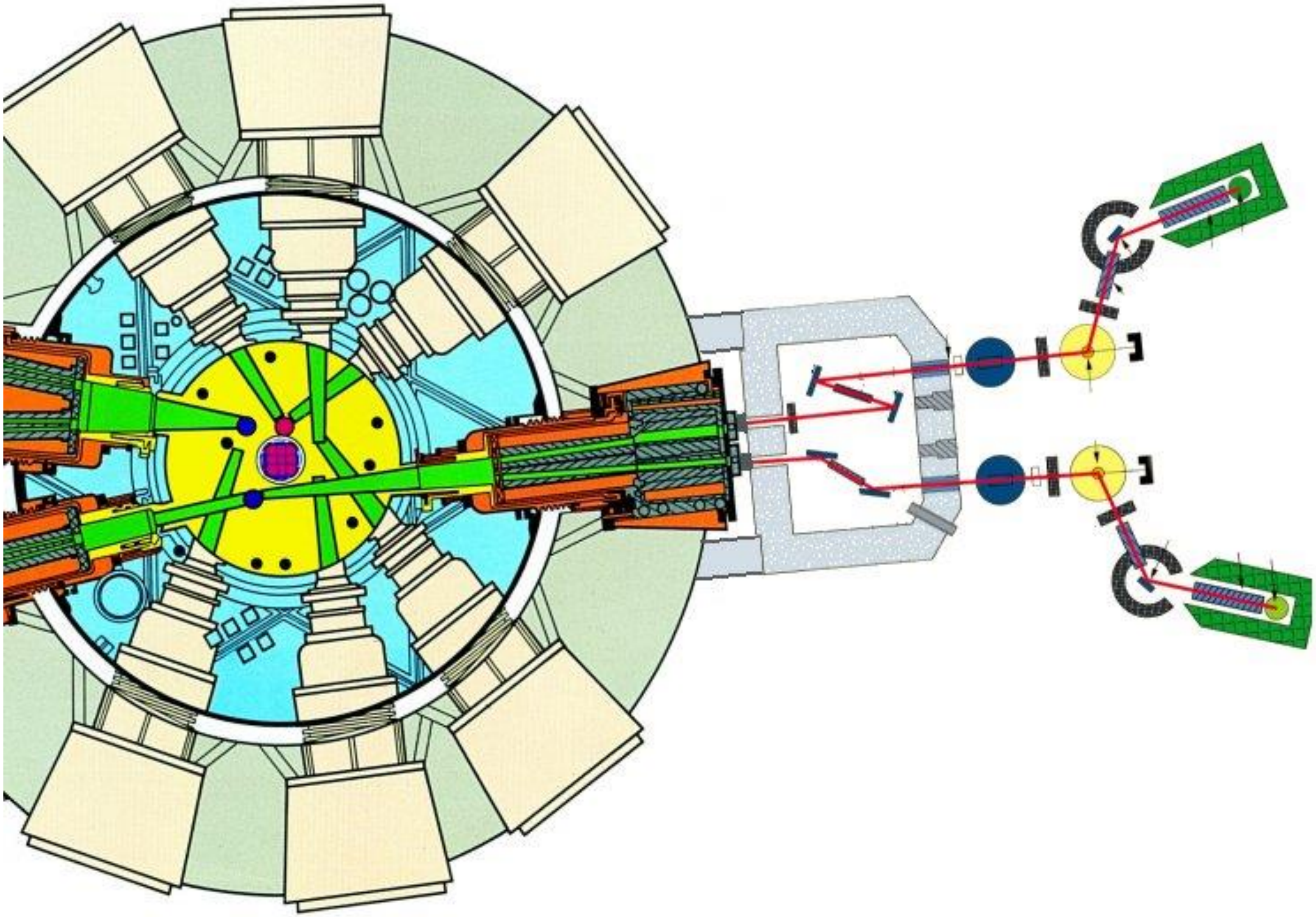




Producing neutrons



Producing neutrons

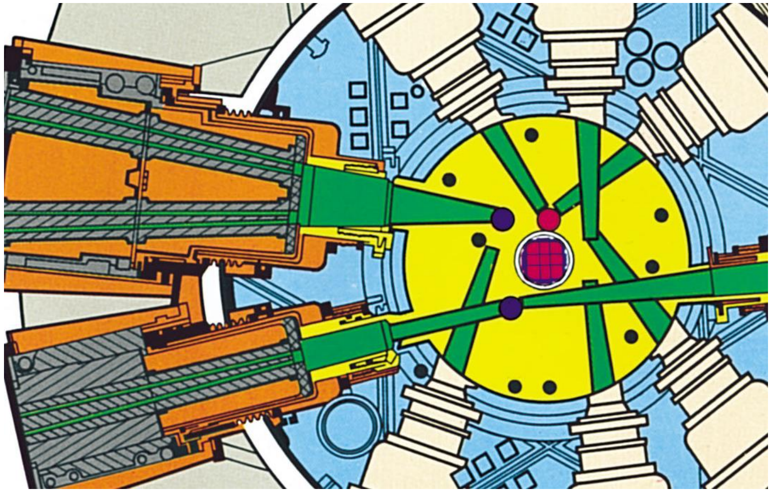
Producing neutrons

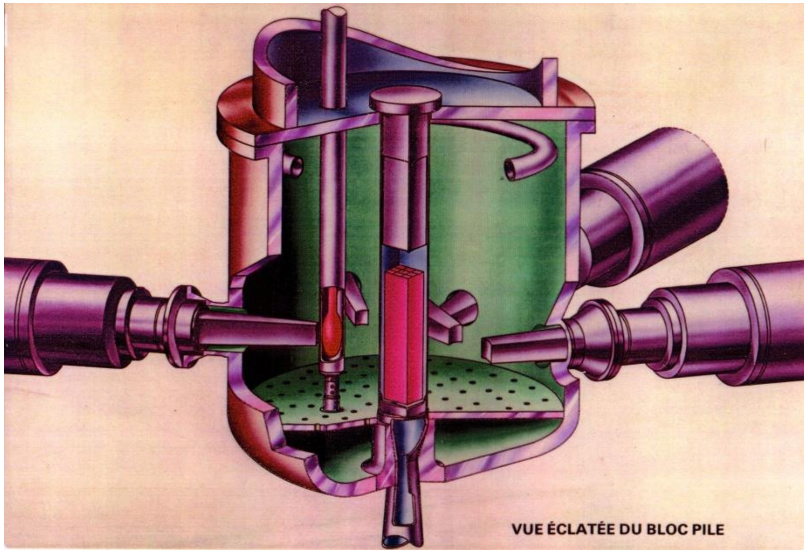
Orphée reactor

4 thermal beam tubes

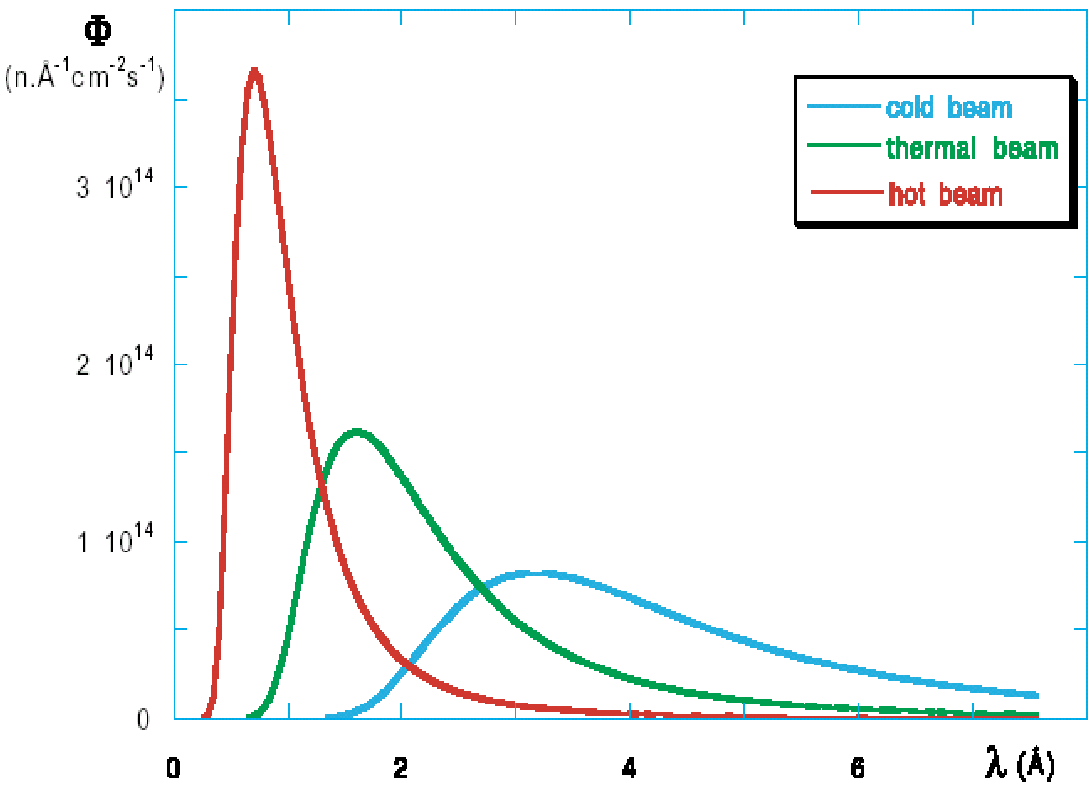
2 hot beam tubes

3 cold beam tubes





Producing neutrons



Differential flux (per Å) given by local moderators

<u>Hot neutrons</u>
Structure of liquids and crystals

<u>Thermal neutrons</u>
Condensed matter

<u>Cold neutrons</u>
Condensed matter and soft matter

Neutron-matter interaction

Strong Interaction : the neutron enters into the nucleus, forms a composite nucleus and eventually evaporates.

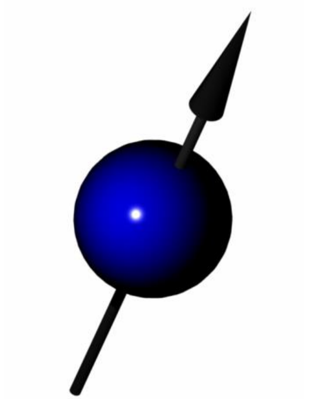
$$\hat{V}_n(\vec{r}) = \frac{2\pi\hbar^2}{M} b\delta(\vec{r} - \vec{R}).$$

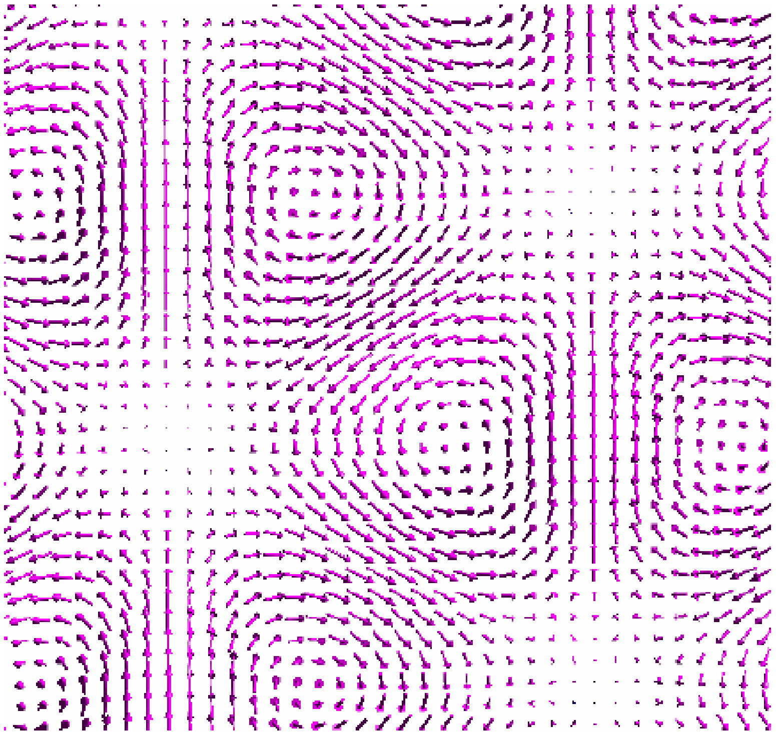
b = « scattering length »
positive or negative
depends on the isotope

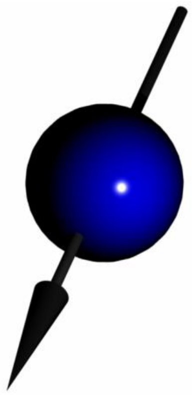
Neutron-matter interaction

Dipolar magnetic Interaction between the spin of the neutron and the assembly of spins within the sample

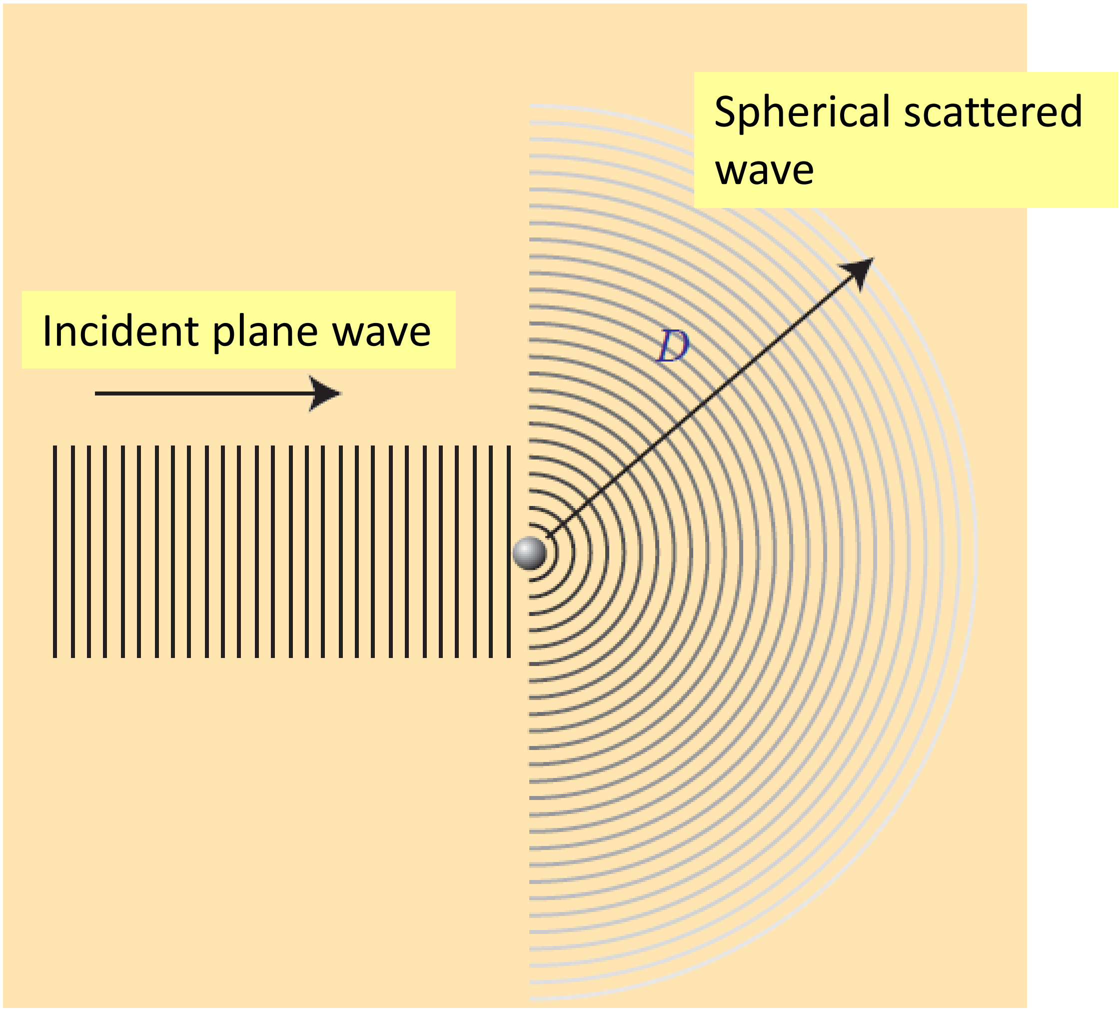
$$E_{ne} = -\mu_n . B_e \qquad B_e(R) = \frac{\mu_o}{4\pi}\left(\mathrm{rot}(\frac{\mu_e \times R}{R^3}) - e\ v_e \times \frac{R}{R^3}\right)$$



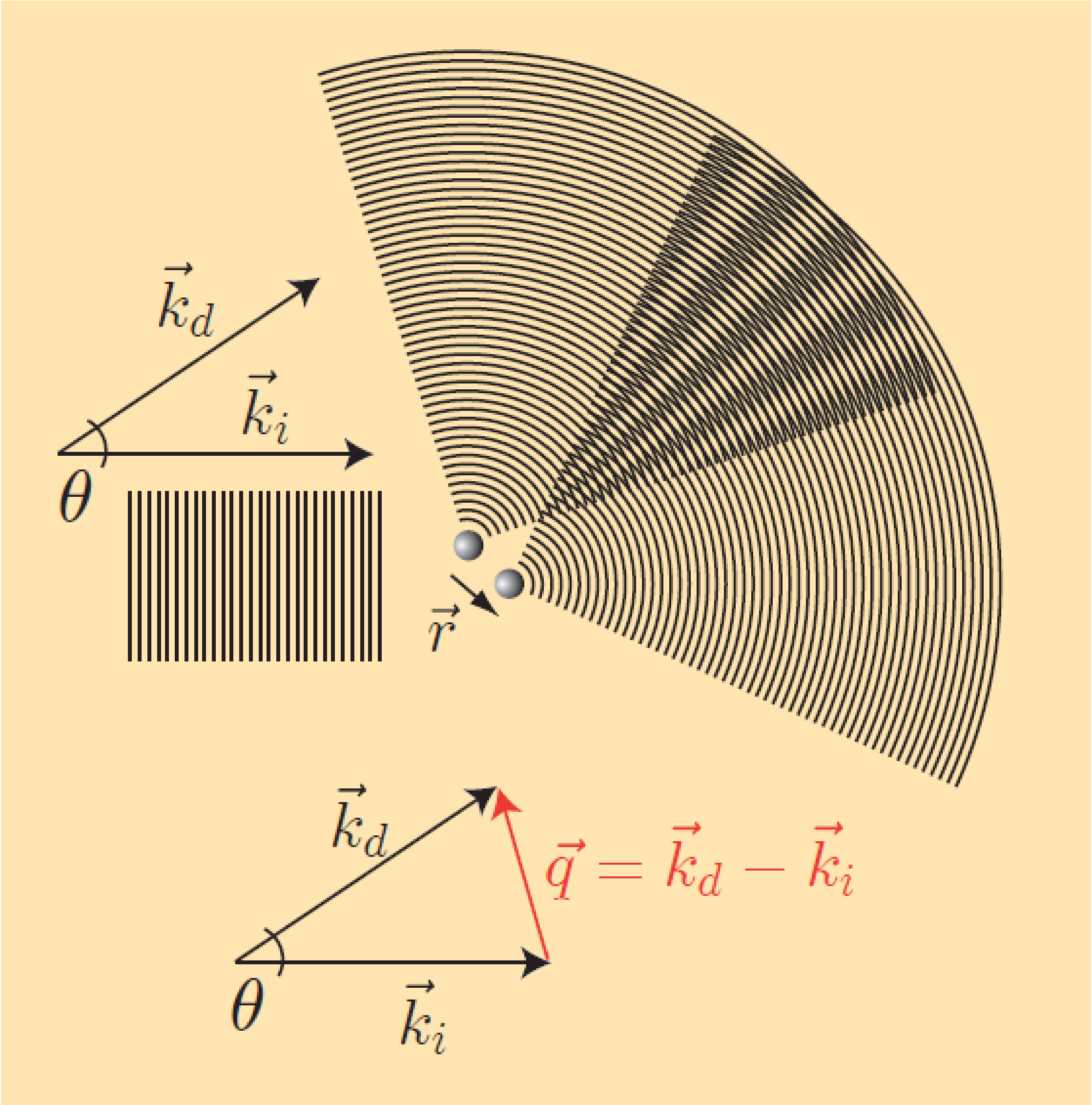




Diffraction



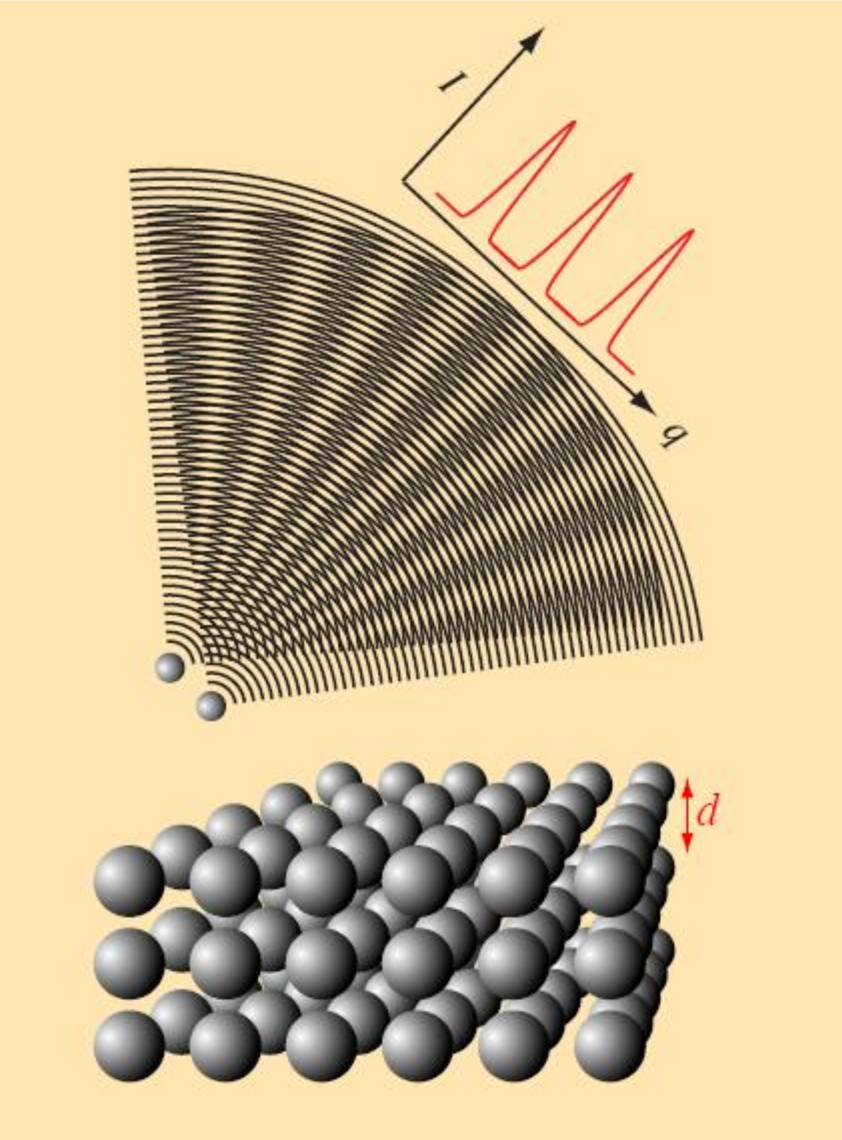
Diffraction



Phase difference

$$\Delta\phi = \vec{r}(\vec{k}_d - \vec{k}_i)$$

Diffraction



Sum of partial waves

Incident amplitude

$$\psi = \frac{\psi_0}{D} \times \sum_{i=1}^{n} b_i e^{iqr_i}$$

D = distance to the detector

Scattered intensity : $\psi^2 = \psi\psi^*$

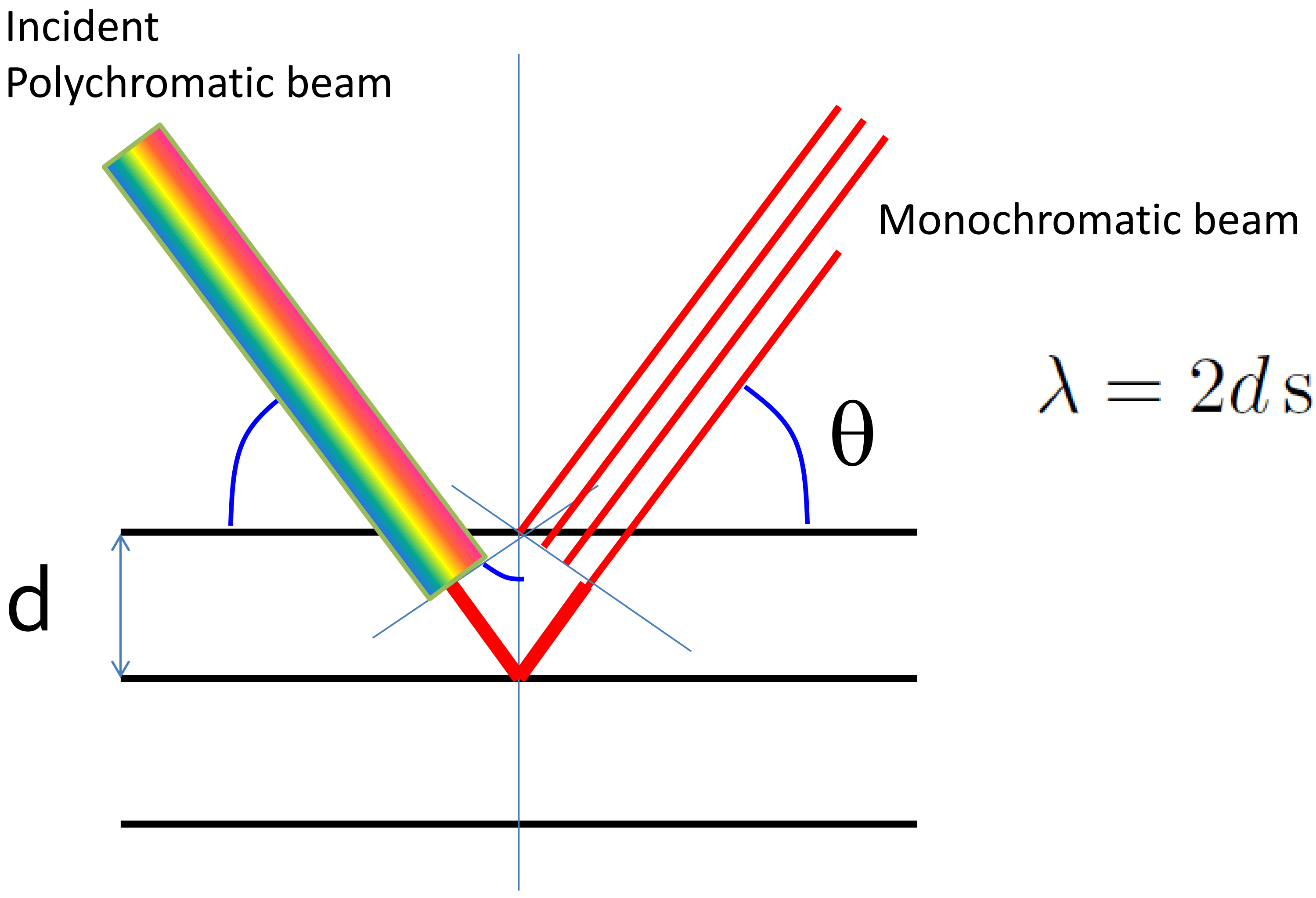
$$\psi^2 = \frac{\psi_0^2}{D^2} \times \left[\sum_i^n b_i e^{iqr_i} \times \sum_j^n b_j e^{-iqr_j} \right]$$

$$= \frac{\psi_0^2}{D^2} \times \sum_i^n \sum_j^n b_i b_j e^{iq(r_i - r_j)}$$

CONSEQUENCE 1:

