MAGNETIC INELASTIC NEUTRON SCATTERING

Sylvain PETIT
Laboratoire Léon Brillouin
CEA – CNRS – Université Paris-Saclay
91190 Gif sur Yvette, France
sylvain.petit@cea.fr

SwedNess/NNSP 2017 neutron school

Société Française de la Neutronique

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

Articles by

M. Enderle (ILL)

S. Raymond (CEA, Grenoble)

PSI Master School 2017

[1] P.W. Anderson, Phys. Rev. 83, 1260 (1951)

[2] R. Kubo, Phys. Rev. 87, 568 (1952)

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

[4] D.C. Mattis, *Theory of Magnetism I*, Springer Verlag, 1988

[5] R.M. White, *Quantum Theory of Magnetism*, Springer Verlag, 1987

[6] A. Auerbach, *Interacting electrons and Quantum Magnetism*, Springer Verlag, 1994.

Outline

- Introduction
- Cross section

- Localized excitations : Crystal field modes
- « Nearly » localized modes : excitons
- Collective excitations : spin waves

- Quantum magnets (spin ½ and spin 1)
- Metals

Outline

- Introduction
- Cross section

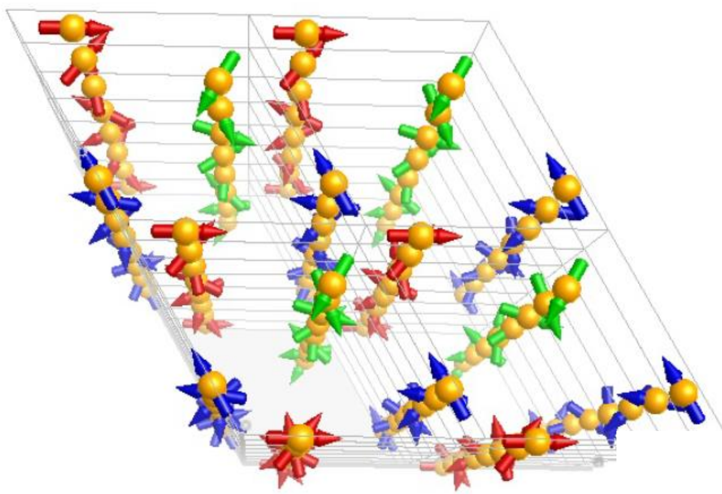
- Localized excitations : Crystal field modes
- « Nearly » localized modes : excitons
- Collective excitations : spin waves

$\Delta S=1$
Collective excitations
Well defined modes

- Quantum magnets (spin ½ and spin 1)
- Metals

$\Delta S=1/2$
Collective excitations
Continuum

Possible emergence of bound states and of well defined modes



Crystalline and magnetic structures

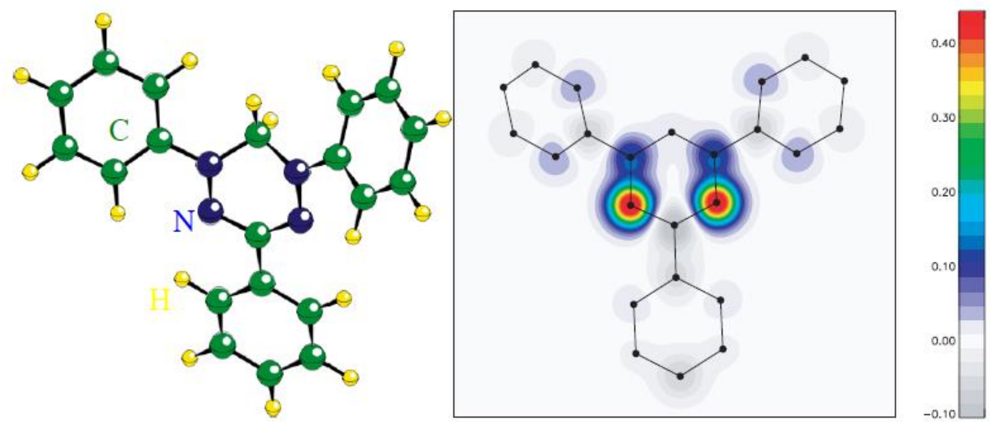


Fig. 4. View of the TPV molecule (left) and experimental magnetization distribution (right) as measured by polarized neutron diffraction.

Why neutrons ?

The structure factor provides information about the structure:

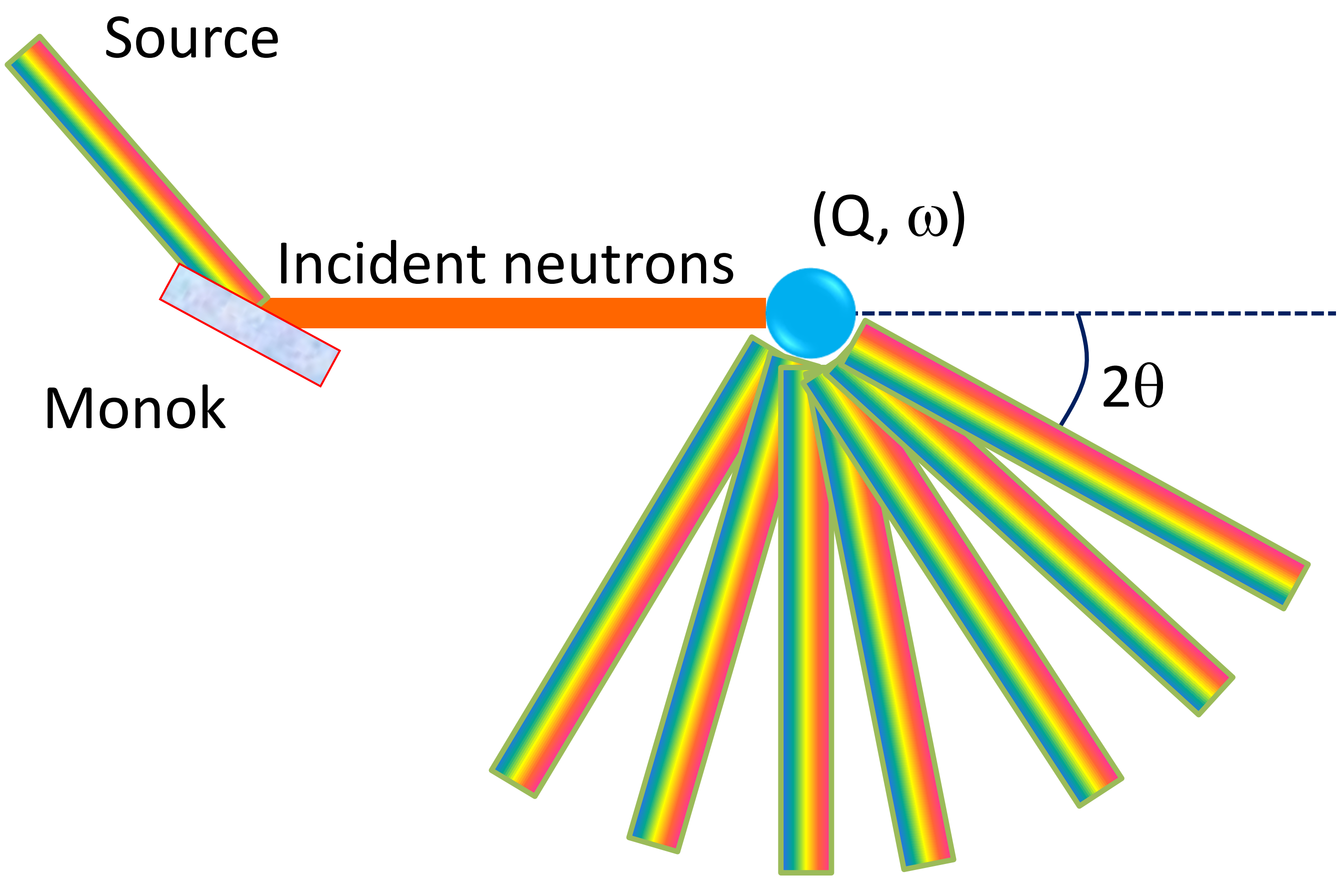
$$F_N(\mathbf{Q}) = \sum_\ell b_\ell \, e^{i\mathbf{Q}\mathbf{r}_\ell} \, e^{-W_\ell}$$

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

But what about the Hamiltonian of the system ?
What about the terms that stabilize the structure ?

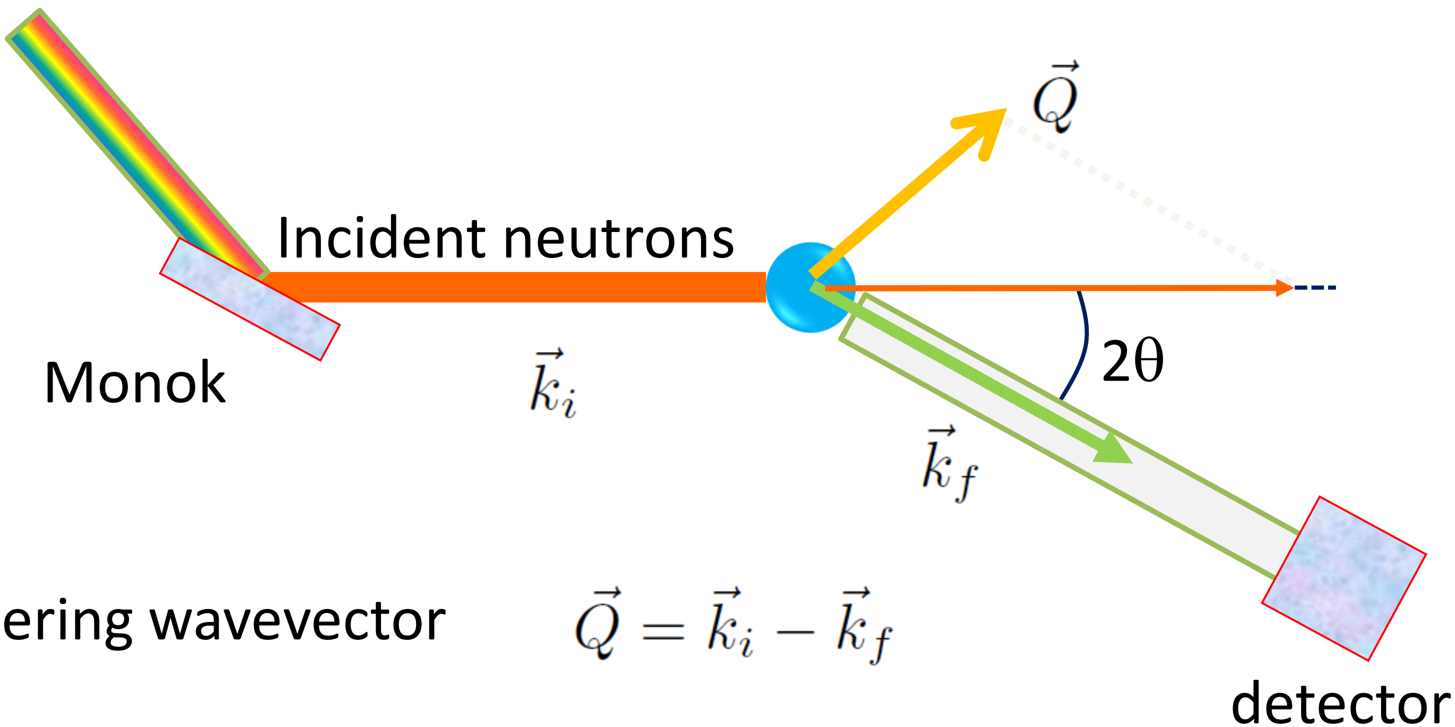
Investigate the excited states:
spectroscopy as a function of Q and ω

Inelastic scattering



For a given θ, the neutrons gain or loose an energy ω and a momemtum Q depending on the scattering process.

