Summary

Spin waves : excited states of the Heisenberg Hamiltonian

Approximations

1) Ordered phase
2) Small deviations around the ordered moment, large S, low T

Calculation : equation of motion (linear set of L coupled equations)

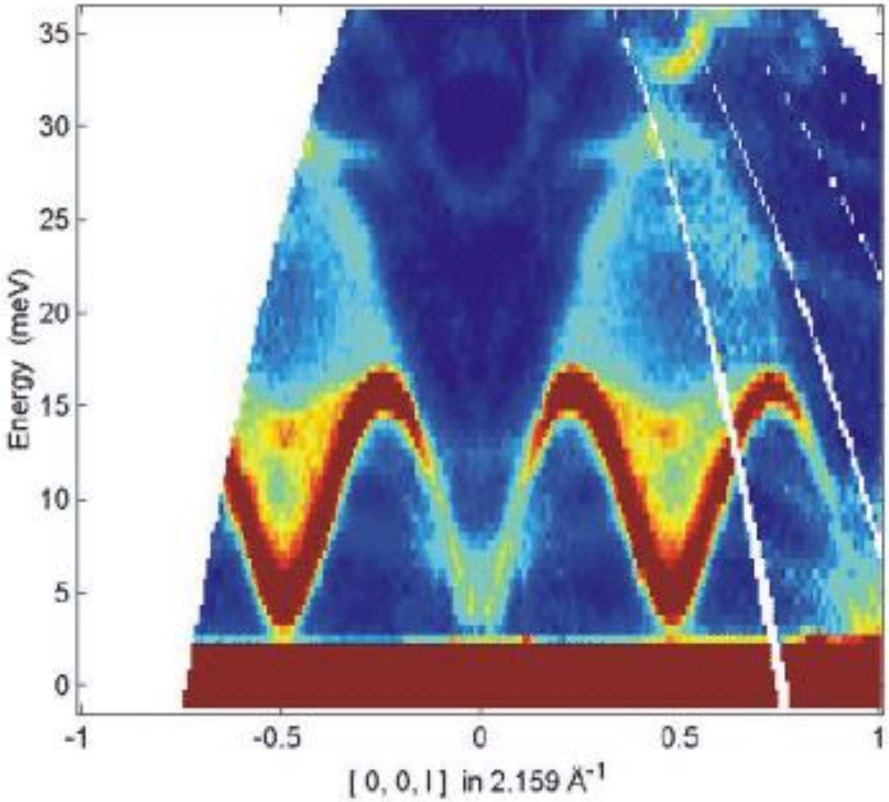
L ions in the magnetic unit cell : L spin waves branches

Quasi independent modes (bosons) , and role of quantum fluctuations in low dimension

Part II

How to measure spin excitations in (Q,ω) space ?

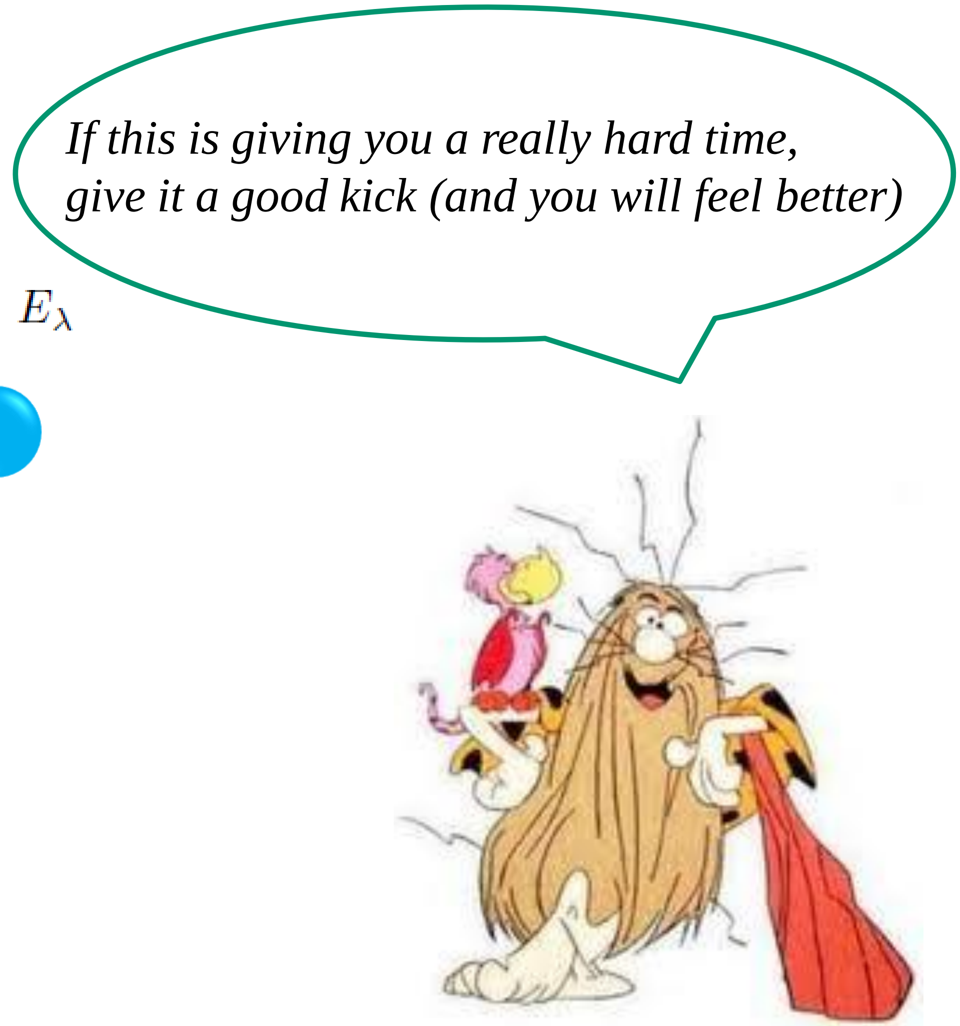
Neutron spectroscopy



M. Arai et al, Phys Rev. Lett. 77, 3649 (1996)

Basic idea

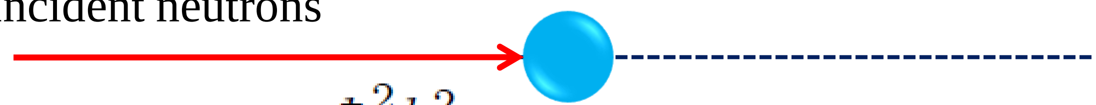
$|\lambda\rangle \quad E_\lambda$



Basic idea

$|\lambda\rangle \quad E_\lambda$

Incident neutrons

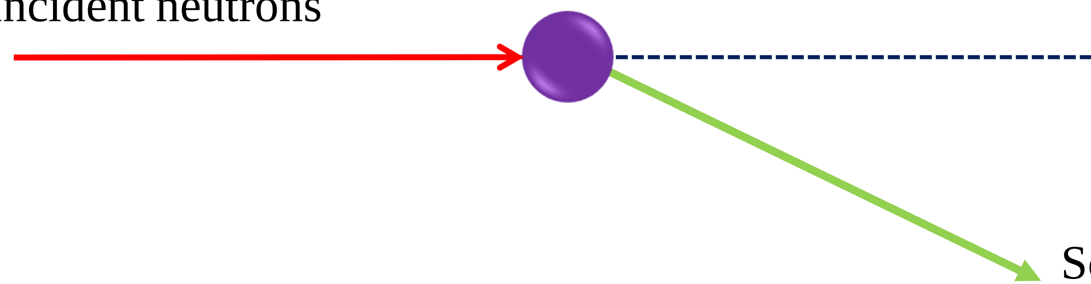


$$|\vec{k}_i, \sigma_i\rangle \qquad E_i = \frac{\hbar^2 k_i^2}{2m_n}$$

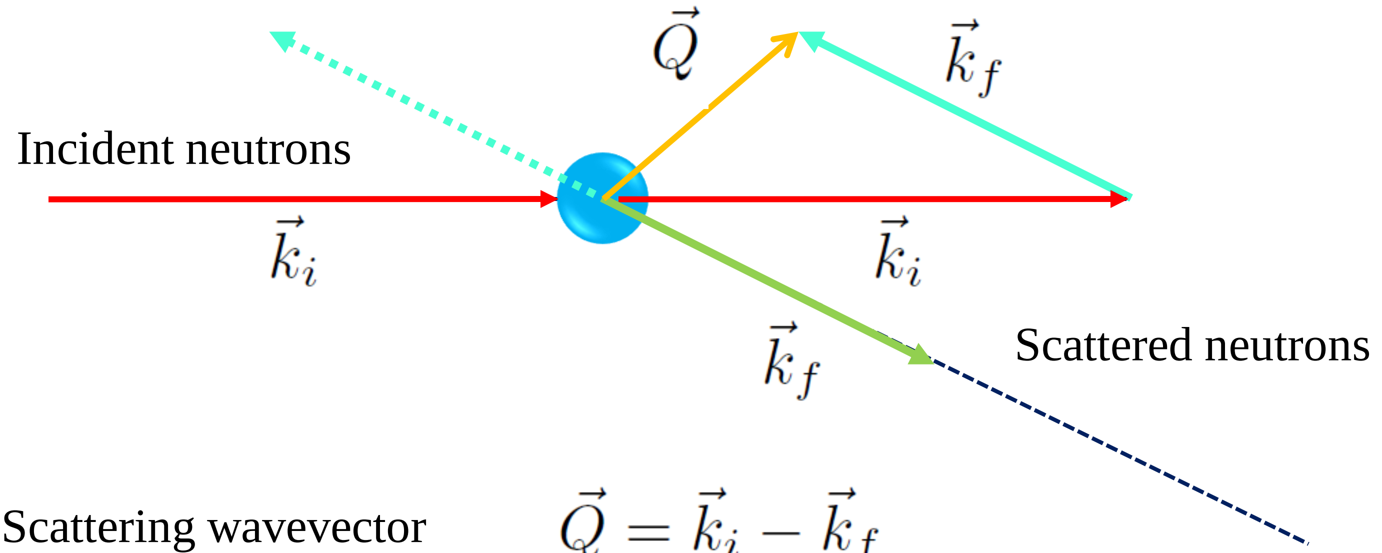
wavevector spin energy

Neutrons are plane waves

Basic idea


$|\lambda'\rangle \quad E_{\lambda'}$
Incident neutrons
Scattered neutrons
$|\vec{k}_f, \sigma_f\rangle \quad E_f = \dfrac{\hbar^2 k_f^2}{2m_n}$

Basic idea



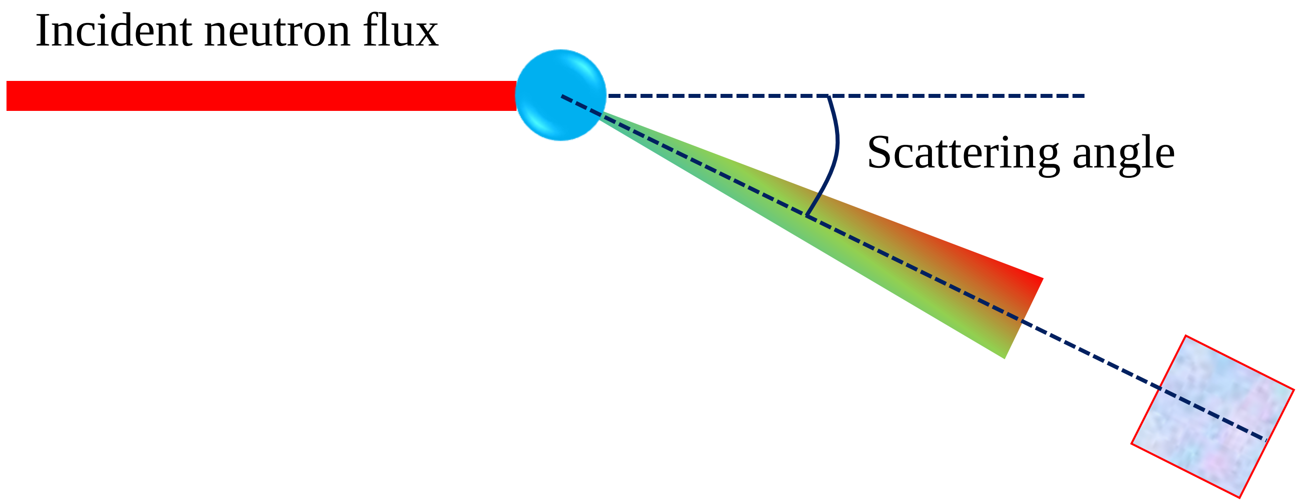
Scattering wavevector $\vec{Q} = \vec{k}_i - \vec{k}_f$

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

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

Basic idea

Scattered neutrons in a given solid angle can :



1. gain energy (up to infinity)

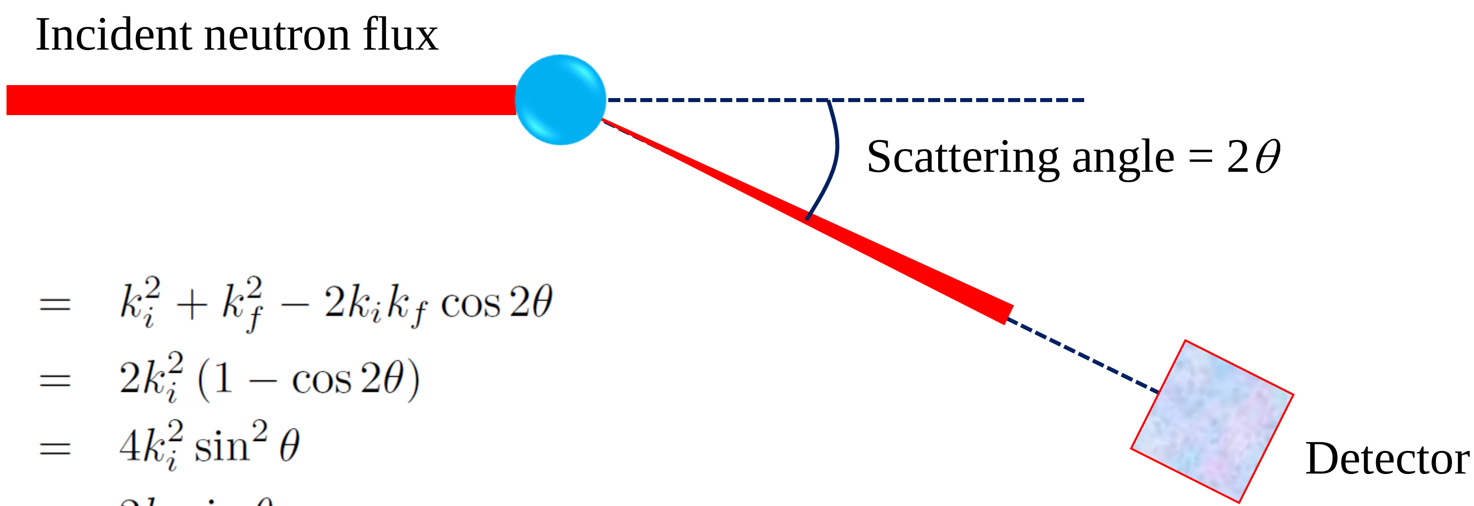
2. loose energy (up to E_i)

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

$$-\infty \leq \hbar\omega \leq E_i$$

Elastic scattering

The neutrons keep their energy : « elastic scattering »



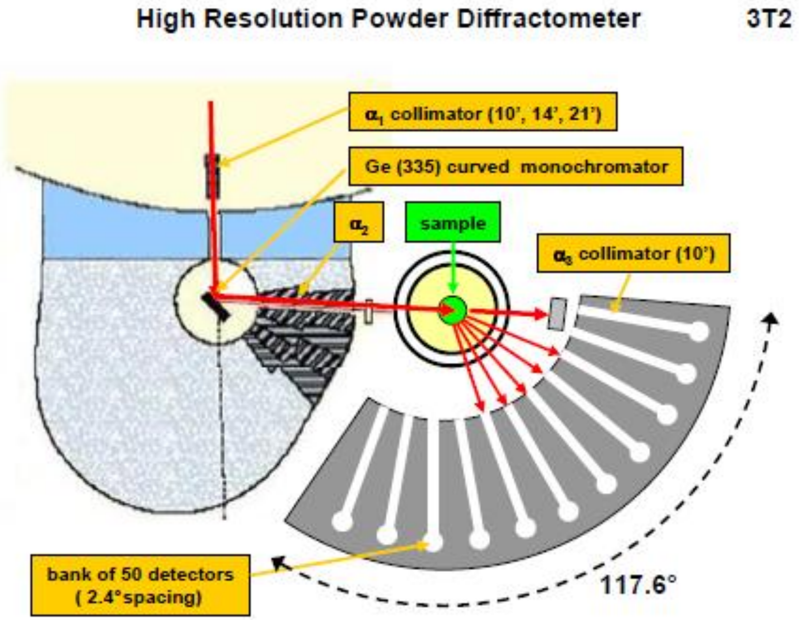
$$
\begin{aligned}
Q^2 &= k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta \\
&= 2k_i^2 \left(1 - \cos 2\theta\right) \\
&= 4k_i^2 \sin^2 \theta \\
Q &= 2k_i \sin \theta
\end{aligned}
$$