It is possible to prepare a monochromatic neutron beam

Monochromatic neutron Flux



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

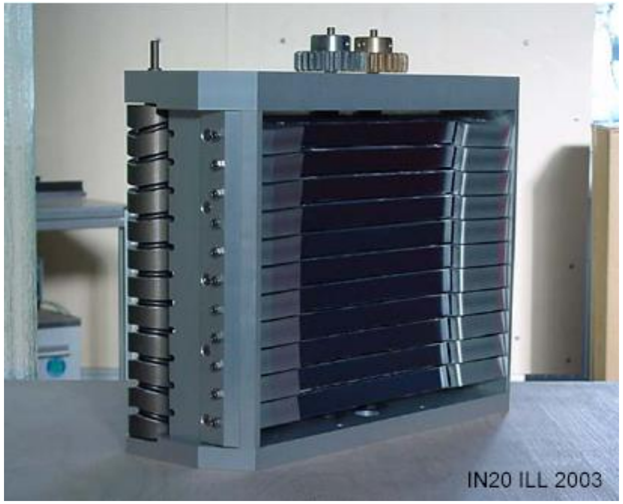
Monochromatic neutron Flux

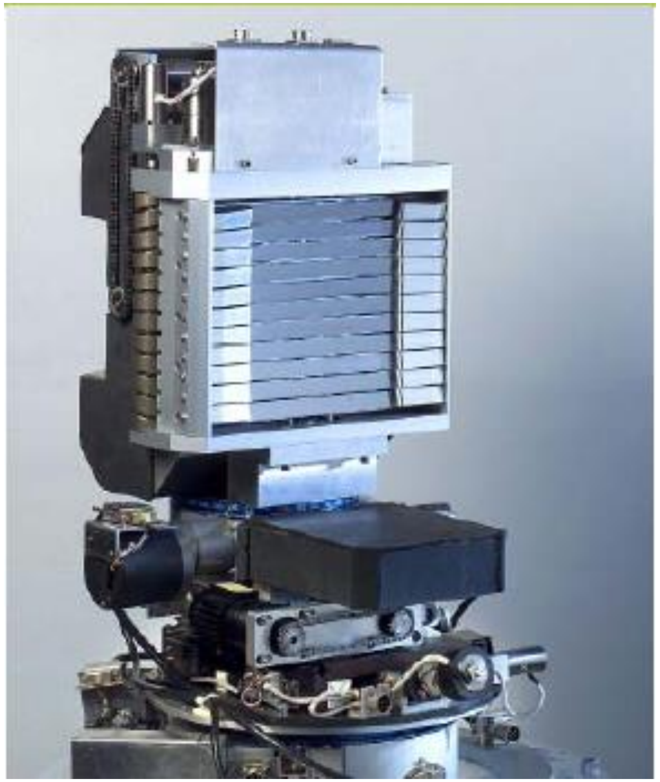
Graphite

Cu

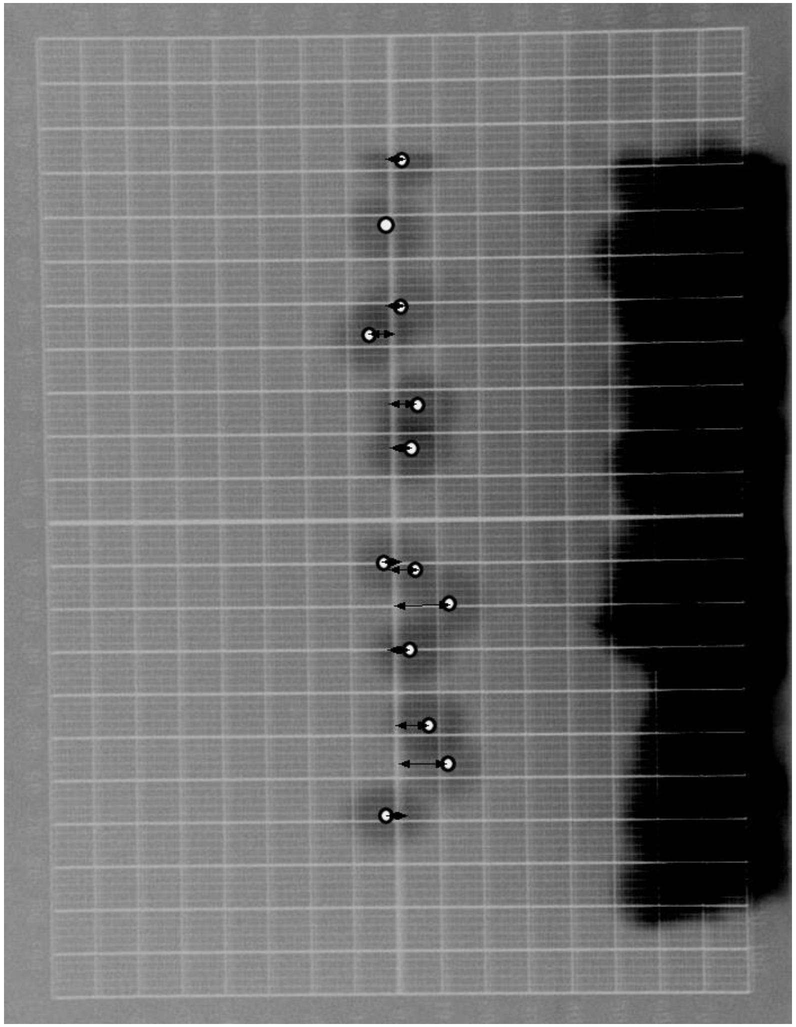
- Low absorption
- Strong reflectivity
- d-spacing
- Single crystals
- « low » Cost

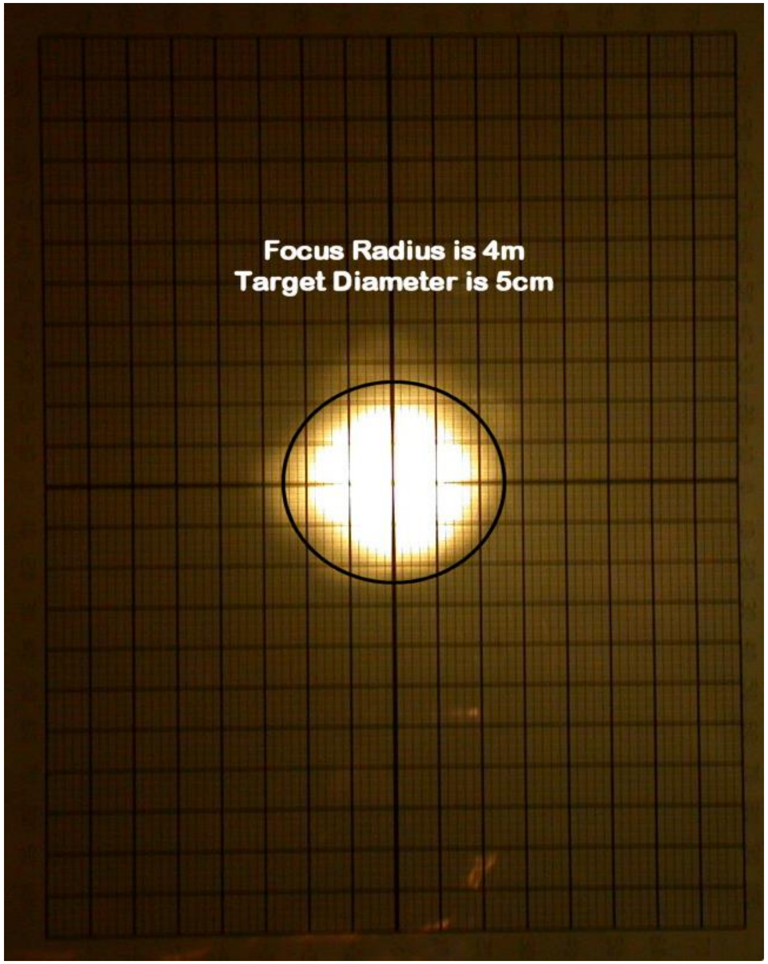
Monochromatic neutron Flux





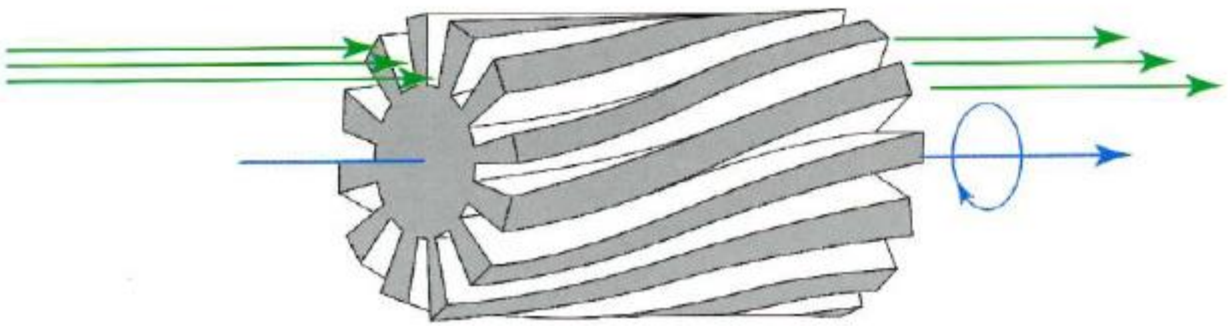
Monochromatic neutron Flux





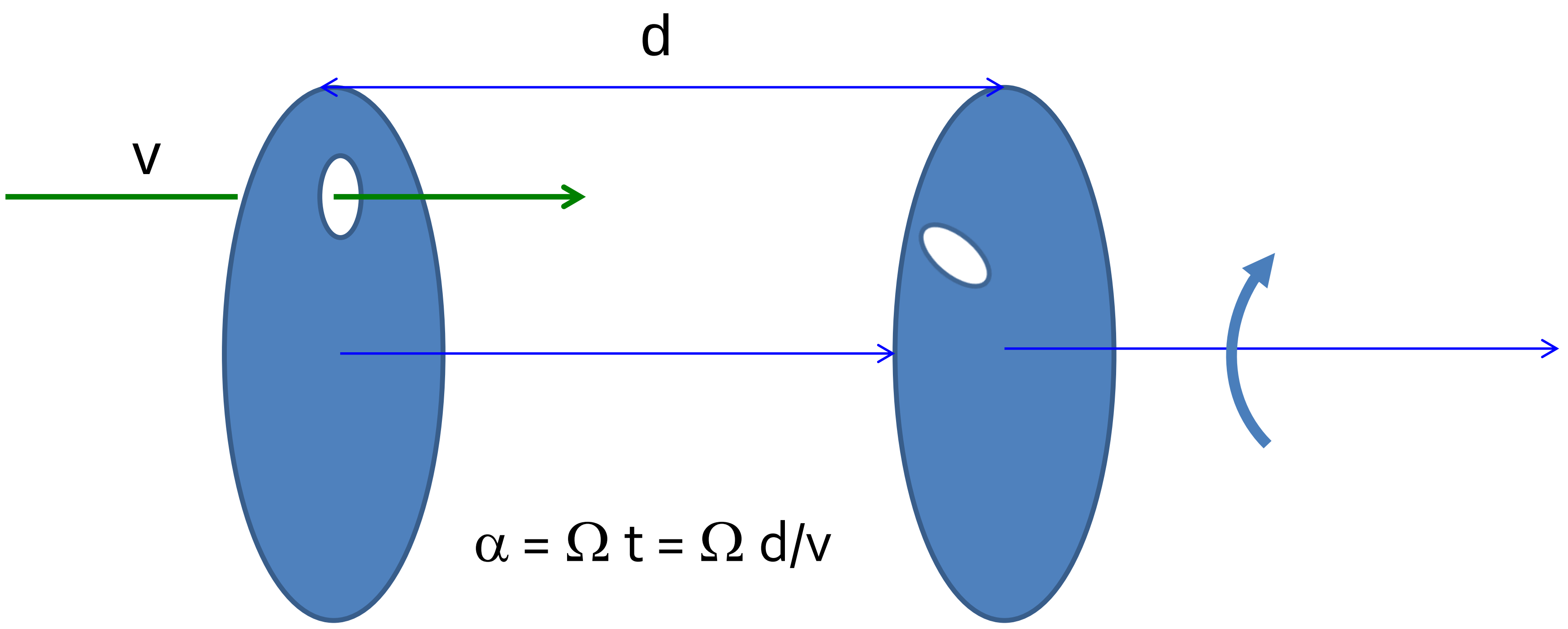
Monochromatic neutron Flux

Velocity selector



Monochromatic neutron Flux

Disk Chopper

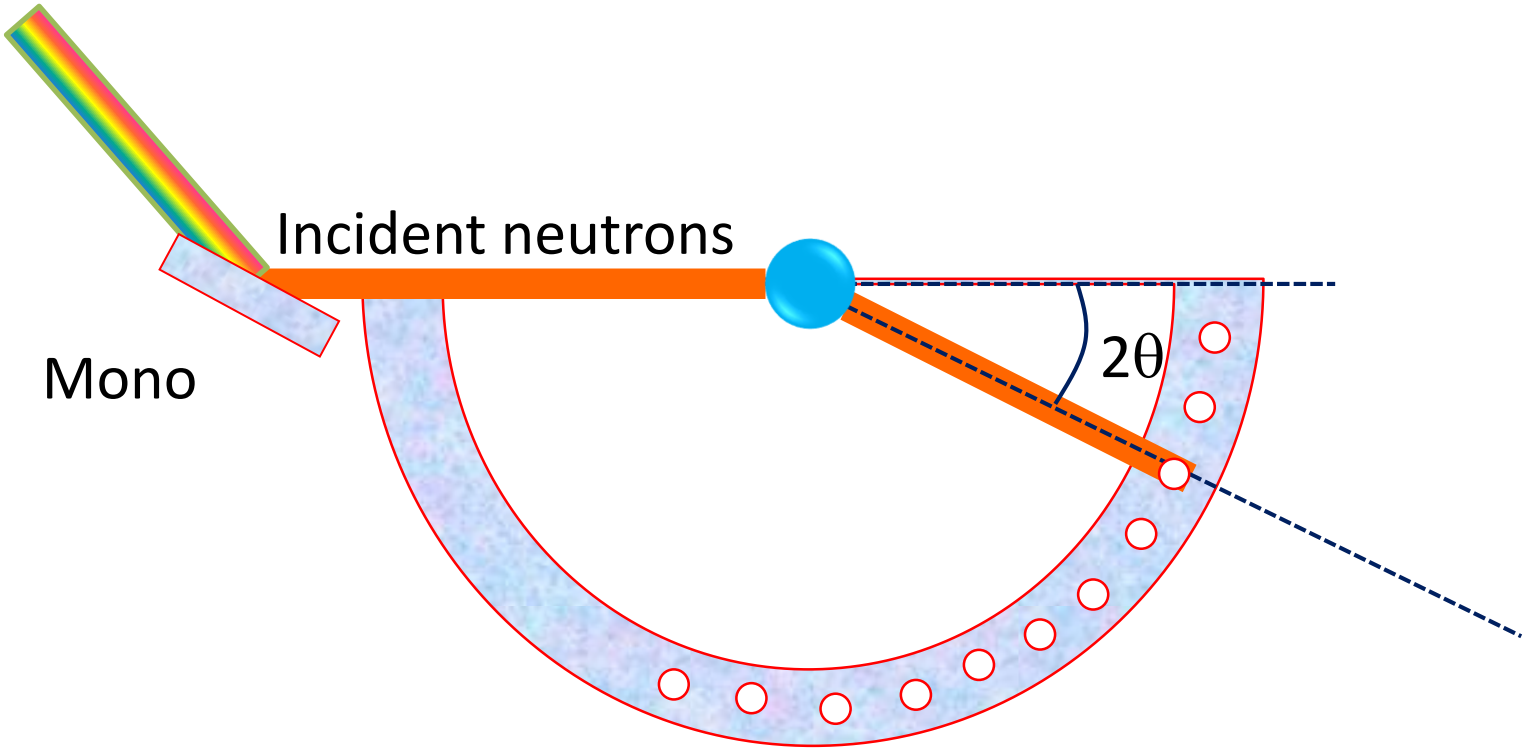


Produces a pulse of a given energy

CONSEQUENCE 2

diffractometer

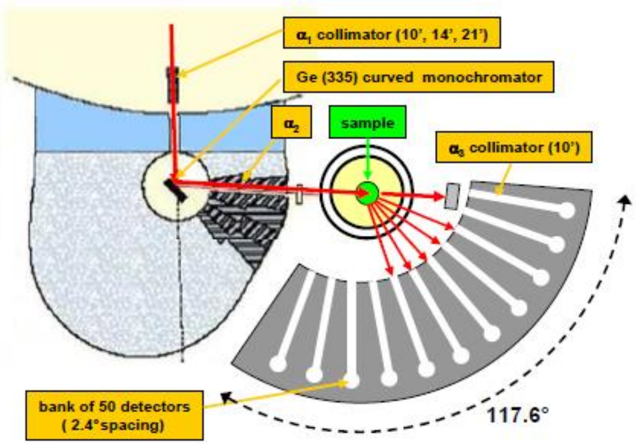
Powder diffractometer

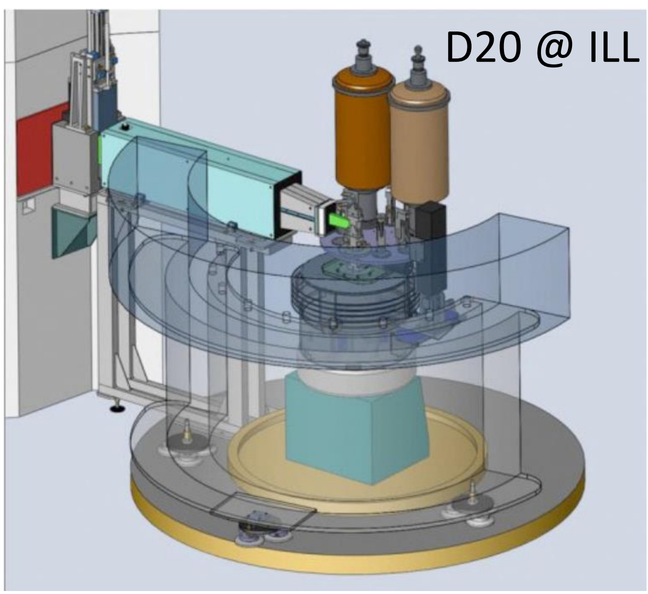


Powder diffractometer

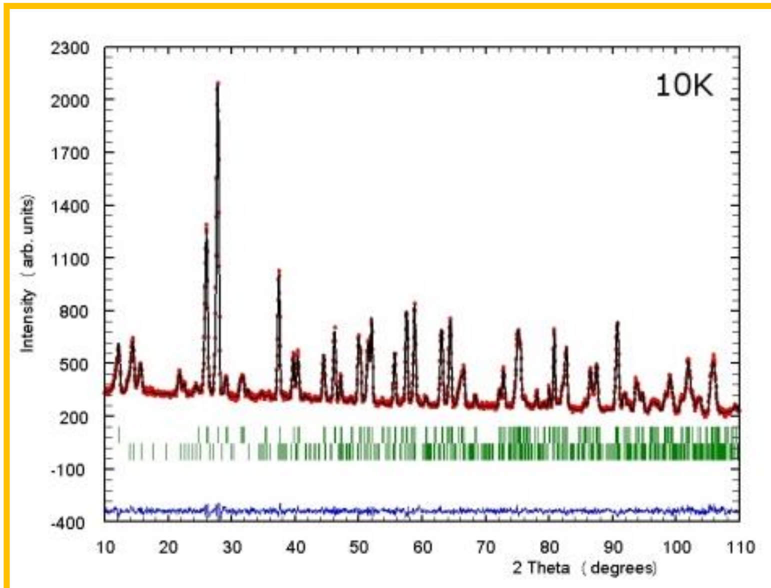
High Resolution Powder Diffractometer

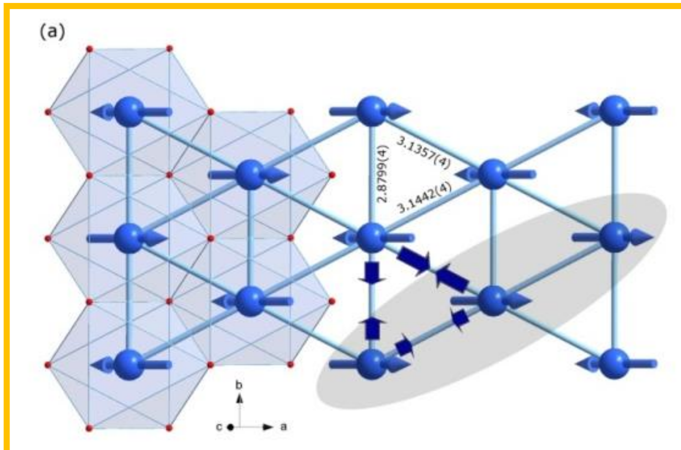
3T2 @ LLB



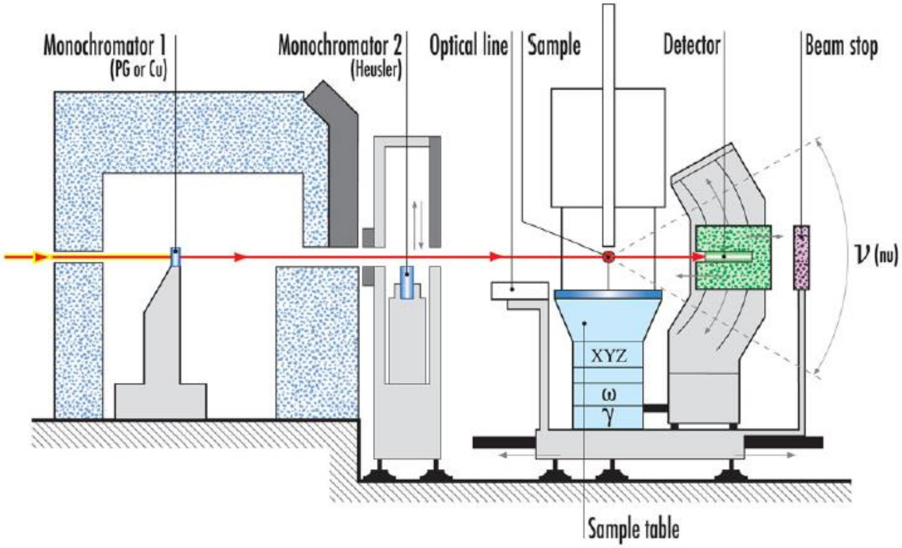


Pcr file !!



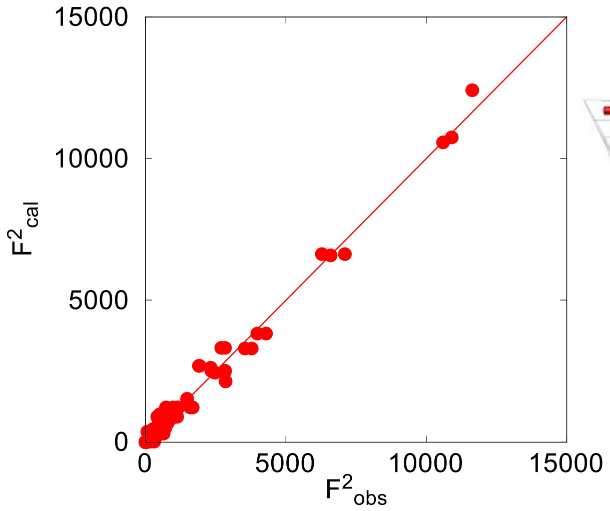


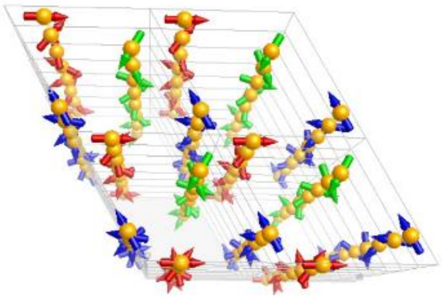
Xcrystal diffractometer

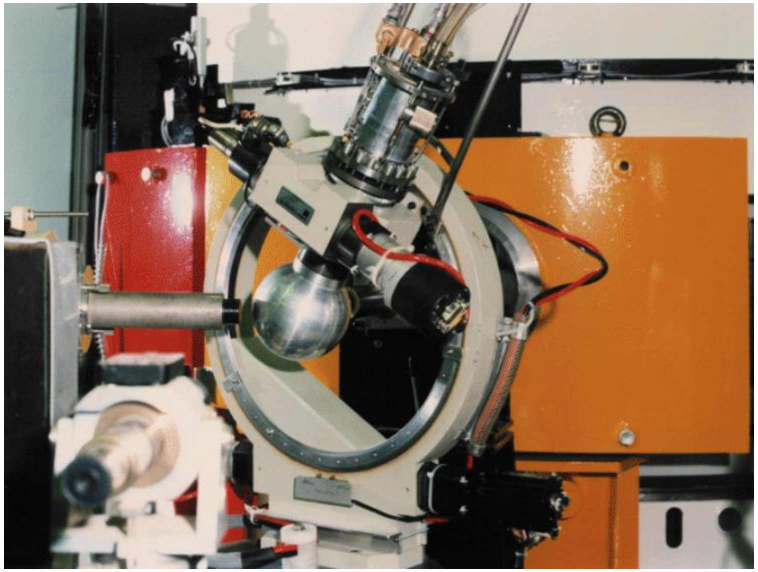


D23 (CRG CEA, ILL)
Lifting arm

D9 (ILL)
Eulerian cradle







SANS

Compressibility, form of macromolecules, interfaces :
Sticks, spheres, …

C. Loupiac et al. (2002), *Eur. J. Biochem.*, 269 : 4731-4737 (PACE, LLB, Saclay)

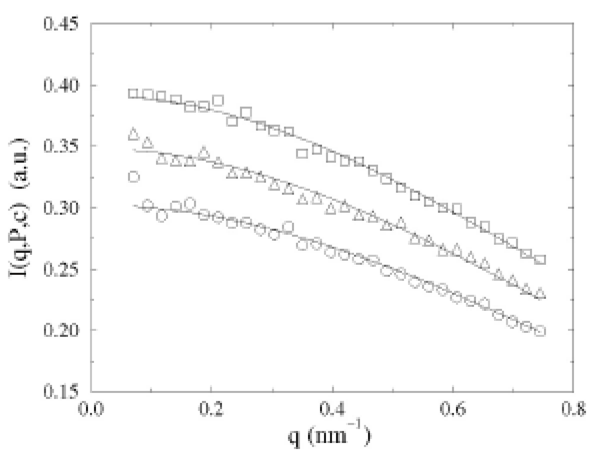


Fig. 1. Scattering spectra $I(q,P,c)$ of MbN_3 at p^2H 6.6, as a function of the wave-number transfer *q*. The measurements were performed at room temperature. The protein concentration, *c*, at atmospheric pressure is 11.7 mg·cm^{-3} and the pressures, *P*, are: 54 (○), 154 (△), and 302 (□) MPa. Fits of Eqn 6 to the data are shown as full lines.