Inelastic scattering

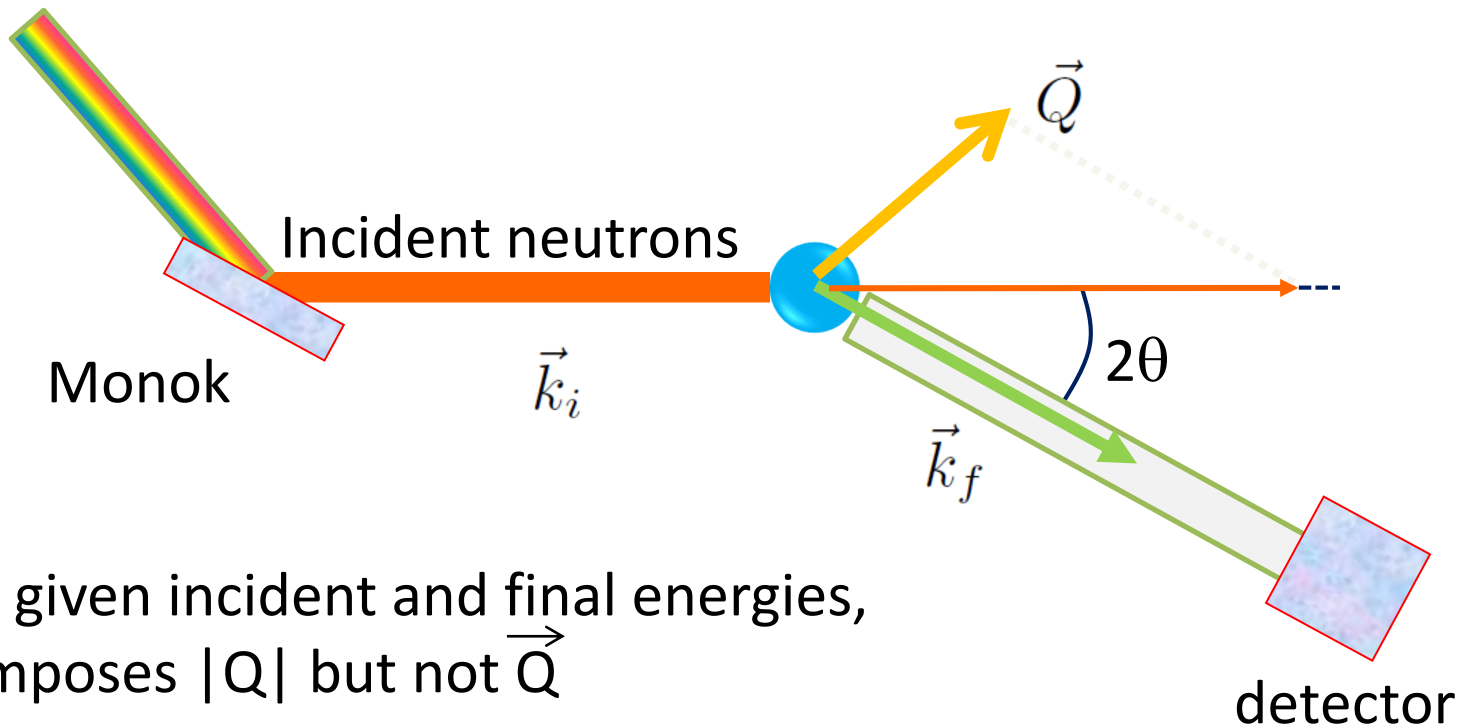


Scattering wavevector $\vec{Q} = \vec{k}_i - \vec{k}_f$

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

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

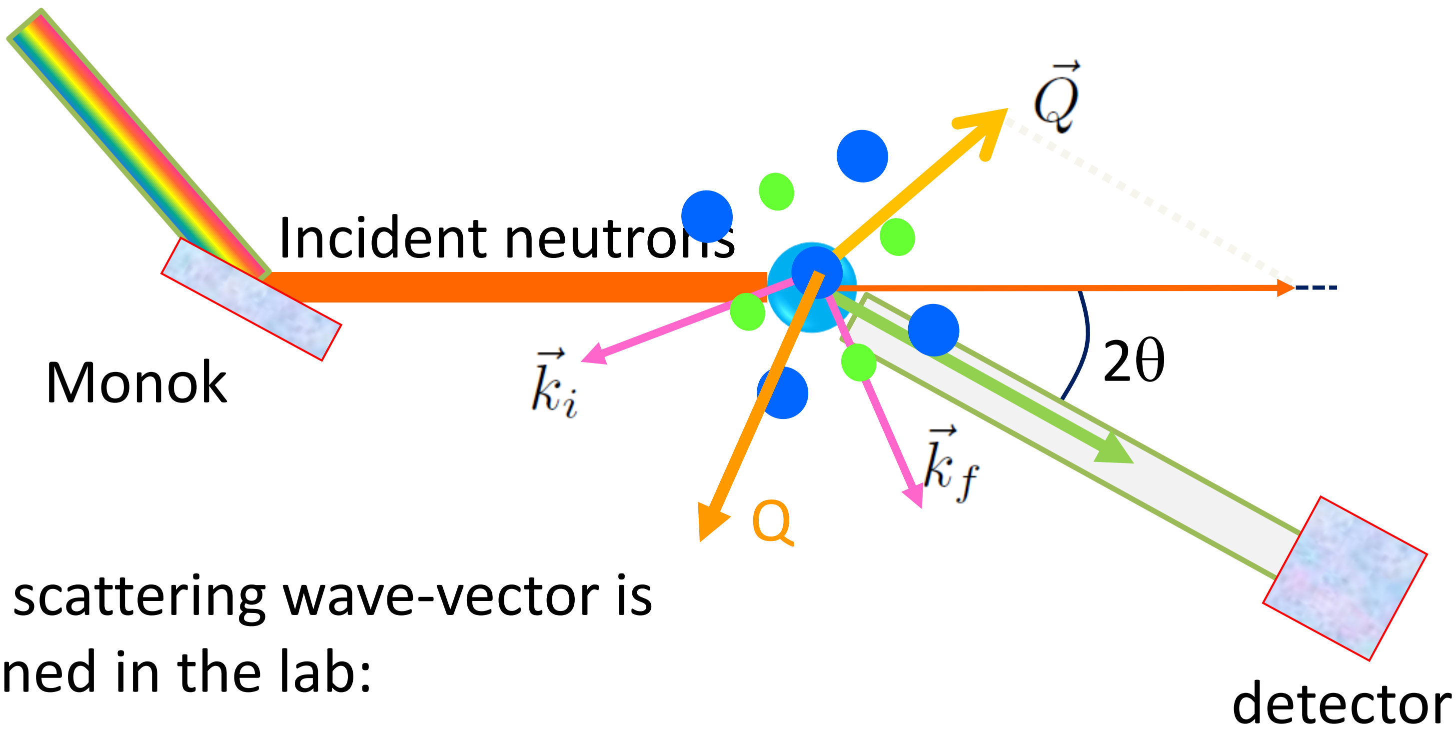
Inelastic scattering



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

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

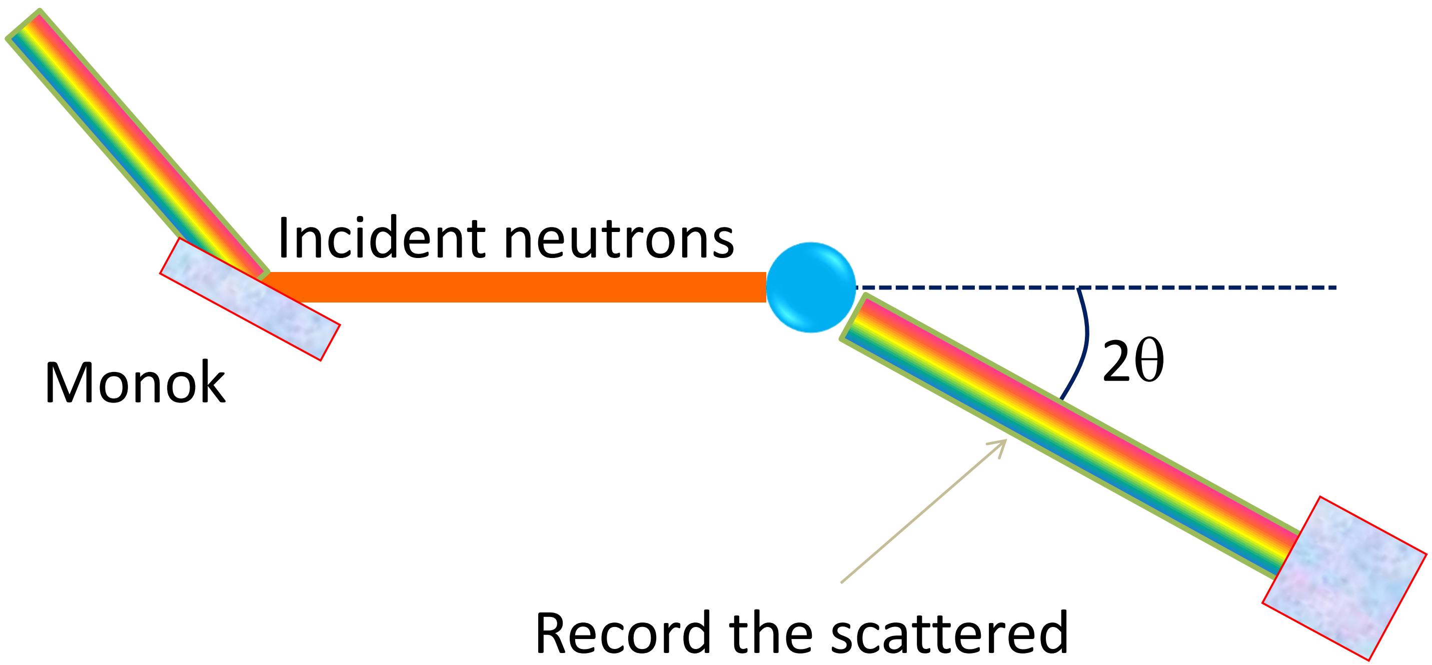
Inelastic scattering



The scattering wave-vector is defined in the lab:

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

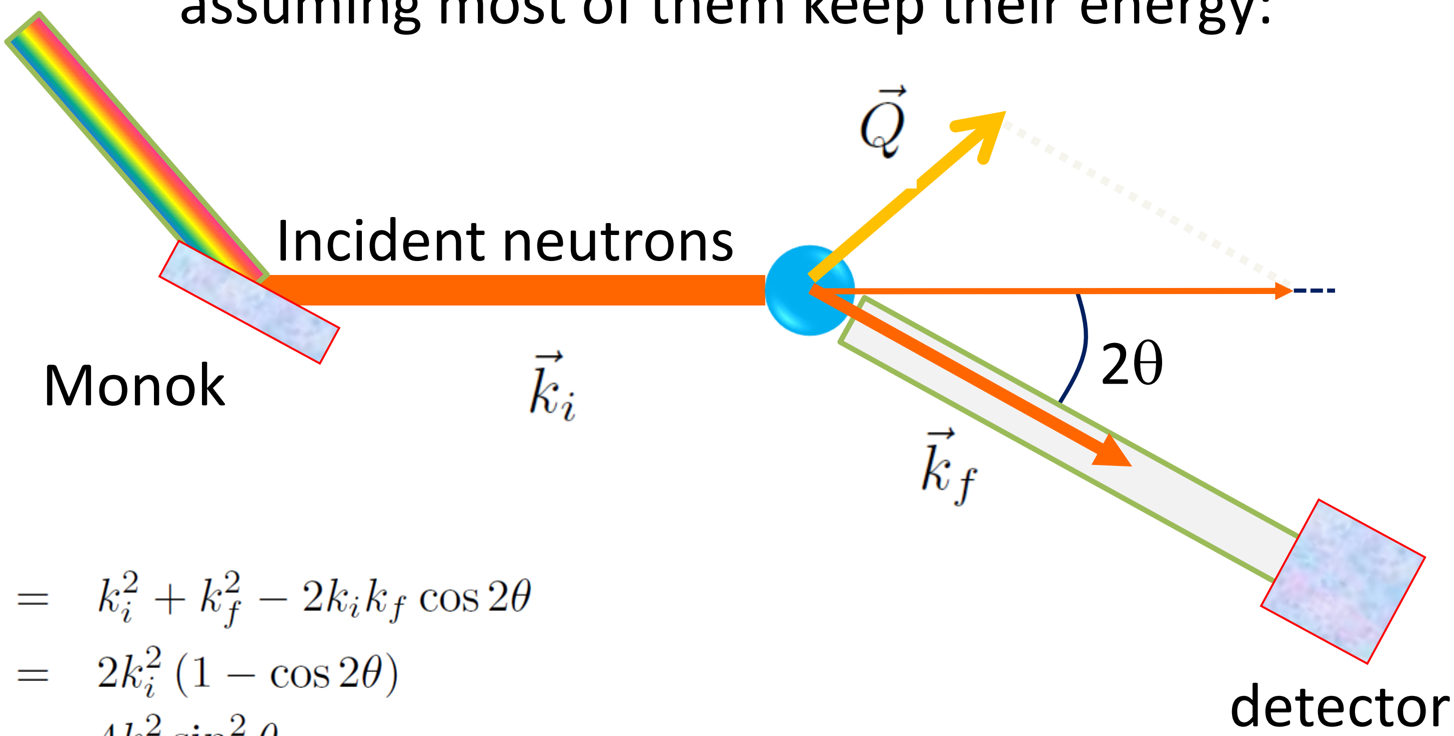
« Elastic » scattering



Record the scattered neutrons whatever their final energy

« Elastic » scattering

assuming most of them keep their energy:

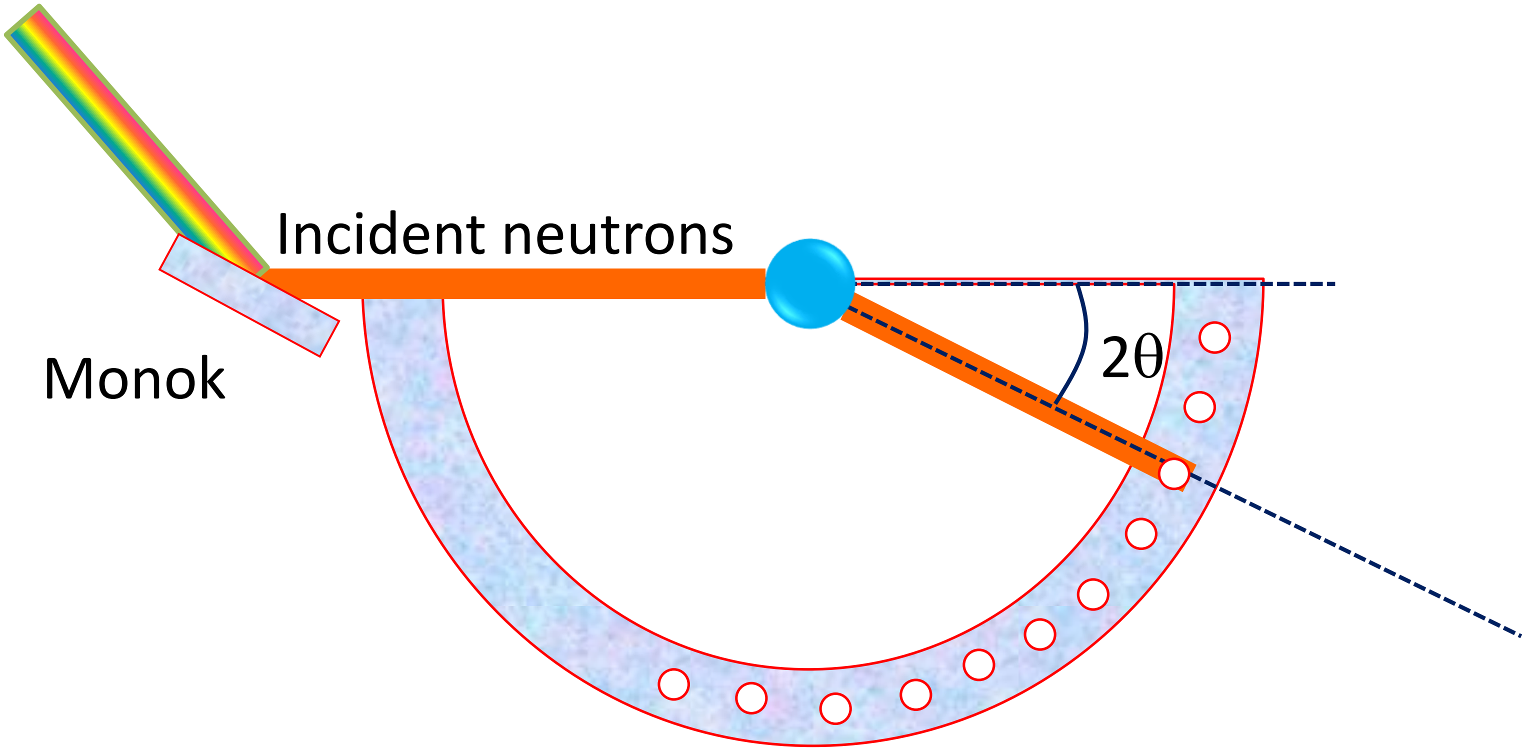


$$
\begin{aligned}
Q^2 &= k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta \\
&= 2k_i^2 (1 - \cos 2\theta) \\
&= 4k_i^2 \sin^2 \theta \\
Q &= 2k_i \sin \theta \\
\frac{2\pi}{d} &= 2\frac{2\pi}{\lambda} \sin \theta
\end{aligned}
$$