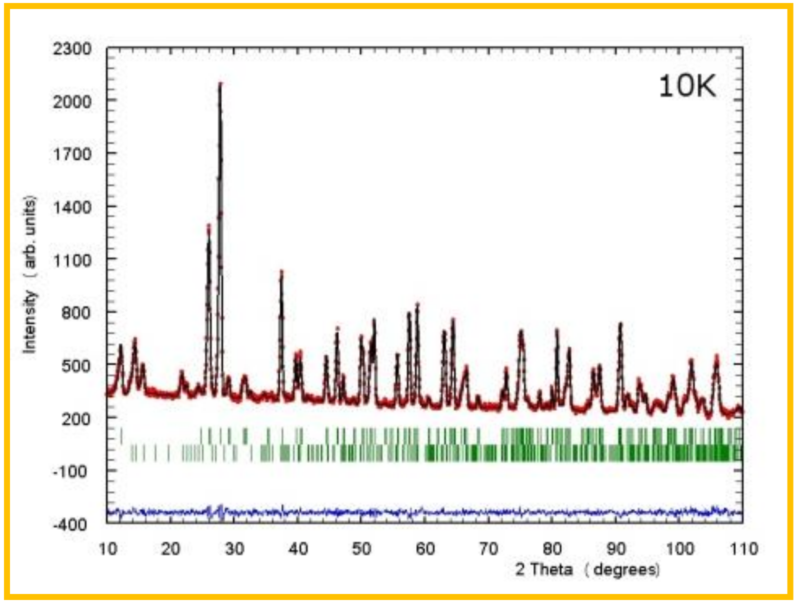
$$
\frac{2\pi}{d} = 2\frac{2\pi}{\lambda} \sin \theta
$$

$$
\boxed{\lambda = 2d \sin \theta}
$$

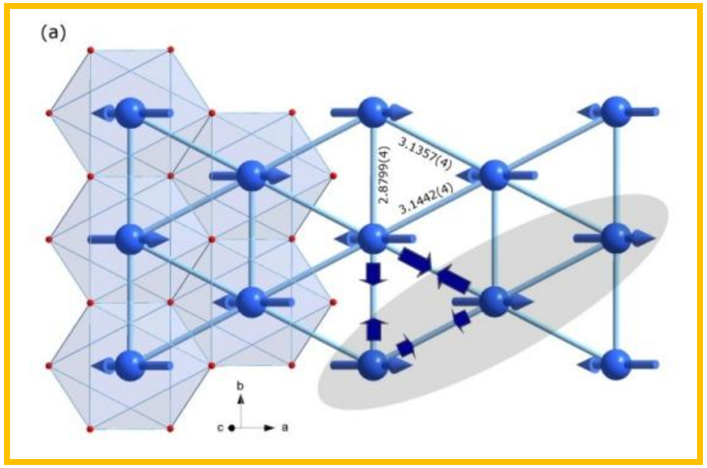
This is the Bragg law !!

Elastic scattering



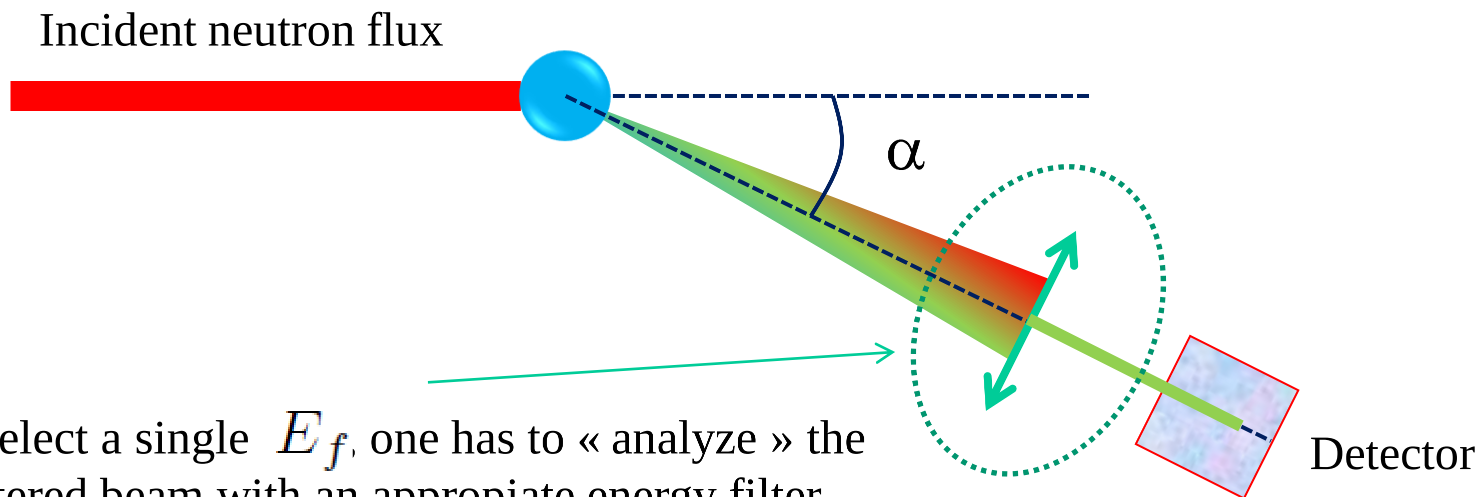


Unit cell and space group



Inelastic scattering

The neutrons gain or loose energy : « inelastic scattering »

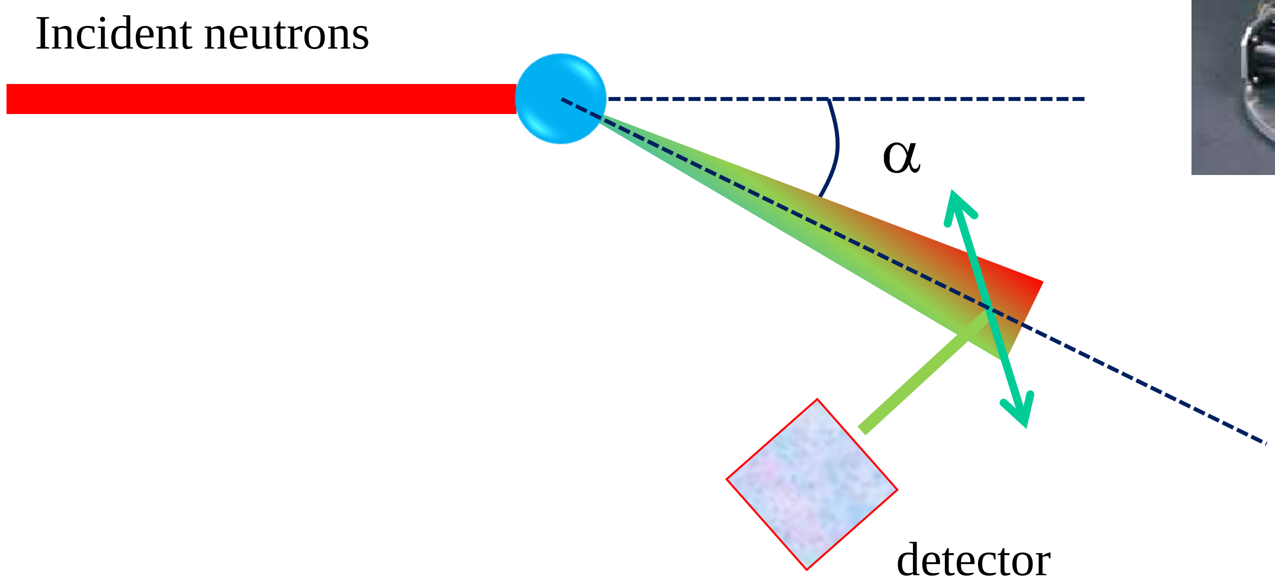


To select a single E_f, one has to « analyze » the scattered beam with an appropiate energy filter

Once E_i and E_f are chosen, select $|\vec{Q}|$ by varying α

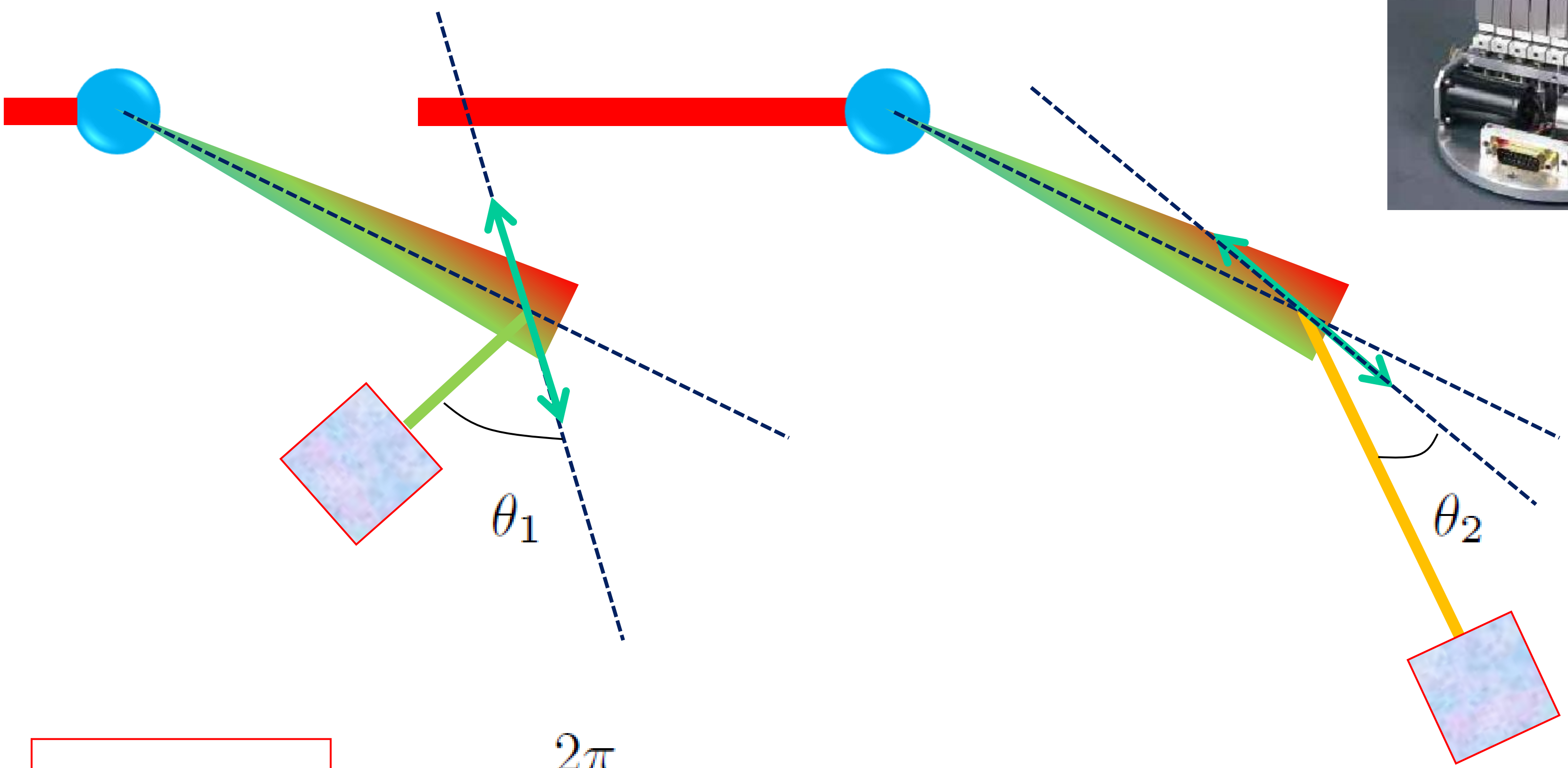
Inelastic scattering

Case 1 : single crystal analyzer



Inelastic scattering

Case 1 : single crystal analyzer (Bragg law)

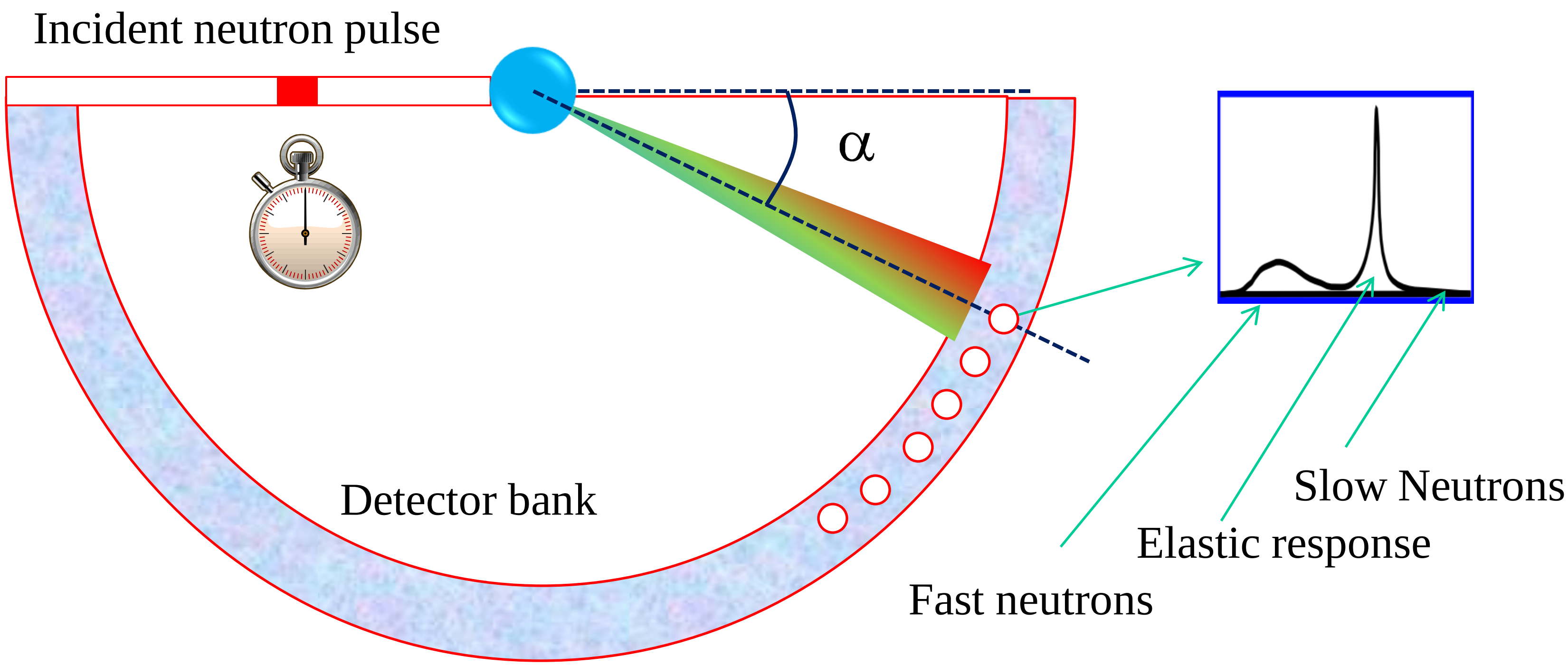


$$\boxed{\lambda = 2d\sin\theta} \qquad k_f = \frac{2\pi}{\lambda_f} \qquad \theta_1 \geq \theta_2 \qquad E_1 \leq E_2$$