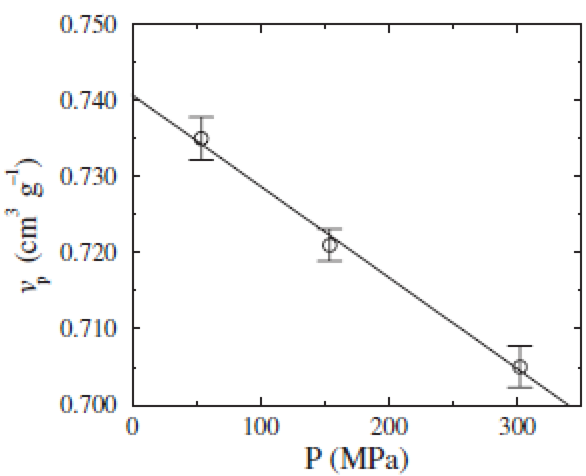


Fig. 7. Partial specific volume $v_p(P)$, of MbN_3 as a function of pressure, *P*. The almost linear variation of $v_p(P)$ with *P* allows the isothermal compressibility of the hydrated protein to be computed: $\kappa_{T,p}$ = (1.6 ± 0.1) 10^{-4} MPa^{-1}.

Take-home message (until now)…

1. Neutron diffraction allows to study crystalline and magnetic structures.

2. Any kind of materials (condensed or soft matter : macromolecules, biologie, crystals ..)

3. sensible to the volume and not to the surface

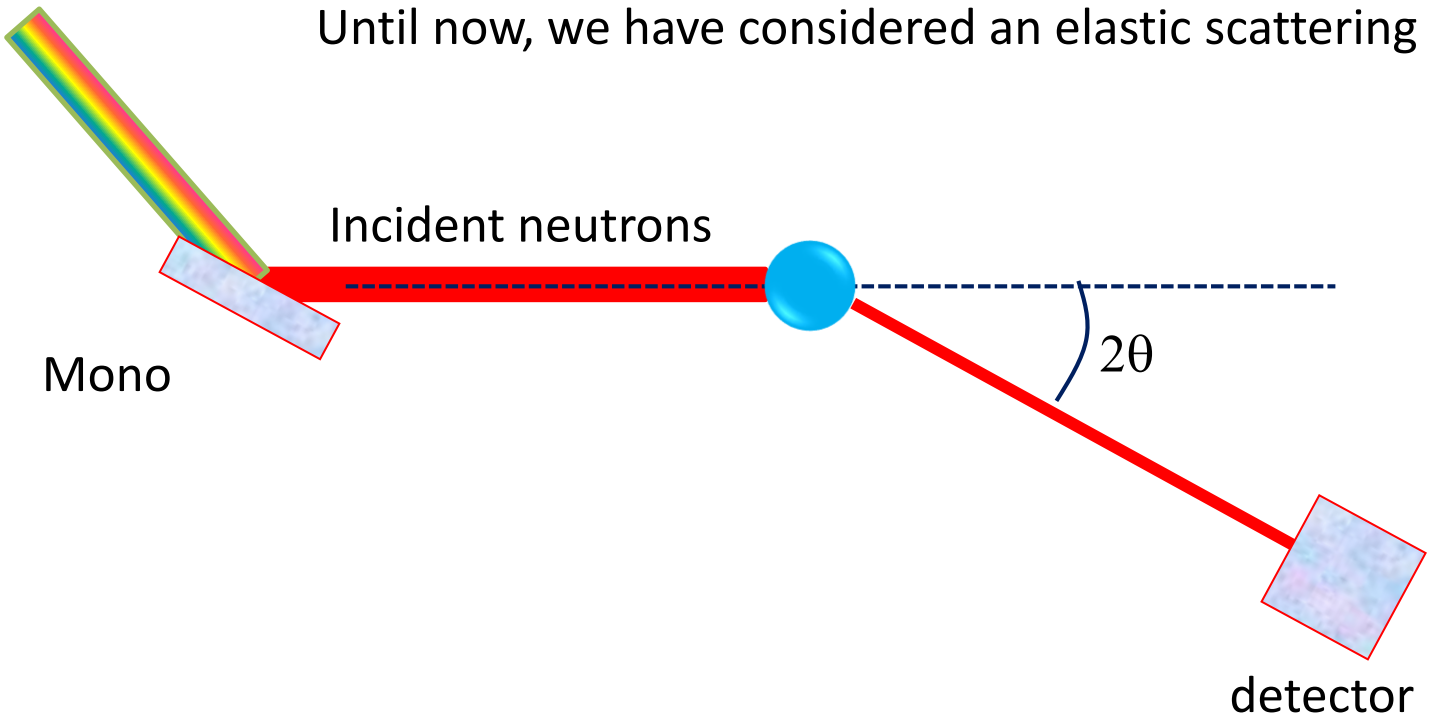
4. Do not destroy the sample (but radioactive)

CONSEQUENCE 3

Inelastic neutron scattering

Inelastic neutron scattering

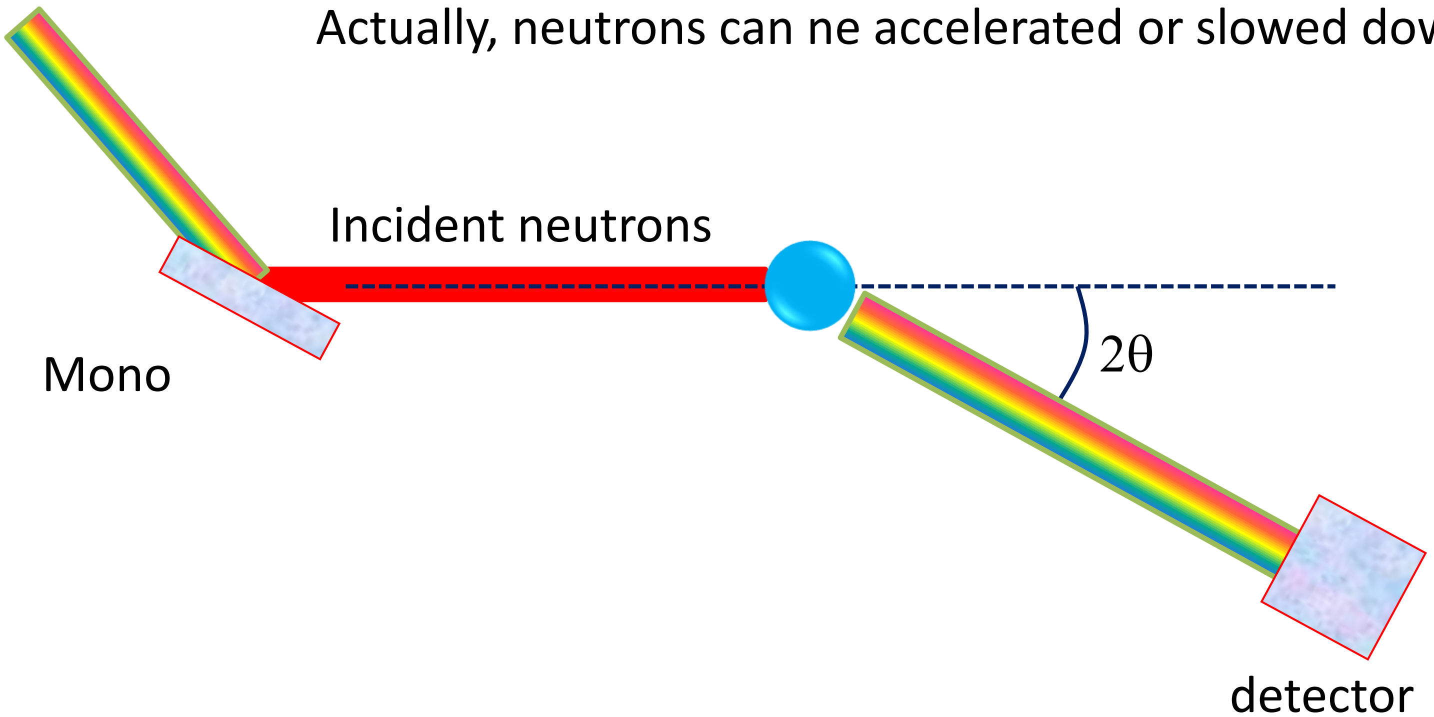
Until now, we have considered an elastic scattering



neutrons in the detector ≈ $\dfrac{\partial^2\sigma}{\partial\Omega\partial E}$

Inelastic neutron scattering

Actually, neutrons can ne accelerated or slowed down :



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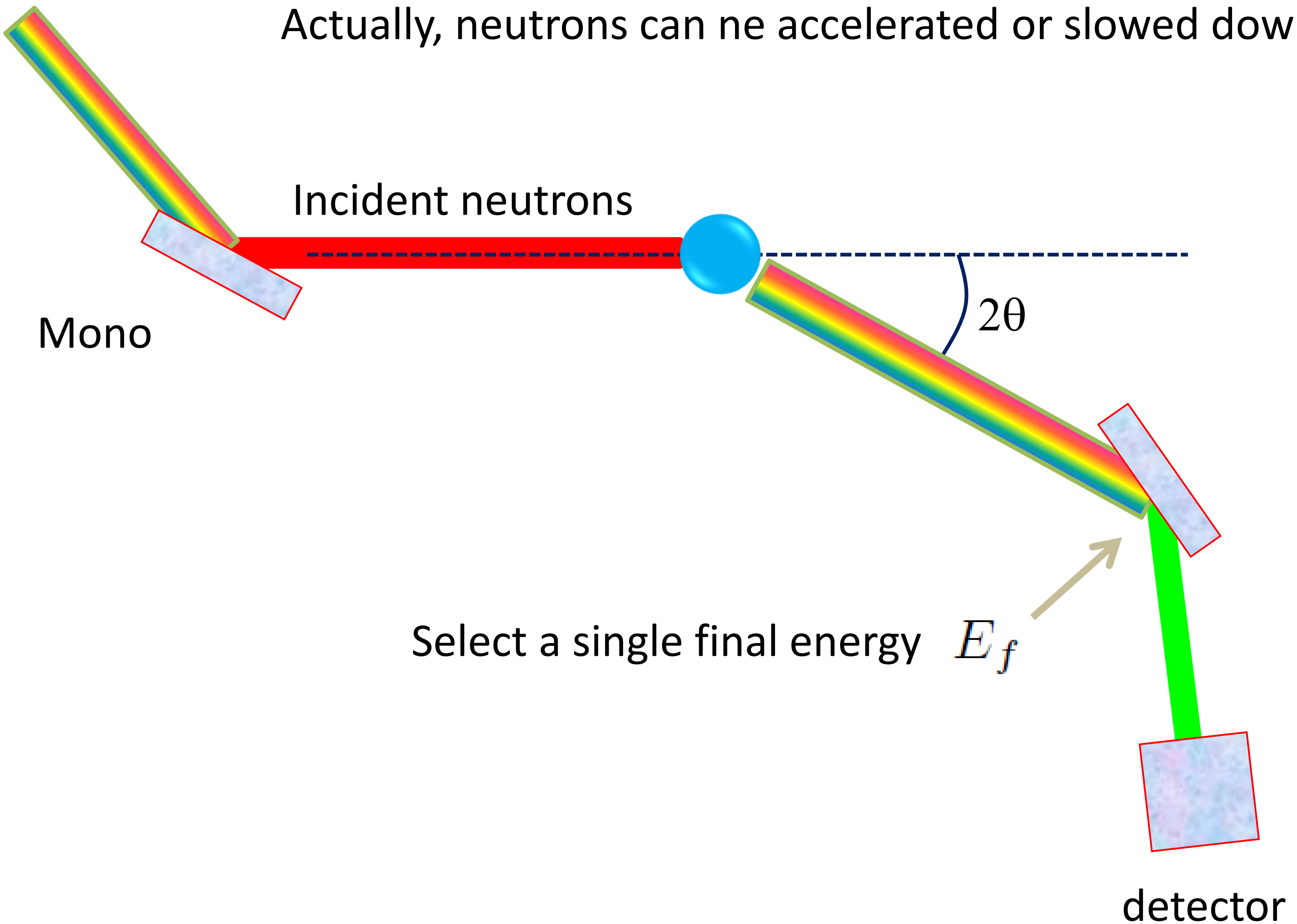
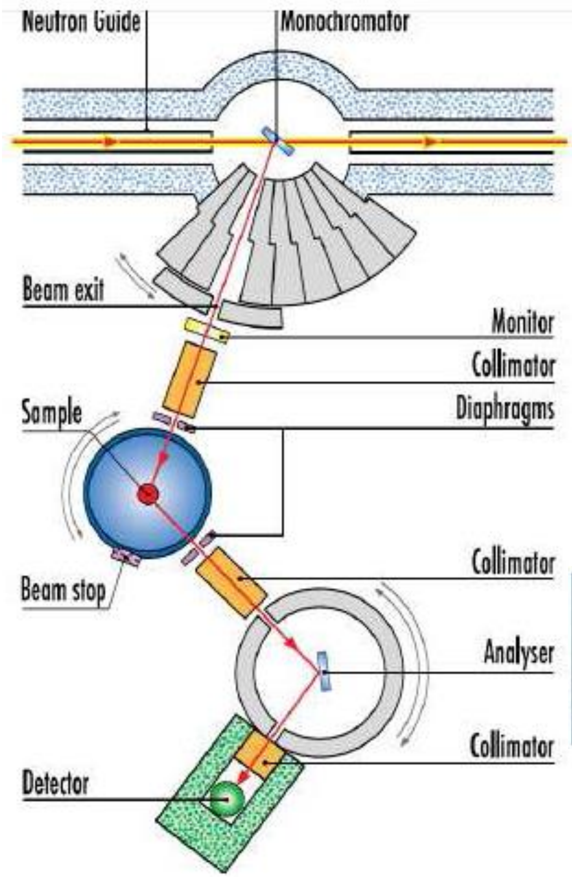
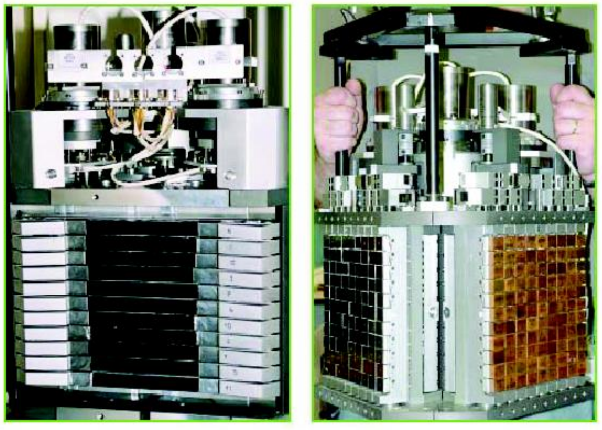


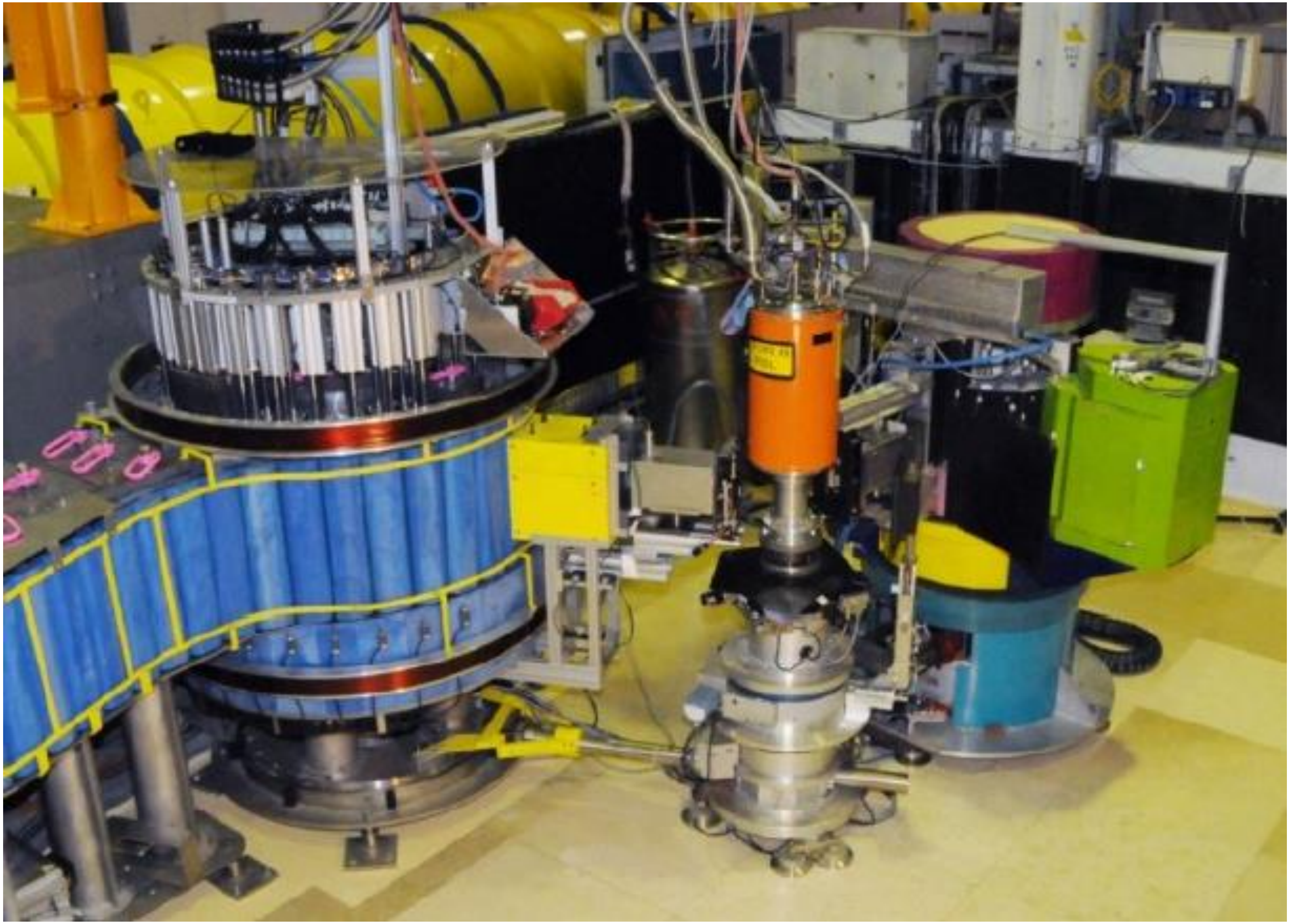
Inelastic neutron scattering

Triple axis spectrometer
(IN8, IN20, IN12, IN22 @ ILL)





Inelastic neutron scattering



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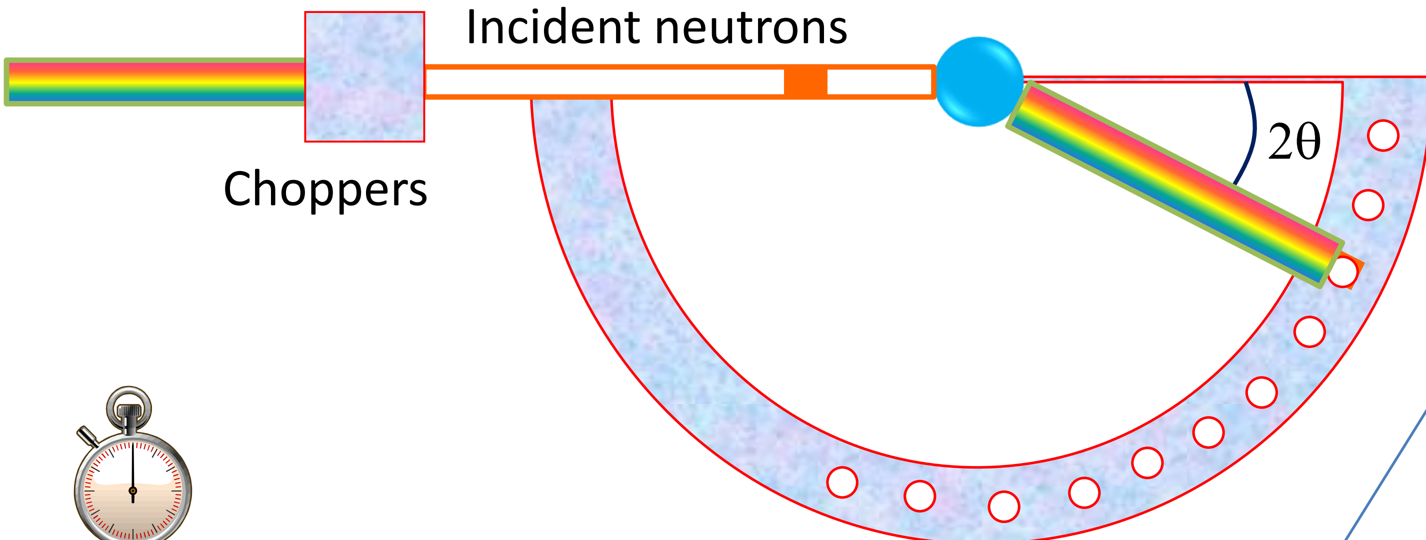
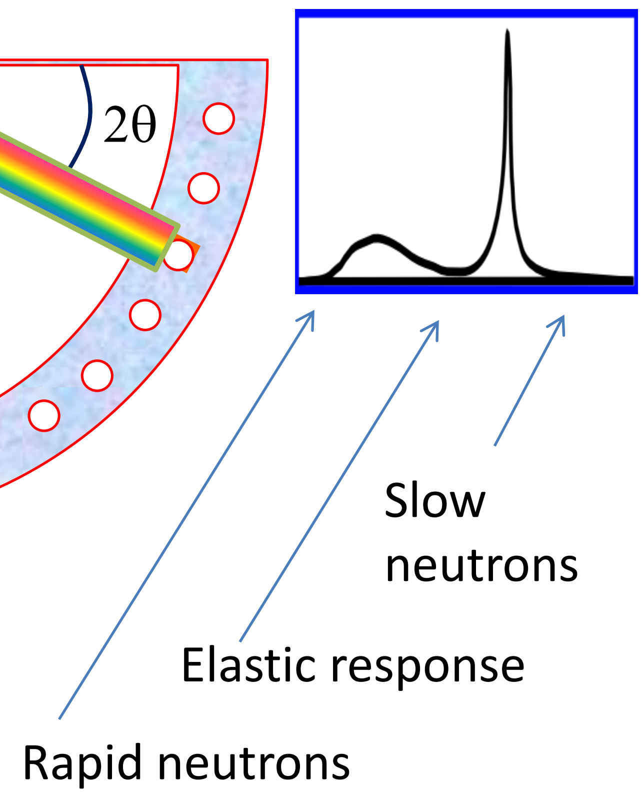