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

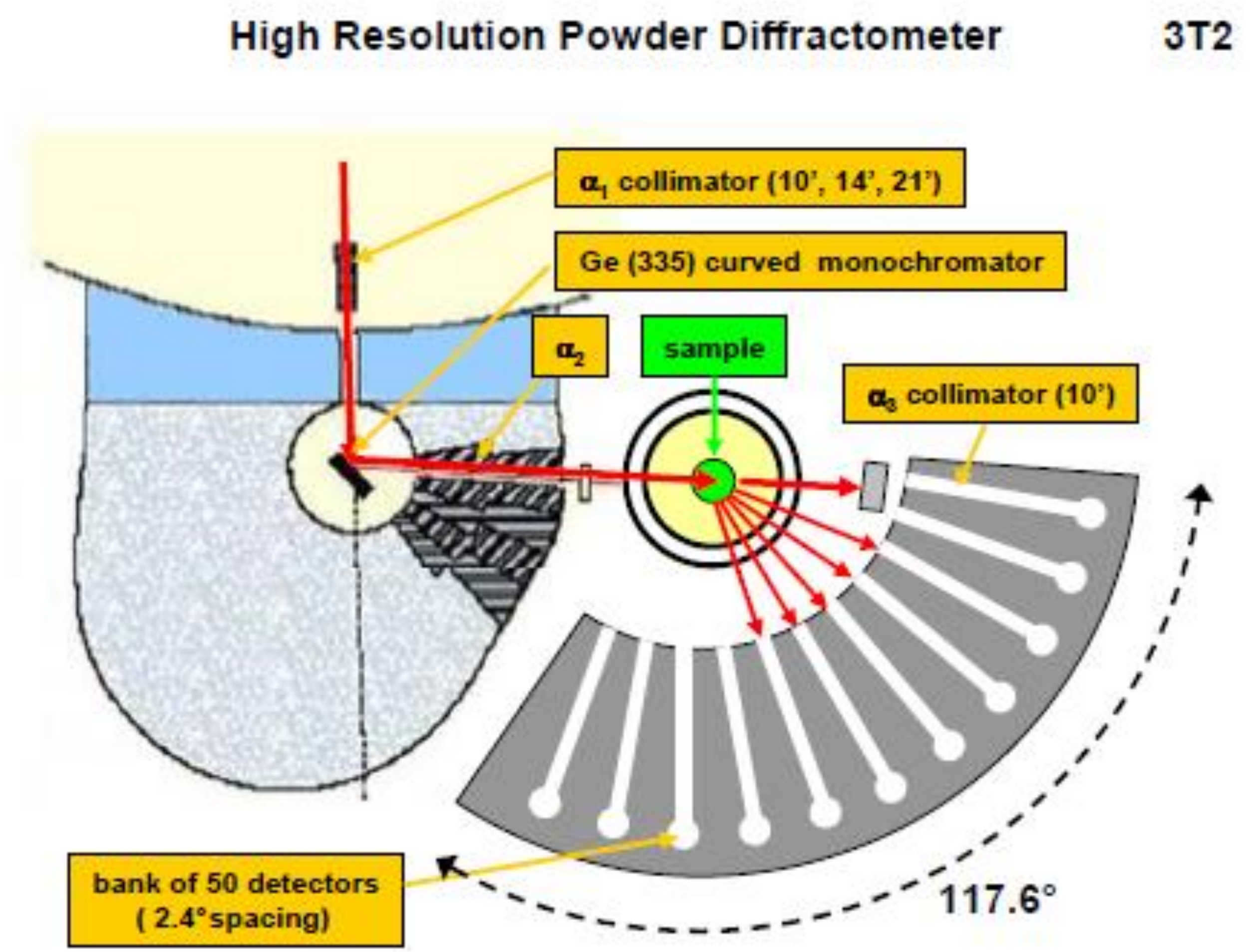
Bragg law

« Elastic » scattering

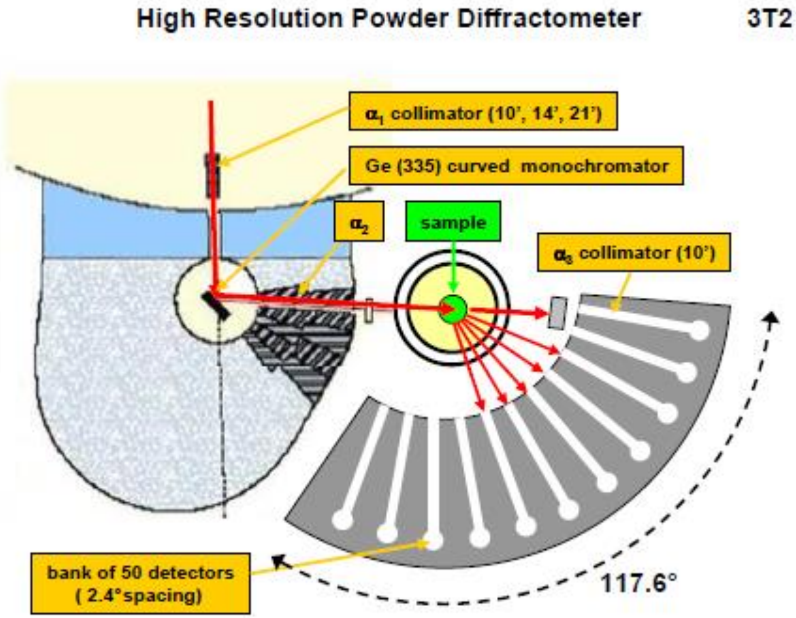


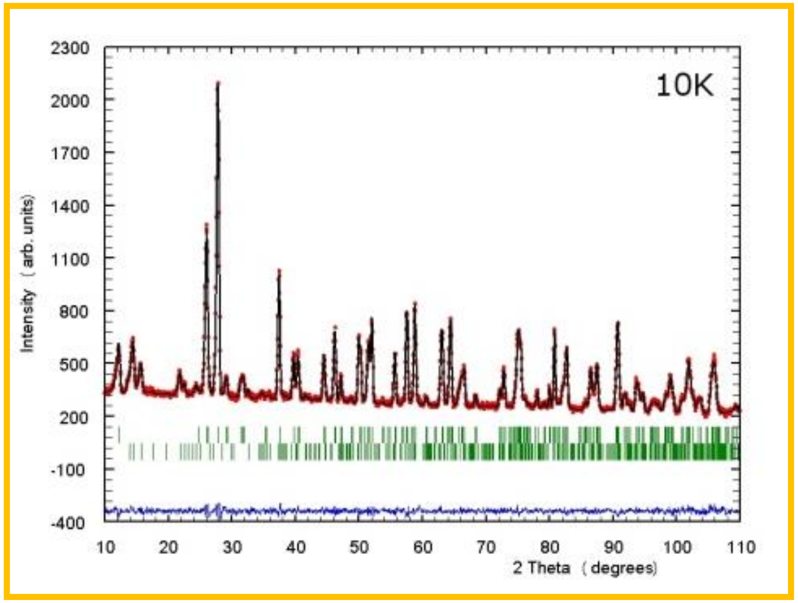
We still have to rotate the sample even if the experiment looks more efficient with a detector bank