Inelastic scattering

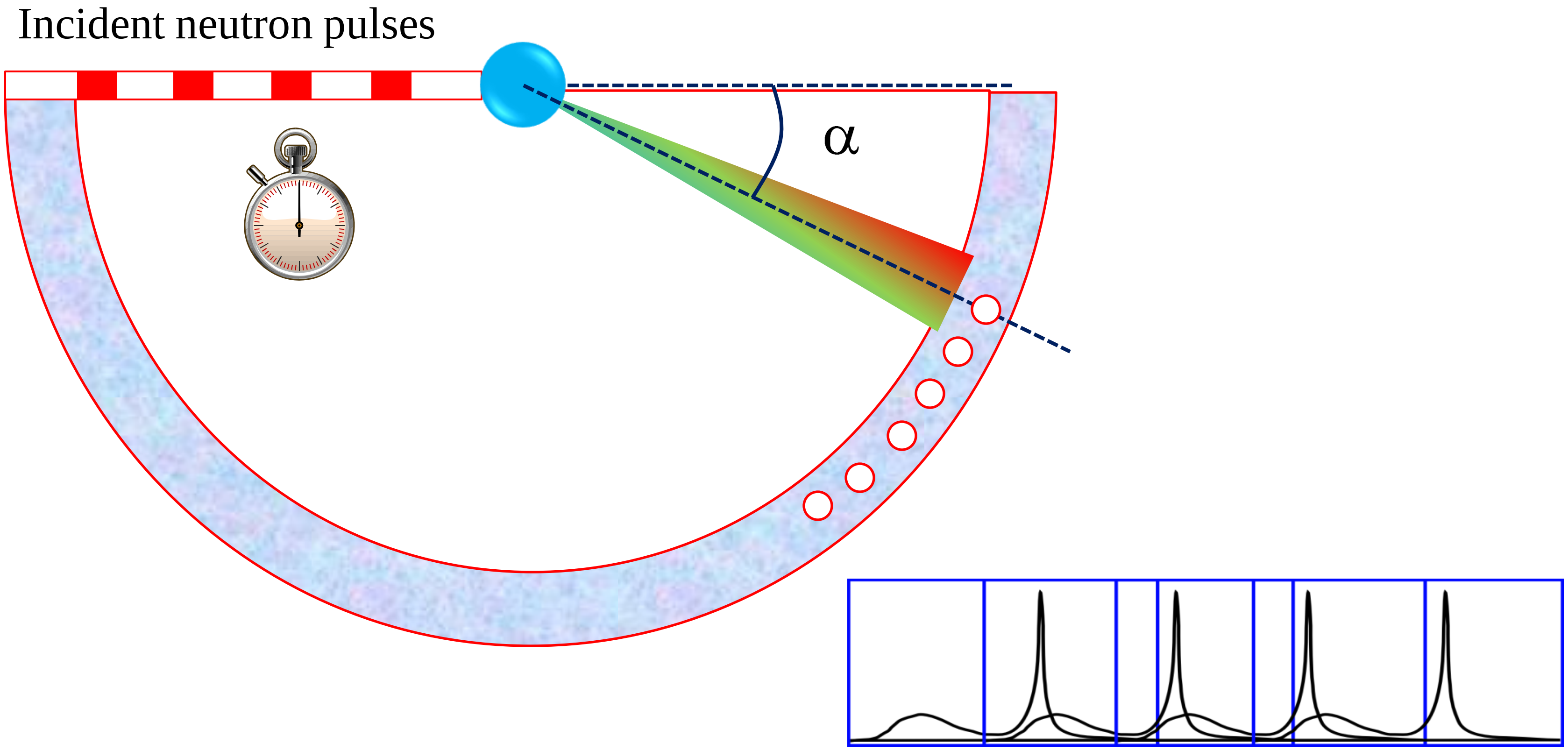
Case 2 : « time of flight » analyzer

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



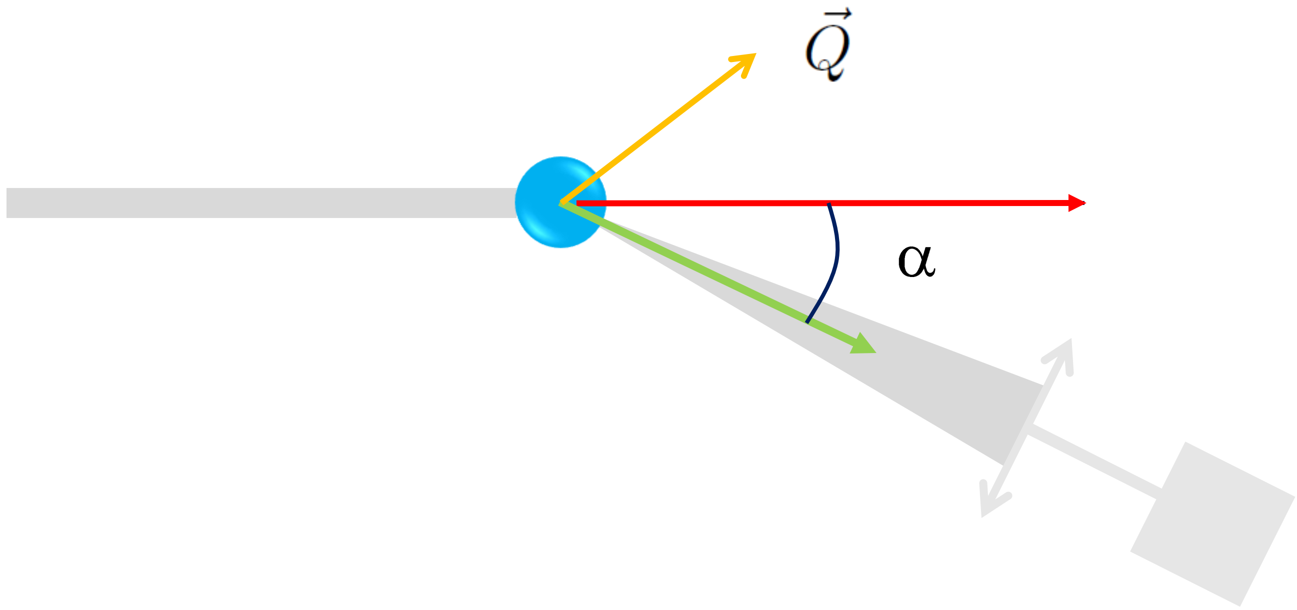
Inelastic scattering

Case 2 : « time of flight » analyzer



Inelastic scattering

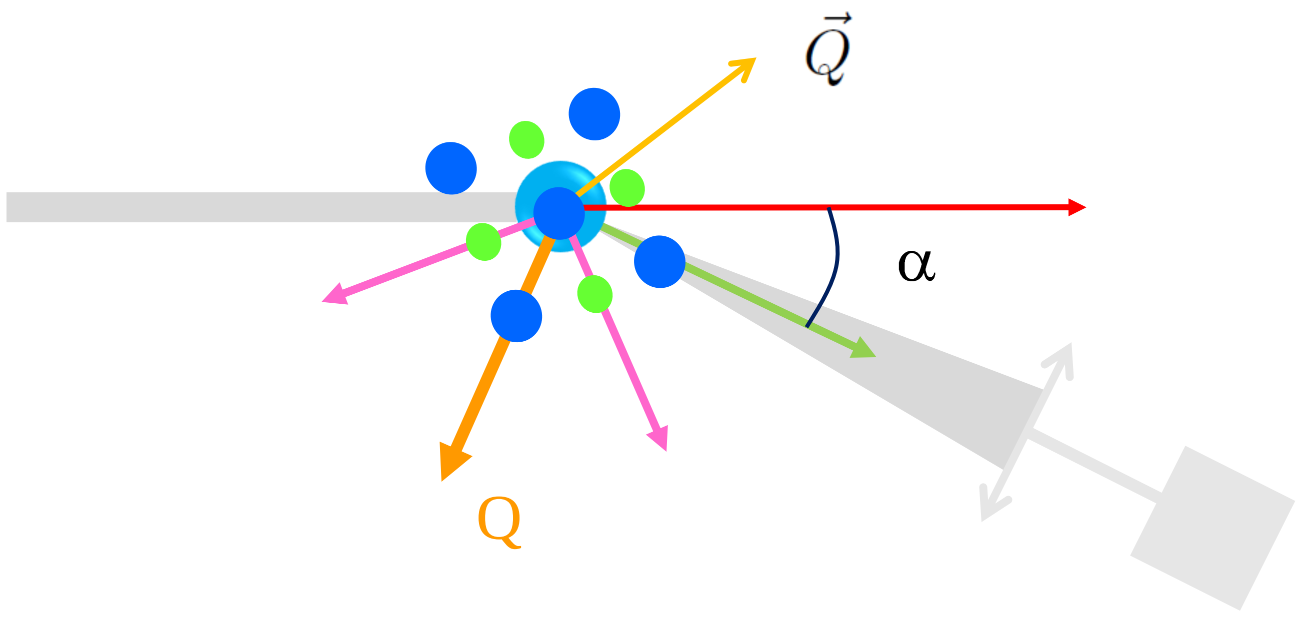
How to select the wavevector for a given energy transfer ?



There is just one known wavevector in the lab geometry : $\vec{Q} = \vec{k}_i - \vec{k}_f$

Inelastic scattering

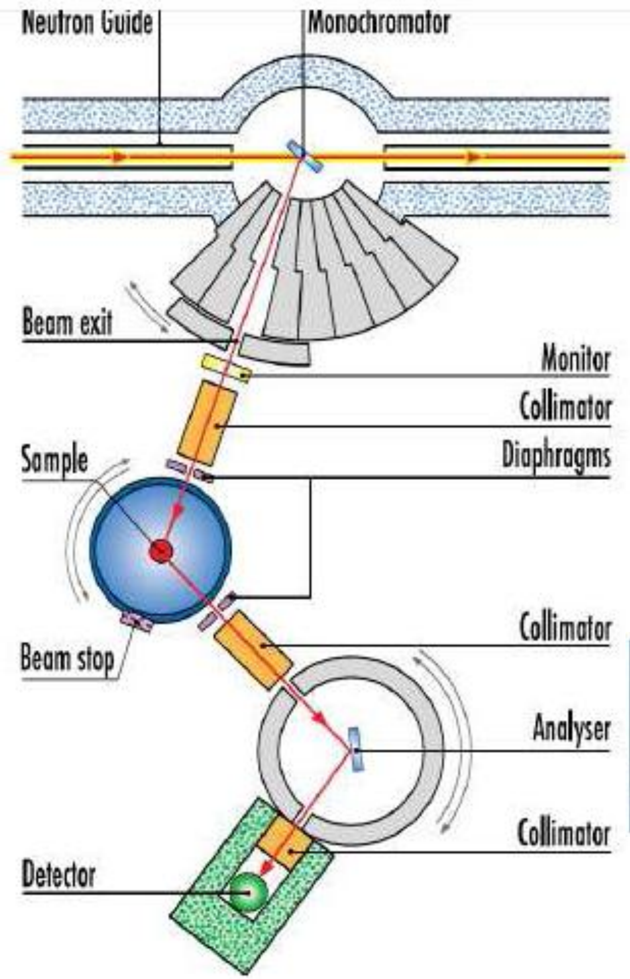
How to select the wavevector for a given energy transfer ?

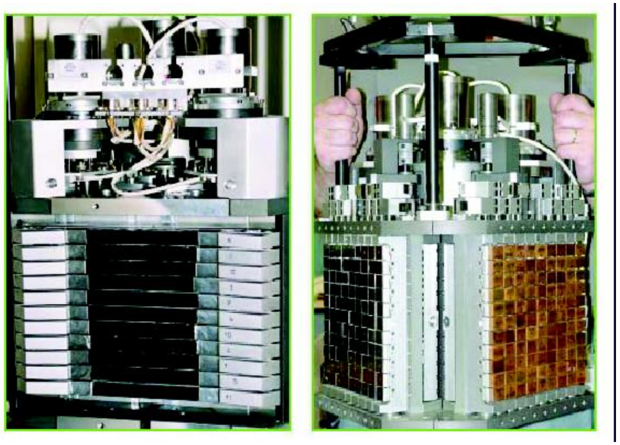


Rotate the sample to let coincide Q with Q

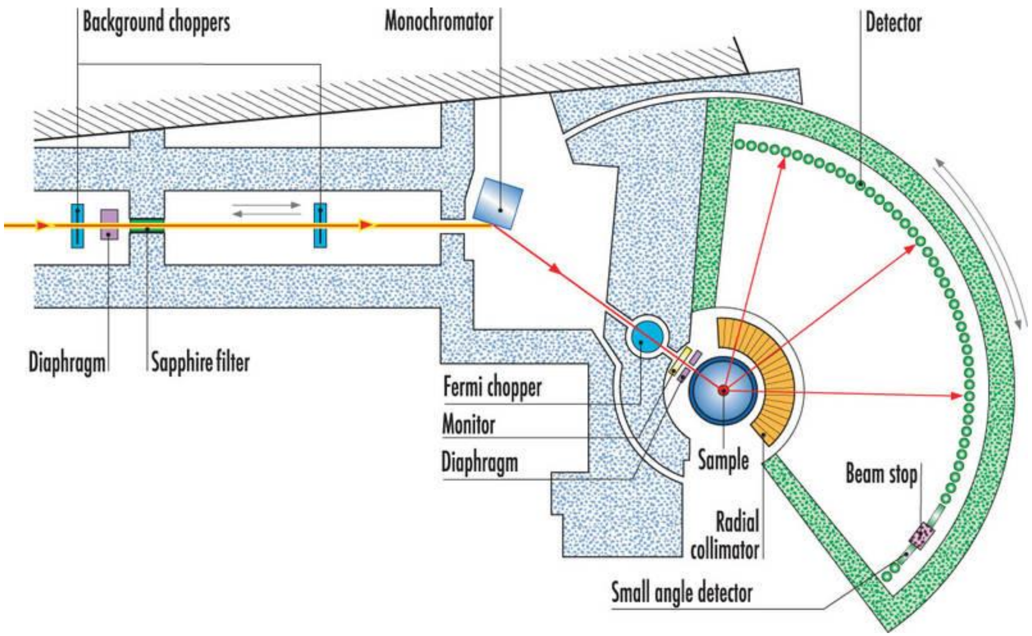
Useless if the sample is a powder, mandatory if the sample is a single crystal

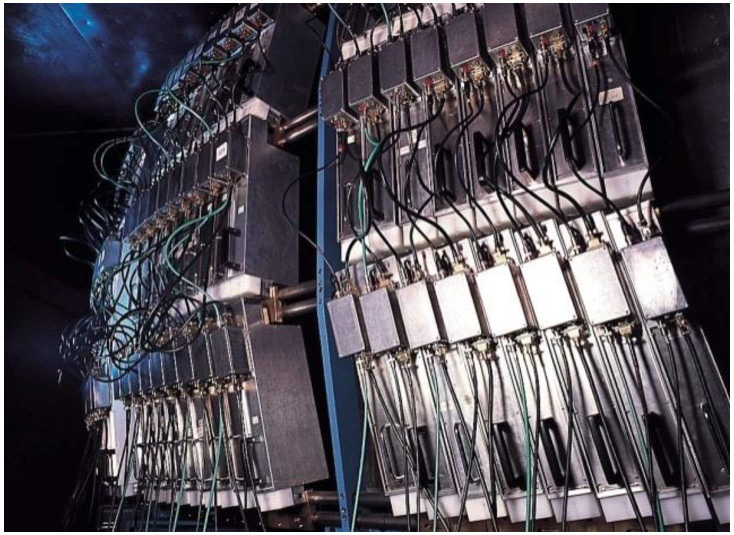
Triple axis


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



Time of flight





Neutron sources

In Europe:

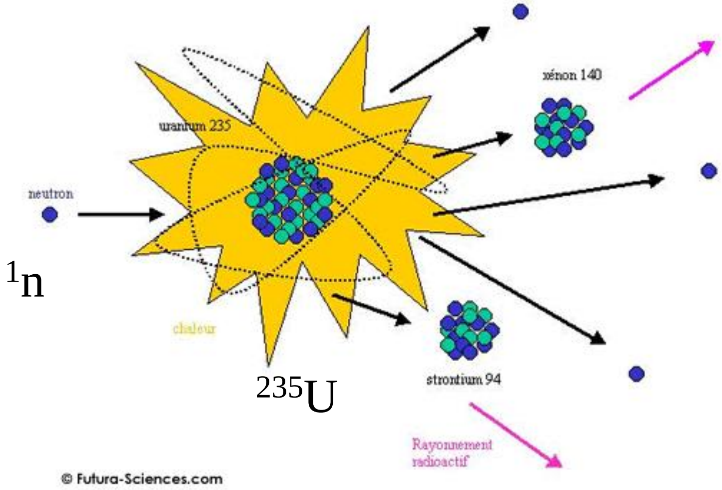
Reactors
ILL-Grenoble (France)
LLB-Saclay (France)
FRMII-Munich (Germany)
HMI-Berlin (Germany)

Spallation sources
ISIS-Didcot (UK)
PSI-Villigen (Switzerland)

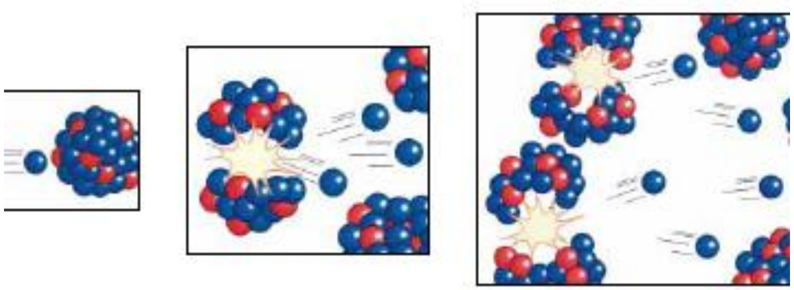
But also:

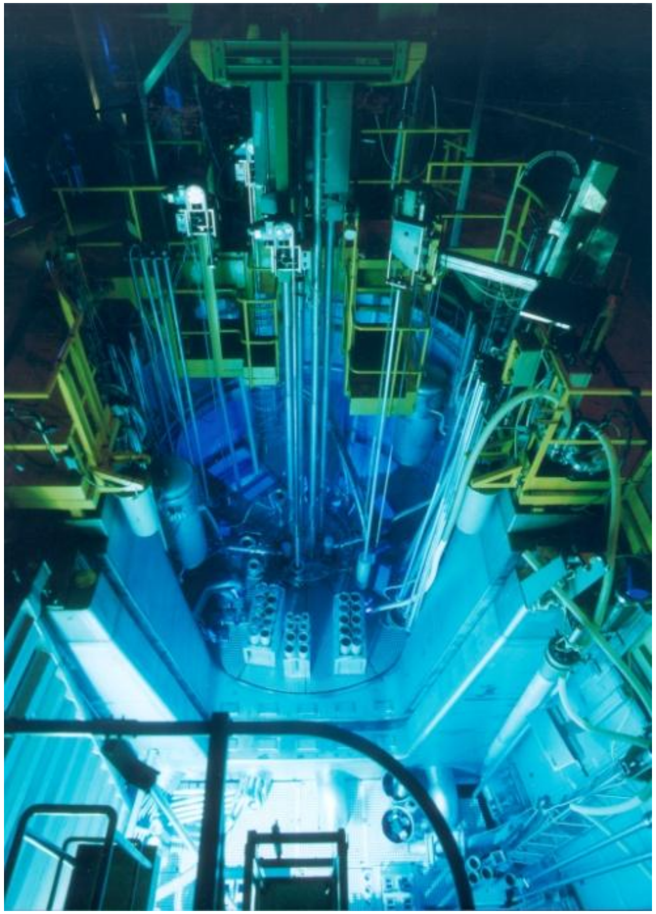
Dubna (Russia), JPARC (Japan)
SNS, DOE labs (USA), ANSTO (Australia)
Canada, India, …