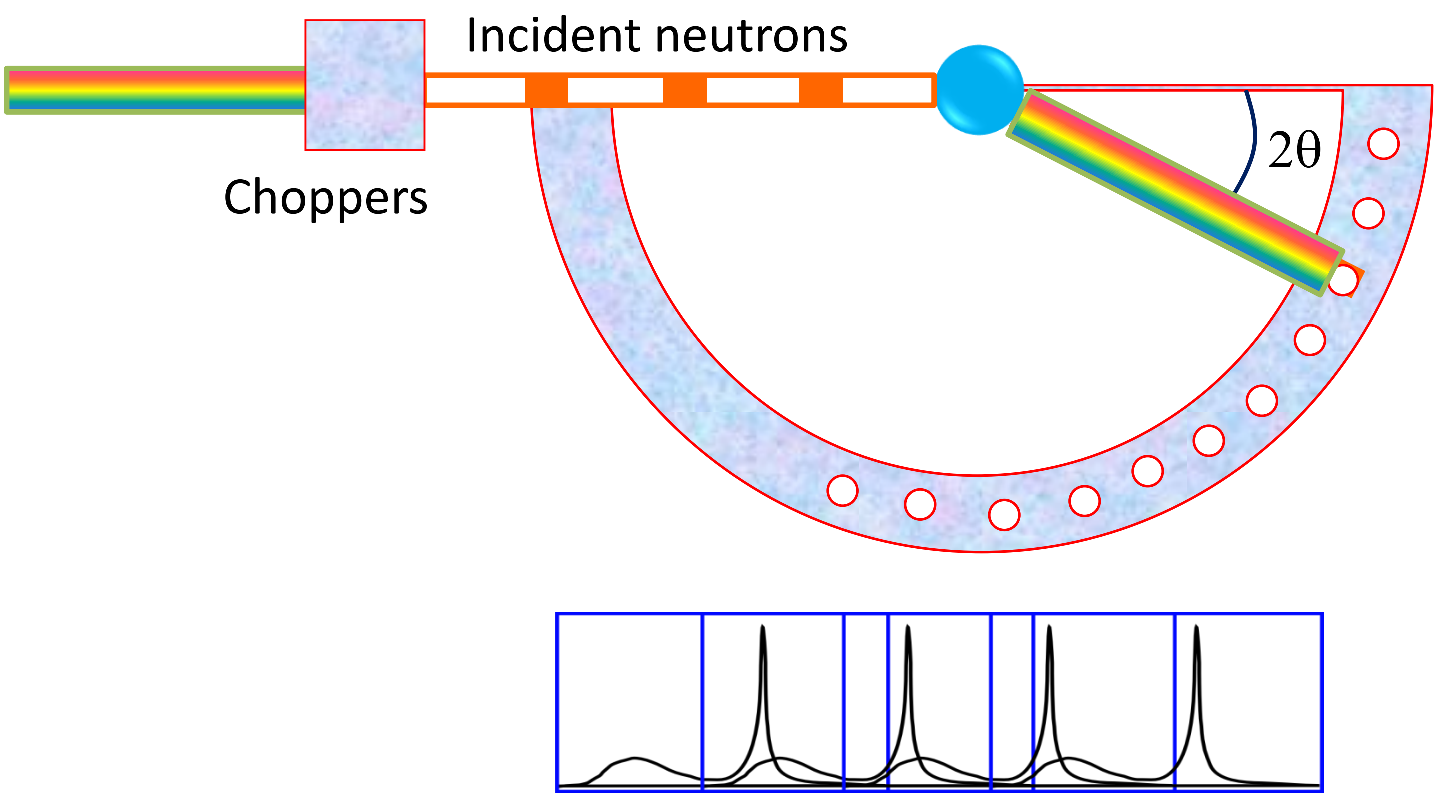


Select the final energy using the Time of flight

$$t = \frac{d}{\sqrt{2E_f / m_n}}$$

Inelastic neutron scattering

The incident beam
must be chopped



Inelastic neutron scattering

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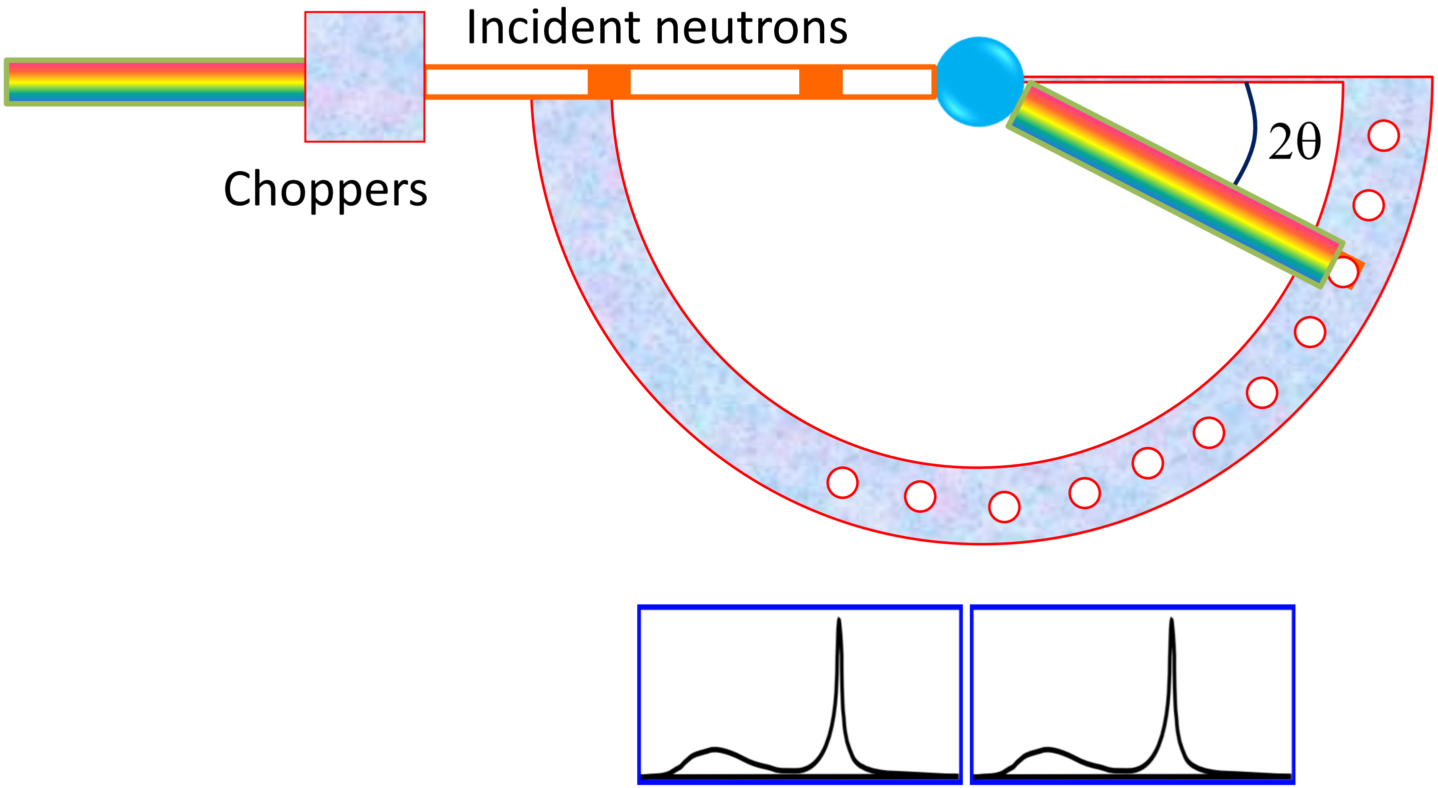
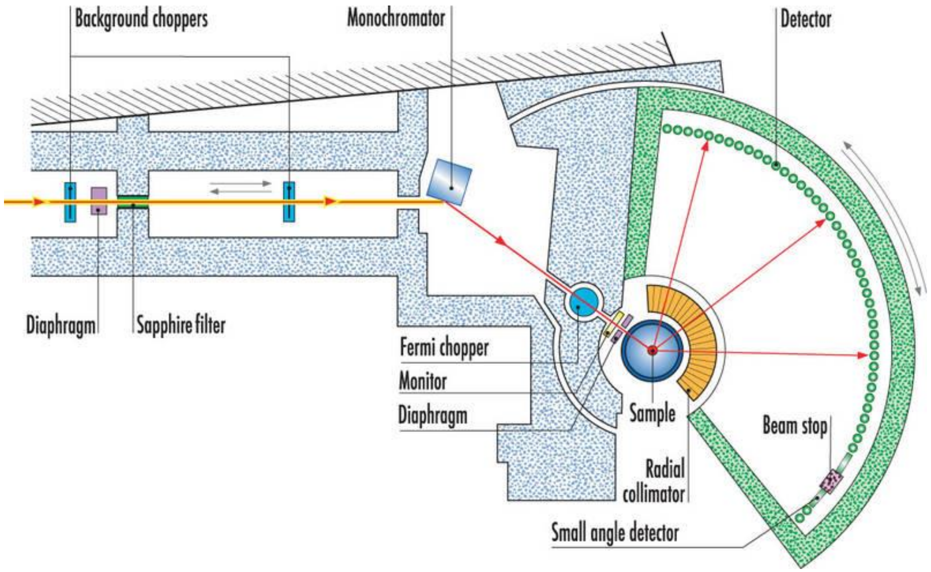
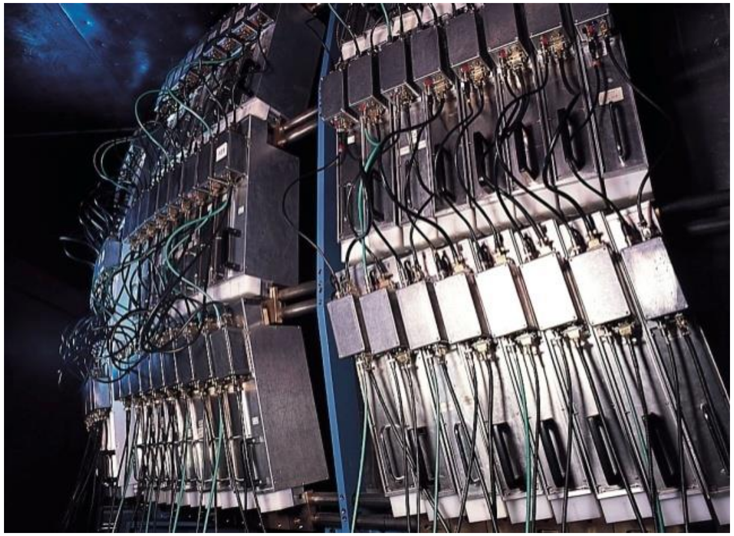


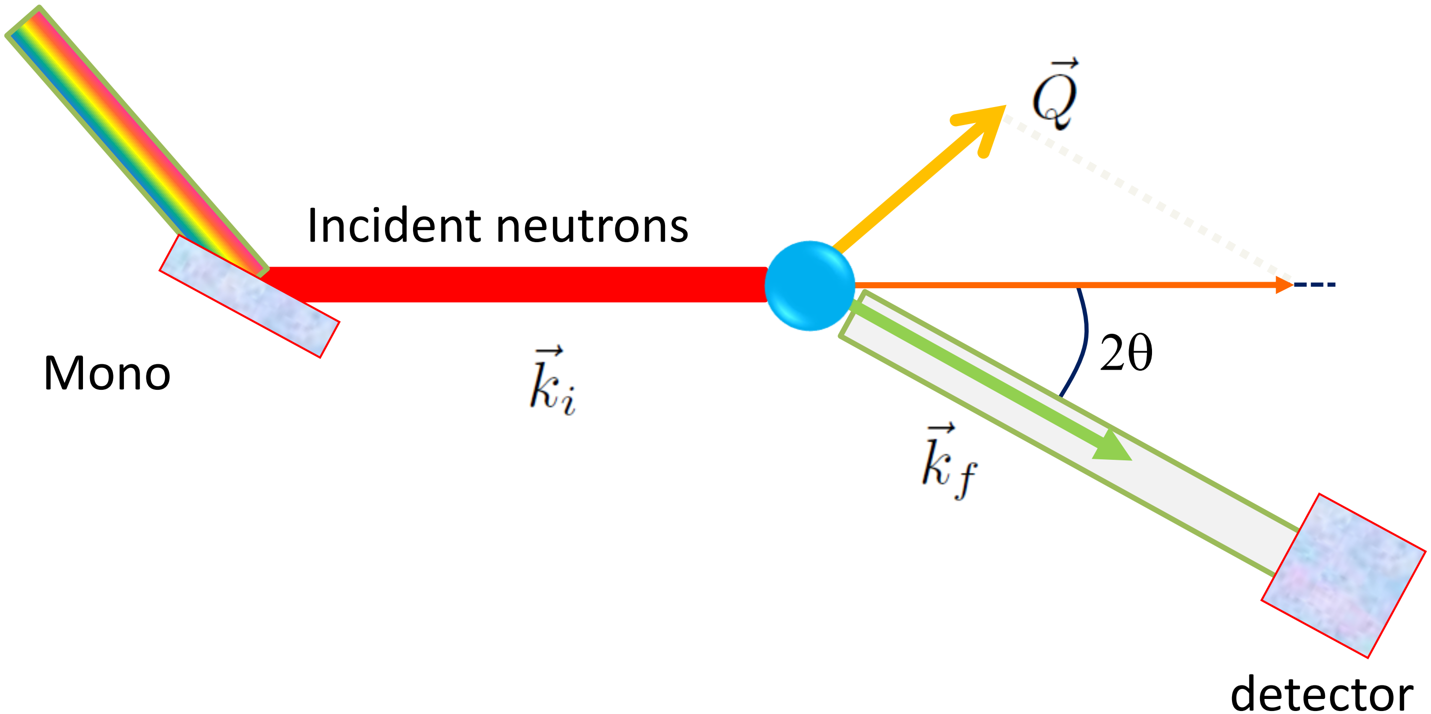
Inelastic neutron scattering

Time of flight spectrometers
IN4, IN5, IN6-Sharp @ ILL
FOCUS @ PSI
...





Comments

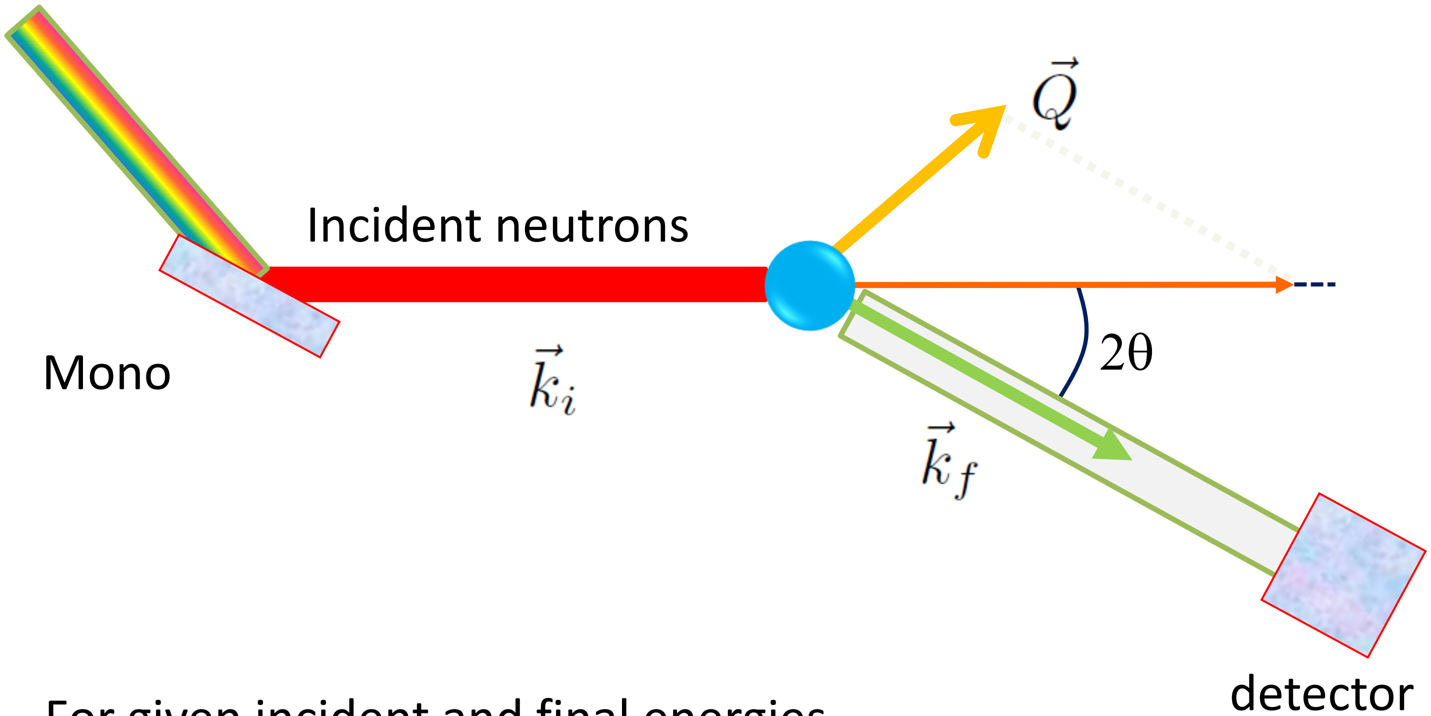


Scattering vector $$\vec{Q} = \vec{k}_i - \vec{k}_f$$

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

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

Comments

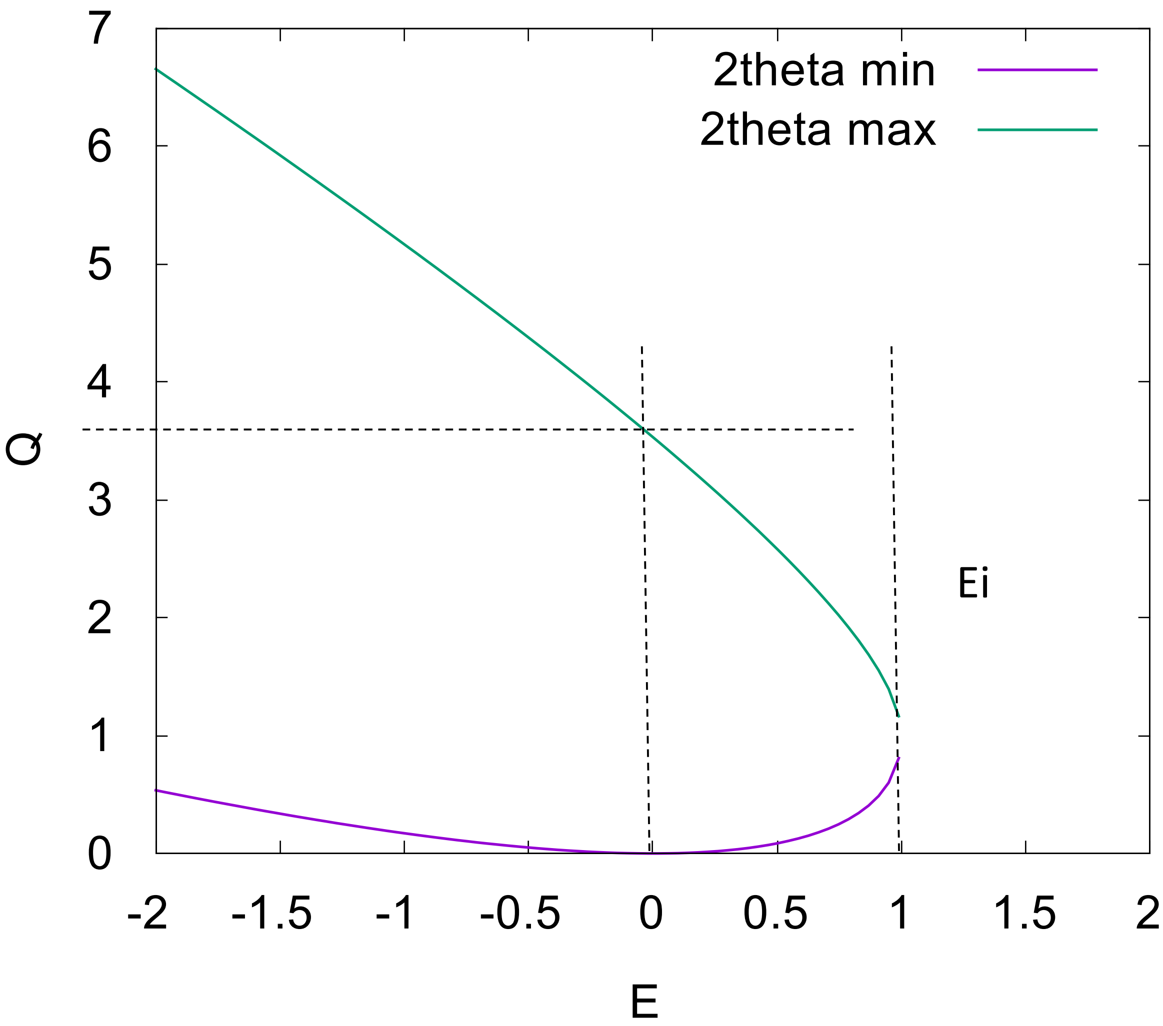


For given incident and final energies,
θ imposes |Q| but NOT the vecteur Q

$$\vec{Q} = \vec{k}_i - \vec{k}_f \qquad \hbar\omega = k_i^2 - k_f^2$$

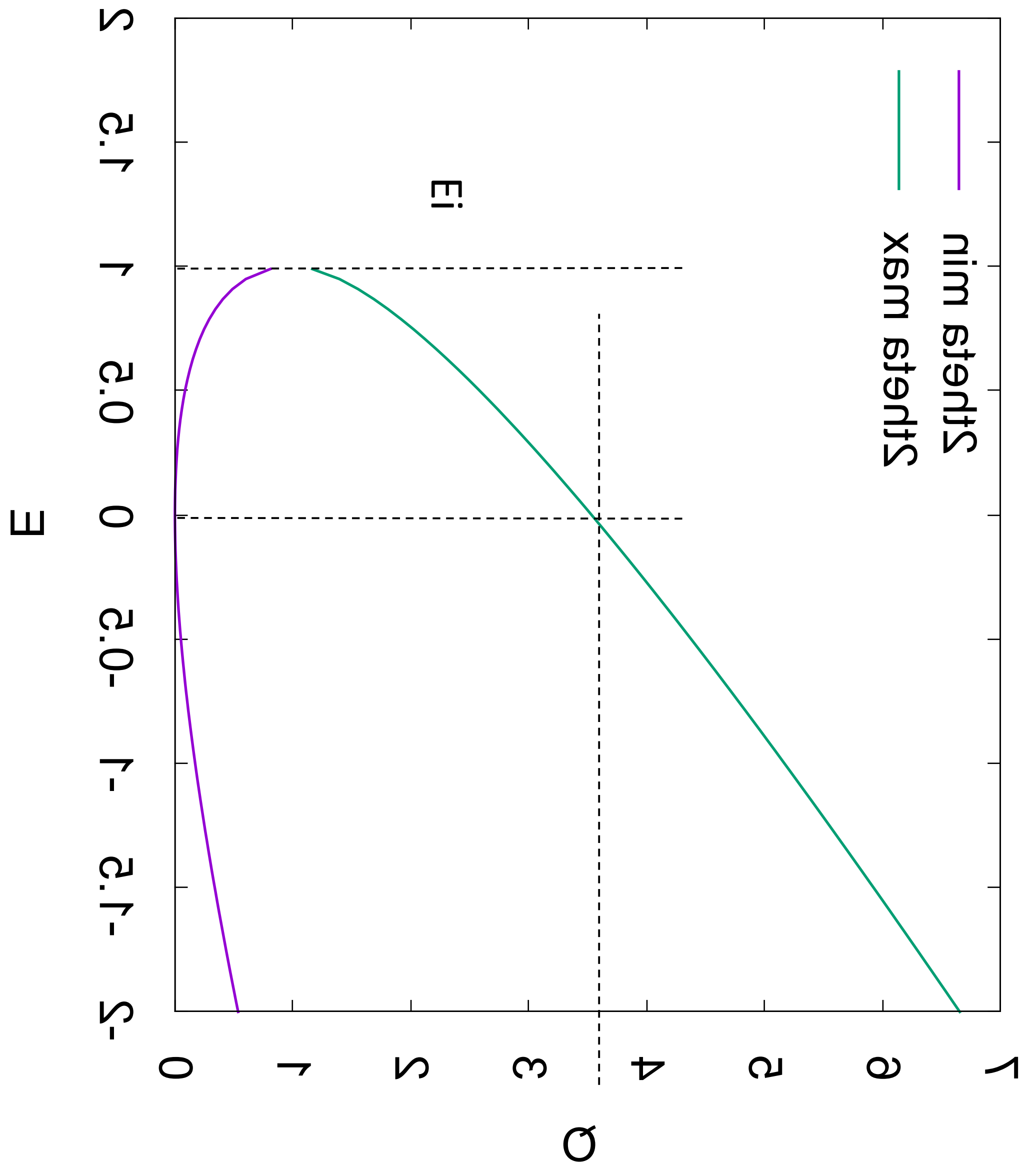
$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta$$

Comments



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

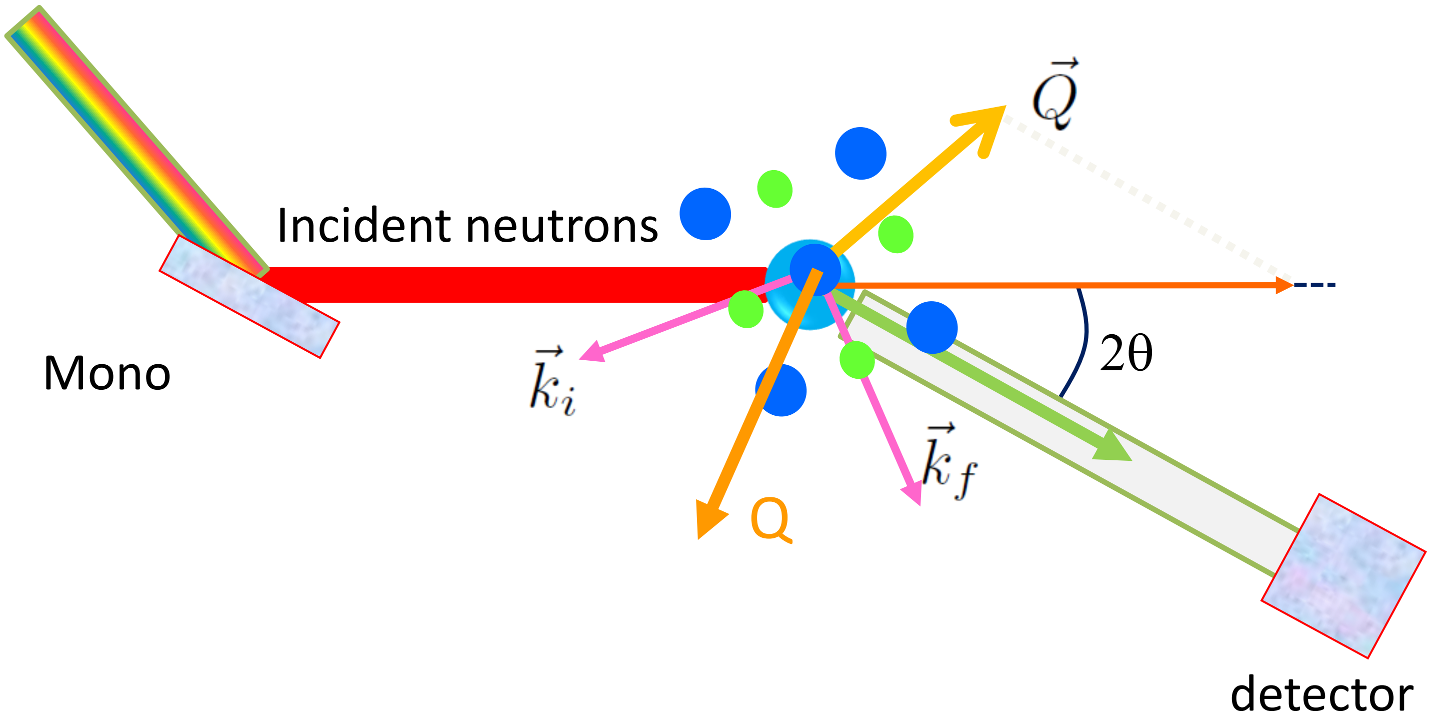
$$
\frac{2\pi}{d} = 2\frac{2\pi}{\lambda}\sin\theta
$$



$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta$$
$$= 2k_i^2 (1 - \cos 2\theta)$$
$$= 4k_i^2 \sin^2 \theta$$
$$Q = 2k_i \sin \theta$$

$$\frac{2\pi}{d} = 2\frac{2\pi}{\lambda} \sin \theta$$

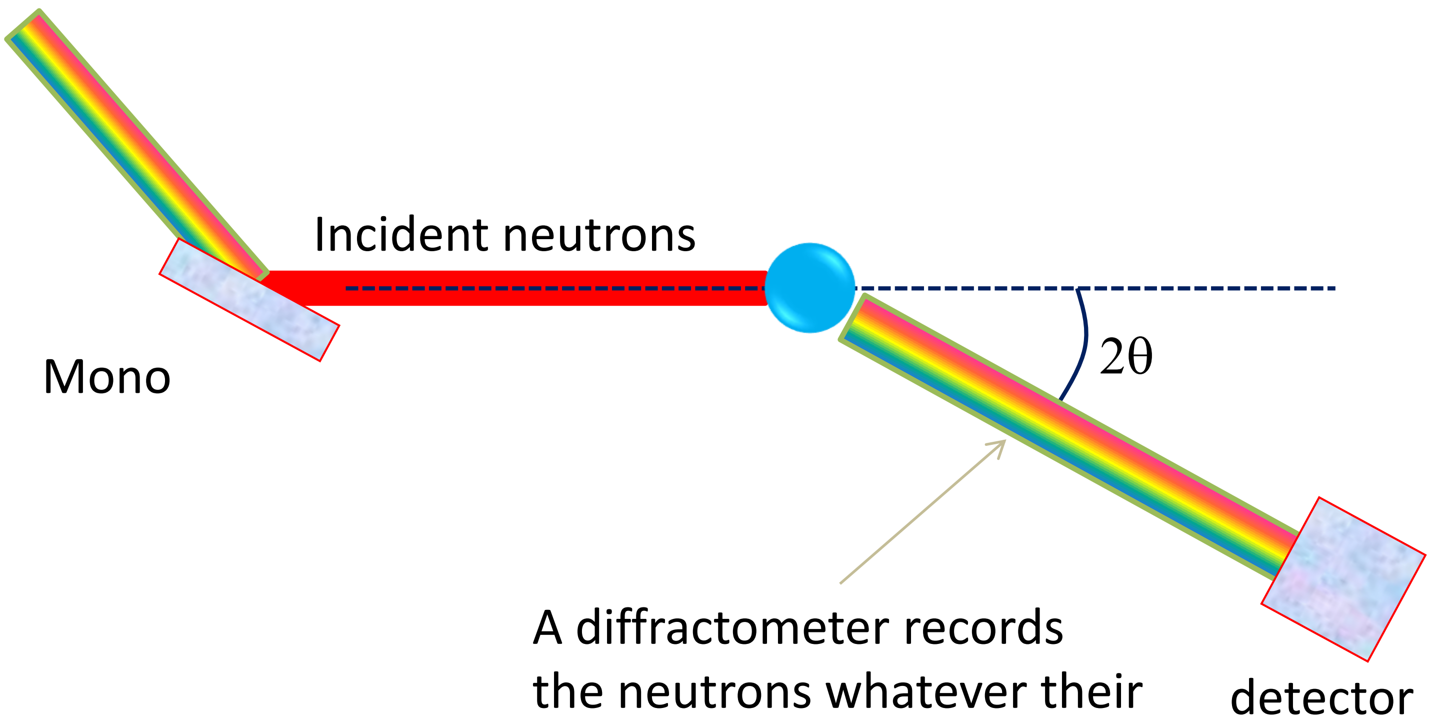
Comments



The scattering wavevector is defined in the lab frame :