« Elastic » scattering

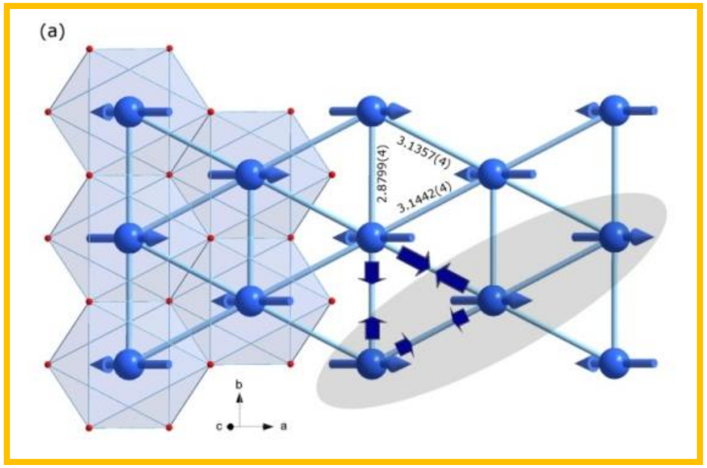


« Elastic » scattering



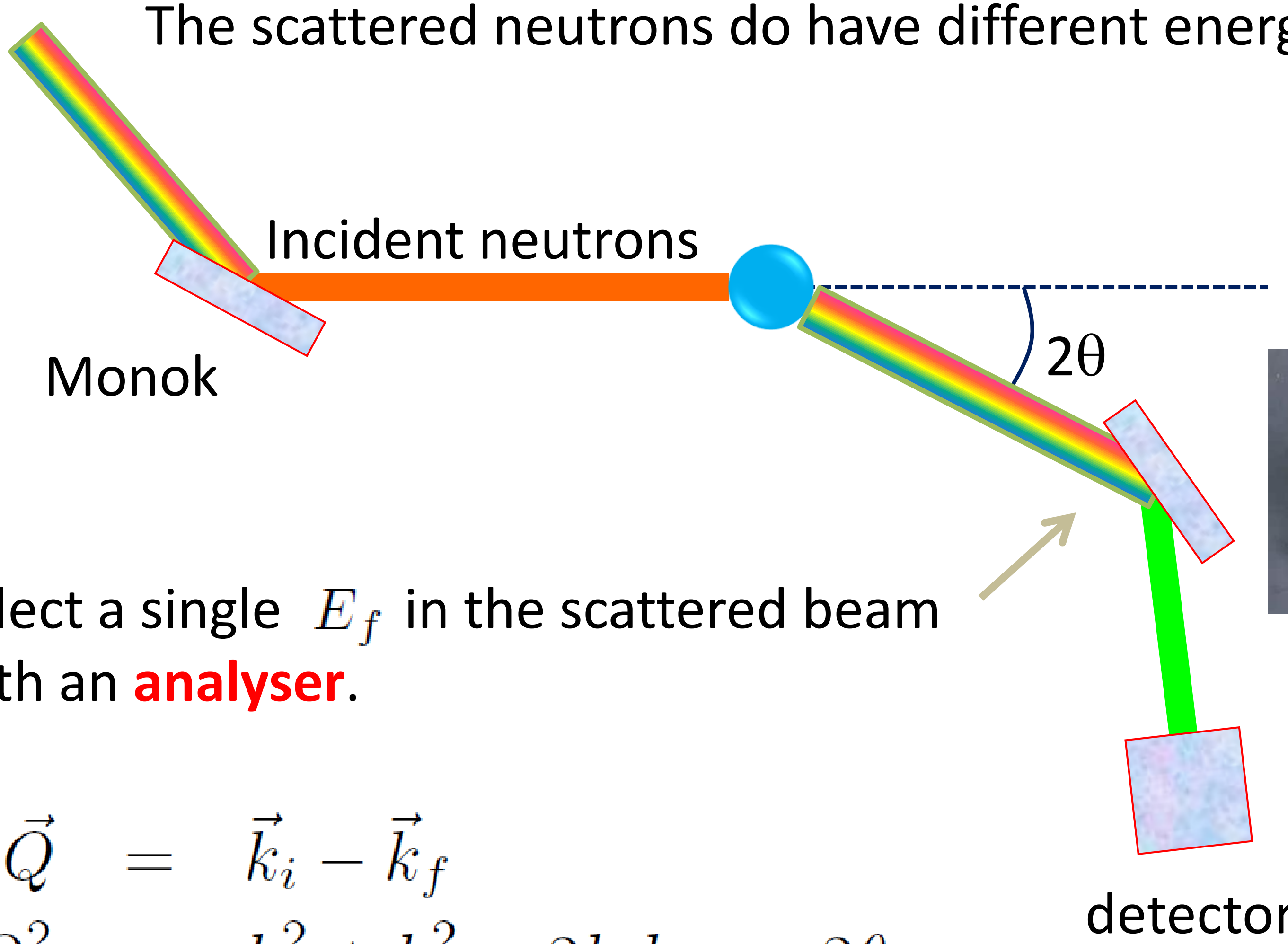


Unit cell and space group



Inelastic scattering TAS

The scattered neutrons do have different energies

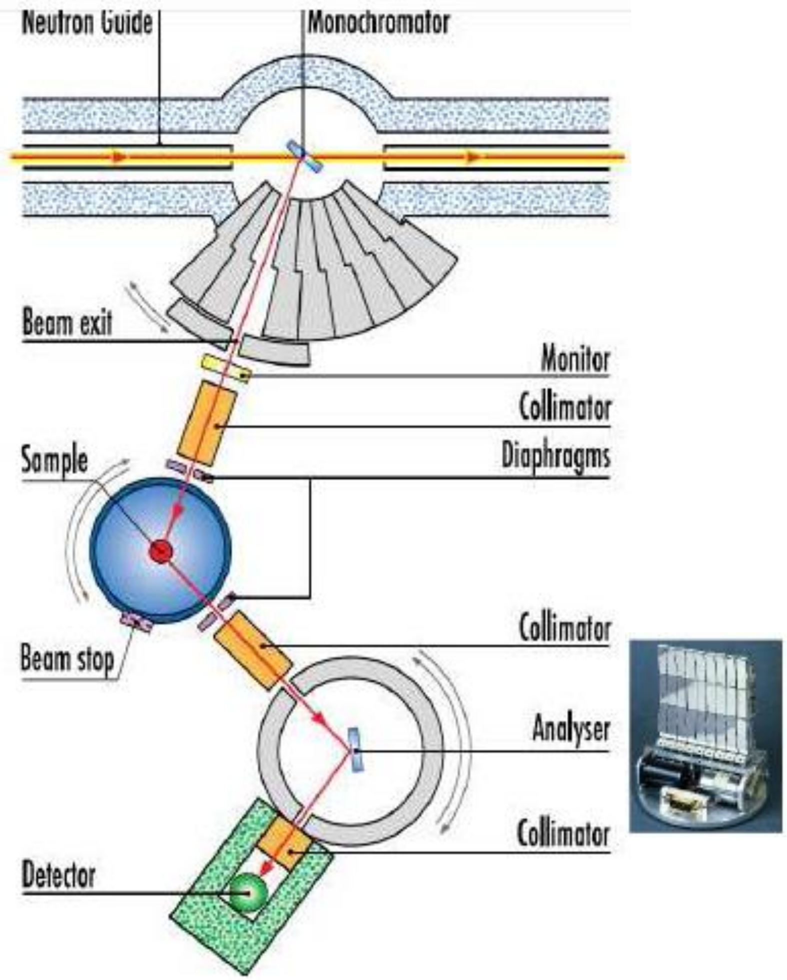


Select a single E_f in the scattered beam with an **analyser**.

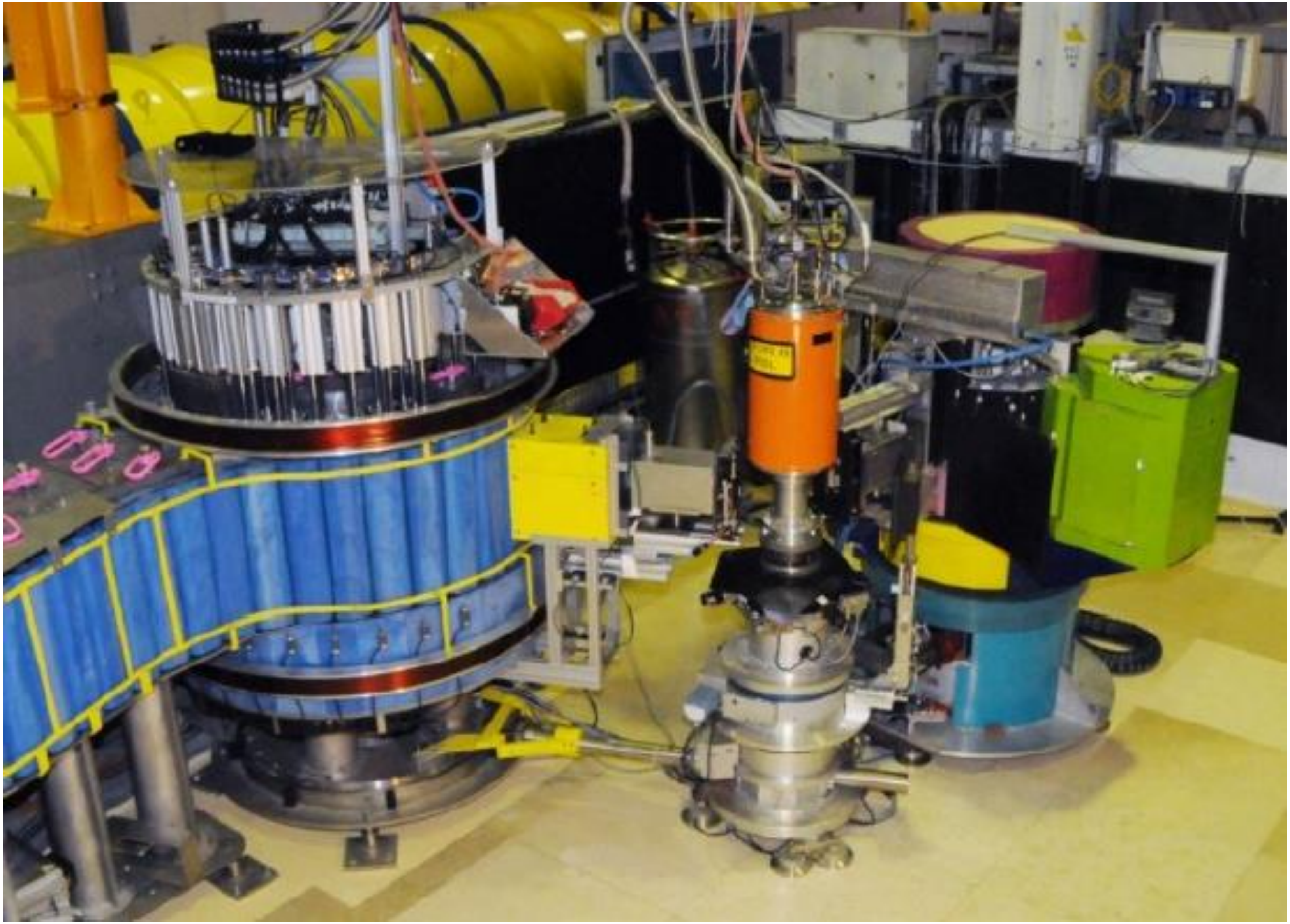
$$\vec{Q} = \vec{k}_i - \vec{k}_f$$

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

Inelastic scattering TAS

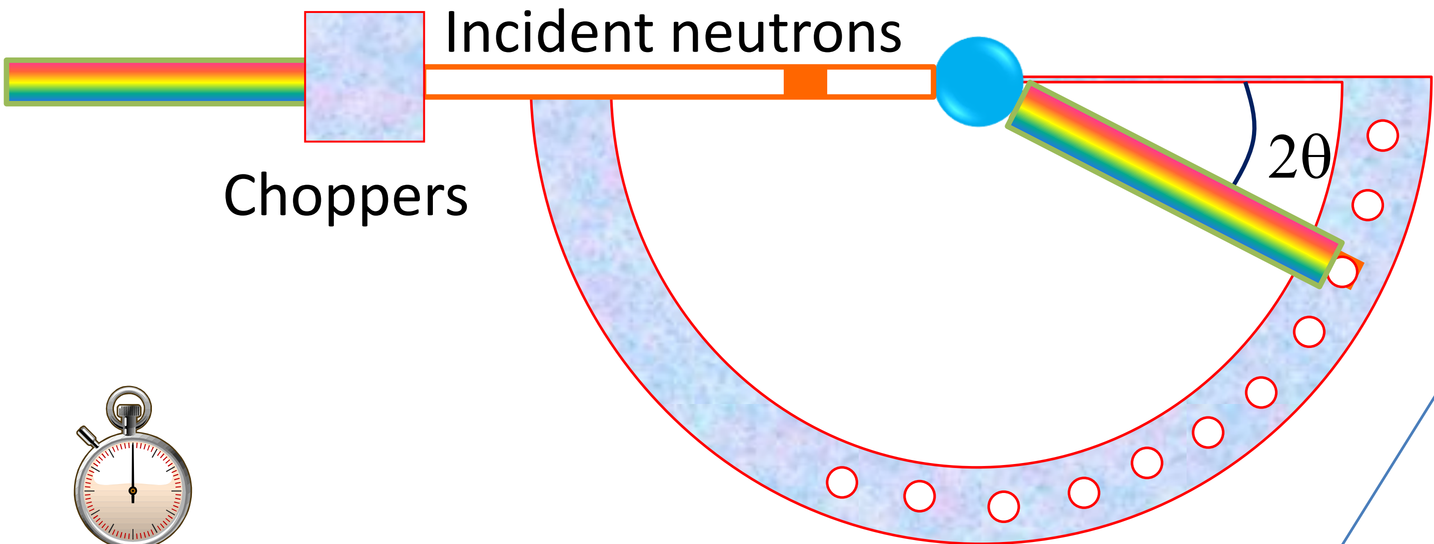


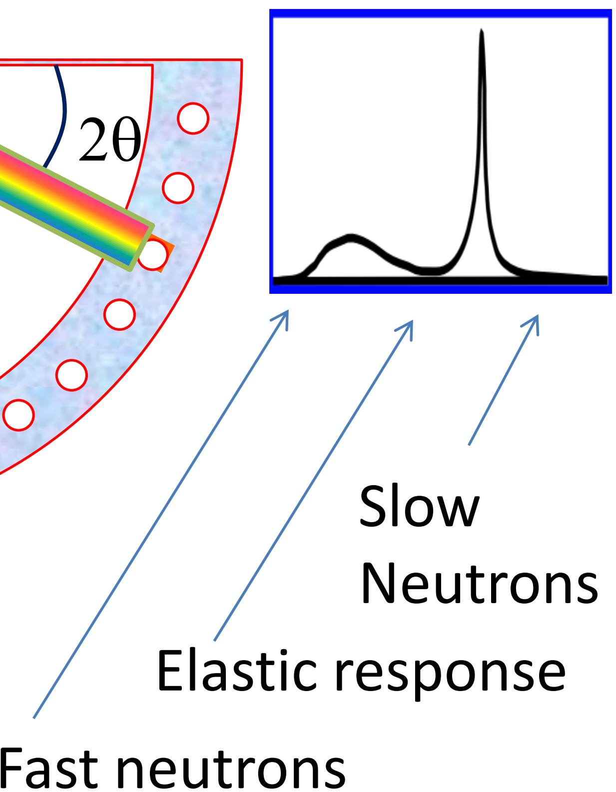
Inelastic scattering TAS



Inelastic scattering TOF

The scattered neutrons do have different energies



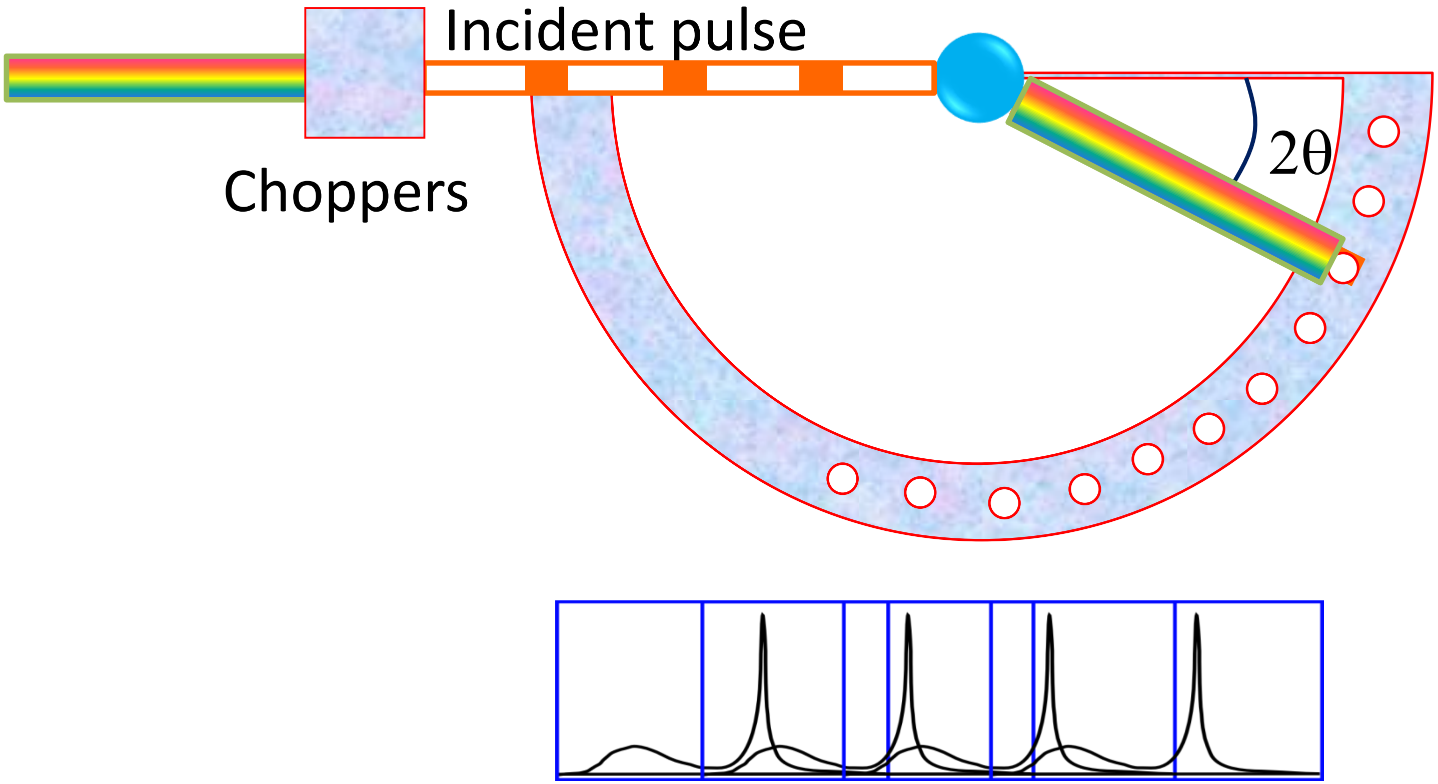


Select a single E_f in the scattered beam via the « time of flight »