Fission

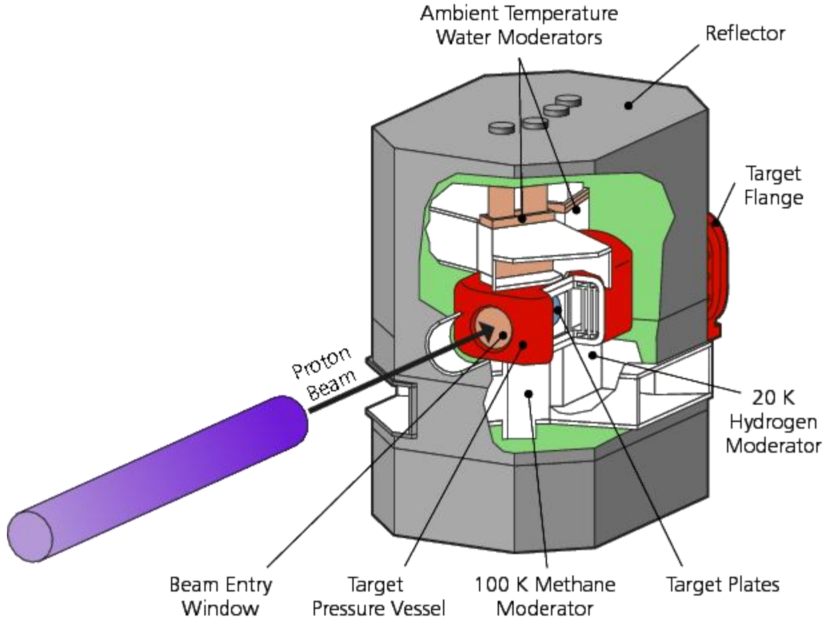


neutron
uranium 235
xénon 140
chaleur
strontium 94
Rayonnement
radioactif
© Futura-Sciences.com
^{1}n
^{235}U

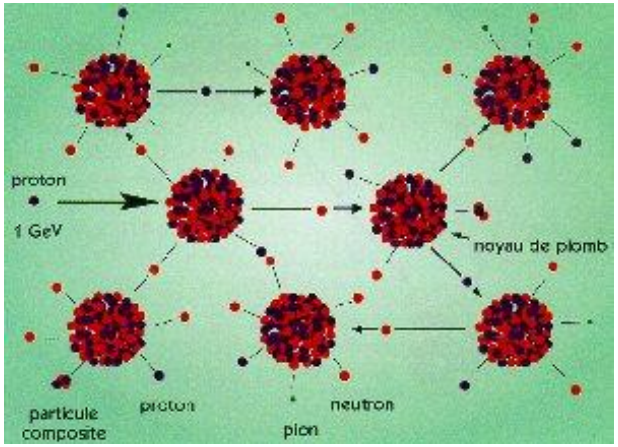




Spallation

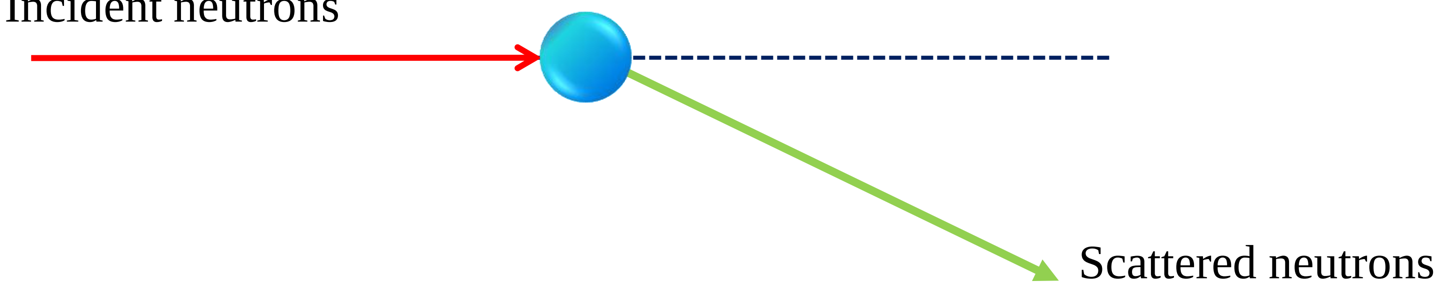


Ambient Temperature
Water Moderators
Reflector
Target
Flange
Proton
Beam
20 K
Hydrogen
Moderator
Beam Entry
Window
Target
Pressure Vessel
100 K Methane
Moderator
Target Plates



proton
1 GeV
noyau de plomb
particule
composite
proton
neutron
pion

Cross section



Each individual process is characterized by a transition probability (« Fermi Golden Rule ») :

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

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

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

Cross section

Probability for the incident neutron to be in the σ_i spin state

Density of accessible states 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

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

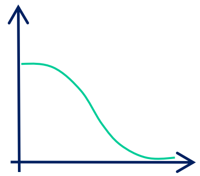
Probability for the target to be in the initial state λ

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

Cross section

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

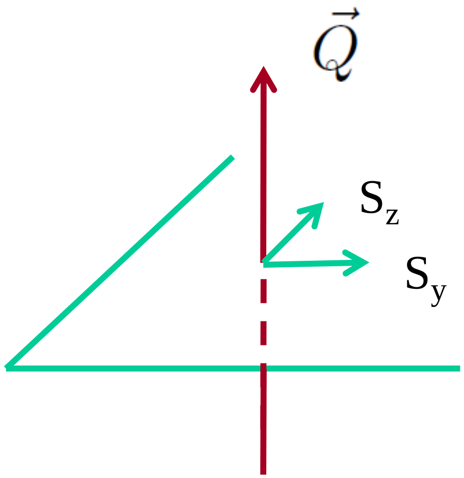
$$\vec{S}_{\perp,i}\, \vec{S}_{\perp,j}(t) = \sum_{\alpha,\beta} S_{\alpha,i} \left(\delta_{\alpha,\beta} - \frac{Q_\alpha\, Q_\beta}{Q^2} \right) S_{\beta,j}(t)$$

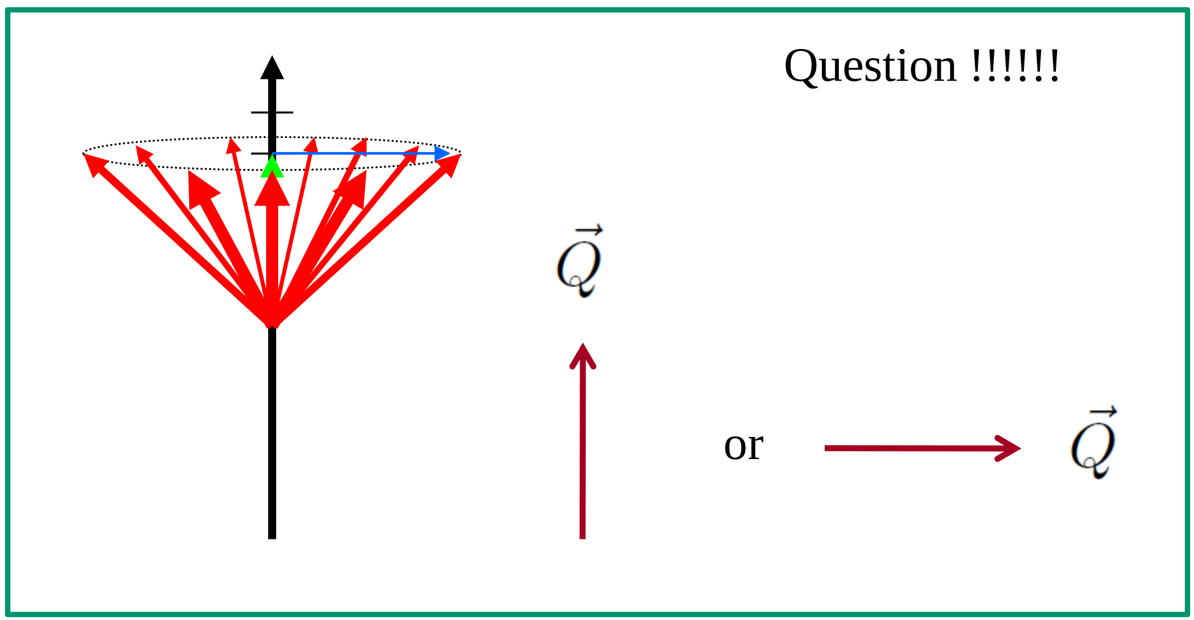
Cross section

What does that mean ?

$$\vec{S}_{\perp,i}\,\vec{S}_{\perp,j}(t) = \sum_{\alpha,\beta} S_{\alpha,i}\left(\delta_{\alpha,\beta} - \frac{Q_\alpha\,Q_\beta}{Q^2}\right) S_{\beta,j}(t)$$

Selects correlaton between spin components perpendicular to $\vec{Q}$





Ferromagnet

Neutron cross section

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\,(\gamma r_o)^2\ f(Q)\ f^*(Q)\ e^{-2W}\ \int_{-\infty}^{+\infty} dt\ \langle \vec{S}_{\perp,Q}\ \vec{S}_{\perp,-Q}(t)\ \rangle\ e^{-i\omega t}$$

From linear spin wave theory :

$$\begin{aligned}
\langle S^x_k\ S^x_{-k}(t)\rangle &= \frac{S}{2}\left(n_B(\omega_k)e^{+i\omega_k t}\ +\ (1+n_B(\omega_k))e^{-i\omega_k t}\right)\\
\langle S^y_k\ S^y_{-k}(t)\rangle &= \frac{S}{2}\left(n_B(\omega_k)e^{+i\omega_k t}\ +\ (1+n_B(\omega_k))e^{-i\omega_k t}\right)\\
\langle S^z_k\ S^z_{-k}(t)\rangle &= \langle S\rangle^2
\end{aligned}$$

$$\int dt\ e^{i\omega t} = \delta(\omega) \qquad \int dt\ e^{i(\omega-\omega_k)t} = \delta(\omega-\omega_k)$$

Ferromagnet

Dynamical structure factor
(Form factor, Debye, geometry)

Detailed balance

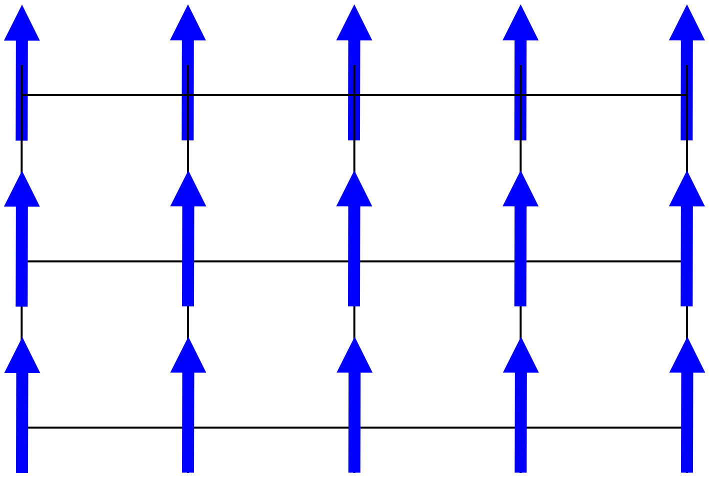
$$\mathcal{S}(Q,\omega) = S\left(1+\frac{Q^z\,Q^z}{Q^2}\right)\sum_{q,\tau}\{n_B(\omega_k)\delta(\omega+\omega_k)\,\delta_{Q,-k+\tau} + (1+n_B(\omega_k))\delta(\omega-\omega_k)\,\delta_{Q,k+\tau}\}$$
$$+\left(1-\frac{Q^z\,Q^z}{Q^2}\right)\langle S\rangle^2\,\delta(\omega)\sum_{\tau}\delta_{Q,\tau}$$

Dirac functions along
the dispersion

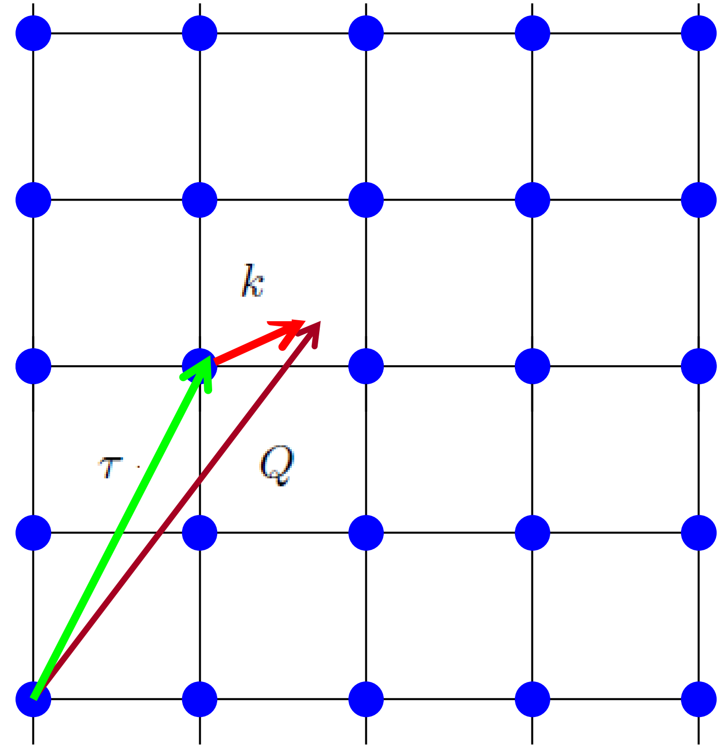
Ferromagnet

$$Q = \tau + k$$

Real space



Reciprocal space

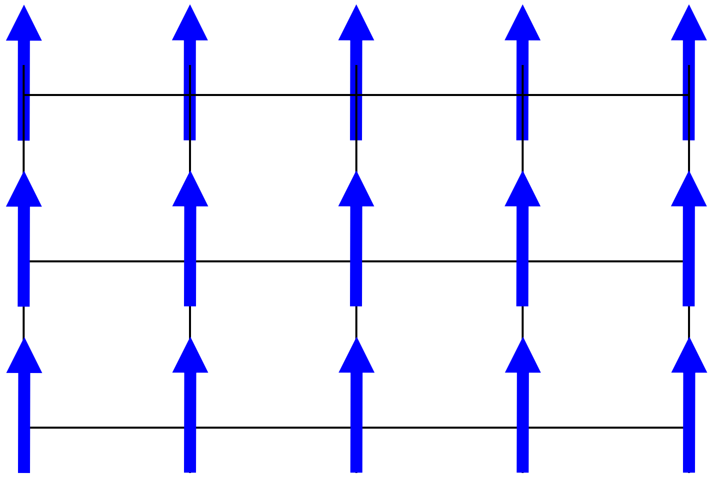


τ = Bragg peak position
k = wavevector in the first Brillouin zone

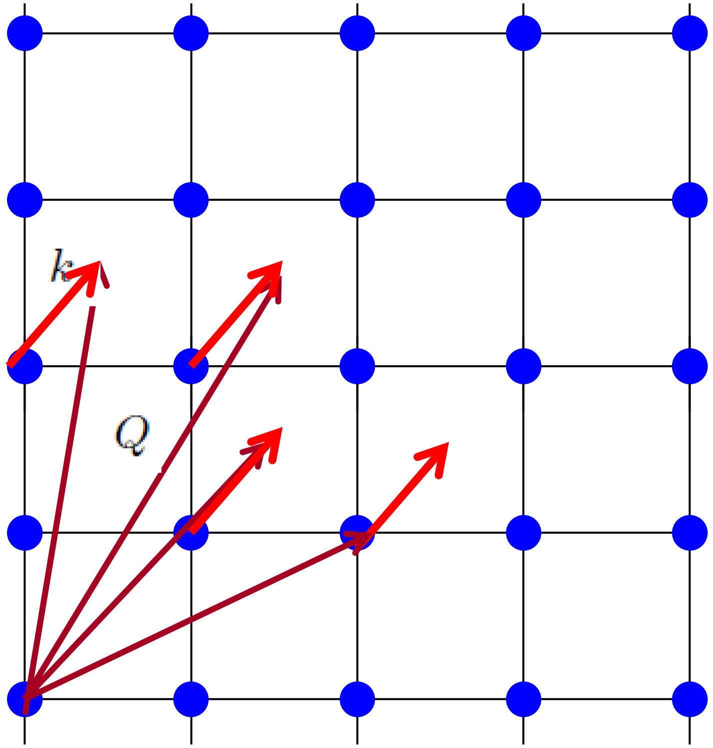
Ferromagnet

$$Q = \tau + k$$

Real space



Reciprocal space

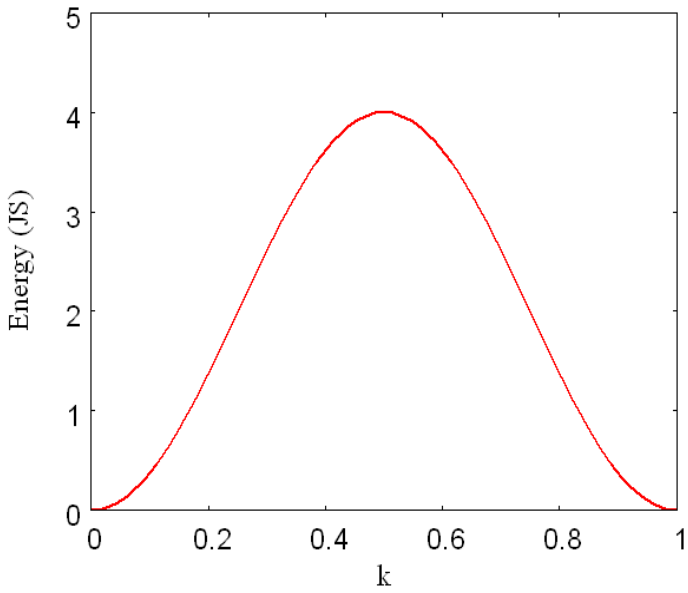


!