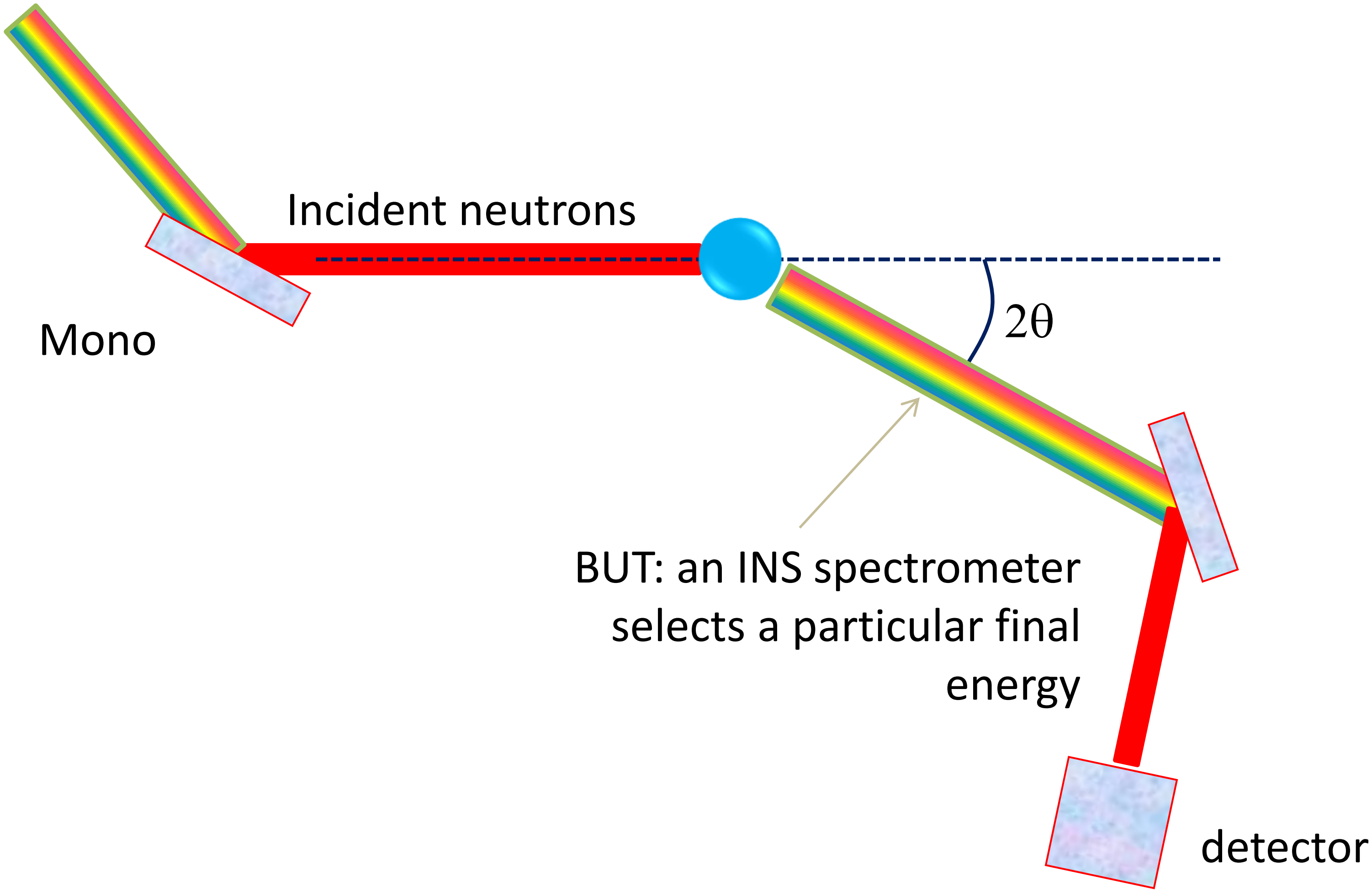
$$\vec{Q} = \vec{k}_i - \vec{k}_f$$

Comments



A diffractometer records the neutrons whatever their final energy

Comments


Incident neutrons
Mono
2θ
BUT: an INS spectrometer selects a particular final energy
detector

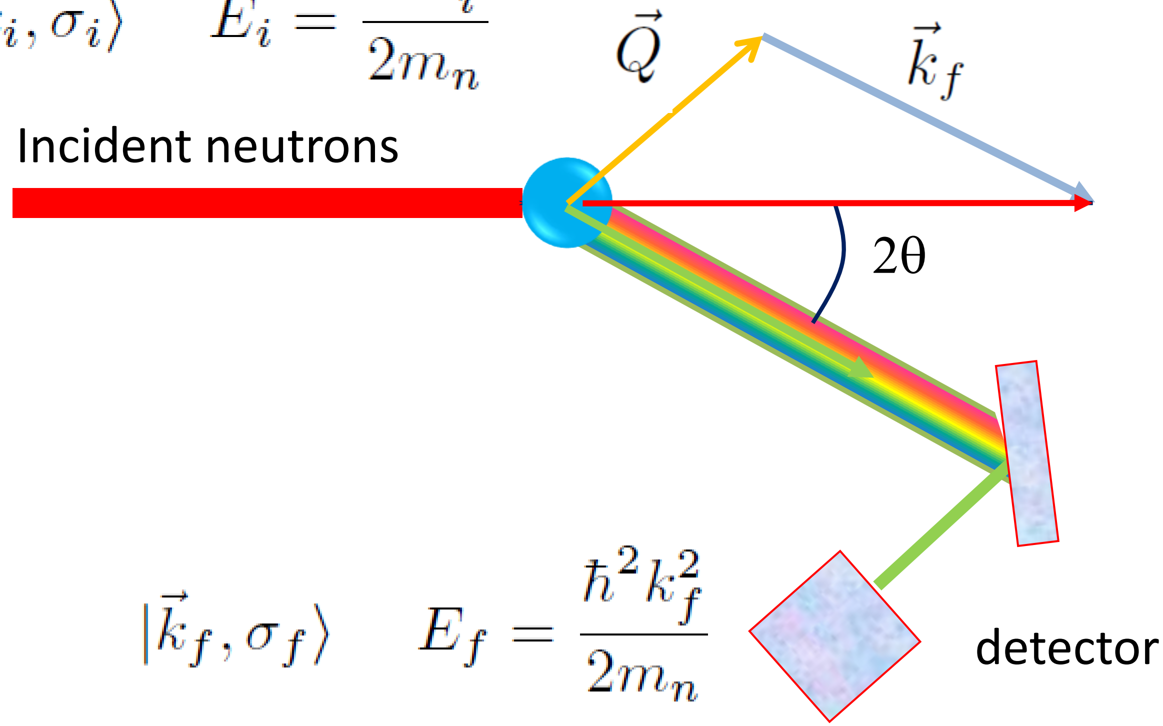
What about the neutron spin ?

Is it possible to manipulate it ?

What about novel information ?

Neutron spin

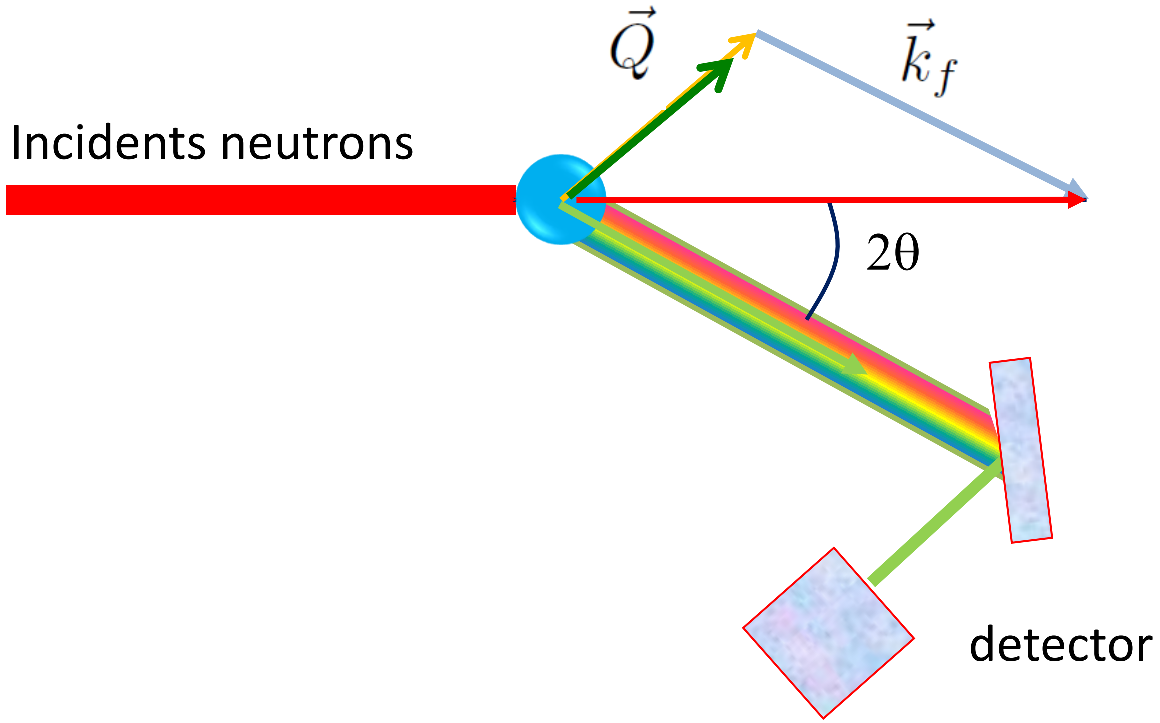
$$|\vec{k}_i, \sigma_i\rangle \qquad E_i = \frac{\hbar^2 k_i^2}{2m_n}$$



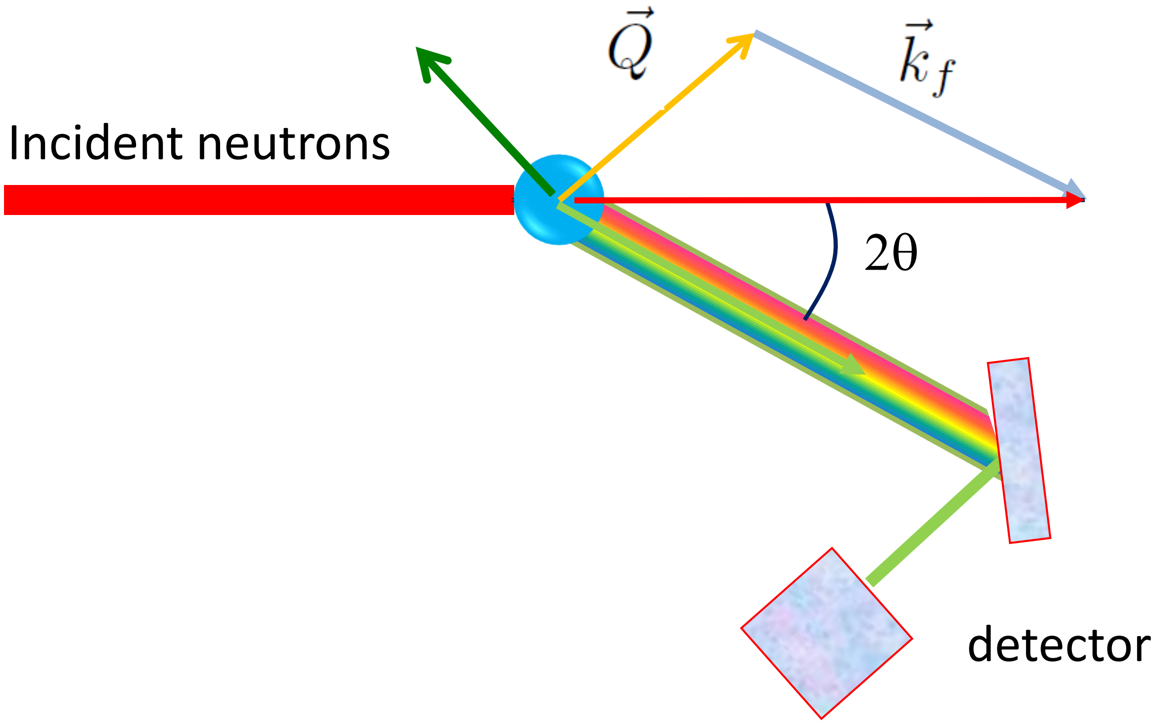
$$|\vec{k}_f, \sigma_f\rangle \qquad E_f = \frac{\hbar^2 k_f^2}{2m_n}$$

$\frac{\partial^2 \sigma}{\partial \Omega \partial E}$ depends on the direction of the neutron spin

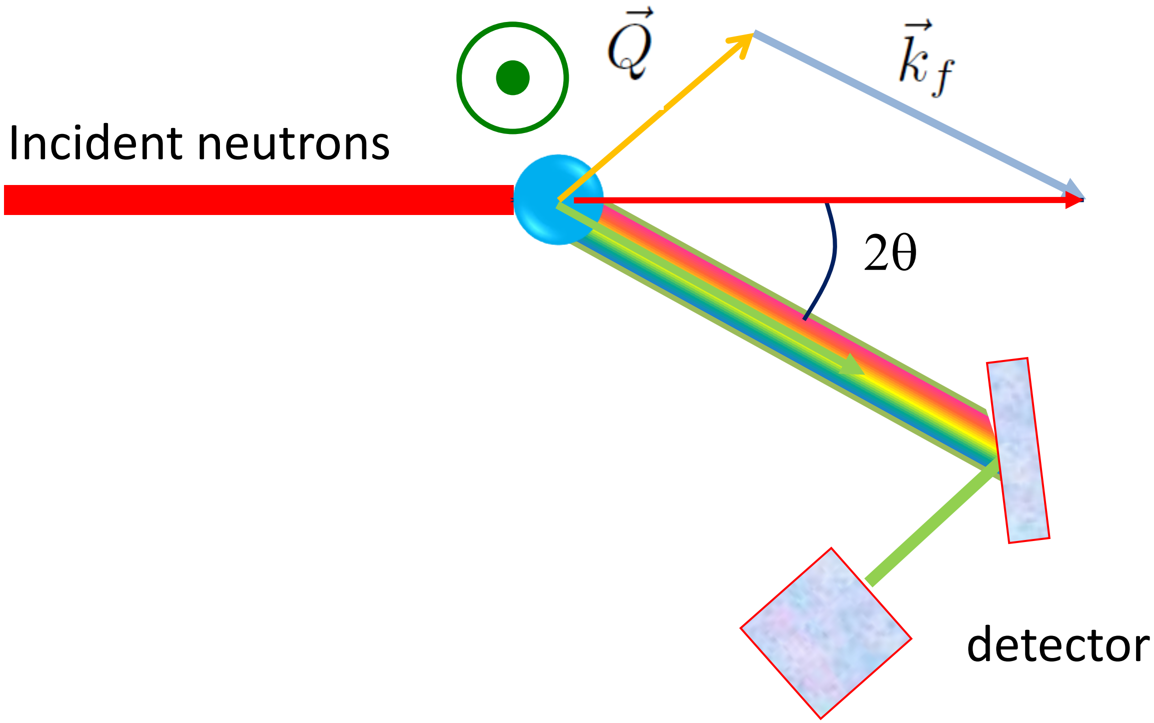
Neutron spin



Neutron spin is parallel to Q (« x »)

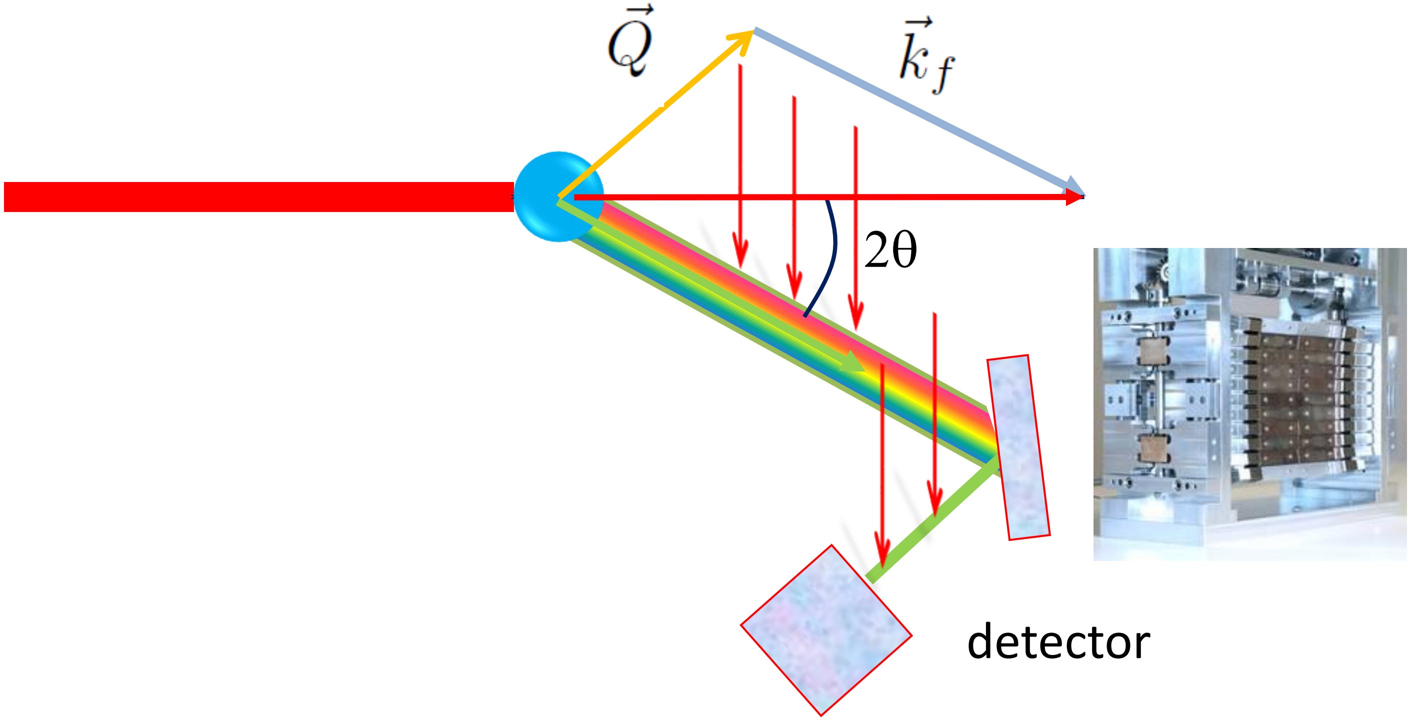


Neutron spin is perpendicular to Q in the scattering plane (« y »)

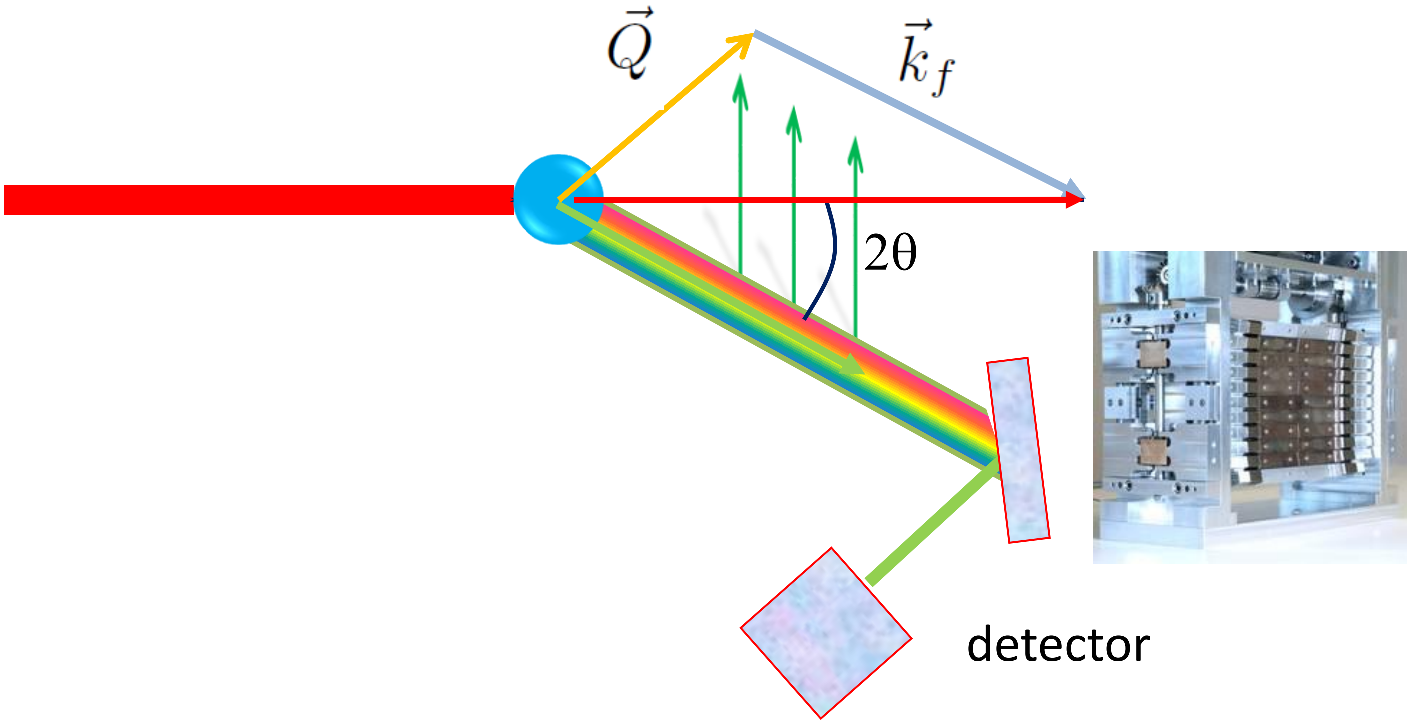


Neutron spin is perpendicular to Q AND to the scattering plane (« z »)

Neutron spin

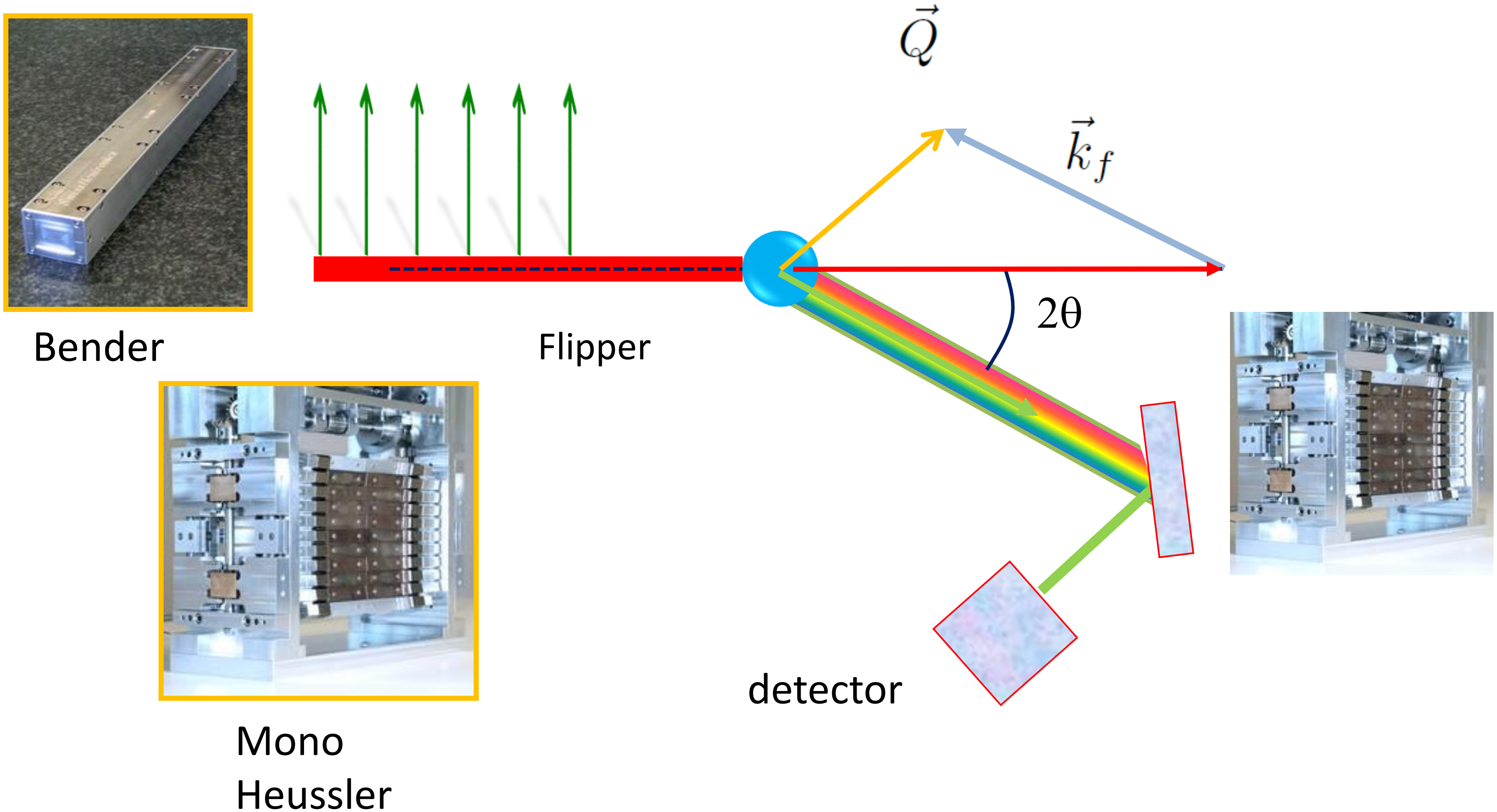


The analyser also seletcs the final spin direction (= polarisation) by projecting onto a given direction (fixed by the set up = a strong magnetic field)

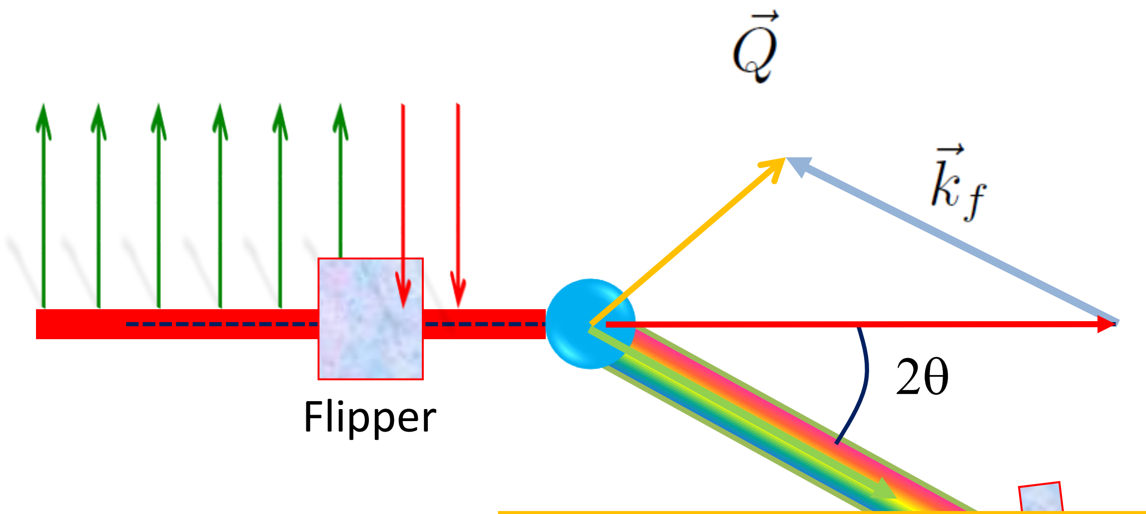


The analyser also seletcs the final spin direction (= polarisation) by projecting onto a given direction (fixed by the set up = a strong magnetic field)