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

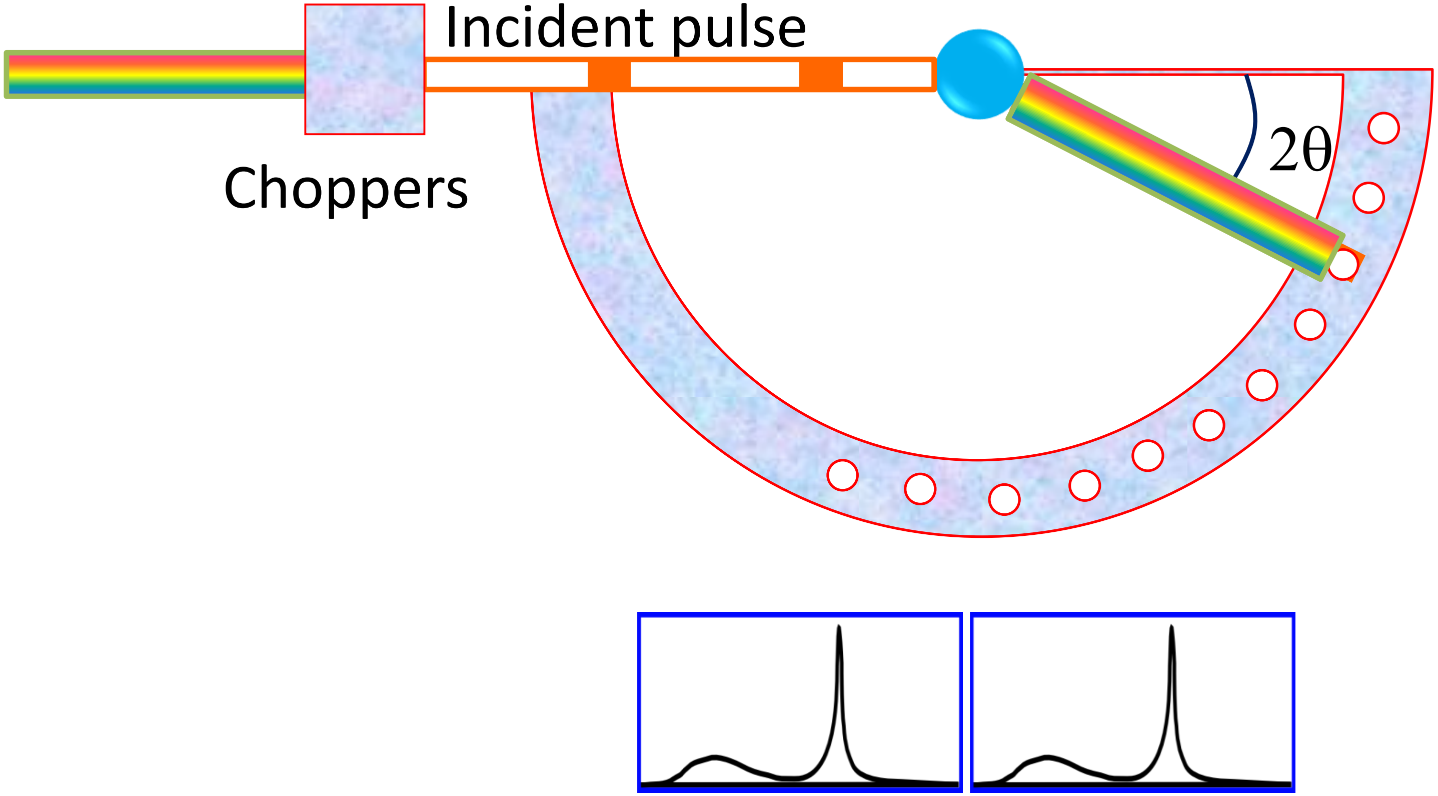
Inelastic scattering TOF

Chop the beam



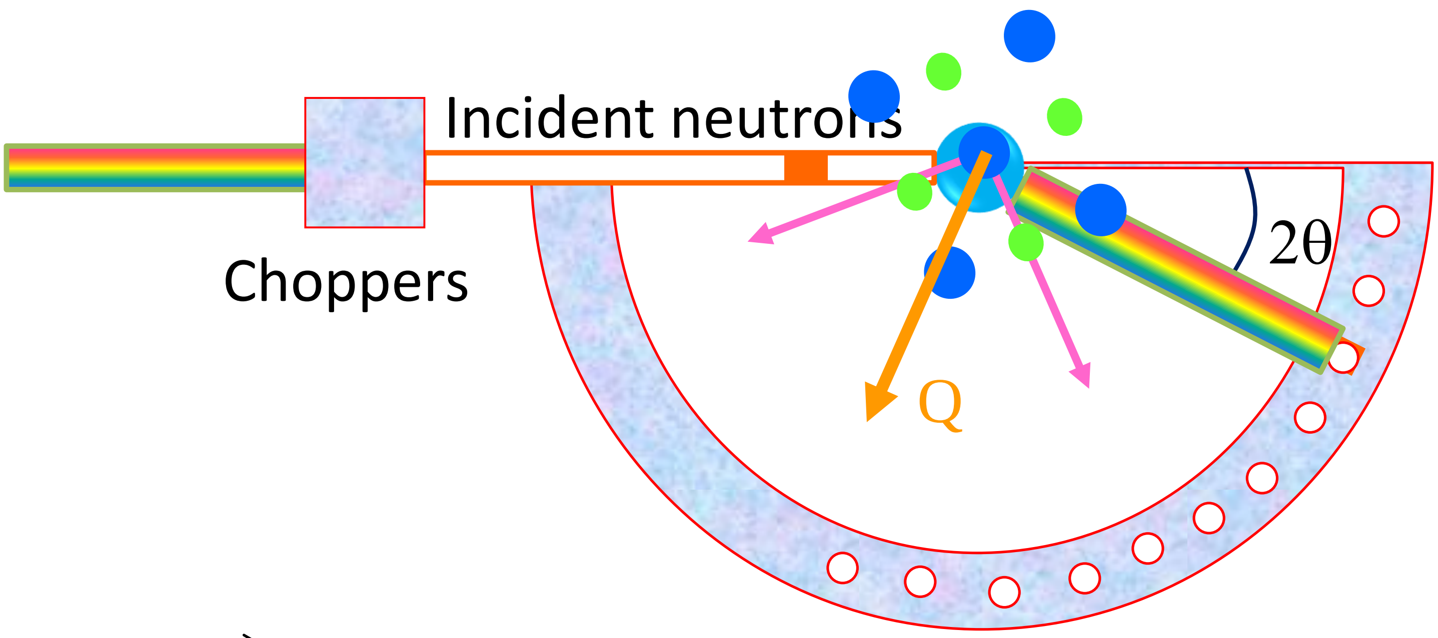
Inelastic scattering TOF

Chop the beam



Inelastic scattering TOF

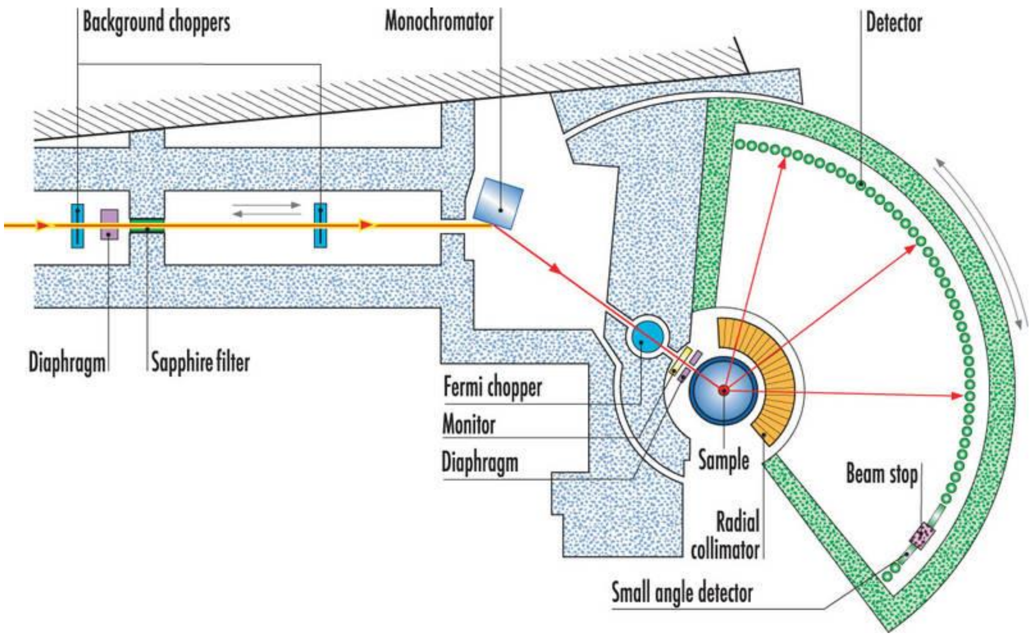
The scattered neutrons do have different energies

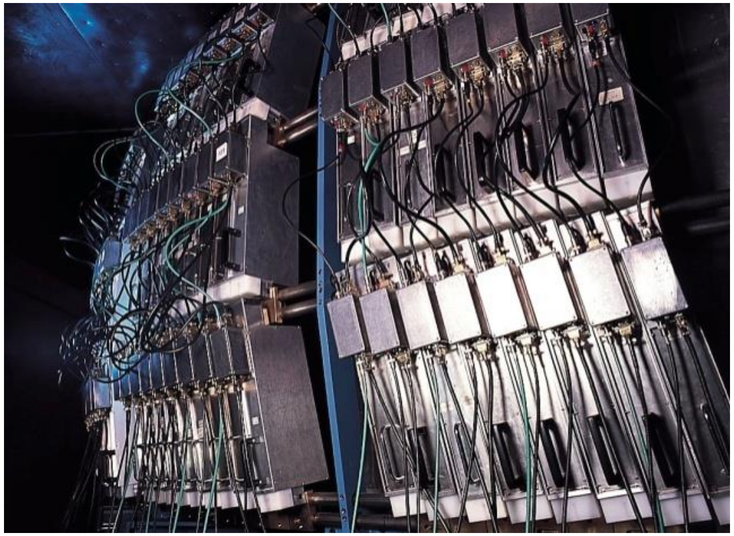


Select $\overrightarrow{Q}$ by rotating the sample

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

Inelastic scattering TOF





Cross section

- The spin (and orbital motion) of unpaired electrons creates a dipolar field.
- The spin of the neutron interacts with this field.

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\,(\gamma r_o)^2 \sum_{m,n} \int_{-\infty}^{+\infty} dt \sum_{a,b} \langle S_m^a \left(\delta_{a,b} - \frac{\mathbf{Q}^a\mathbf{Q}^b}{\mathbf{Q}^2}\right) S_n^b(t)\; e^{i\mathbf{Q}\ \mathbf{R}_m}\; e^{-i\mathbf{Q}\ \mathbf{R}_n(t)}\rangle\; e^{-i\omega t}$$

Cross section

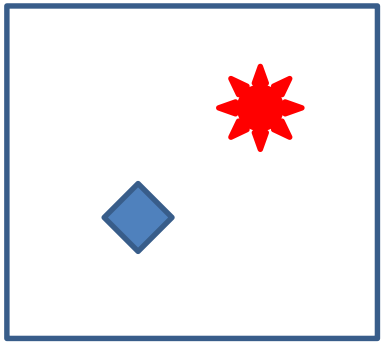
- The spin (and orbital motion) of unpaired electrons creates a dipolar field.

- The spin of the neutron interacts with this field.