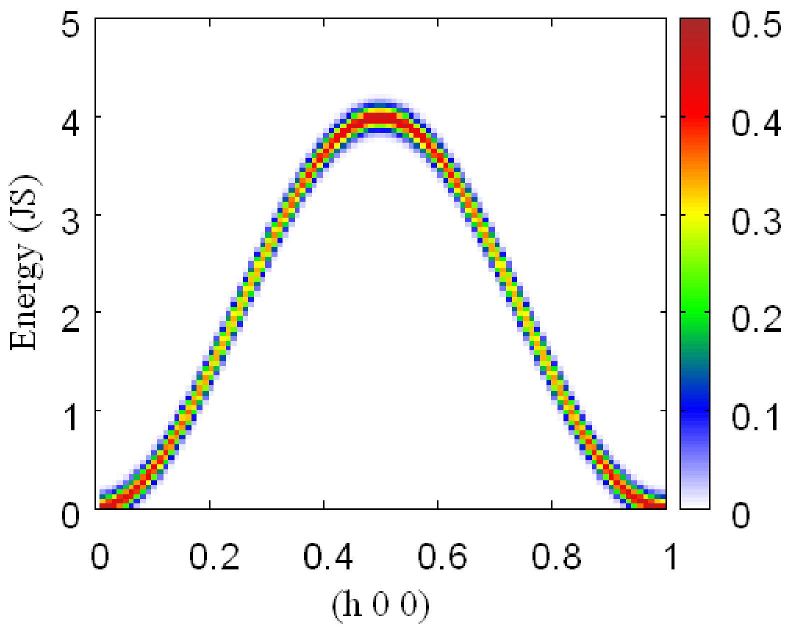
These points are equivalent regarding the dispersion relation
But have different neutron cross sections

Ferromagnet

Dispersion relation



Intensity superimposed on the dispersion relation, depends on the neutron cross section



Ferromagnet

Dynamical structure factor
(Form factor, Debye, geometry)

Dirac functions along
the dispersion

$$\mathcal{S}(Q,\omega) \;=\; S\left(1+\frac{Q^z\,Q^z}{Q^2}\right)\sum_{q,\tau}\{n_B(\omega_k)\delta(\omega+\omega_k)\;\delta_{Q,-k+\tau}\;+\;(1+n_B(\omega_k))\delta(\omega-\omega_k)\;\delta_{Q,k+\tau}\}$$
$$+\left(1-\frac{Q^z\,Q^z}{Q^2}\right)\langle S\rangle^2\;\delta(\omega)\sum_{\tau}\;\delta_{Q,\tau}$$

Static response (Bragg peak)
With a different geometrical factor
(best seen if $Q^z = 0$, in contrats with
the inelastic response)

Detailed balance

General case

Dynamical structure factor (inlcudes Form factor, Debye, geometry)

Dirac functions : intensity different from zero along the dispersion

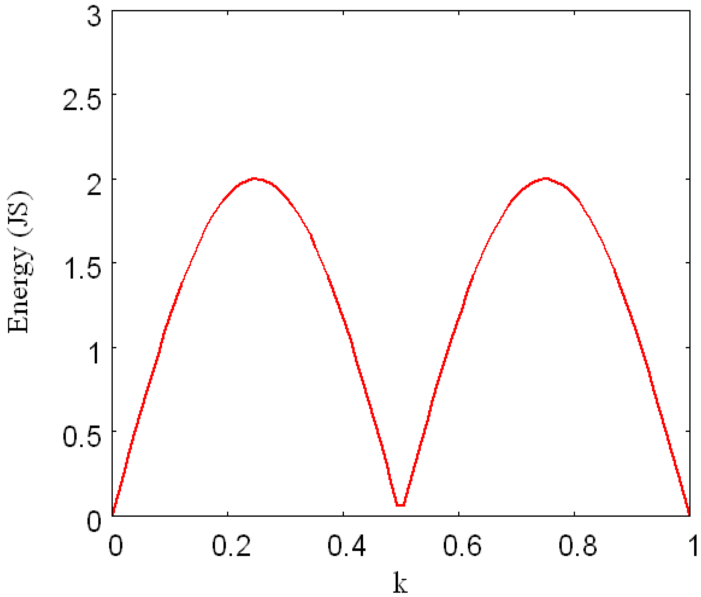
$$\mathcal{S}(Q,\omega) \sim F(Q) \sum_s \sum_{k,\tau} \{ n_B(\omega_{k,s}) \delta(\omega + \omega_{k,s}) \ \delta_{Q,-k+\tau} + (1 + n_B(\omega_{k,s})) \delta(\omega - \omega_{k,s}) \ \delta_{Q,k+\tau} \}$$

Sum over all spin wave modes

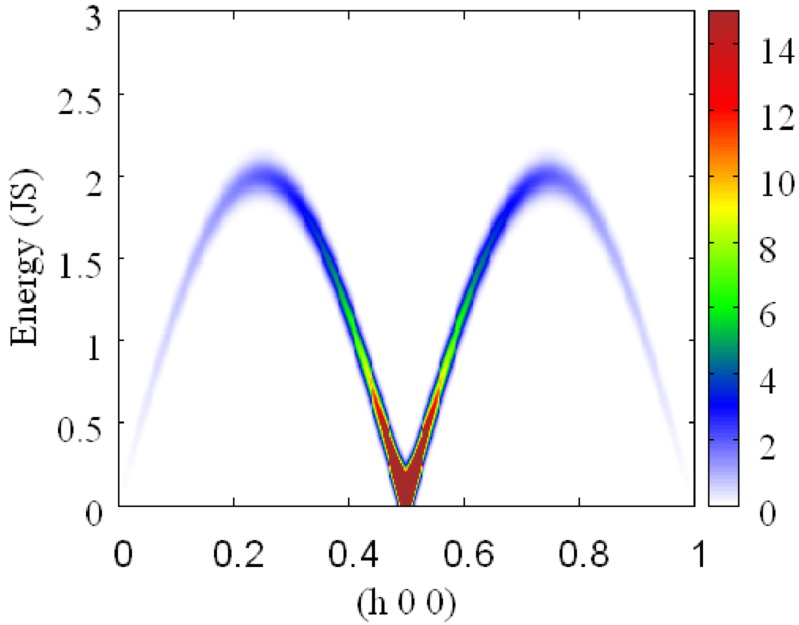
Detailed balance

Antiferromagnet

Dispersion relation



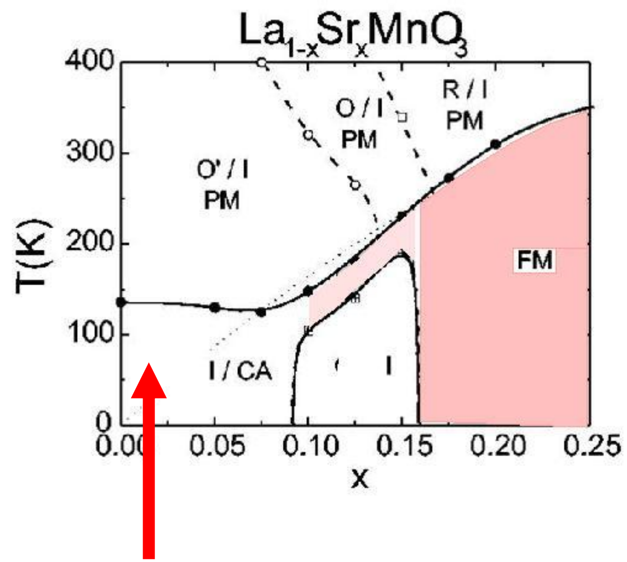
Intensity superimposed on the dispersion relation, depends on the neutron cross section

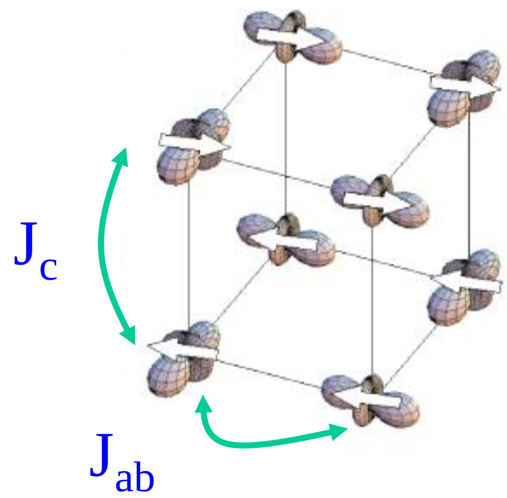


Example 1

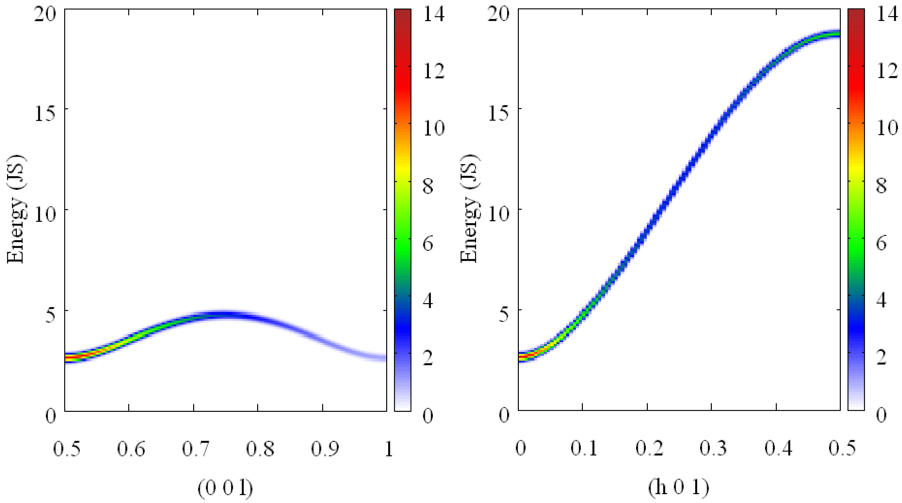
Spin dynamics in $LaSrMnO_3$

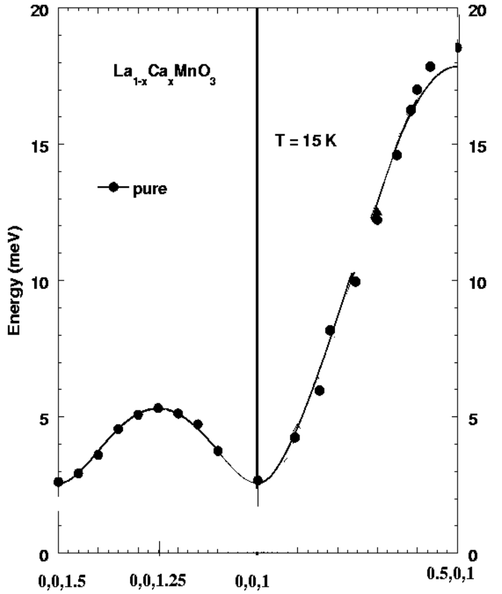
Example : manganites





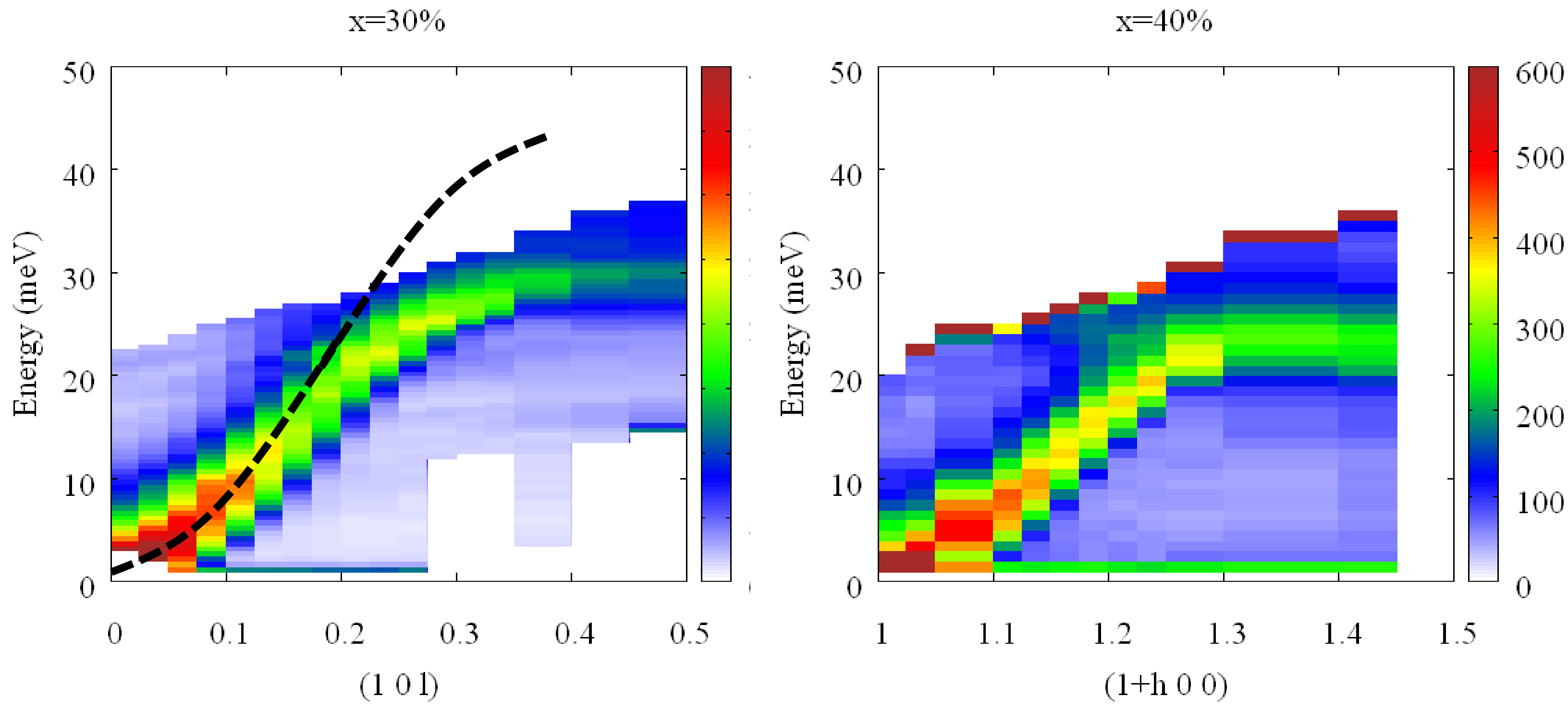
J_c ~ 0.5 meV
J_{ab} ~ -0.8 meV





Example : manganites

In the metallic state : spin wave in a metal ?