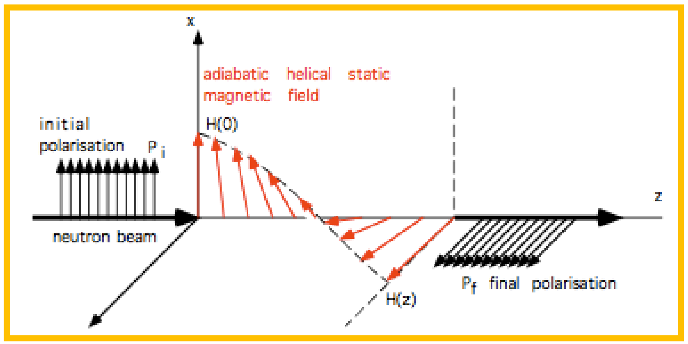
Example



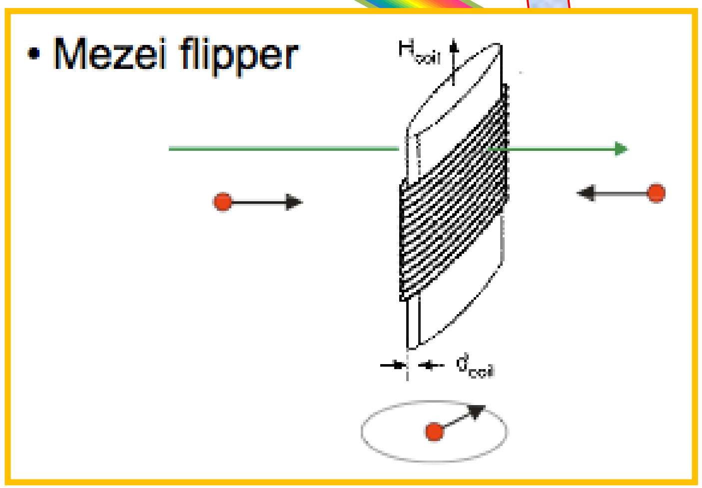
Example



$\vec{Q}$
$\vec{k}_f$
2θ
Flipper

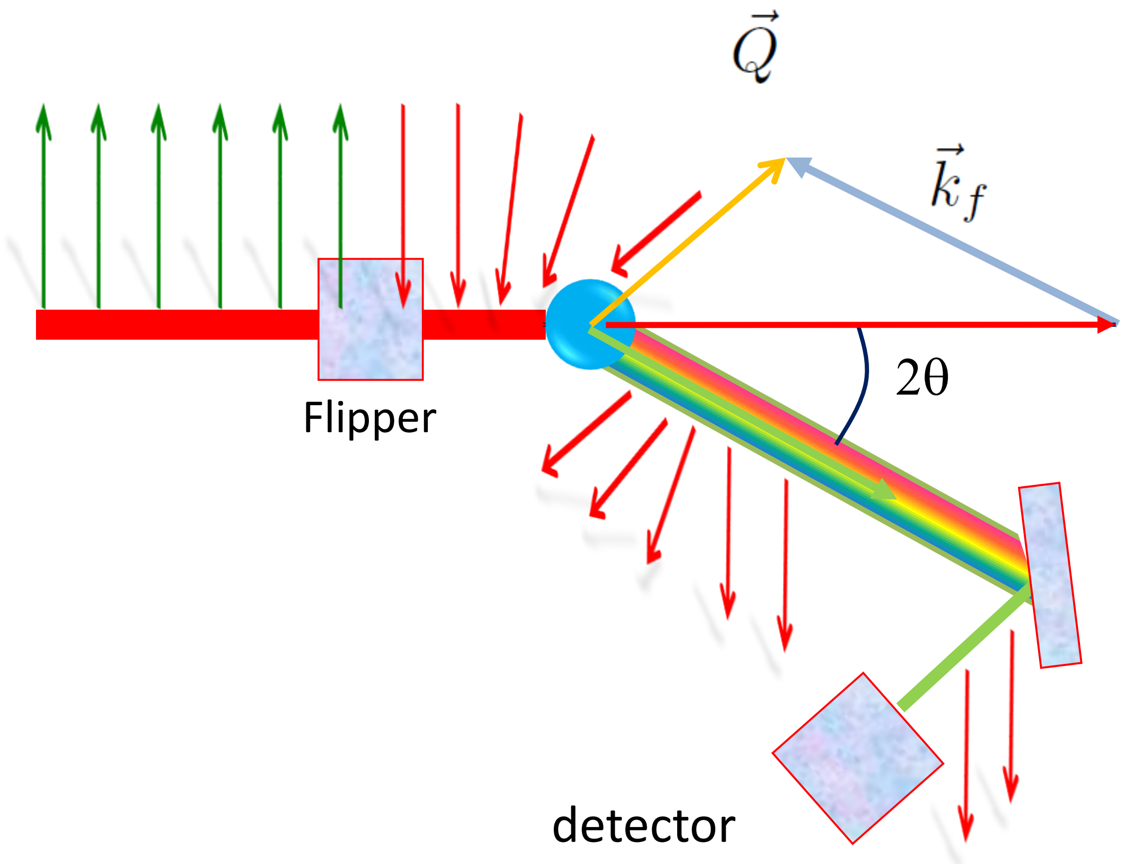


x
adiabatic helical static
magnetic field
initial
polarisation P i
H(0)
z
neutron beam
H(z)
Pf final polarisation



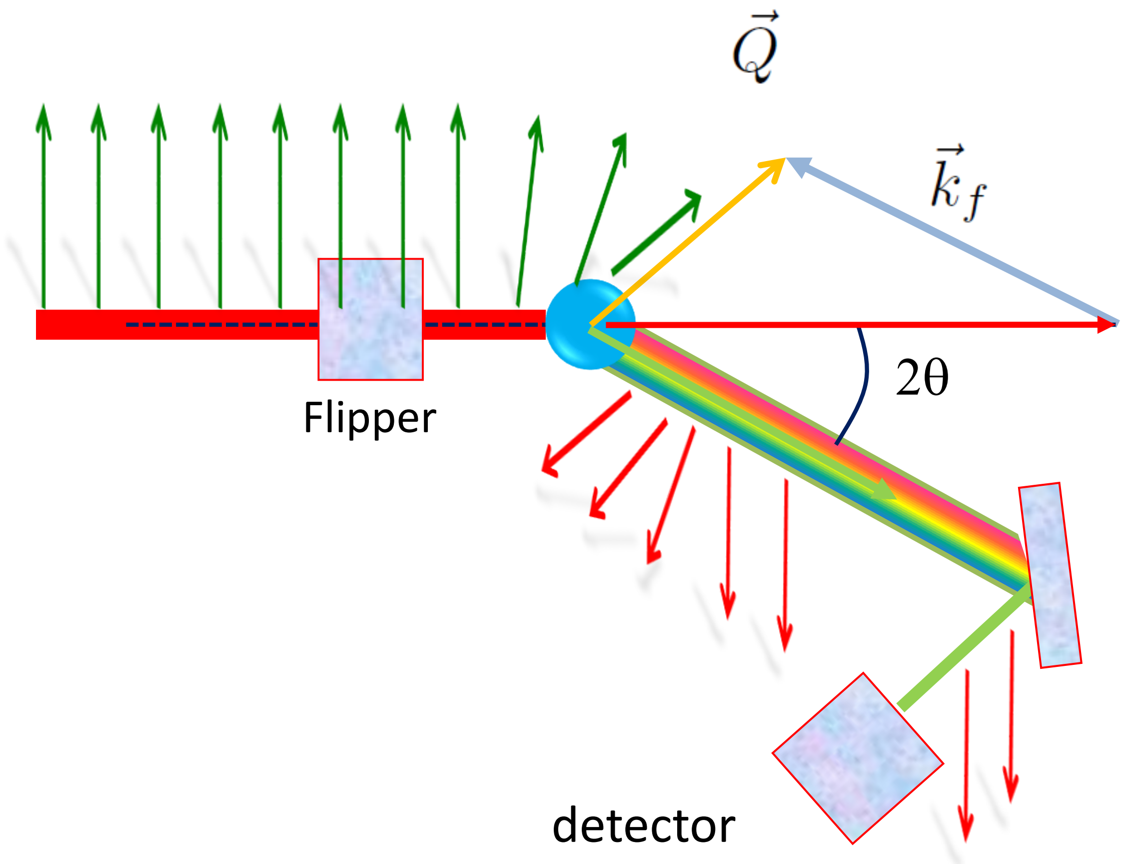
• Mezei flipper
Hcoil
dcoil

Example



Channel I^{--}_x

Example



Channel I^{+-}_{x}

Novel information

$$I_x^{++} \propto |N|^2$$
$$I_x^{--} \propto |N|^2$$
$$I_x^{+-} \propto |M_\perp|^2 - M_{ch}$$
$$I_x^{-+} \propto |M_\perp|^2 + M_{ch}$$
$$I_y^{++} \propto |N|^2 + |M_\perp^y|^2 + R_y$$
$$I_y^{--} \propto |N|^2 + |M_\perp^y|^2 - R_y$$
$$I_y^{+-} \propto |M_\perp^z|^2$$
$$I_y^{-+} \propto |M_\perp^z|^2$$
$$I_z^{++} \propto |N|^2 + |M_\perp^z|^2 + R_z$$
$$I_z^{--} \propto |N|^2 + |M_\perp^z|^2 - R_z$$
$$I_z^{+-} \propto |M_\perp^y|^2$$
$$I_z^{-+} \propto |M_\perp^y|^2$$

Depending on the incident and scattered neutron spin, new correlation functions appear

M^z

Novel information

$$I_x^{++} \propto |N|^2$$
$$I_x^{--} \propto |N|^2$$
$$I_x^{+-} \propto |M_\perp|^2 - M_{ch}$$
$$I_x^{-+} \propto |M_\perp|^2 + M_{ch}$$
$$I_y^{++} \propto |N|^2 + |M_\perp^y|^2 + R_y$$
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$$I_z^{+-} \propto |M_\perp^y|^2$$
$$I_z^{-+} \propto |M_\perp^y|^2$$

Depending on the incident and scattered neutron spin, new correlation functions appear

M^y

Novel information

$$I_x^{++} \propto |N|^2$$
$$I_x^{--} \propto |N|^2$$
$$I_x^{+-} \propto |M_\perp|^2 - M_{ch}$$
$$I_x^{-+} \propto |M_\perp|^2 + M_{ch}$$
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$$I_y^{--} \propto |N|^2 + |M_\perp^y|^2 - R_y$$
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$$I_z^{--} \propto |N|^2 + |M_\perp^z|^2 - R_z$$
$$I_z^{+-} \propto |M_\perp^y|^2$$
$$I_z^{-+} \propto |M_\perp^y|^2$$

Depending on the incident and scattered neutron spin, new correlation functions appear

$M=M^z+M^y$

Novel information

$$I_x^{++} \propto |N|^2$$
$$I_x^{--} \propto |N|^2$$
$$I_x^{+-} \propto |M_\perp|^2 - M_{ch}$$
$$I_x^{-+} \propto |M_\perp|^2 + M_{ch}$$
$$I_y^{++} \propto |N|^2 + |M_\perp^y|^2 + R_y$$
$$I_y^{--} \propto |N|^2 + |M_\perp^y|^2 - R_y$$
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$$I_z^{+-} \propto |M_\perp^y|^2$$
$$I_z^{-+} \propto |M_\perp^y|^2$$

Depending on the incident and scattered neutron spin, new correlation functions appear

« Interference term »

$$M_{ch} = i\,(M_\perp^{z*} M_\perp^y - M_\perp^{y*} M_\perp^z)$$
$$R_y = N\ M_\perp^{y*} + N^*\ M_\perp^y$$
$$R_z = N\ M_\perp^{z*} + N^*\ M_\perp^z$$

Flipping ratio technique

$$I_x^{++} \propto |N|^2$$
$$I_x^{--} \propto |N|^2$$
$$I_x^{+-} \propto |M_\perp|^2 - M_{ch}$$
$$I_x^{-+} \propto |M_\perp|^2 + M_{ch}$$
$$I_y^{++} \propto |N|^2 + |M_\perp^y|^2 + R_y$$
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$$I_z^{++} \propto |N|^2 + |M_\perp^z|^2 + R_z$$
$$I_z^{--} \propto |N|^2 + |M_\perp^z|^2 - R_z$$
$$I_z^{+-} \propto |M_\perp^y|^2$$
$$I_z^{-+} \propto |M_\perp^y|^2$$

$$R = \frac{|N|^2 + |\boldsymbol{M}_\perp|^2 + P(NM_\perp^{Z*} + N^* M_\perp^Z)}{|N|^2 + |\boldsymbol{M}_\perp|^2 - P(NM_\perp^{Z*} + N^* M_\perp^Z)}$$

The incident beam is up or down. The strong H field imposes the « z » channel.
All neutrons are recorded whatever the final spin direction.

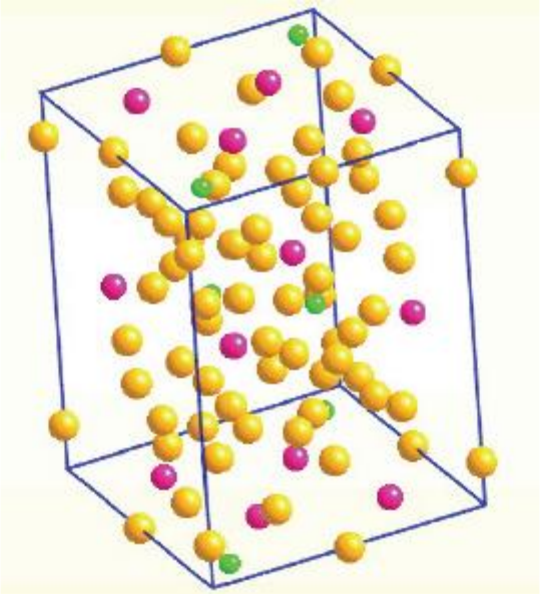
- ILL : D3, D23
- LLB : 5C1, 6T2

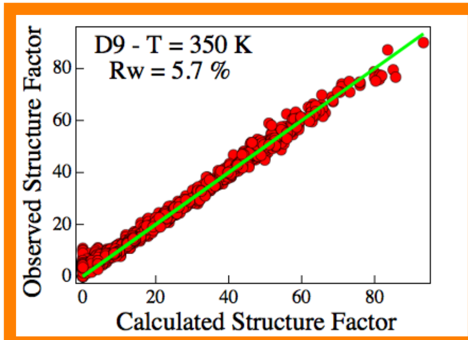
Flipping ratio technique

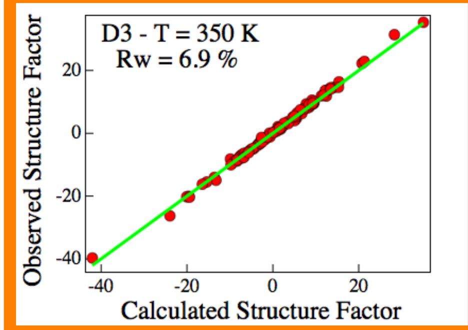
$$R = \frac{|N|^2 + |\boldsymbol{M}_\perp|^2 + P(NM_\perp^{Z*} + N^* M_\perp^Z)}{|N|^2 + |\boldsymbol{M}_\perp|^2 - P(NM_\perp^{Z*} + N^* M_\perp^Z)}$$

Improves precision since R depends linearly on Mz

$Nd_2Fe_{14}B$

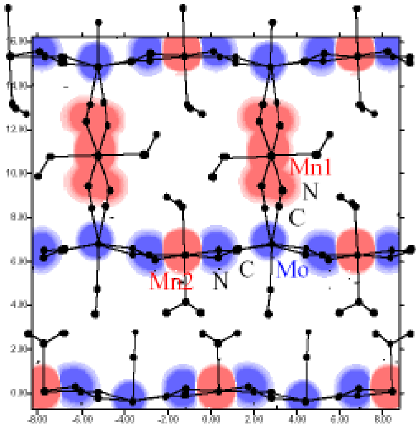






T(K)	350 K	400 K	450 K
Nd1	1.62(3)	1.37(3)	1.13(2)
Nd2	1.65(3)	1.50(3)	1.24(2)
Fe1	1.95(5)	1.80(5)	1.63(4)
Fe2	2.16(5)	2.23(5)	1.99(4)
Fe3	2.12(4)	2.02(4)	1.78(3)
Fe4	3.01(4)	2.99(4)	2.79(4)
Fe5	2.29(2)	2.18(3)	2.04(2)
Fe6	2.31(3)	2.19(4)	2.03(3)

Magnetization density



Mn(CN)Mo (B. Gillon et al, Inorg. Chem. **46**, 1090 (2007).

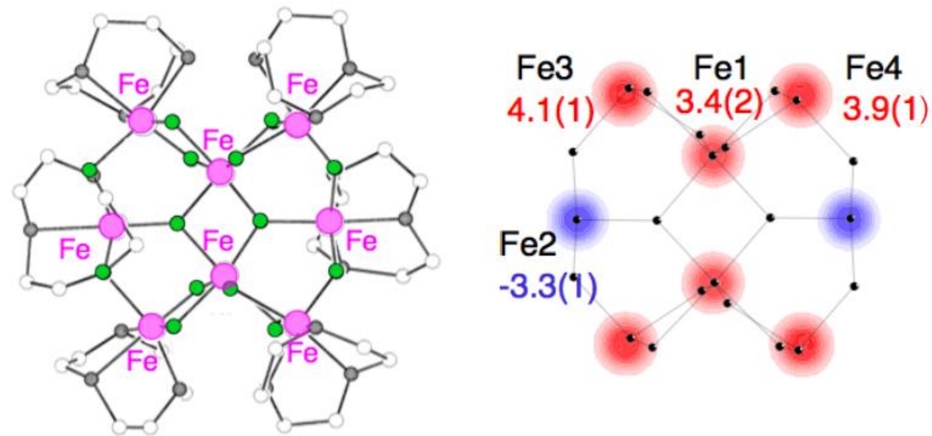


Fig. 3. View of the Fe_8 molecular cluster (left) and experimental magnetization distribution (right) as measured by polarized neutron diffraction. From Ref. [23].

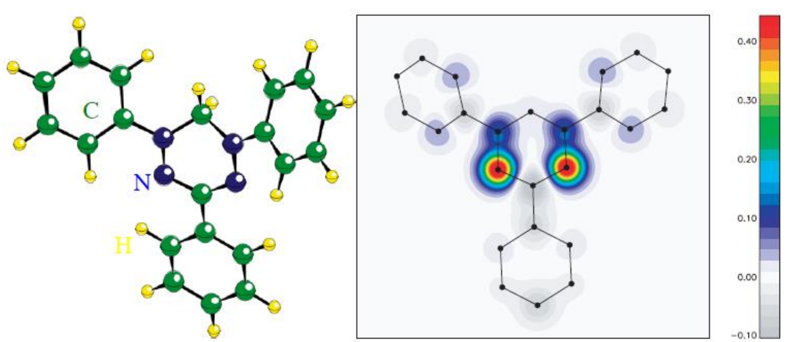


Fig. 4. View of the TPV molecule (left) and experimental magnetization distribution (right) as measured by polarized neutron diffraction.

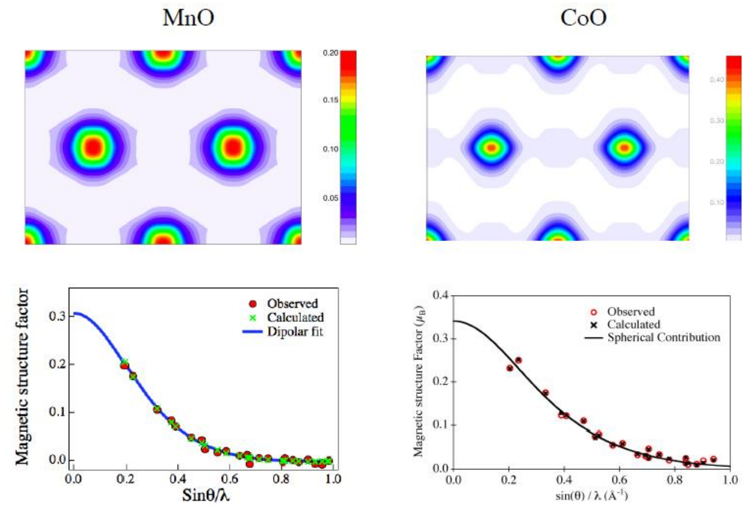


Fig. 5. Magnetization distribution in direct space (up) and form factor in reciprocal space (bottom) for MnO (left) and CoO (right). From Refs. [19] and [30].

Local susceptibility

Shape of the local anisotropy tensor

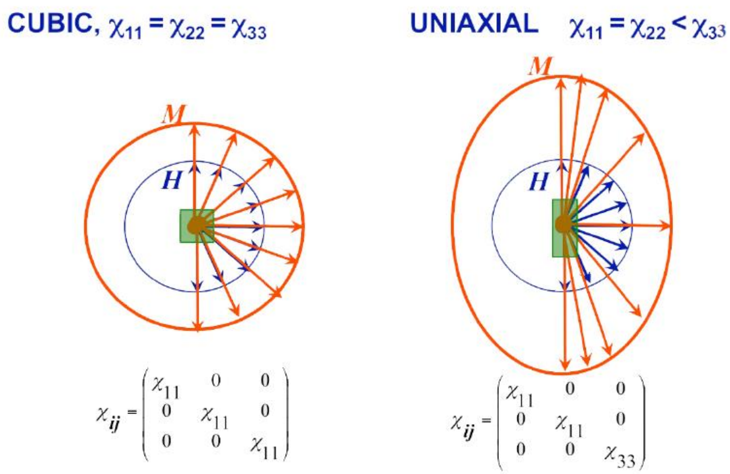
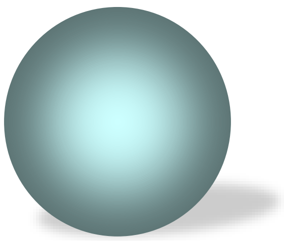
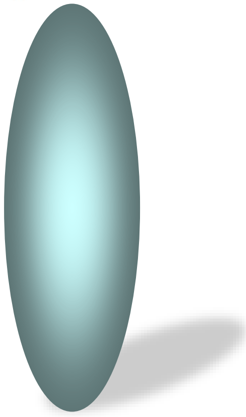
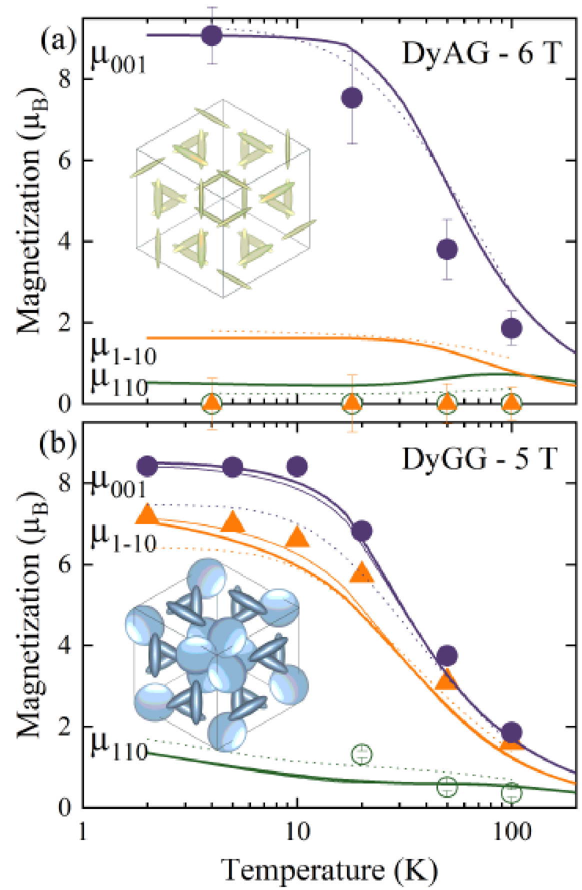


Fig. 6. Atomic site susceptibility tensor.