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

1. Spins reside on a lattice
2. Decoupled from atomic displacements

$$\mathbf{R}_m(t) = \mathbf{R}_i^o + \mathbf{r}_\ell$$



Cross section

Classical radius of electrons

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

$$\times \int_{-\infty}^{+\infty} dt\, \langle \sum_{a,b} S_{i\ell}^a \left(\delta_{a,b} - \frac{\mathbf{Q}^a\mathbf{Q}^b}{\mathbf{Q}^2}\right) S_{j\ell'}^b(t)\rangle\; e^{-i\omega t}$$

Cross section

Unit cell position

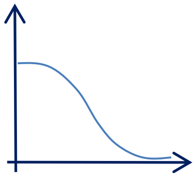
Classical radius of electrons

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

$$\times \int_{-\infty}^{+\infty} dt\, \langle \sum_{a,b} S^a_{i\ell} \left(\delta_{a,b} - \frac{\mathbf{Q}^a\mathbf{Q}^b}{\mathbf{Q}^2}\right) S^b_{j\ell'}(t)\rangle\, e^{-i\omega t}$$

Cross section

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



Unit cell position

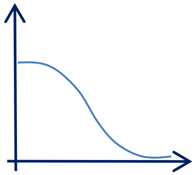
Classical radius of electrons

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

$$\times \int_{-\infty}^{+\infty} dt\ \langle \sum_{a,b} S_{i\ell}^a \left(\delta_{a,b} - \frac{\mathbf{Q}^a \mathbf{Q}^b}{\mathbf{Q}^2} \right) S_{j\ell'}^b(t) \rangle\ e^{-i\omega t}$$

Cross section

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



Atomic positions within the unit cell

Unit cell position

Classical radius of electrons

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}(\gamma r_o)^2 \sum_{i,j} e^{i\mathbf{Q}\,(\mathbf{R}_i^o-\mathbf{R}_j^o)} \sum_{\ell,\ell'} f_\ell(\mathbf{Q})\; f^*_{\ell'}(\mathbf{Q})\; e^{i\mathbf{Q}\,(\mathbf{r}_\ell-\mathbf{r}_{\ell'})}\; e^{-W_\ell-W_{\ell'}}$$

$$\times\int_{-\infty}^{+\infty} dt\,\langle\sum_{a,b} S^a_{i\ell}\left(\delta_{a,b}-\frac{\mathbf{Q}^a\mathbf{Q}^b}{\mathbf{Q}^2}\right) S^b_{j\ell'}(t)\rangle\; e^{-i\omega t}$$

Cross section

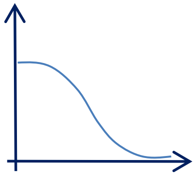
Form factor of unpaired electrons in a given orbital (tabulated)

Atomic positions within the unit cell

Unit cell position

Debye-waller factor (thermal motion of the ions)

Classical radius of electrons

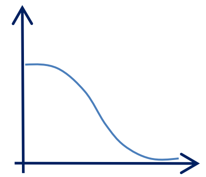


$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}\,(\gamma r_o)^2 \sum_{i,j} e^{i\mathbf{Q}\,(\mathbf{R}_i^o-\mathbf{R}_j^o)} \sum_{\ell,\ell'} f_\ell(\mathbf{Q})\; f^*_{\ell'}(\mathbf{Q})\; e^{i\mathbf{Q}\,(\mathbf{r}_\ell-\mathbf{r}_{\ell'})}\; e^{-W_\ell-W_{\ell'}}$$

$$\times \int_{-\infty}^{+\infty} dt\, \langle \sum_{a,b} S^a_{i\ell} \left(\delta_{a,b} - \frac{\mathbf{Q}^a\mathbf{Q}^b}{\mathbf{Q}^2}\right) S^b_{j\ell'}(t)\rangle\; e^{-i\omega t}$$

Cross section

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



Atomic positions within the unit cell

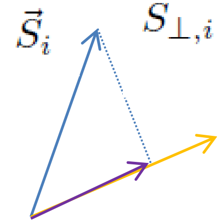
Debye-waller factor (thermal motion of the ions)

Unit cell position

Classical radius of electrons

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

$$\times \int_{-\infty}^{+\infty} dt\; \langle \sum_{a,b} S_{i\ell}^a \left(\delta_{a,b} - \frac{\mathbf{Q}^a\mathbf{Q}^b}{\mathbf{Q}^2}\right) S_{j\ell'}^b(t)\rangle\; e^{-i\omega t}$$



- Spin-spin correlation function
- spin components perp to Q (dipolar interaction)

$$\sum_{a,b} S_m^a \left(\delta_{a,b} - \frac{\mathbf{Q}^a\mathbf{Q}^b}{\mathbf{Q}^2}\right) S_n^b(t) = \mathbf{S}_{\perp,m} \cdot \mathbf{S}_{\perp,n}(t)$$

Frozen spins (Cross section)

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

Assume frozen spins (independent on time)

Frozen spins (Cross section)

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

Assume frozen spins (independent on time)

↓

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

This is noting but the magnetic structure factor

1. Crystal field

- Electrostatic interaction between a magnetic ion and its neighbors.

- A common example is the splitting in t2g and eg levels for d orbitals.

- The crystal field (along with spin-orbit) is also very important for rare-earth (4f electron) compounds.

- The local symmetry is lower than the spherical symmetry of the “free” magnetic ion → aspherical potential written in spherical harmonics or, in practice, using Stevens operators.

$$\mathcal{H} = \sum_{a,b} B_{ab} O_{ab}$$

1. Crystal field

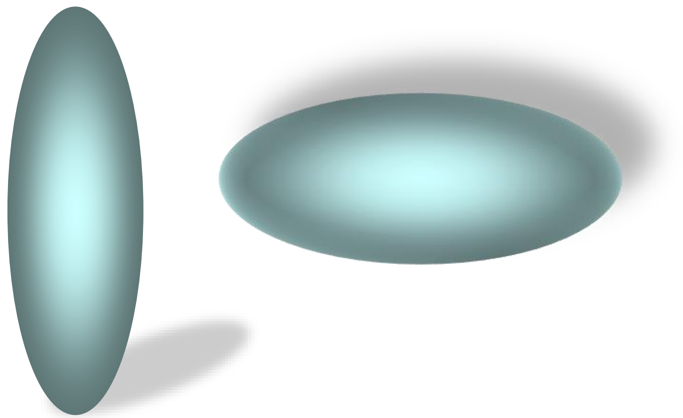
$$
\begin{aligned}
B_{20} &= 3\ J_z^2 - J(J+1) \\
B_{22} &= (J_+^2\ +\ J_-^2)/2 \\
B_{40} &= 35\ J_z^4 + (-30J(J+1)+25)\ J_z^2 + (-6J(J+1)+3J^2(J+1)^2)I \\
B_{42} &= \ldots
\end{aligned}
$$

$$\mathcal{H} = \sum_{a,b} B_{ab} O_{ab}$$

Electronic levels and wavefunctions

$$E_n, \quad |n\rangle$$

$$|n\rangle = \sum_{i=-J,J} a_i |J_z = i\rangle$$



« Anisotropy » of magnetic moments

Expand the thermal average using eigen-states and eigen-energies:

$$
\begin{aligned}
\int_{-\infty}^{+\infty} dt \, \langle \mathbf{S}_{\perp,i\ell} \, \mathbf{S}_{\perp,j\ell'}(t) \rangle \, e^{-i\omega t} &= \int_{-\infty}^{+\infty} dt \sum_{n} p_n \, \langle n | \mathbf{S}_{\perp,i\ell} \, \mathbf{S}_{\perp,j\ell'}(t) | n \rangle \, e^{-i\omega t} \\
&= \int_{-\infty}^{+\infty} dt \sum_{n,m} p_n \, \langle n | \mathbf{S}_{\perp,i\ell} | m \rangle \langle m | \mathbf{S}_{\perp,j\ell'}(t) | n \rangle \, e^{-i\omega t} \\
&= \int_{-\infty}^{+\infty} dt \sum_{n,m} p_n \, \langle n | \mathbf{S}_{\perp,i\ell} | m \rangle \langle m | e^{i\mathcal{H}t} \mathbf{S}_{\perp,j\ell'} e^{-i\mathcal{H}t} | n \rangle \, e^{-i\omega t} \\
&= \int_{-\infty}^{+\infty} dt \sum_{n,m} p_n \, \langle n | \mathbf{S}_{\perp,i\ell} | m \rangle \langle m | \mathbf{S}_{\perp,j\ell'} | n \rangle \, e^{i(E_m - E_n) - i\omega t} \\
&= \sum_{n,m} p_n \, \langle n | \mathbf{S}_{\perp,i\ell} | m \rangle \langle m | \mathbf{S}_{\perp,j\ell'} | n \rangle \, \delta(E_m - E_n - \omega)
\end{aligned}
$$

$$
p_n = \frac{e^{-E_n/k_{\mathrm{B}}T}}{\sum_m e^{-E_m/k_{\mathrm{B}}T}}
$$

Boltzmann factor

Matrix element of the transition between two electronic levels

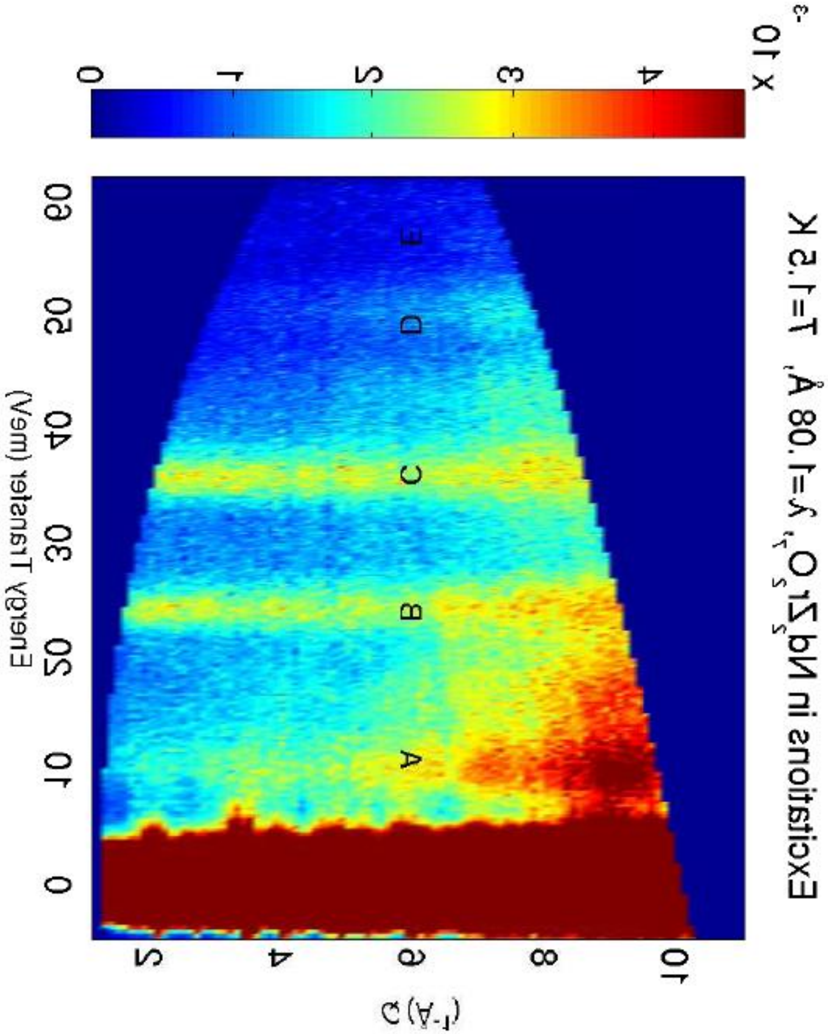
Not zero when the energy transfer matches the transition

1. Crystal field

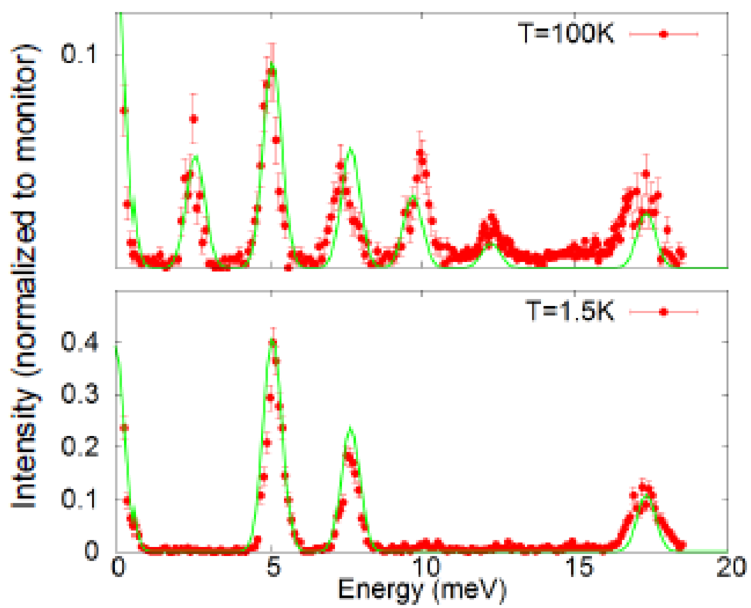
$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{k_f}{k_i}(\gamma r_o)^2 \sum_{i,j} e^{i\mathbf{Q}\,(\mathbf{R}_i^o-\mathbf{R}_j^o)} \sum_{\ell,\ell'} f_\ell(\mathbf{Q})\, f_{\ell'}^*(\mathbf{Q})\, e^{i\mathbf{Q}\,(\mathbf{r}_\ell-\mathbf{r}_{\ell'})}\, e^{-W_\ell-W_{\ell'}}$$
$$\times \sum_{n,m}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

- Series of modes describing the transitions between different electronic levels, provided the matrix elements are not zero.
- Temperature dependence related to Boltzmann factors.
- Does not depend on Q (except form factor)