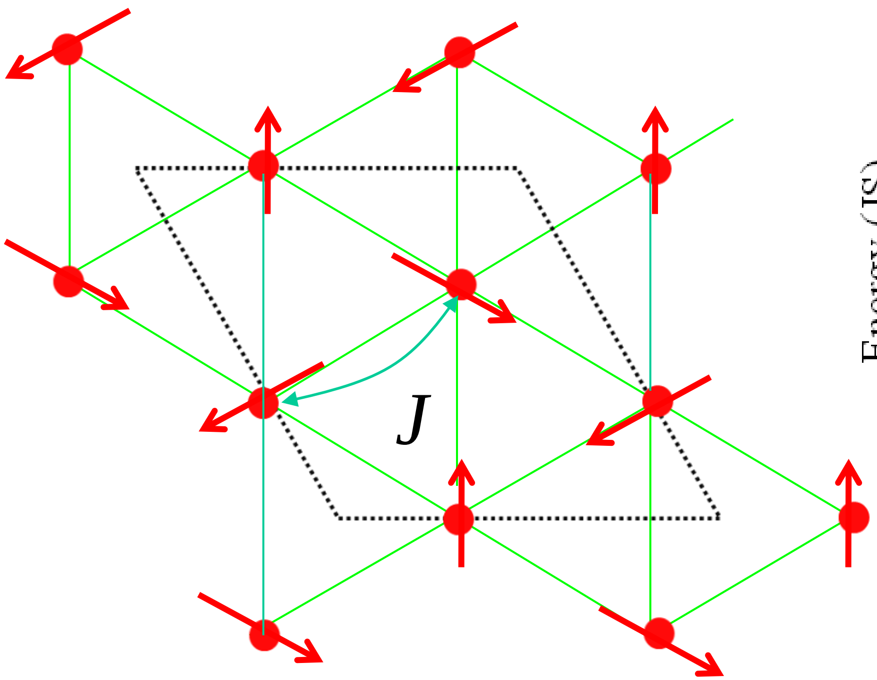


Example 2

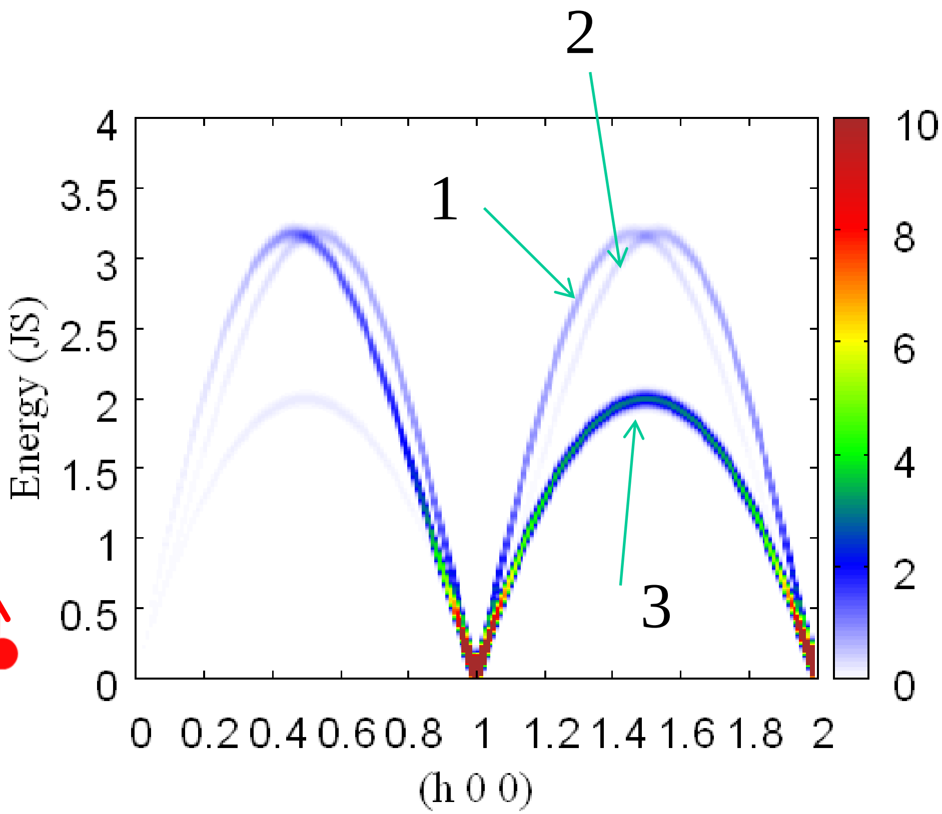
Spin dynamics in triangular lattice

Example : triangular lattice

$$H = \sum_{m,n,i,j} \vec{S}_{m,i} J_{m,i,n,j} \vec{S}_{n,j}$$



120° Néel order



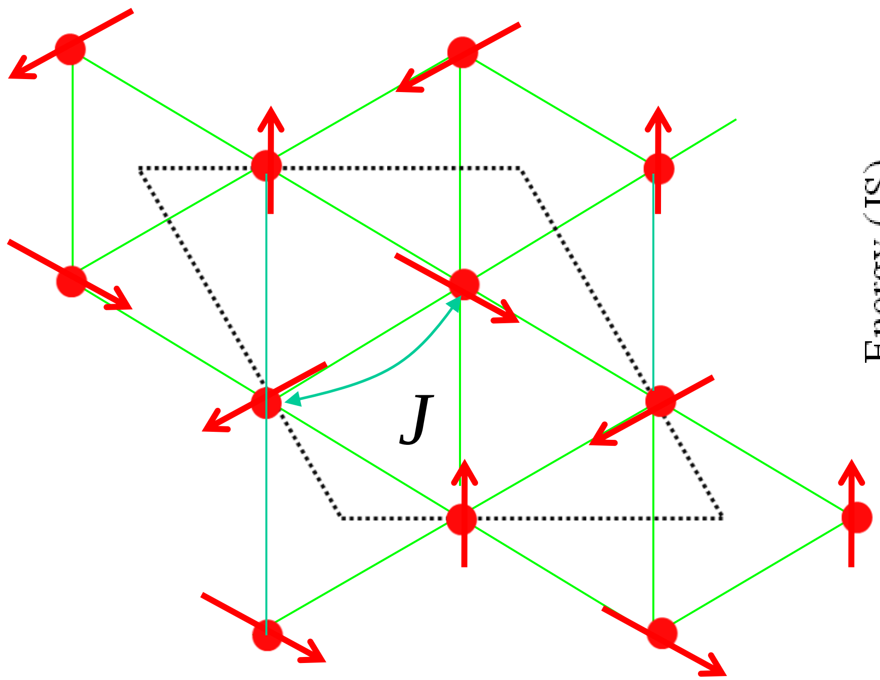
3 spins per unit cell : 3 branches

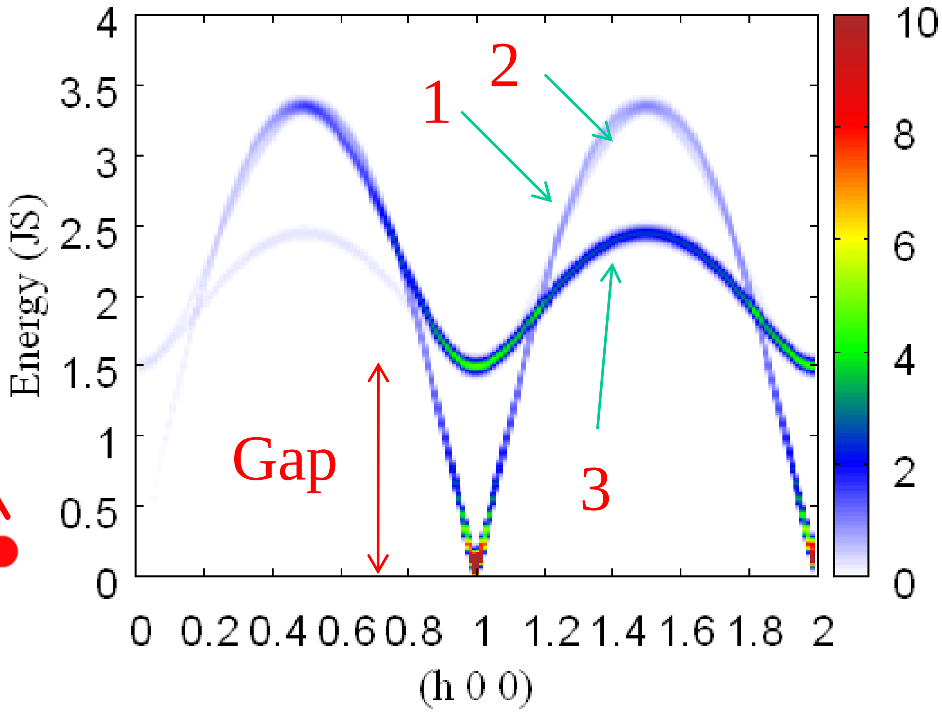
1, 2 : correlations between in plane spin components

3 : correlations between out of plane spin components

Example : triangular lattice

$$H = \sum_{m,n,i,j} \vec{S}_{m,i} J_{m,i,n,j} \vec{S}_{n,j} + \sum_{m,i} D_i \left(\vec{S}_{m,i}.\vec{n}_i \right)^2$$



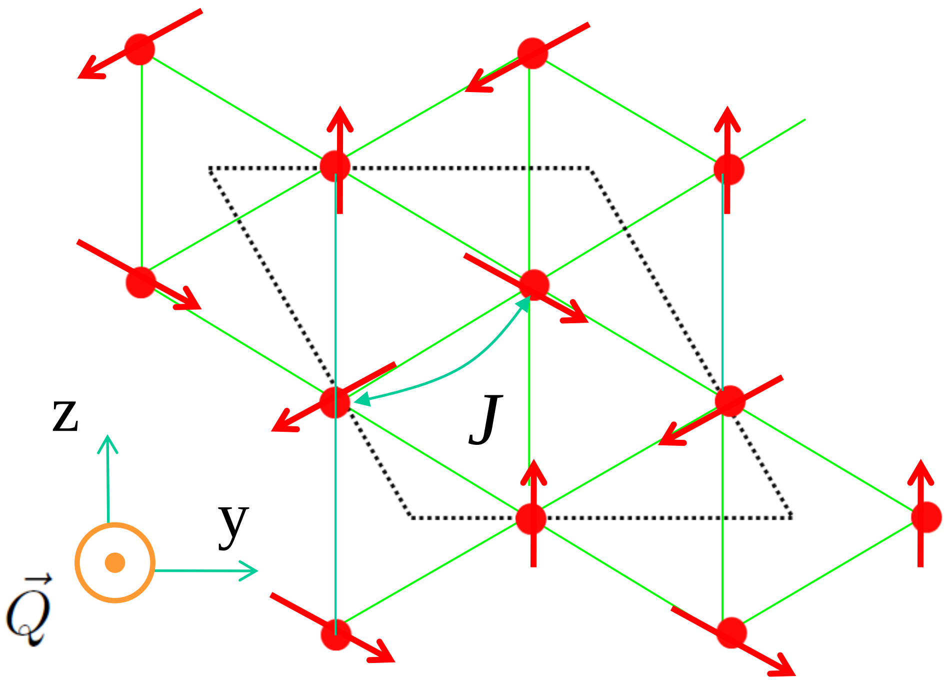


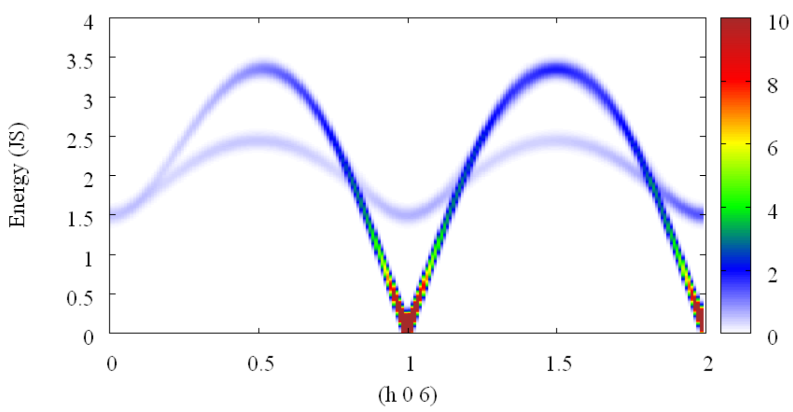
3 spins per unit cell : 3 branches

1, 2 : correlations between in plane spin components

3 : correlations between out of plane spin components

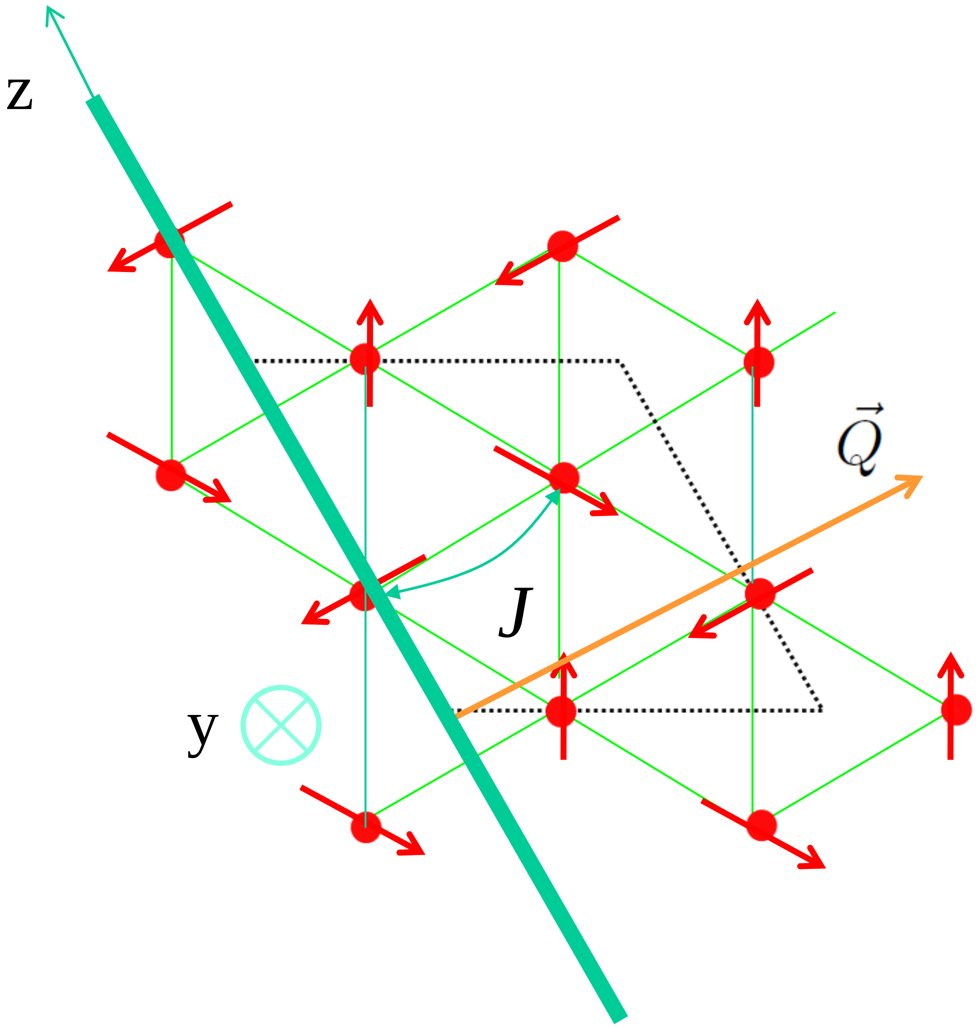
Example : triangular lattice

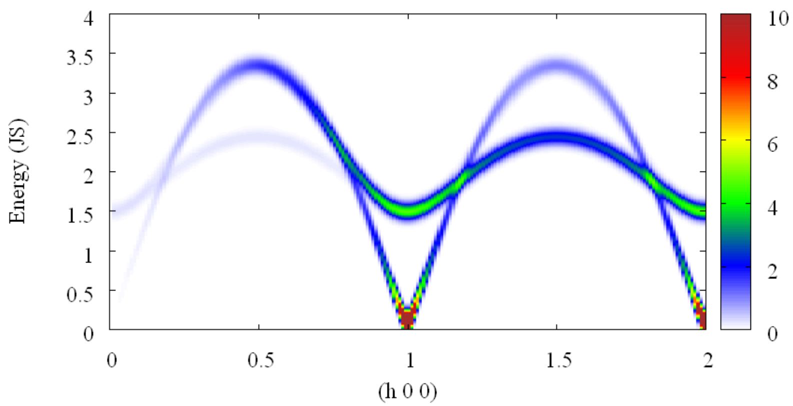




Q perpendicular to the hexagonal (yz) plane allows observing the branches corresponding to correlations between in plane spin components