Kibalin et al, PRR (2020)

Chirality

$$I_x^{++} \propto |N|^2$$

$$I_x^{--} \propto |N|^2$$

$$I_x^{+-} \propto |M_\perp|^2 - M_{ch}$$

$$I_x^{-+} \propto |M_\perp|^2 + M_{ch}$$

$$I_y^{++} \propto |N|^2 + |M_\perp^y|^2 + R_y$$

$$I_y^{--} \propto |N|^2 + |M_\perp^y|^2 - R_y$$

$$I_y^{+-} \propto |M_\perp^z|^2$$

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$$I_z^{++} \propto |N|^2 + |M_\perp^z|^2 + R_z$$

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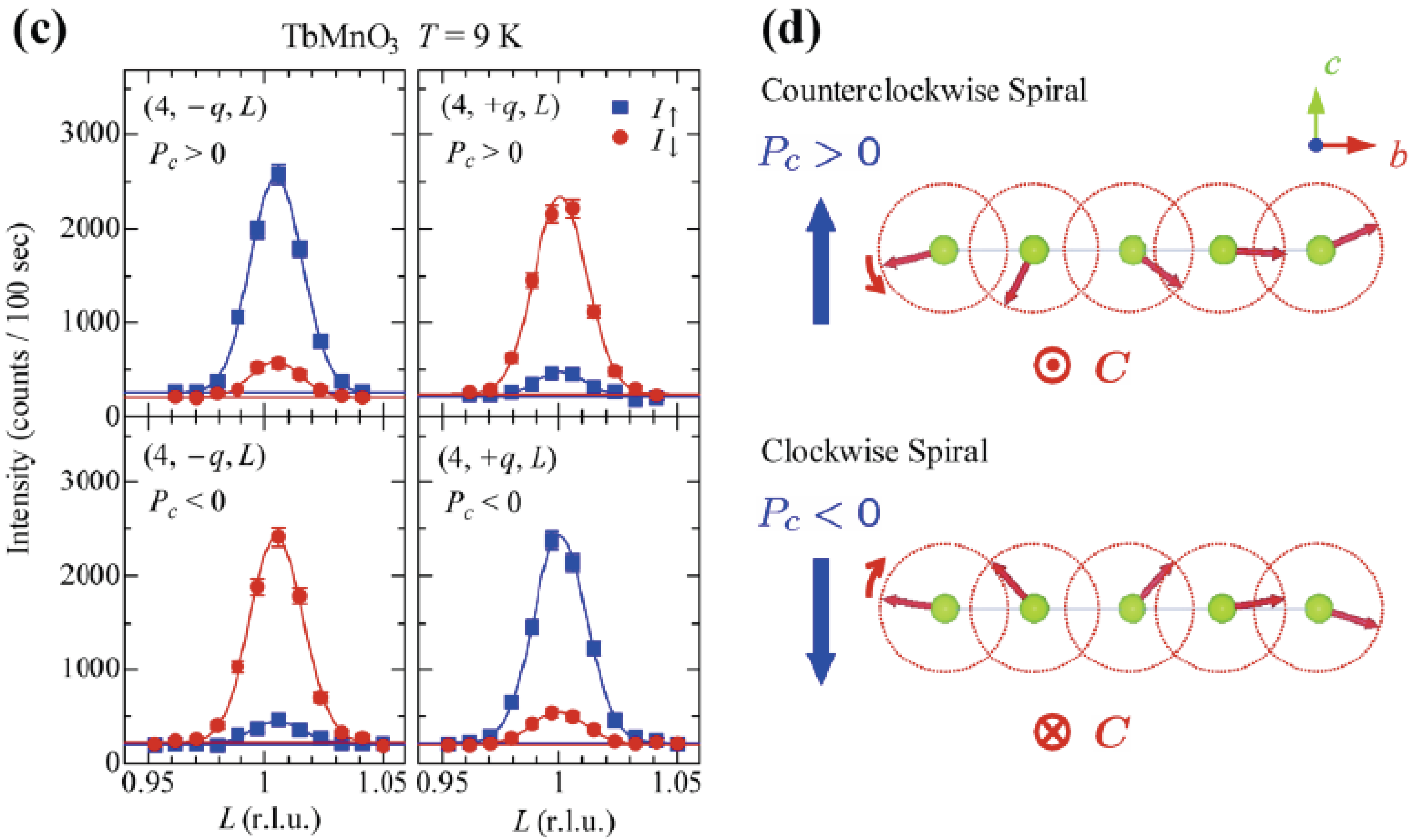
$$I_z^{+-} \propto |M_\perp^y|^2$$

$$I_z^{-+} \propto |M_\perp^y|^2$$

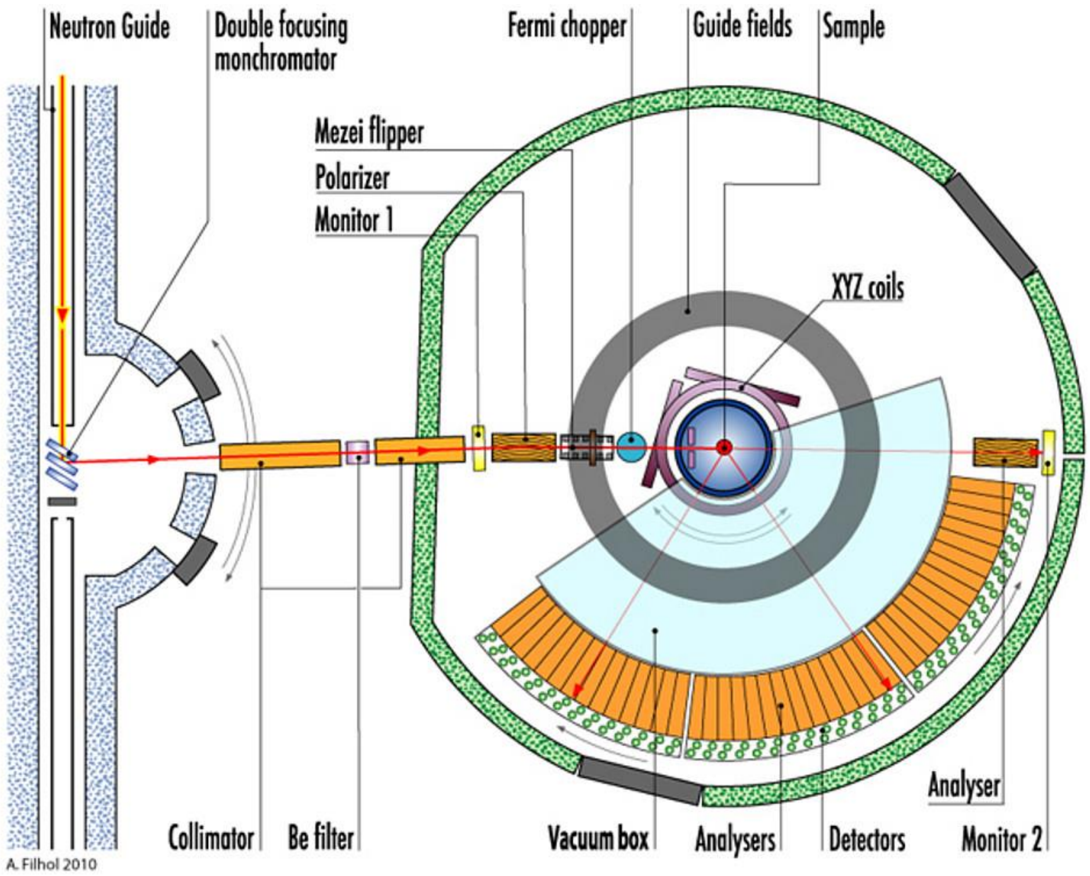
Chiral terms

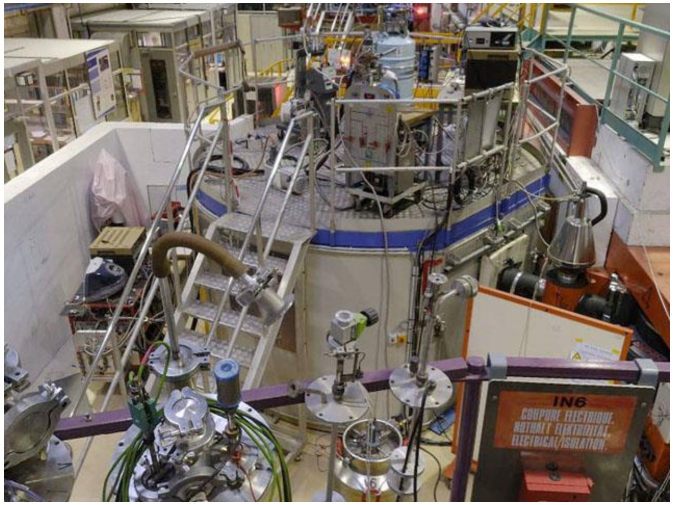
Chirality



(c)
TbMnO3 T = 9 K
(4, −q, L)
Pc > 0
(4, +q, L)
Pc > 0
I↑
I↓
(4, −q, L)
Pc < 0
(4, +q, L)
Pc < 0
Intensity (counts / 100 sec)
3000
2000
1000
0
0.95
1
1.05
L (r.l.u.)
(d)
Counterclockwise Spiral
c
b
Pc > 0
C
Clockwise Spiral
Pc < 0
C

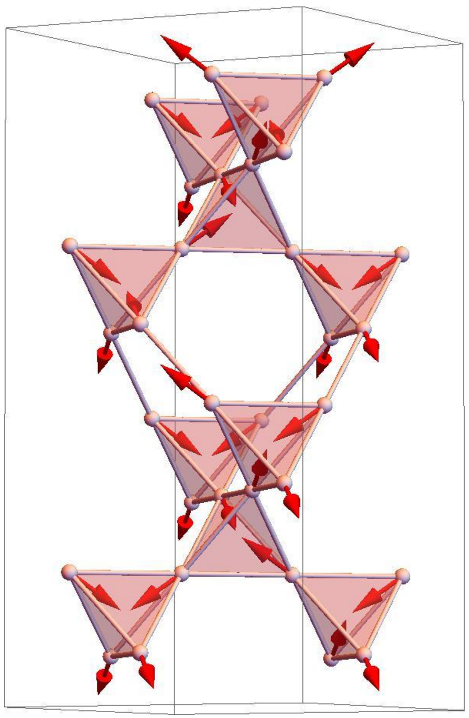
Magnetic diffuse scattering



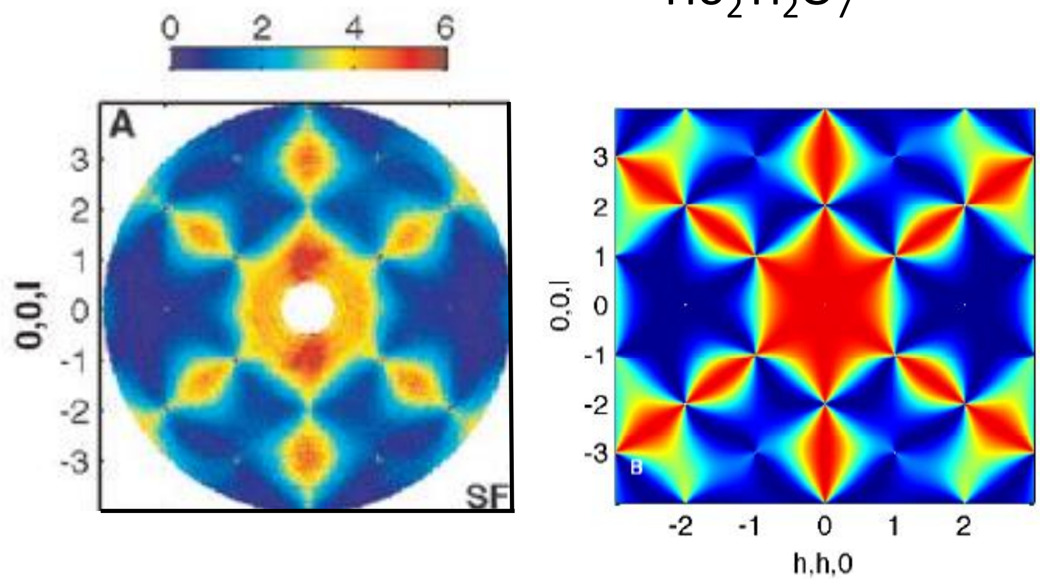


D7@ILL

Magnetic diffuse scattering



$Ho_2Ti_2O_7$



Tom Fennell, Science (2009)

Cross sections,
scattered intensities

Cross sections and scattered intensities

Fermi Golden rule

$$W = \frac{2\pi}{\hbar^2} |\langle k_f, \sigma_f, \lambda' | V | k_i, \sigma_i, \lambda \rangle|^2 \; \delta((E_i + E_\lambda) - (E_f + E_{\lambda'}))$$

Cross sections and scattered intensities

Probability for spin states σ_i

Density of final states with k_f

$$D = \frac{V}{(2\pi)^3}\,\partial\vec{k}_f = \frac{V}{(2\pi)^3}\,\partial\Omega\; k_f^2\;\partial k_f = \frac{V}{(2\pi)^3}\,k_f\,\frac{m_n}{\hbar^2}\,\partial\Omega\;\partial E_f$$

$$\partial\sigma = \frac{1}{F_i}\sum_{\sigma_i,\sigma_f} p_{\sigma_i}\sum_{\lambda,\lambda'} p_\lambda\; W\; D$$

Incident flux

$$F_i = \frac{1}{V}\frac{\hbar k_i}{m_n}$$

Probability for state λ

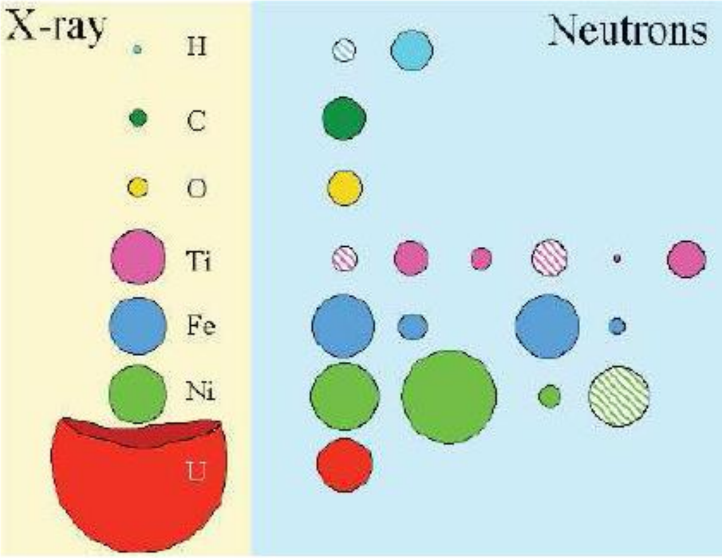
$$p_\lambda = \frac{e^{-E_\lambda/T}}{\sum_\lambda e^{-E_\lambda/T}}$$

Cross sections and scattered intensities

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 sections and scattered intensities

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

Frozen lattice

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

Nothing moves !

Cell

Atom position within the unit cell

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

« simple » definition of the structure factor :

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

Cross sections and scattered intensities

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

Cross sections and scattered intensities

Scattering lengths depend on the isotope

$$\begin{aligned} b_i &= \langle b \rangle + \Delta b_i \\ b_i b_j &= \langle b \rangle^2 + \langle b \rangle (\Delta b_i + \Delta b_j) + \Delta b_i \Delta b_j \end{aligned}$$

$$\begin{aligned} \langle \Delta b_i \rangle &\equiv 0 \\ \langle \Delta b_i \Delta b_j \rangle &= \langle \Delta b_i \Delta b_j \rangle \delta_{i,j} = \langle b^2 \rangle - \langle b \rangle^2 \end{aligned}$$

$$|F(Q)|^2 = \sum_{\ell,\ell'} \langle b \rangle^2 e^{iQ(r_\ell - r_{\ell'})} + N(\langle b^2 \rangle - \langle b \rangle^2)$$

Coherent scattering

Incoherent scattering

Cross sections and scattered intensities

Values of σ_{coh} and σ_{inc}

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

- Difference between H and D used in experiments with soft matter (contrast variation)
- Al used for windows
- V used for sample containers in diffraction experiments and as calibration for energy resolution

Cross sections and scattered intensities

Vibrating lattice

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

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

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

Series expansion

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

Elastic term

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

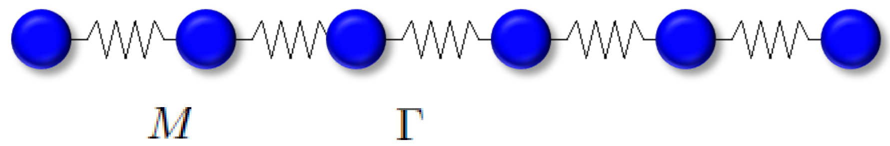
Inelastic term (phonons)

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

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

Collective excitations

Monoatomic chain



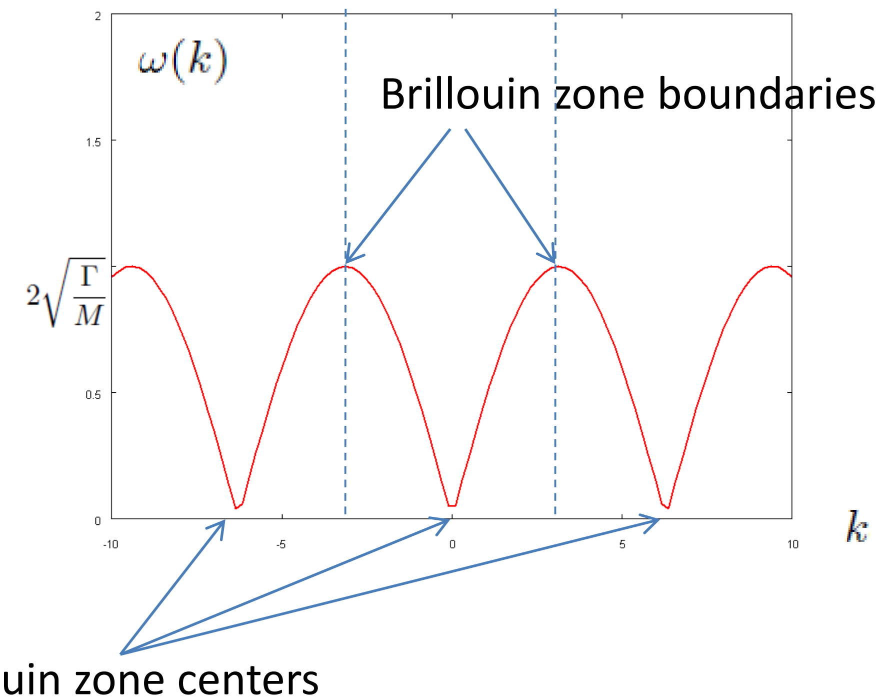
$$M\frac{d^2}{dt^2}u_m \quad = \quad \Gamma\ \left(u_{m+1} + u_{m-1} - 2u_m\right)$$

Fourier transform

$$-M\omega^2 \quad = \quad \Gamma\ \left(e^{ika} + e^{-ika} - 2\right)$$

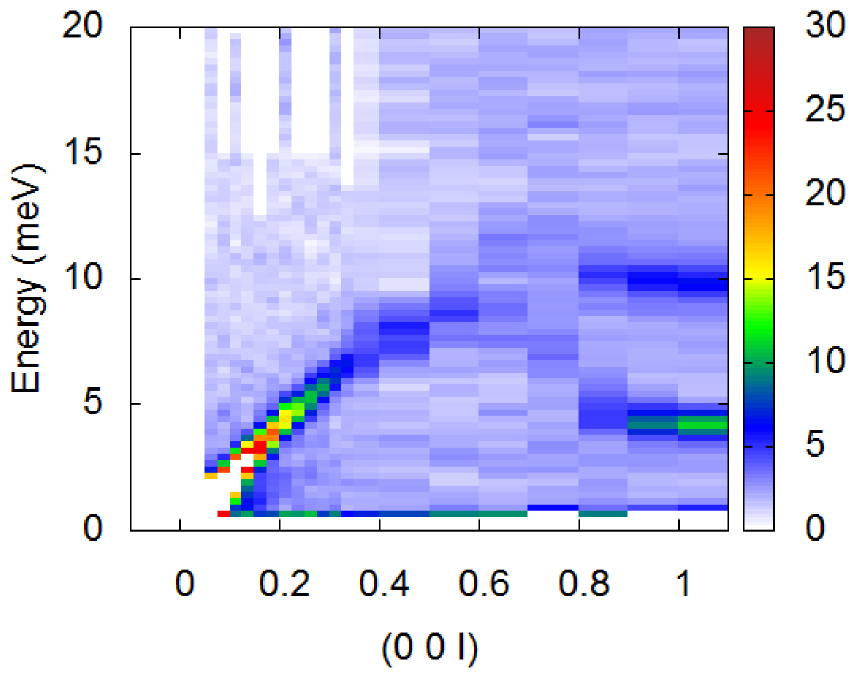
$$\omega(k) = 2\sqrt{\frac{\Gamma}{M}}\sin\frac{ka}{2}$$

Dispersion relation



Collective excitations

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



Direct measurement of the force constants

Cross sections and scattered intensities

Magnetic Interaction between the dipolar field created by both the spins and the 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 sections and scattered intensities

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\,(\gamma r_o)^2 \sum_{i,j} e^{i\mathbf{Q}\,(\mathbf{R}_i^o-\mathbf{R}_j^o)} \sum_{\ell,\ell'} f_\ell(\mathbf{Q})\, f_{\ell'}^*(\mathbf{Q})\, e^{i\mathbf{Q}\,(\mathbf{r}_\ell-\mathbf{r}_{\ell'})}\, e^{-W_\ell-W_{\ell'}}$$
$$\times \int_{-\infty}^{+\infty} dt\, \langle \sum_{a,b} S_{i\ell}^a \left(\delta_{a,b} - \frac{\mathbf{Q}^a\mathbf{Q}^b}{\mathbf{Q}^2}\right) S_{j\ell'}^b(t)\rangle\, e^{-i\omega t}$$

Frozen spins

↓

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\,(\gamma r_o)^2 \sum_{i,j} e^{i\mathbf{Q}\,(\mathbf{R}_i^o-\mathbf{R}_j^o)} \sum_{\ell,\ell'} f_\ell(\mathbf{Q})\, f_{\ell'}^*(\mathbf{Q})\, e^{i\mathbf{Q}\,(\mathbf{r}_\ell-\mathbf{r}_{\ell'})}\, e^{-W_\ell-W_{\ell'}}\, \langle \mathbf{S}_{\perp,i\ell}\, \mathbf{S}_{\perp,j\ell'}\rangle\, \delta(\omega)$$
$$= \frac{k_f}{k_i}\,(\gamma r_o)^2\, |\sum_{i,\ell} e^{i\mathbf{Q}\,(\mathbf{R}_i^o+\mathbf{r}_\ell)}\, f_\ell(\mathbf{Q})\, e^{-W_\ell}\, \mathbf{S}_{\perp,i\ell}|^2\, \delta(\omega)$$

Magnetic structure factor

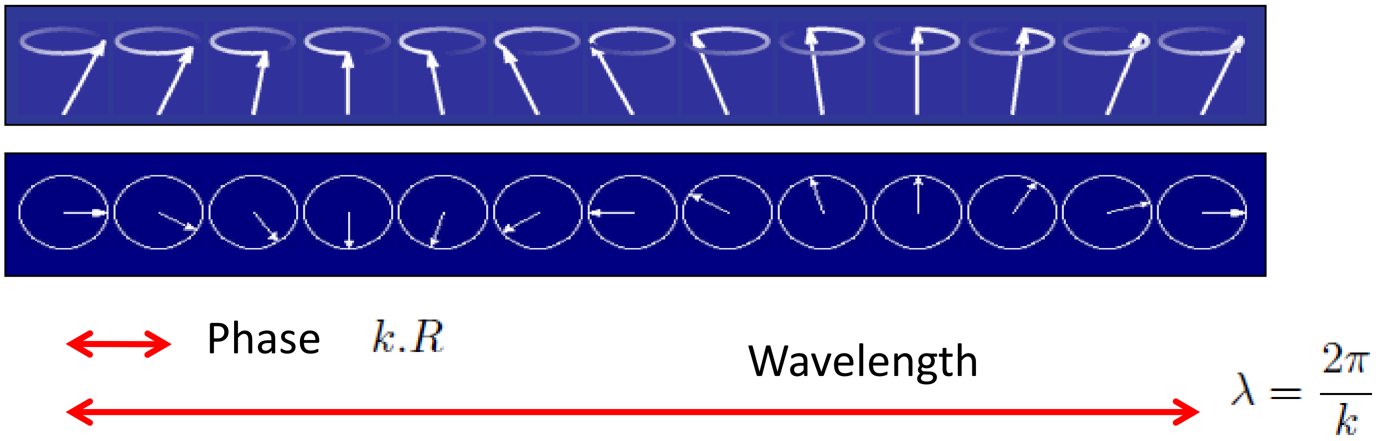
Cross sections and scattered intensities

TWO FORMULAE:

$$F_N(\mathbf{Q}) = \sum_\ell b_\ell \, e^{i\mathbf{Q}\mathbf{r}_\ell} \, e^{-W_\ell}$$

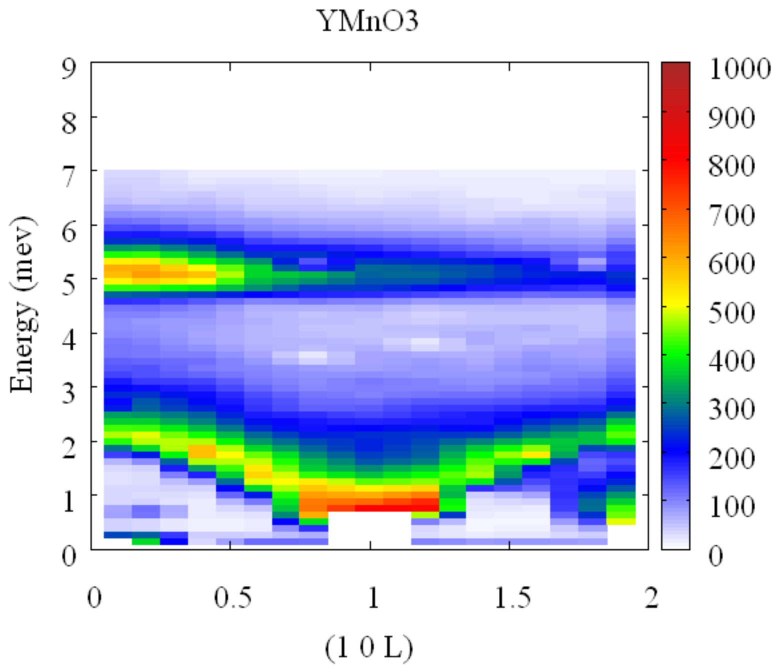
$$\mathbf{F}_M(\mathbf{Q}) = \sum_\ell \mathbf{S}_{\ell\perp} e^{i\mathbf{Q}\mathbf{r}_\ell} \, e^{-W_\ell}$$

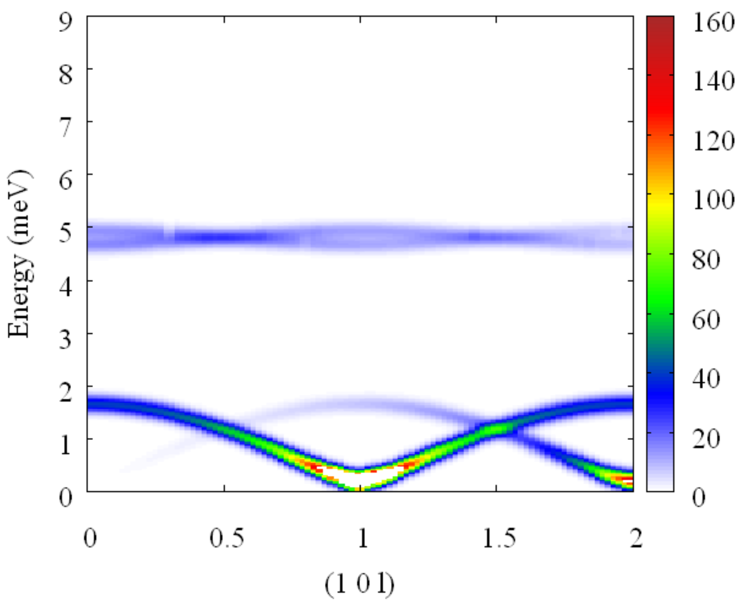
Collective excitations



- Spin waves are precessions of the spins in conventional magnets around the (local) magnetization
- Described by bosonic particles (S=1)
- INS allows one to measure the dispersion of these particles

Collective excitations





Spin wave in spins in multiferroic $YMnO_3$
(meas. and calculations)

Direct measurement of exchange couplings

Take-home message

1. neutron scattering makes it possible to study the correlations in time and space of the particles that constitute matter.