$Nd_2Zr_2O_7$

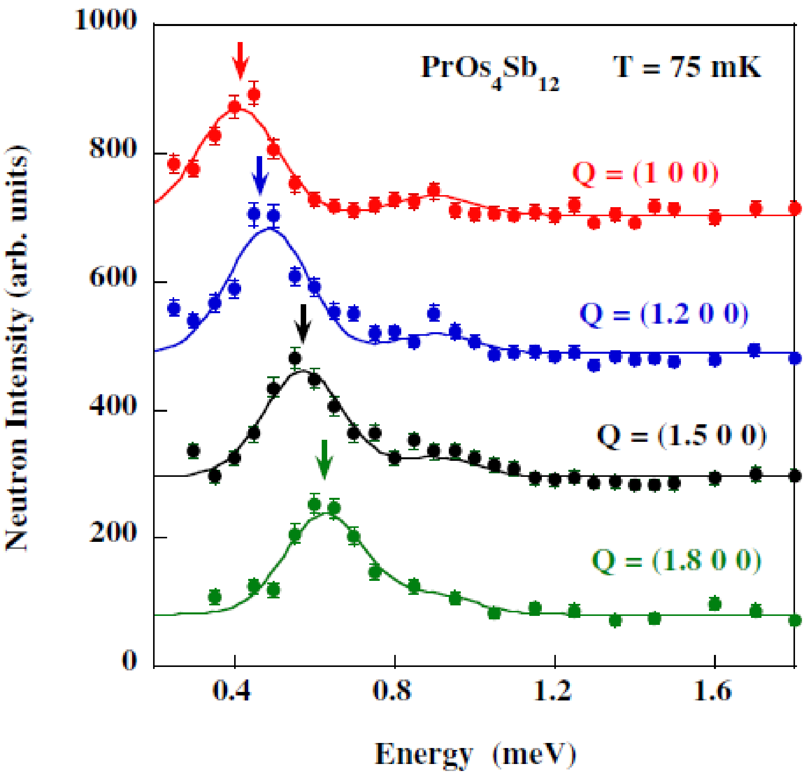


$Er_2Ti_2O_7$



2. Crystal field excitons

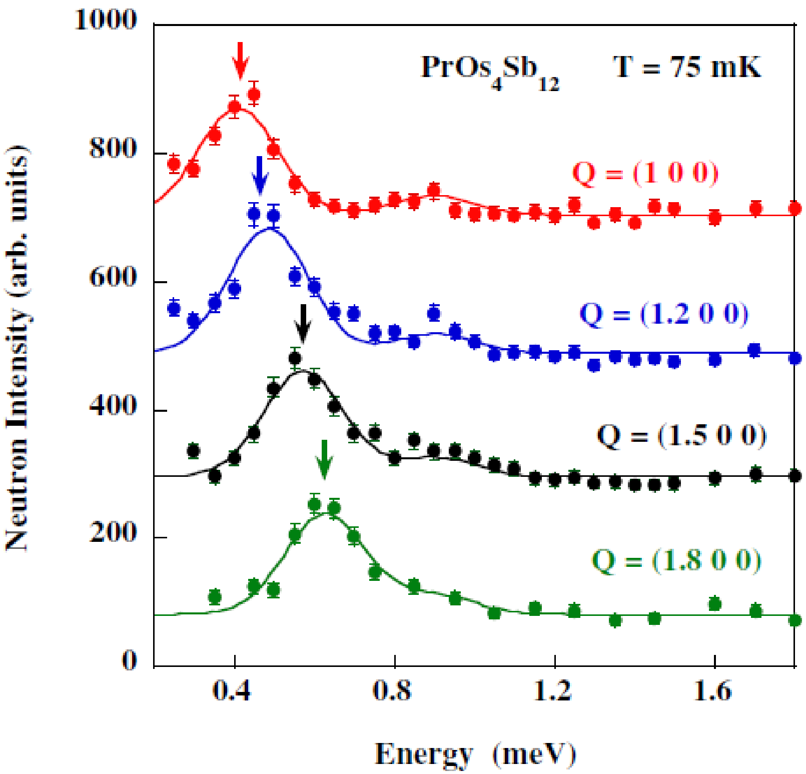
- The transitions can become weakly dispersive because of interactions

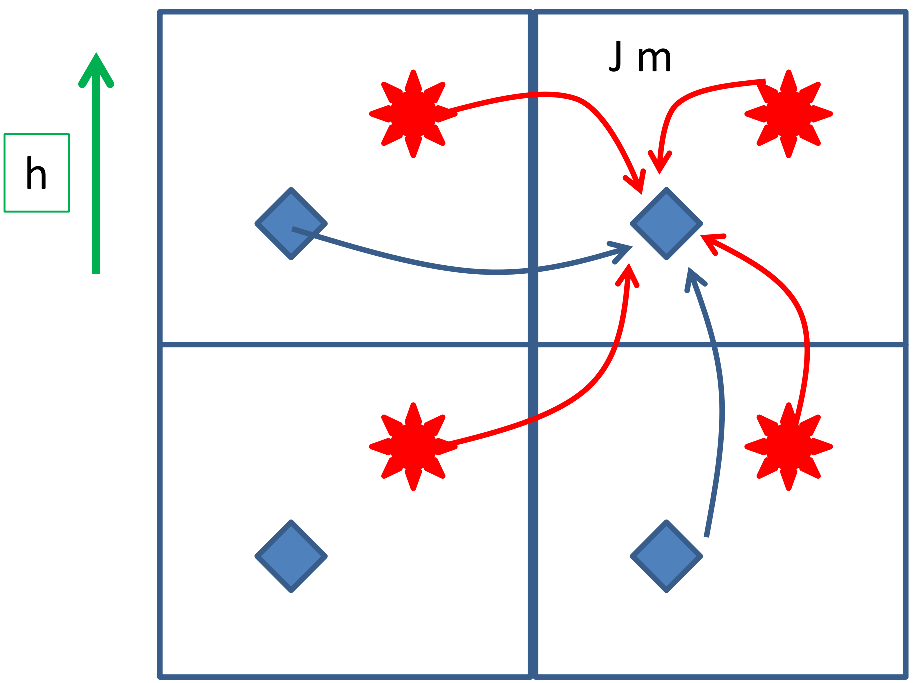


2. Crystal field excitons

- The transitions can become weakly dispersive because of interactions

- Best understood in the **Random Phase approximation** using the concept of generalized susceptibility





2. Crystal field excitons

- The transitions can become weakly dispersive because of interactions.
- Best understood in the **Random Phase approximation** using the concept of generalized susceptibility:

$$m(\mathbf{Q},\omega) = \chi^o(\mathbf{Q},\omega)\, \boxed{h(\mathbf{Q},\omega)} + \chi^o(\mathbf{Q},\omega)\, \boxed{J(\mathbf{Q})\ m(\mathbf{Q},\omega)}$$

$$[1-\chi^o(\mathbf{Q},\omega)\ J(\mathbf{Q})]\ m(\mathbf{Q},\omega) = \chi^o(\mathbf{Q},\omega)\ h(\mathbf{Q},\omega)$$

$$m(\mathbf{Q},\omega) = [1-\chi^o(\mathbf{Q},\omega)\ J(\mathbf{Q})]^{-1}\ \chi^o(\mathbf{Q},\omega)\ h(\mathbf{Q},\omega)$$

$$\chi(\mathbf{Q},\omega) = [1-\chi^o(\mathbf{Q},\omega)\ J(\mathbf{Q})]^{-1}\ \chi^o(\mathbf{Q},\omega)$$

2. Crystal field excitons

- The fluctuation – dissipation theorem relates susceptibility and correlations via the detailed balance factor:

$$\int_{-\infty}^{+\infty} dt\ \langle \mathbf{S}_{\perp,i\ell}\ \mathbf{S}_{\perp,j\ell'}(t)\ \rangle\ e^{-i\omega t} = \left(1 + \frac{1}{\exp \omega/k_B T - 1}\right) \mathrm{Im}\ \chi(Q,\omega)$$

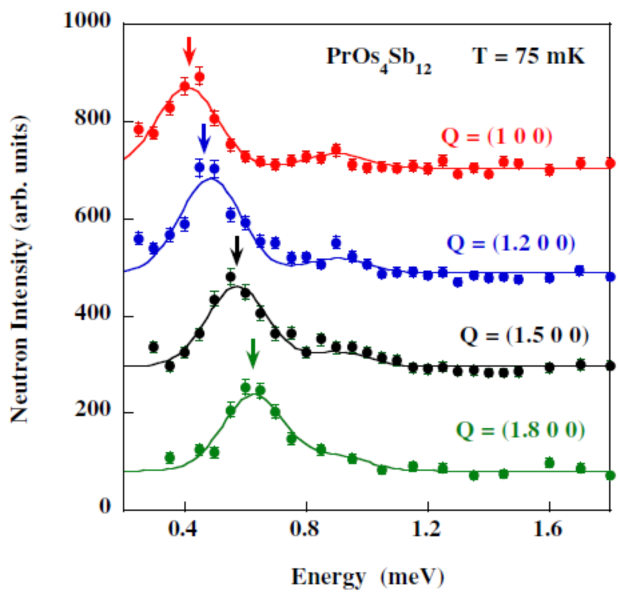
$$\chi = \int_{-\infty}^{+\infty} dt\ i\theta(t)\langle[\mathbf{S}_{\perp,i\ell}\ \mathbf{S}_{\perp,j\ell'}(t)]\rangle\ e^{-i\omega t}$$

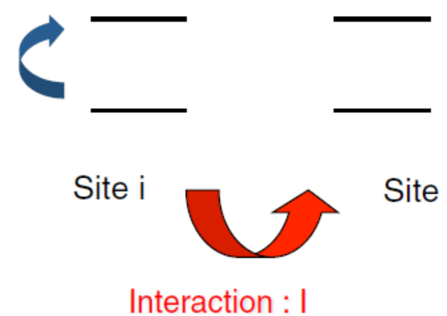
$$\mathrm{Im}\ \chi = \sum_{n,m}(p_n - p_m)\ \langle n|\mathbf{S}_{\perp,i\ell}|m\rangle\langle m|\mathbf{S}_{\perp,j\ell'}|n\rangle\ \delta(E_m - E_n - \omega)$$

$$\chi^o = (p_0 - p_1)\frac{\Delta\ M^2}{\omega^2 - \Delta^2}$$

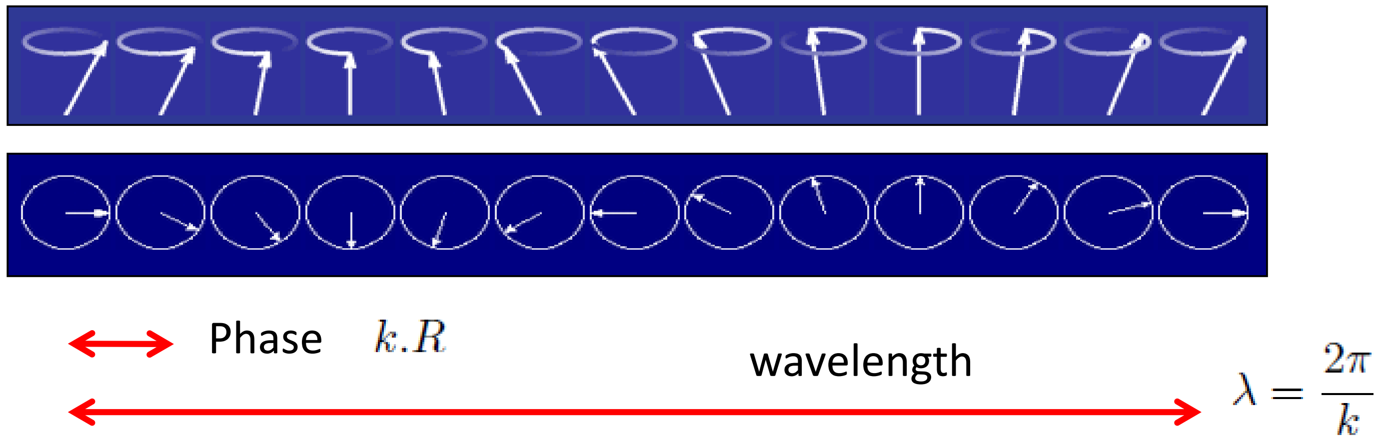
$$\chi = \frac{(p_0 - p_1)\Delta\ M^2}{\omega^2 - \Delta^2 - (p_0 - p_1)\ \Delta\ M^2\ J(\mathbf{Q})}$$

$$\omega = \sqrt{\Delta^2 + (p_0 - p_1)\ \Delta\ M^2\ J(\mathbf{Q})}$$



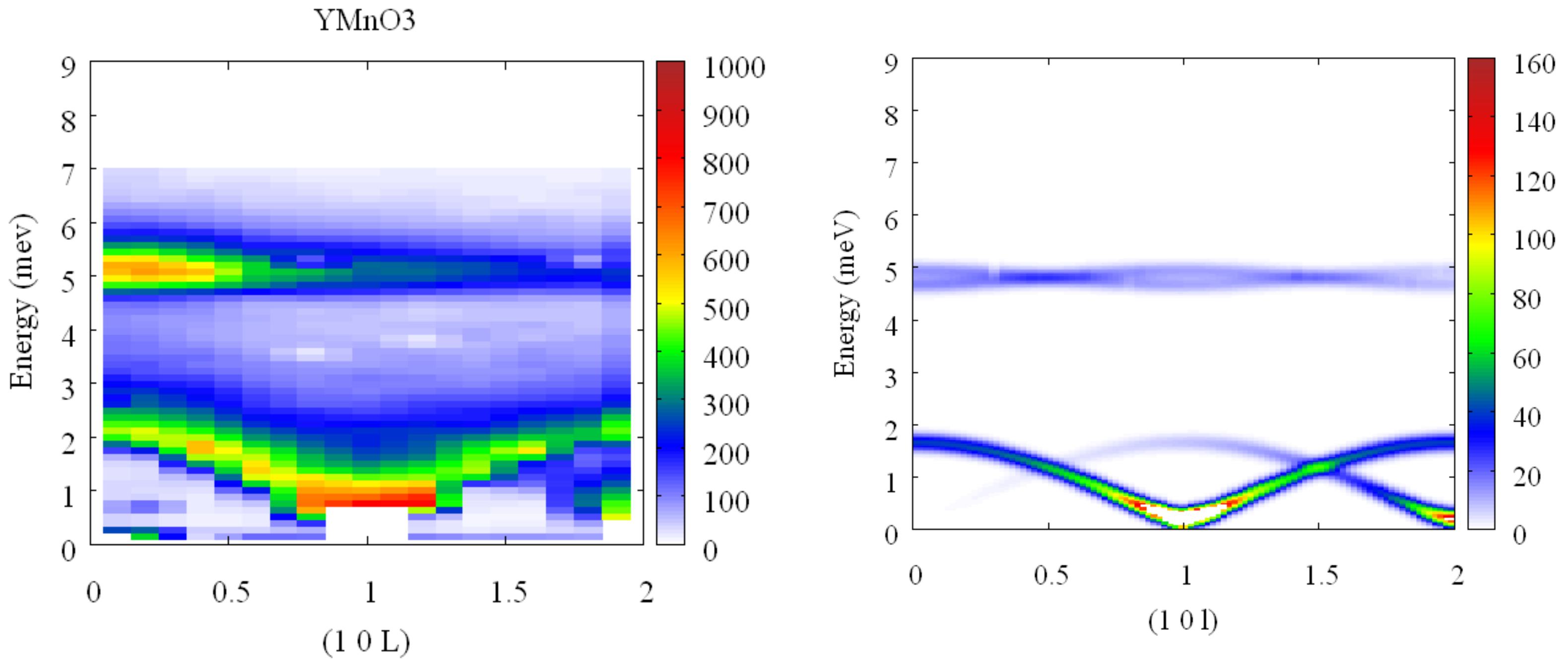


3. Collective excitations : spin waves



- Spin waves are precession modes of the spins in conventional magnets.
- They are described by S=1 bosonic quasiparticles
- INS allows to measure the dispersion of those modes
- With the help of a model, it becomes possible to determine exchange couplings, anisotropies, …
- We need a theory for that …