Example : triangular lattice

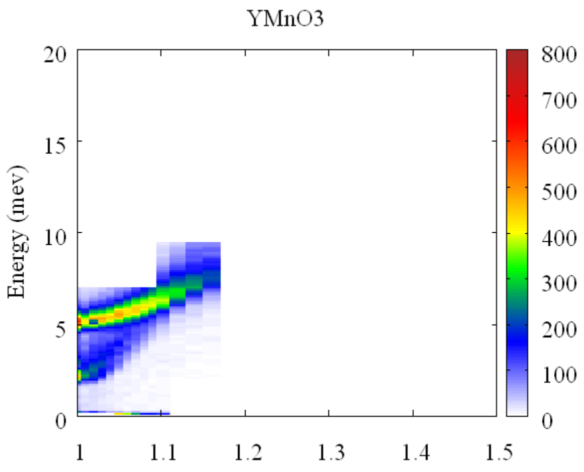


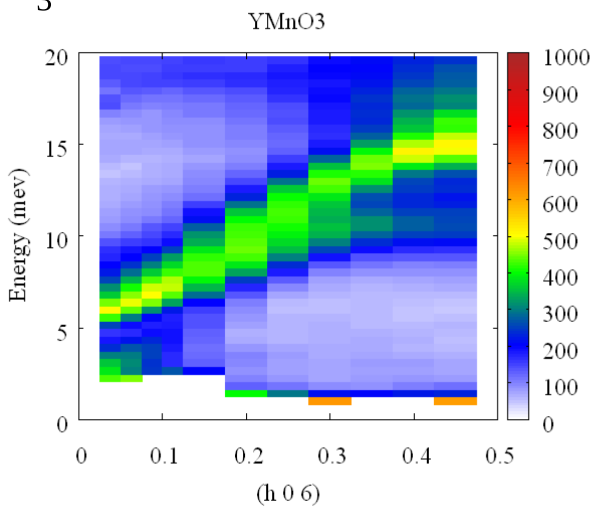


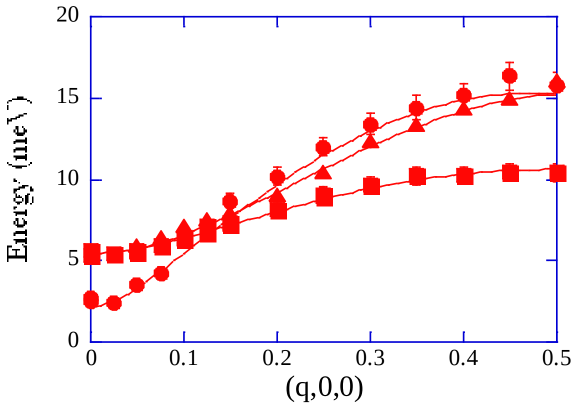
Q in the (yz) hexagonal case, restores intensity on the branch corresponding to correlations between in plane spin components

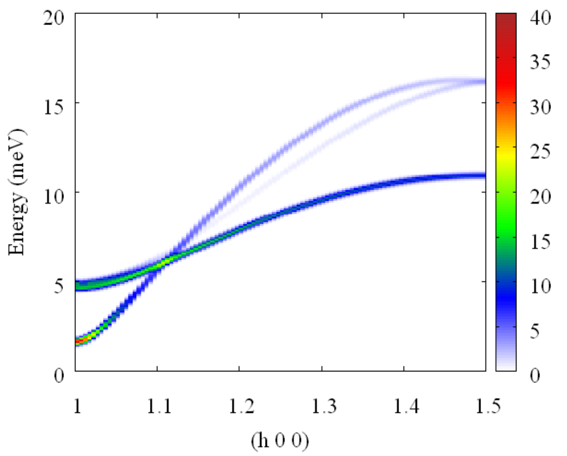
Example : triangular lattice

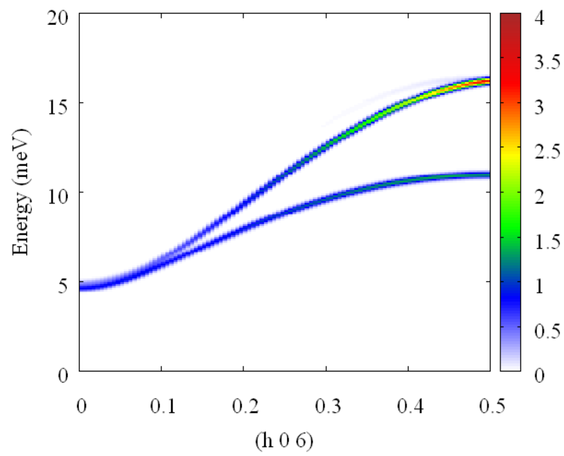
$YMnO_3$









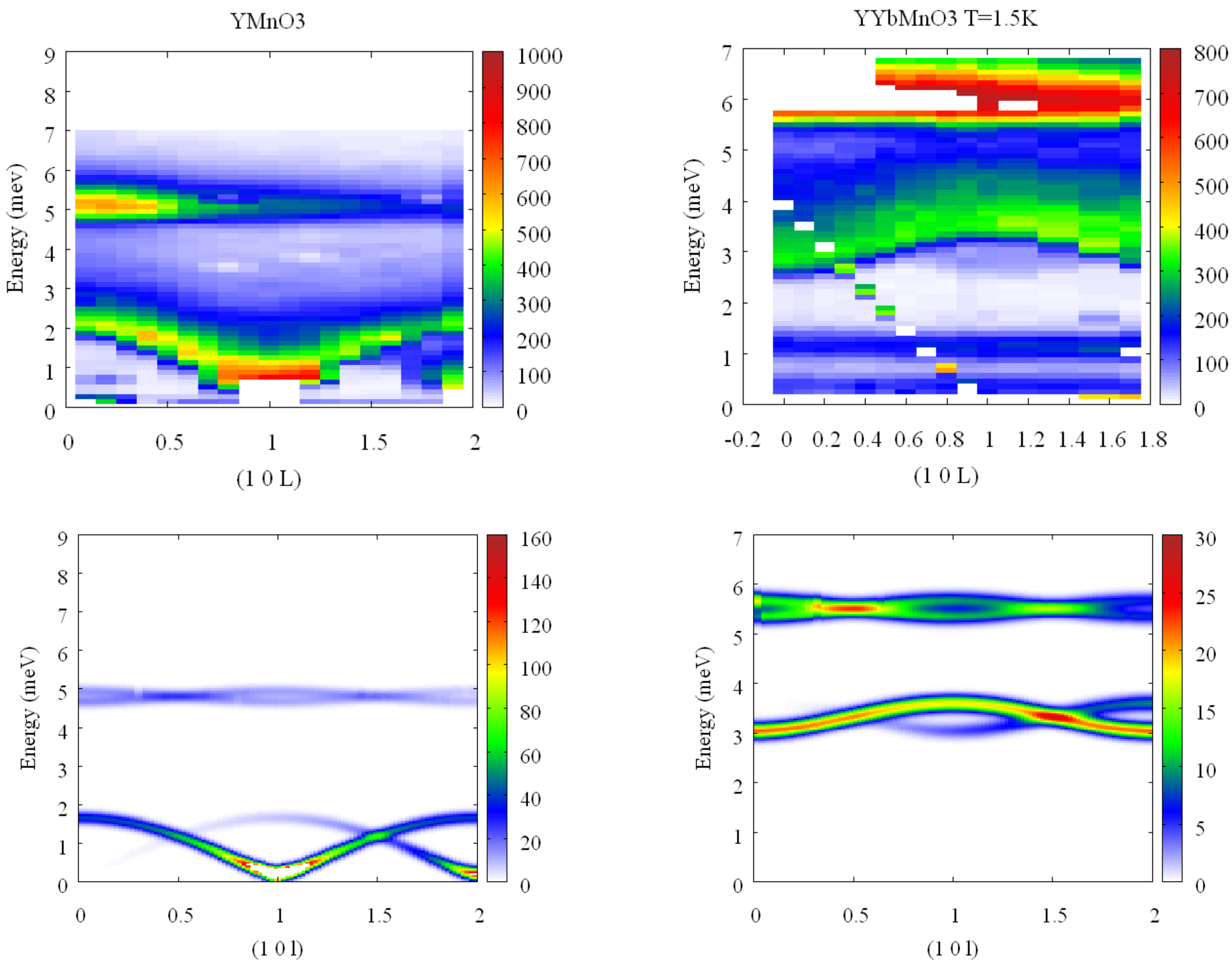


Deduce microscopic parameters from experiments

J = 2.5 meV
D = 0.5 meV

Example : triangular lattice

$YMnO_3$



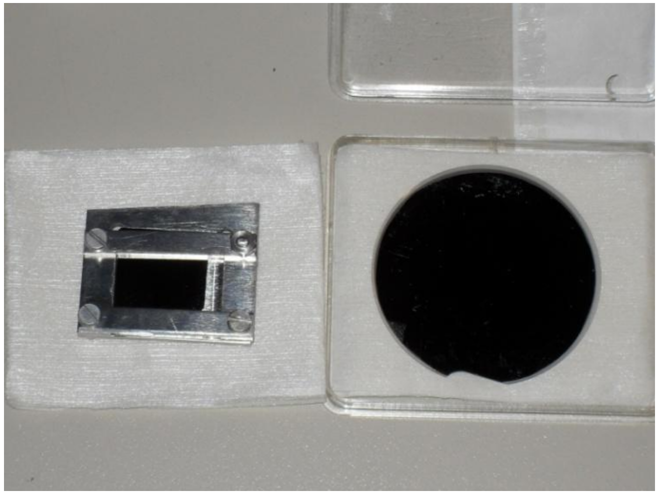
$YbMnO_3$

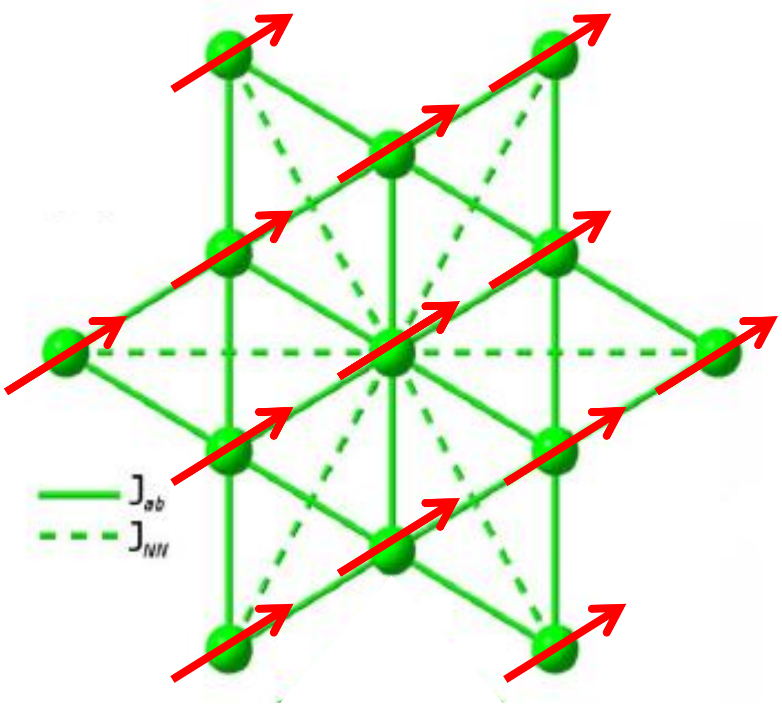
Example 3

Spin dynamics in Dy thin film

Example : Dy thin film

3 μm thick Dy layer (V=~4 mm^3)
Ferromanetic triangular planes
Helicoidal stacking along c

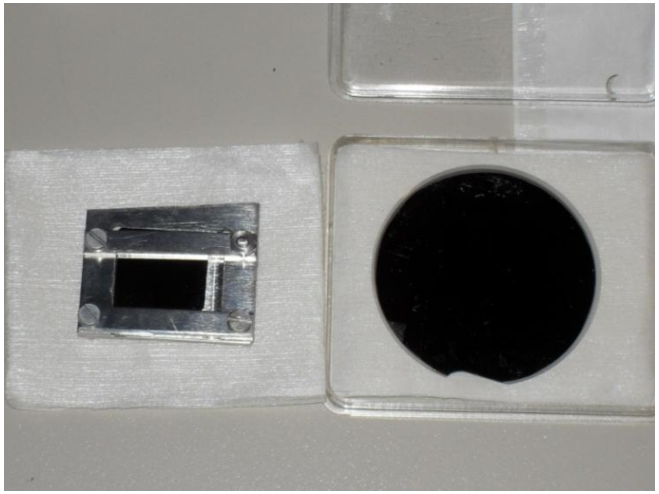


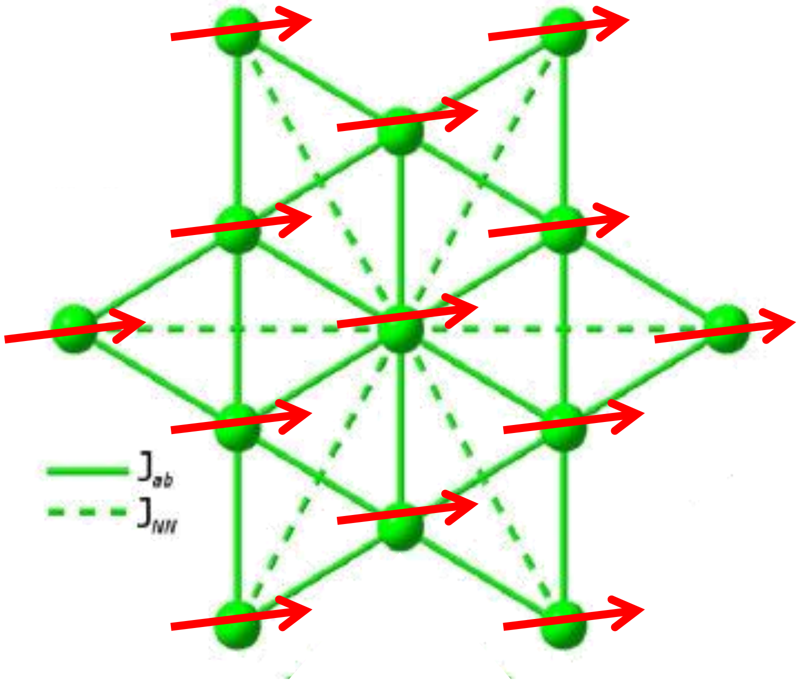


Z=0

Example : Dy thin film

3 μm thick Dy layer (V=~4 mm^3)
Ferromanetic triangular planes
Helicoidal stacking along c

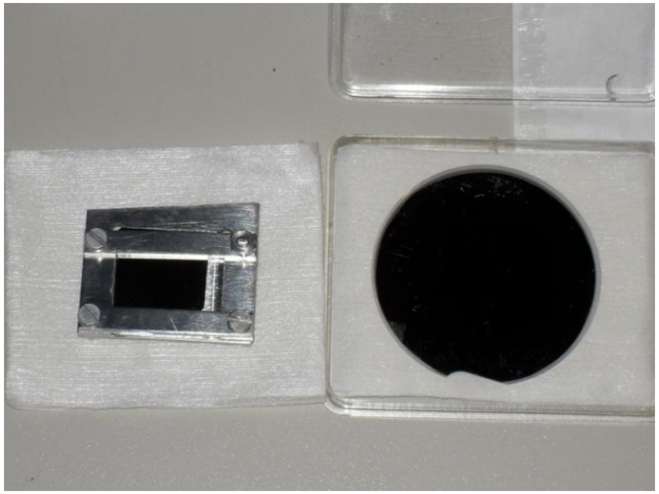


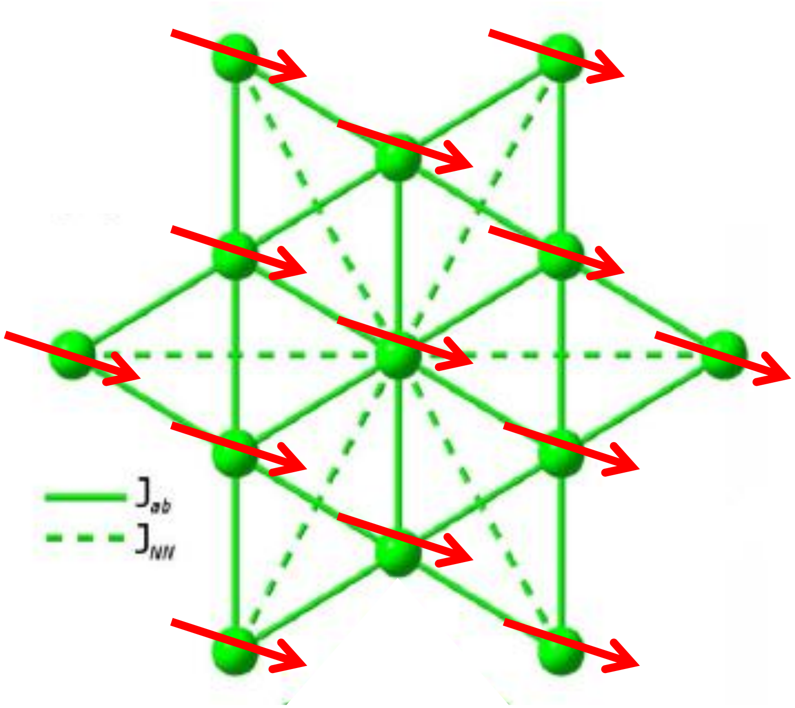


Z=1/2

Example : Dy thin film

3 μm thick Dy layer (V=~4 mm^3)
Ferromanetic triangular planes
Helicoidal stacking along c