2. all kinds of condensed matter systems: macromolecules, biology, crystalline and magnetic structures

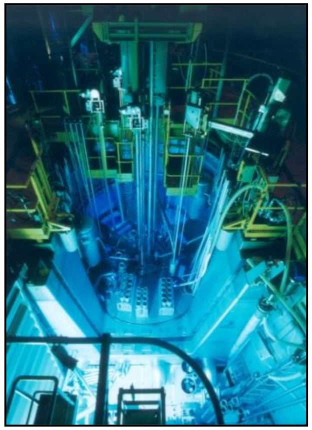
3. does not degrade the sample

4. sensitive to volume, not surface

5. sensitive to magnetism

Magnetic and crystalline structures

Collective excitations



Large scale structures

Reflectometry (surface)

Quasi-elastic scattering

Today : ILL

D20, D2B, D1B

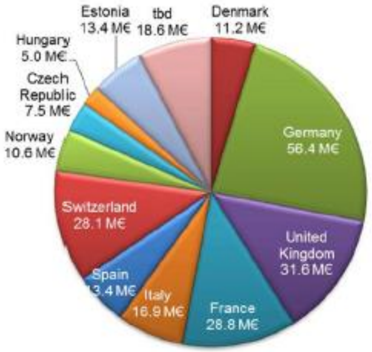
D23, D3, D9, D10

IN4, IN5, IN6

IN8, IN29, IN12, IN22

Tomorrow, ESS (Lund, Suède)

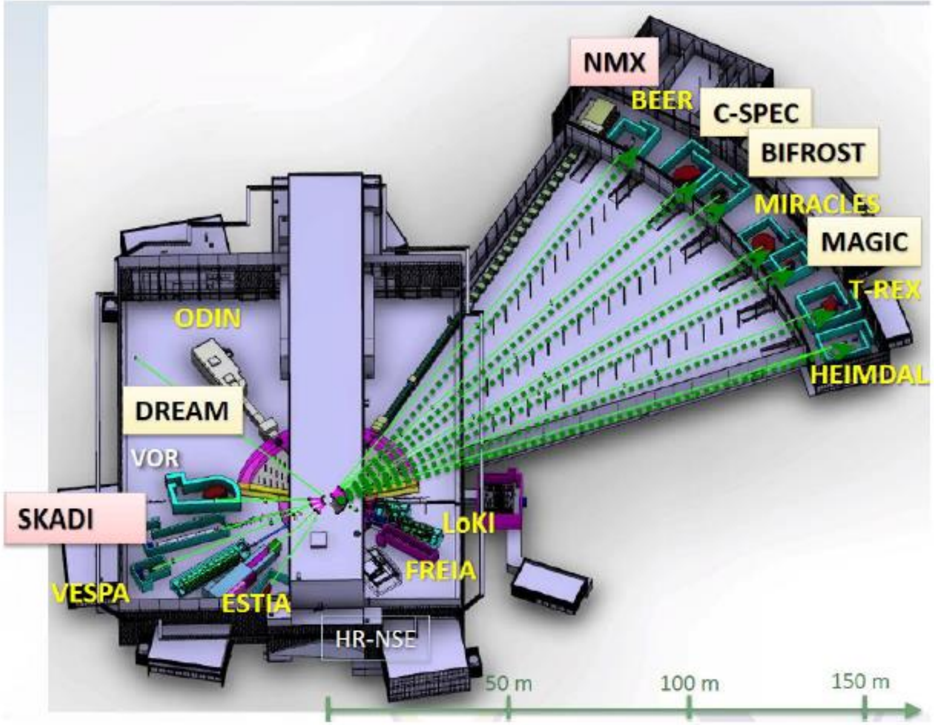
NEW EUROPEAN FACILITY : READY IN 2024 …



LLB teams are heavily involved in several ESS instruments:

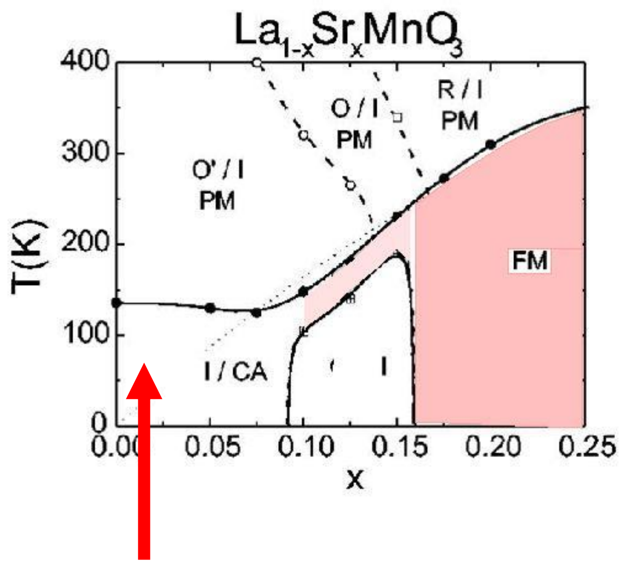
DREAM	diffractometer	24% LLB
SKADI	SANS	50% LLB
MAGIC	diffractometer	59% LLB
BIFROST	INS spectrometer	24% LLB
CSPEC	INS spectrometer	50% LLB
NMX	biocrystallography	15% LLB/IBS

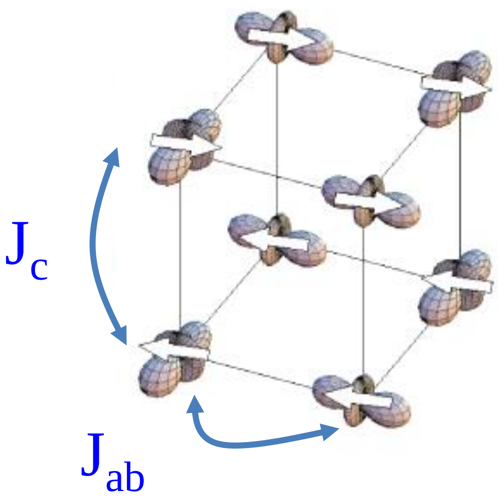
LLB Budget : 35M€ for instruments



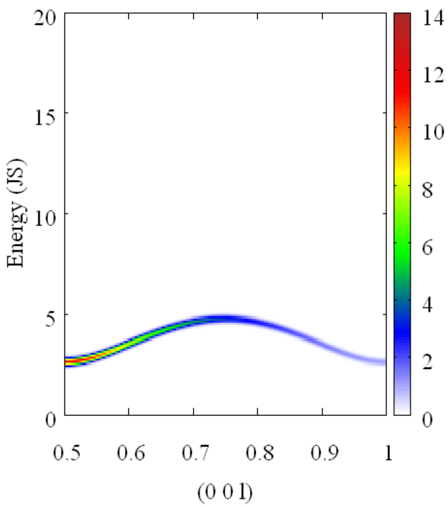
EXAMPLES

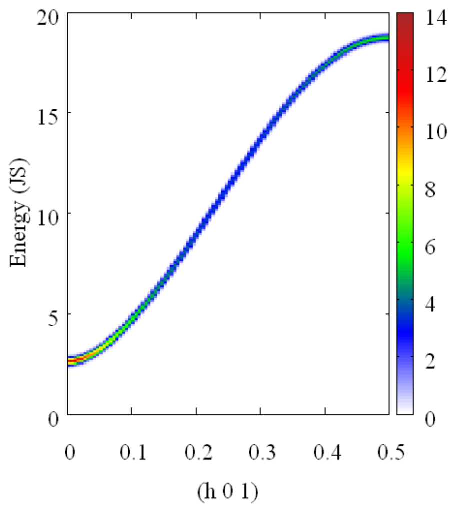
Manganites

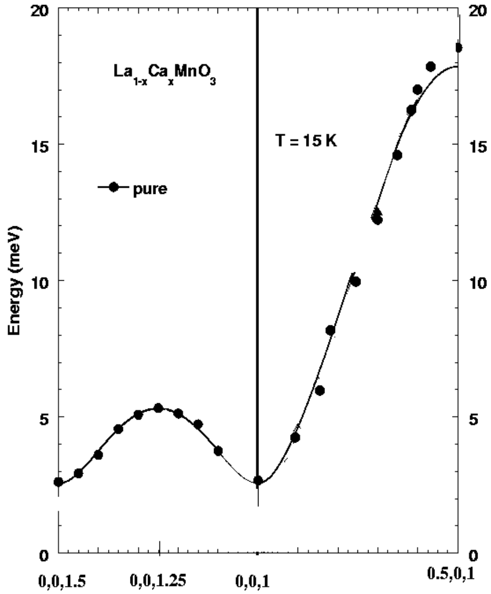




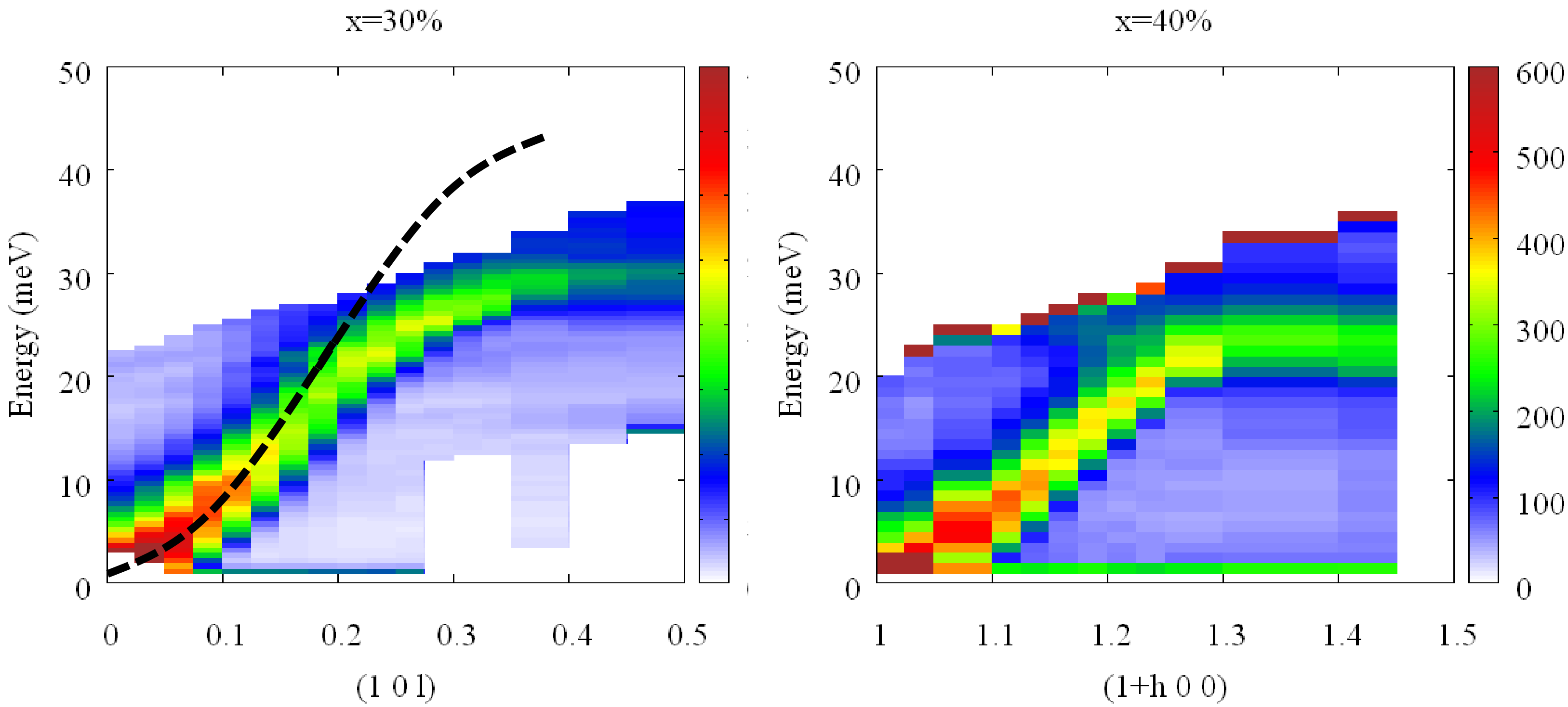
J_c ~ 0.5 meV
J_{ab} ~ -0.8 meV







Spin waves in CMR manganites



Pyrochlore ferromagnet $Yb_2Ti_2O_7$

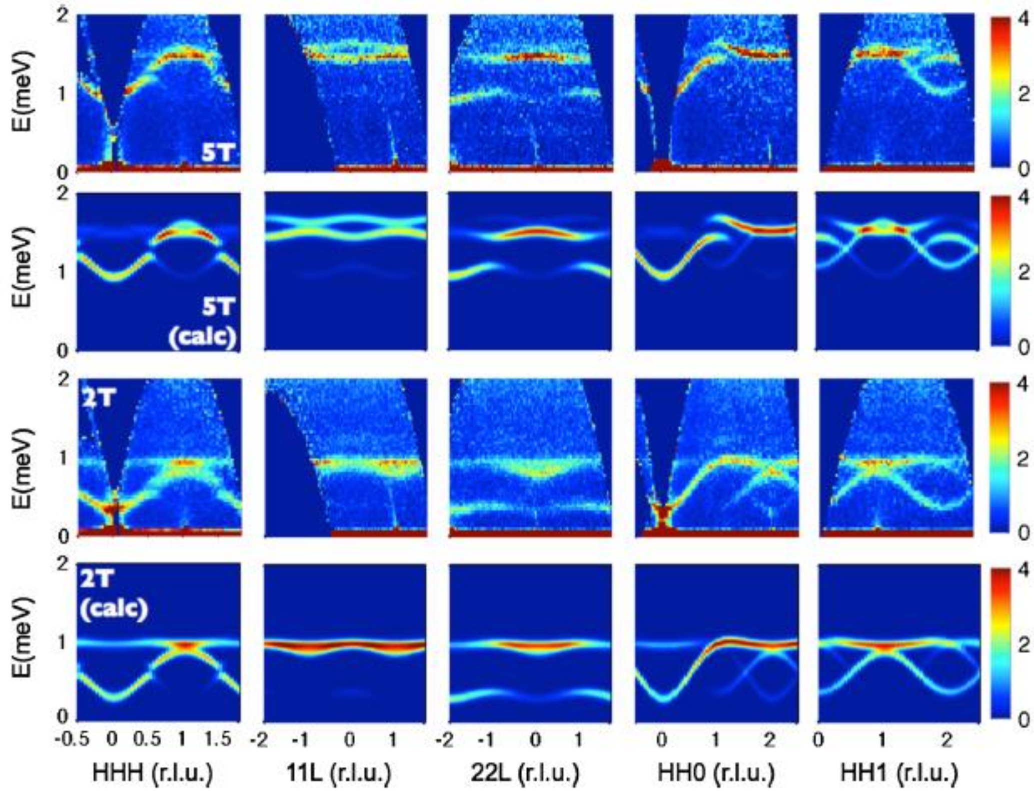


FIG. 1. The measured $S(\mathbf{Q}, \omega)$ at $T = 30$ mK, sliced along various directions in the HHL plane, for both $H = 5$ T (first row) and $H = 2$ T (third row). The second and fourth rows show the calculated spectrum for these two field strengths, based on an anisotropic exchange model with five free parameters (see text) that were extracted by fitting to the 5 T data set. For a realistic comparison to the data, the calculated $S(\mathbf{Q}, \omega)$ is convoluted with a Gaussian of full-width 0.09 meV. Both the 2 T and 5 T data sets, composed of spin wave dispersions along five different directions, are described extremely well by the same parameters. (Note that r.l.u. stands for reciprocal lattice units.)

Pyrochlore antiferromagnet $Er_2Ti_2O_7$

PRL **109**, 167201 (2012) PHYSICAL REVIEW LETTERS week ending 19 OCTOBER 2012

Order by Quantum Disorder in $Er_2Ti_2O_7$

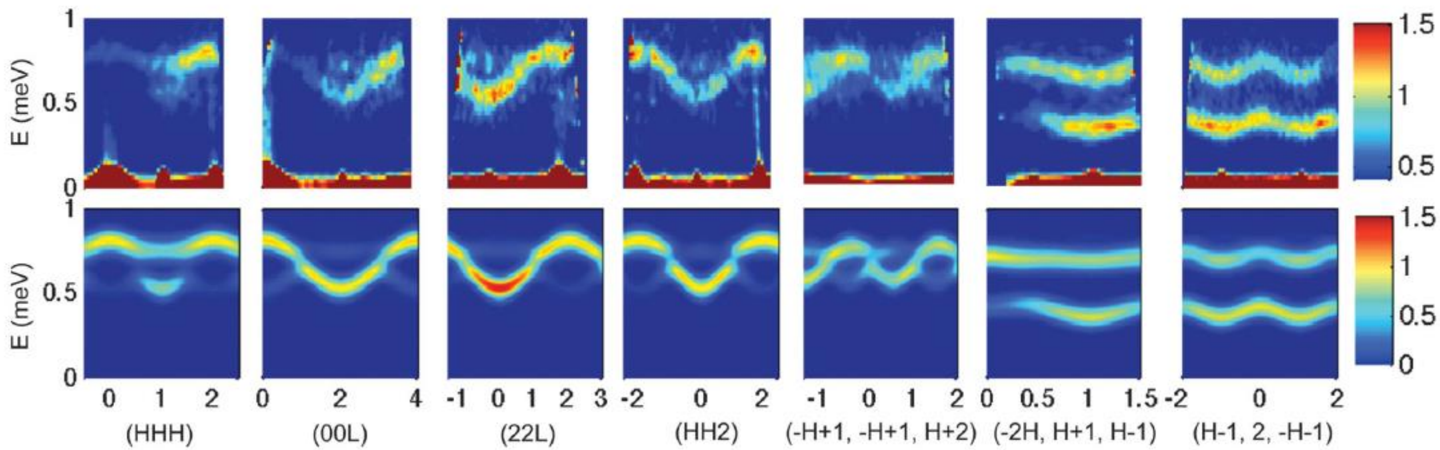
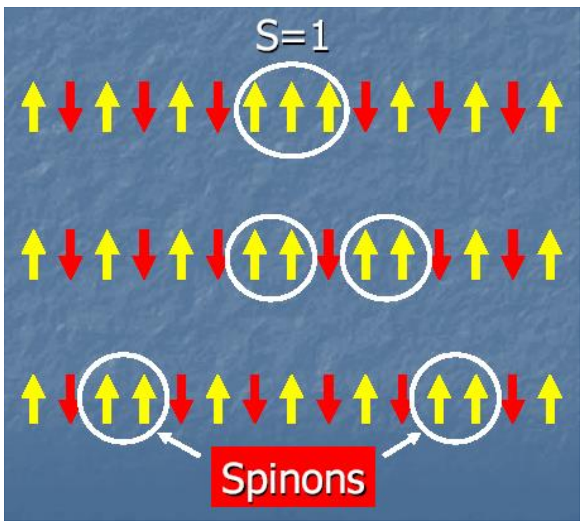


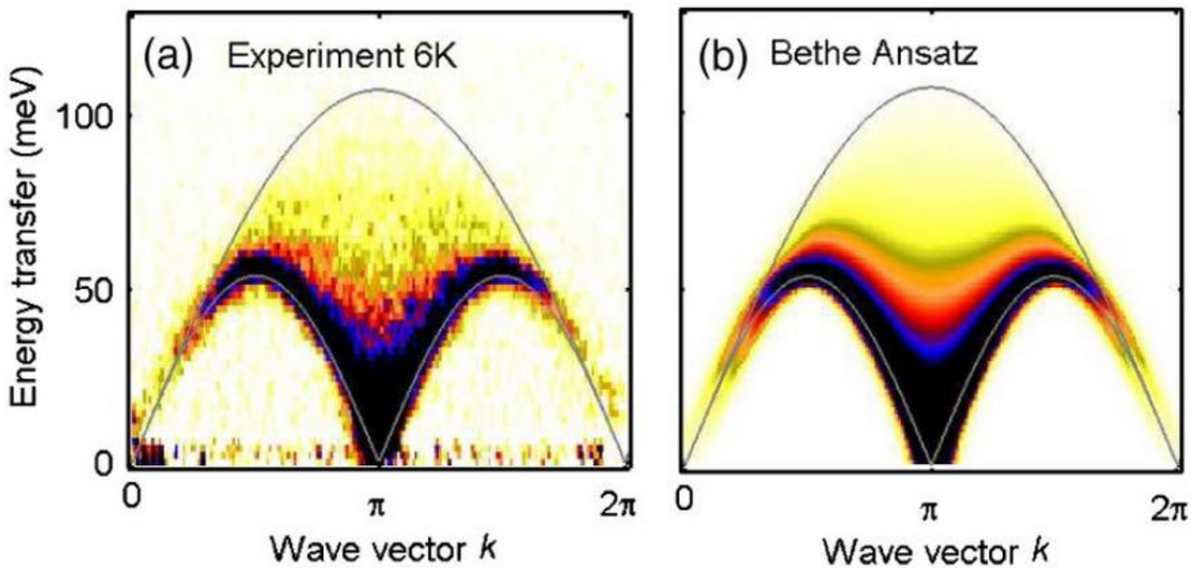
FIG. 1 (color). The measured $S(\mathbf{Q}, \omega)$ at $T = 30$ mK, $H = 3$ T sliced along several directions. The first five columns show $S(\mathbf{Q}, \omega)$ in the *HHL* plane, with the field applied along $[1\bar{1}0]$, while the last two columns show $S(\mathbf{Q}, \omega)$ for the field along [111]. Top row: measured $S(\mathbf{Q}, \omega)$. Bottom row: calculated $S(\mathbf{Q}, \omega)$, based on an anisotropic exchange model with six free parameters (see text) that were extracted by fitting to the measured dispersions.

Quantum magnets

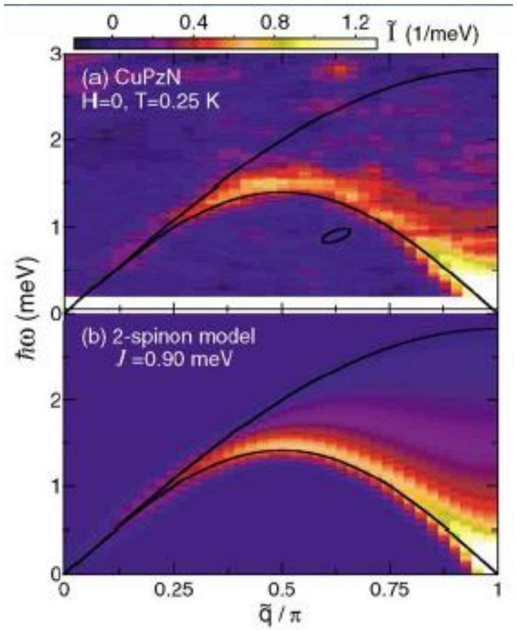
In some systems, quantum fluctuations prevent from long range magnetic ordering. Excitations are not spin waves any more…

Frustration, low dimension, spin =1/2, 1

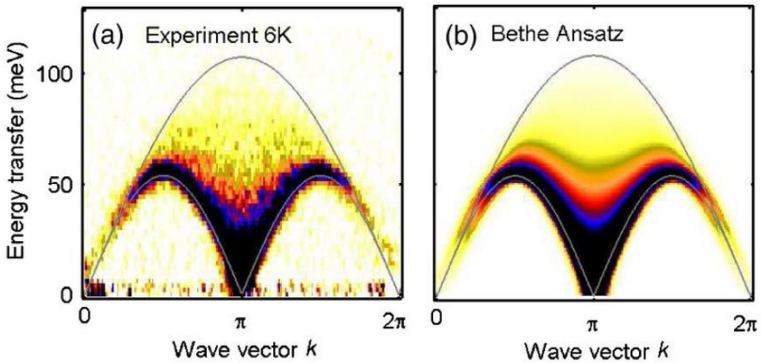




Quantum magnets



CuPzN, Stone et al, PRL 2003



$KCuF_3$, Lake et al, Nature Materials 2005

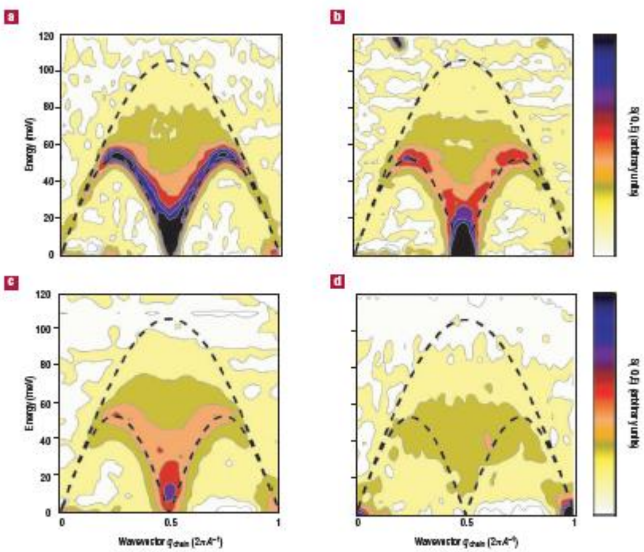
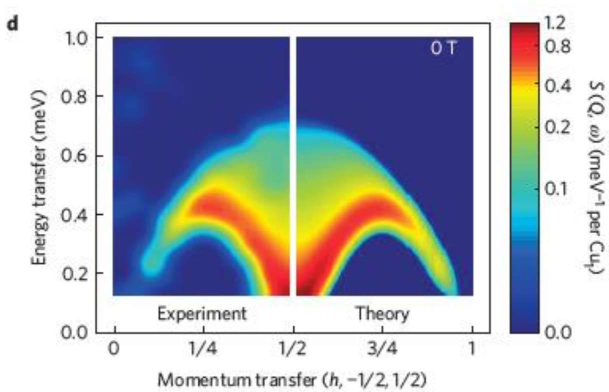


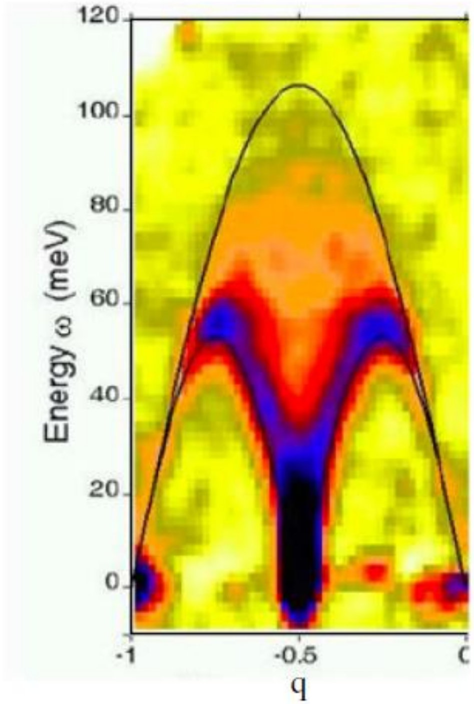
Figure 1 Inelastic neutron scattering data for $KCuF_3$. The data is plotted as a function of E and q parallel to the chains for the temperatures a, $T = 6$ K, b, $T = 50$ K, c, $T = 150$ K and d, $T = 300$ K. The colours indicate the size of the neutron scattering cross-section $S(q,E)$ and the superimposed black dashed lines indicate the region where the multi-spinon continuum is predicted at $T = 0$ K by the Muller Ansatz equation (1). The data was collected using the MAPS time-of-flight spectrometer at ISIS, Rutherford Appleton Laboratory, UK.

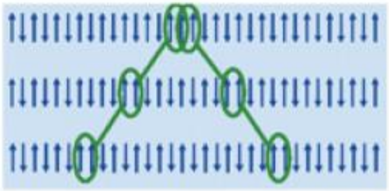


$CuSO_4$ $5D_2O$ Mourigal et al, Nature Physics 2013

Quantum magnets

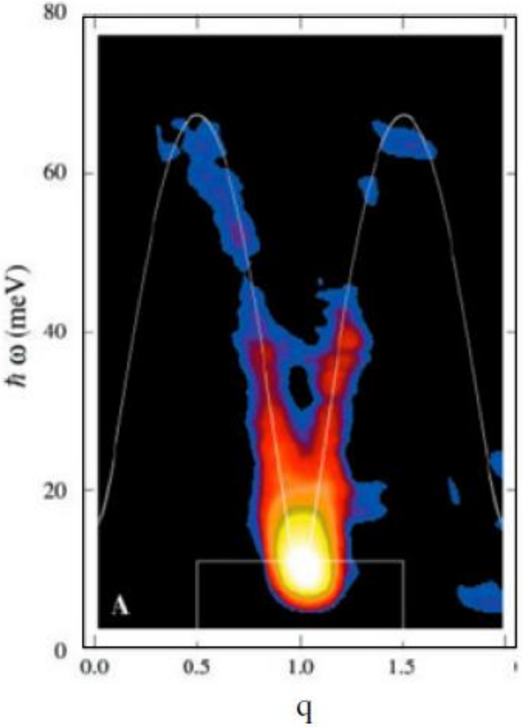
S= ½ $KCuF_3$

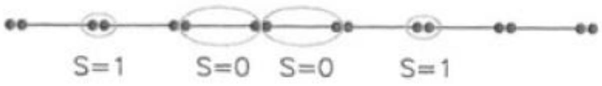




Spin correlations decay as power law
Spin ½ particles : continuum
gapless

S= 1 Y_2BaNiO_5





Spin correlations decay exponentially
Spin 1 excitation : well defined mode
(Haldane) Gapped spectrum