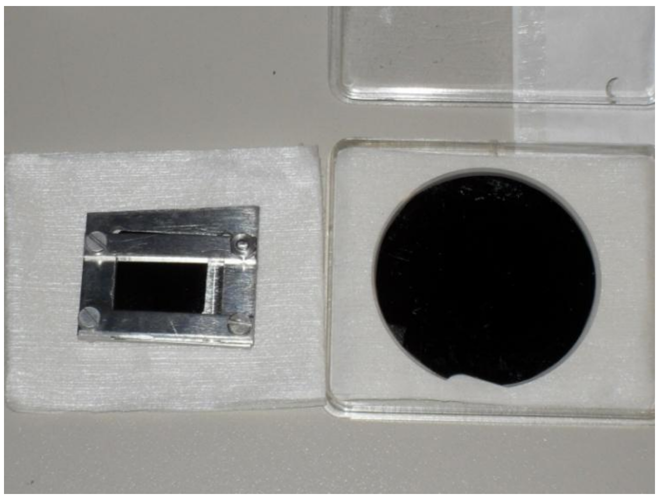


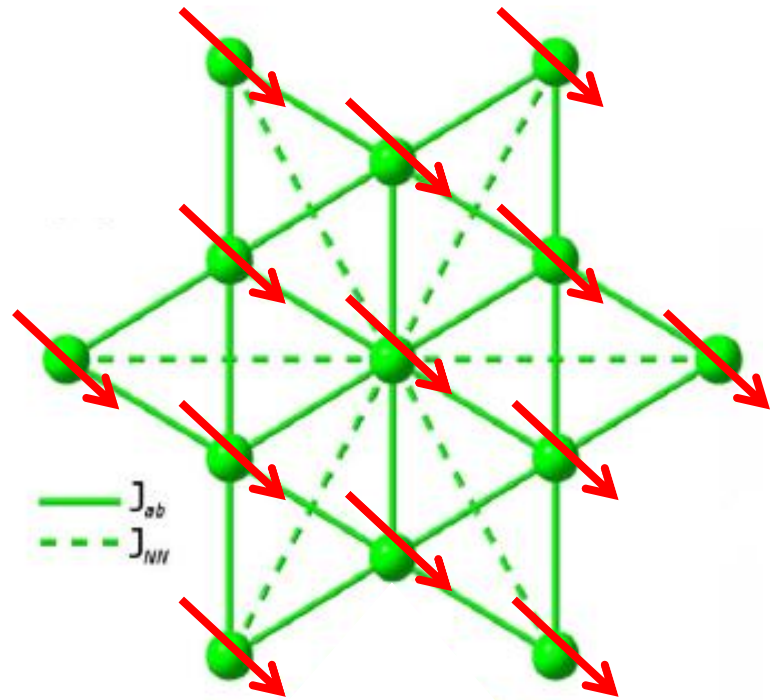


Z=1

Example : Dy thin film

3 μm thick Dy layer (V=~4 mm^3)
Ferromanetic triangular planes
Helicoidal stacking along c

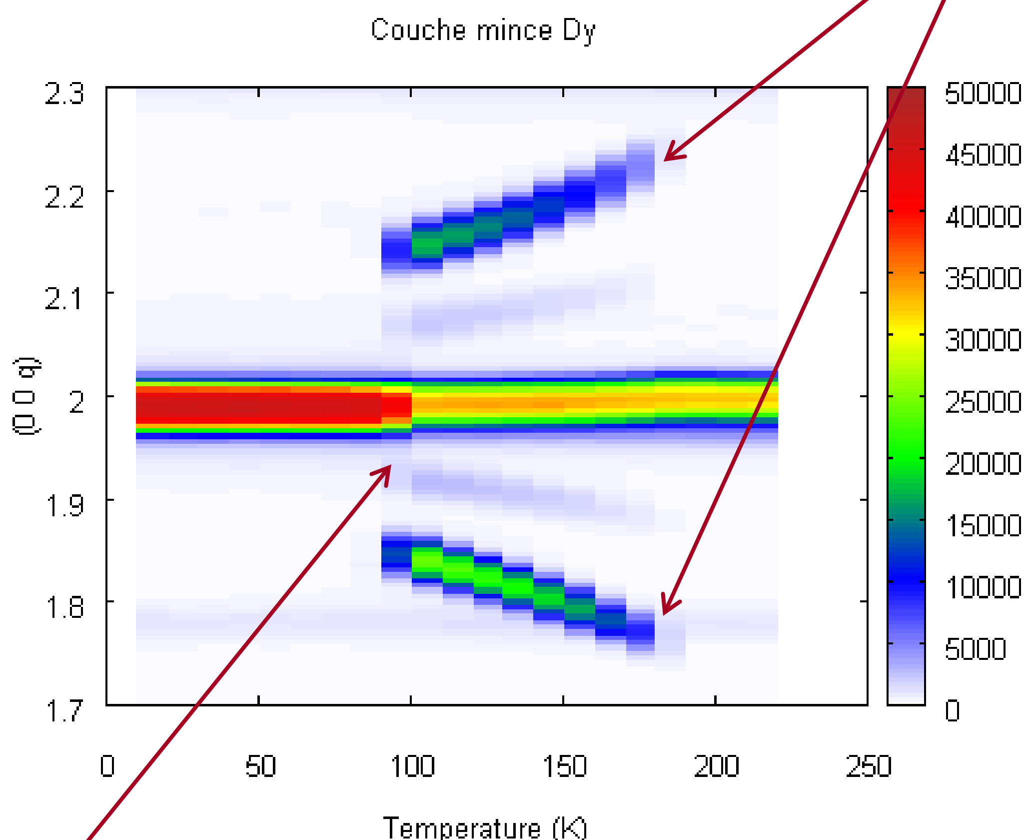




Z=3/2

Example : Dy thin film

Incommensurate magnetic ordering below T_c=170K
Bragg peaks (00 2+/-ε)

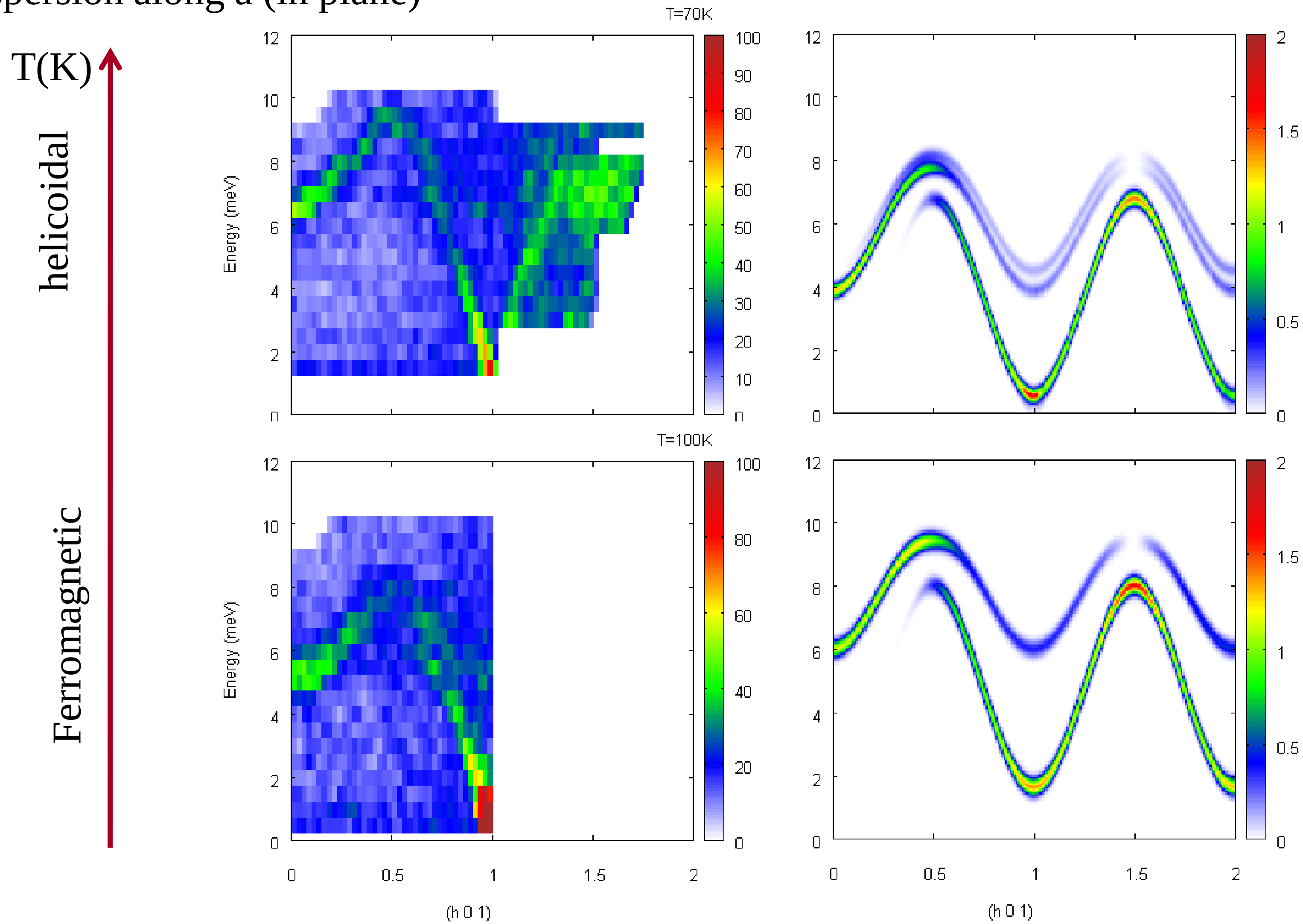


Transition towards a
Ferromagnetic phase below T_N=90K

Mechanism of the transition ?
(Dufour, Dumesnil, *et al*)

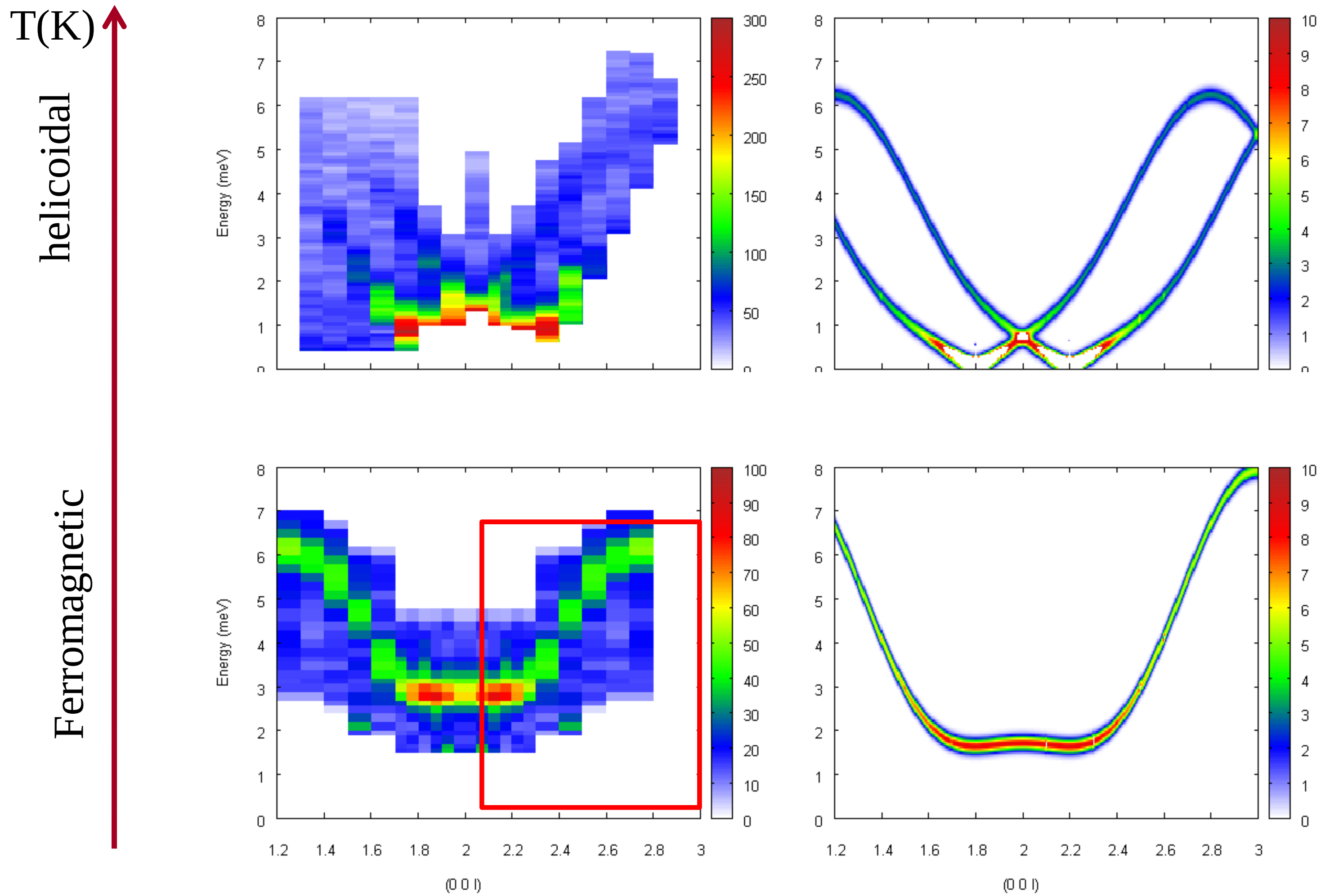
Example : Dy thin film

Dispersion along a (in plane)



Example : Dy thin film

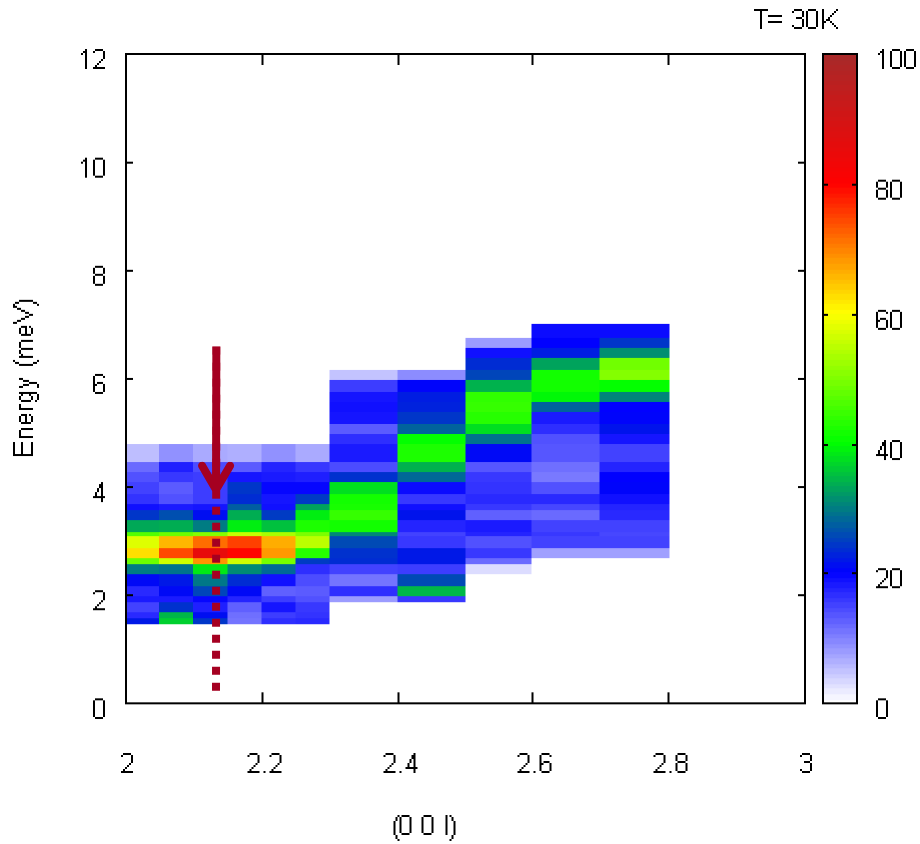
Dispersion along c (perp to the plane)

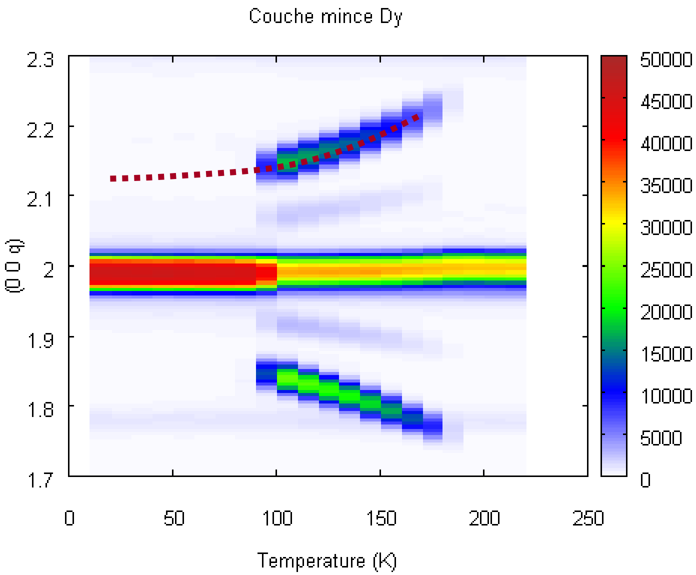


Same exchange parameters but additional anisotropy in the ferromagnetic phase

Example : Dy thin film

Dispersion along c





The minimum of the dispersion is located at incommensurate Q corresponding to the helix : the exchange favors the helicoidal state (***roton-like excitations***) ?
The ferromagnetic phase is likely stabilized by strong anisotropy

Summary

The dispersion of spin waves can be measured in (Q,ω) space by means of inelastic neutron scattering

With the help of a model, it becomes possible to measure physical parameteres as J, D …

Thanks for your attention

Questions ?

References

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[3] T. Oguchi, Phys. Rev 117, 117 (1960)

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