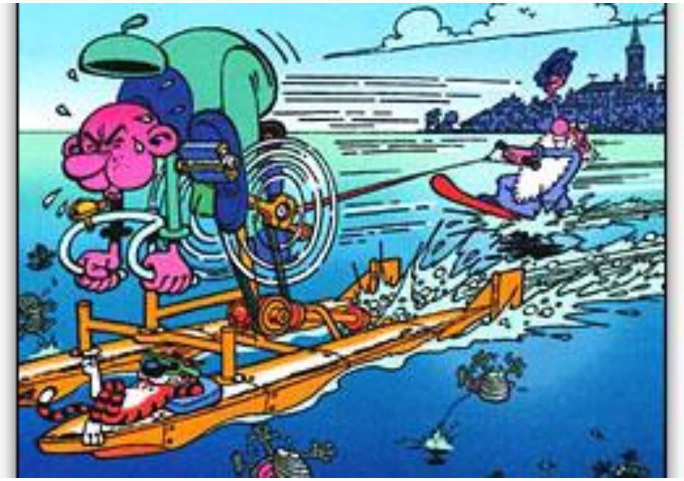
3. Collective excitations : spin waves



Spin wave response (showing the dispersion) in the multiferroic material $YMnO_3$
Right panel shows the modeling

3. Collective excitations : spin waves

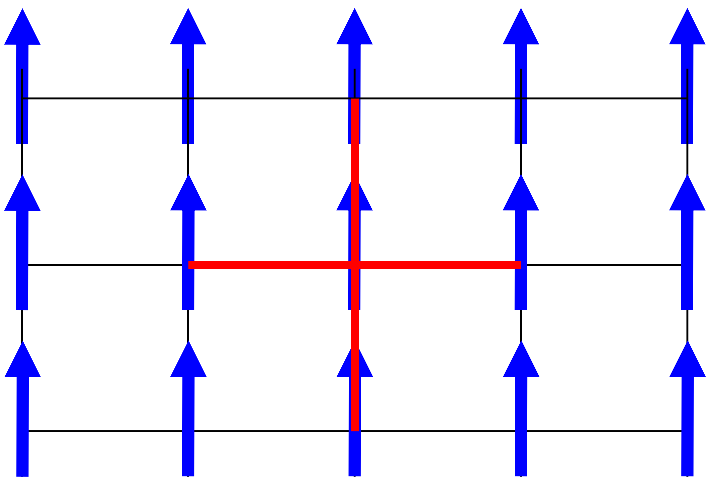
Different attitudes to math and theory ....



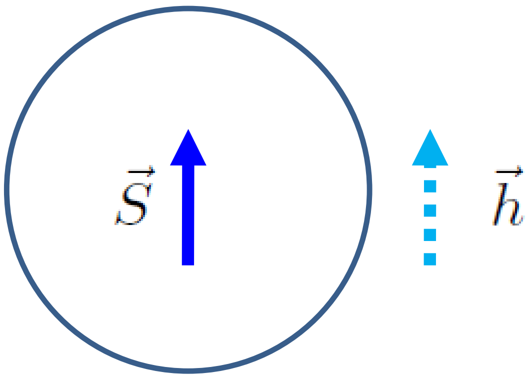
I. Conventional ground state described by molecular field theory
II. « Holstein-Primakov » representation of the spin operators to define deviations away from local magnetization
III. Keeping small deviations only
IV. Solving (Fourier transform, Bogolubov ....) : **free bosons**

3. Collective excitations : spin waves

Heisenberg Hamiltonian $\mathcal{H} = \frac{1}{2} \sum_{m,n} \mathbf{S}_m J_{m,n} \mathbf{S}_n$



$J_{m,n}$

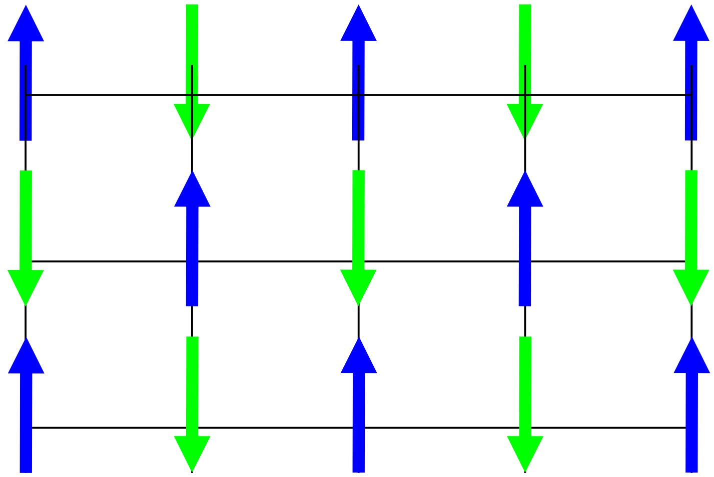


A spin experiences a **molecular field** due to interaction with its neighbours

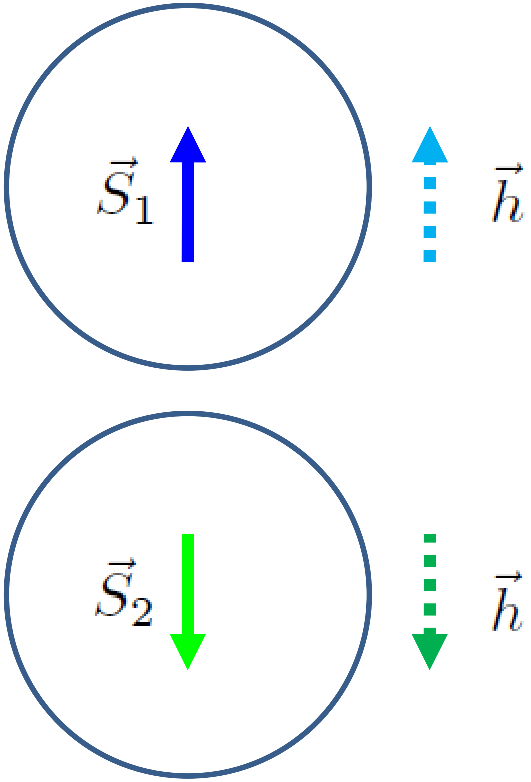
Long range ordering

3. Collective excitations : spin waves

Depending on the nature of the interactions, this molecular field can induce a new periodicity

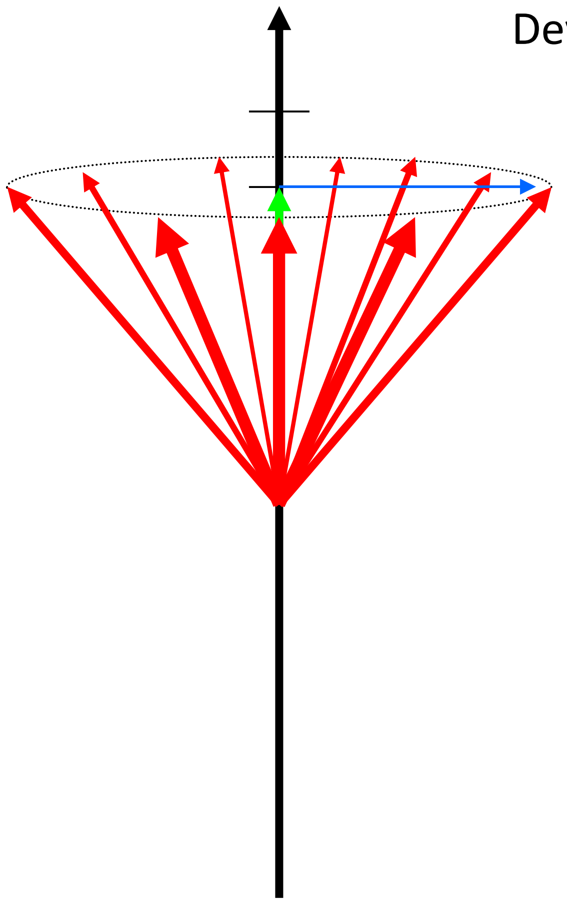


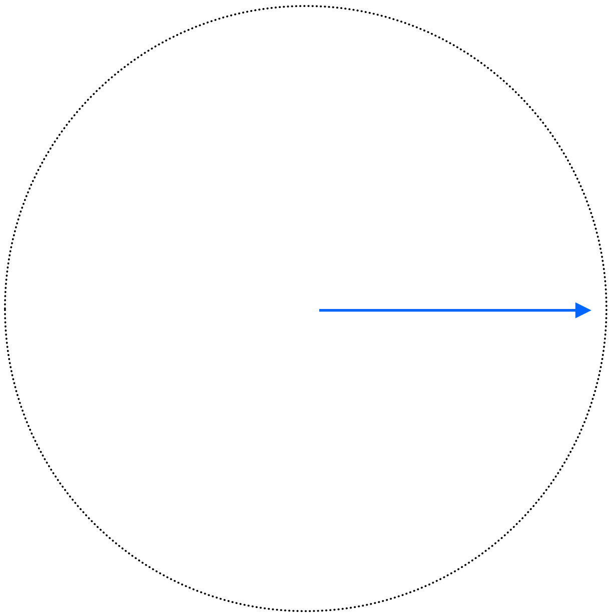
Example : AF ordering



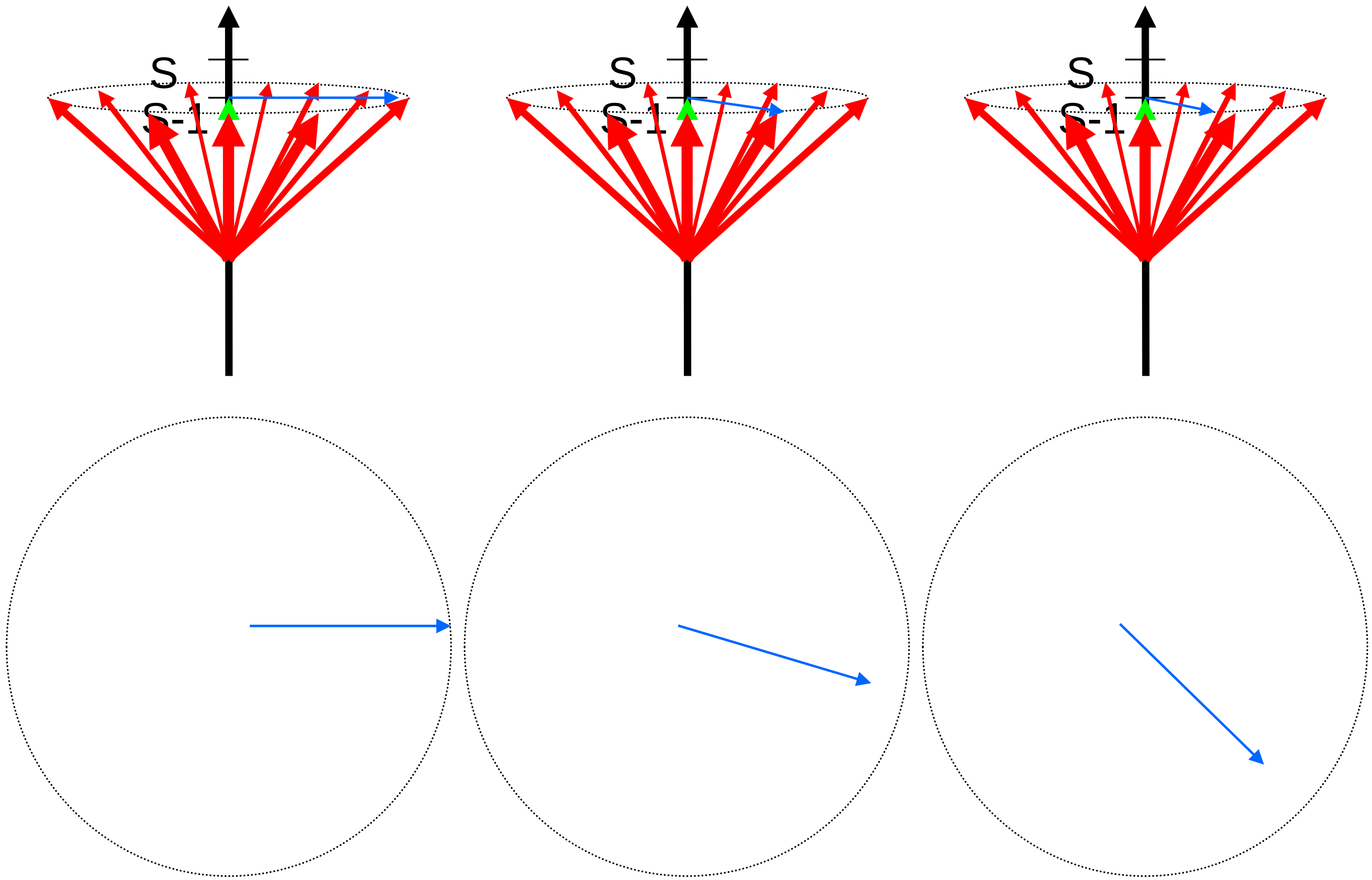
3. Collective excitations : spin waves

Precession of a spin in a magnetic field
Deviation away from local equilibrium magnetization





3. Collective excitations : spin waves



How to describe the « deviation » ?

Polar coordinates:

$$
\begin{aligned}
S_x &= S\cos\theta\sin\phi \\
S_y &= S\sin\theta\sin\phi \\
S_z &= S\cos\phi
\end{aligned}
$$

$$
\begin{aligned}
S^+ &= S\sin\phi\; e^{+i\theta} \\
S^- &= S\sin\phi\; e^{-i\theta}
\end{aligned}
$$

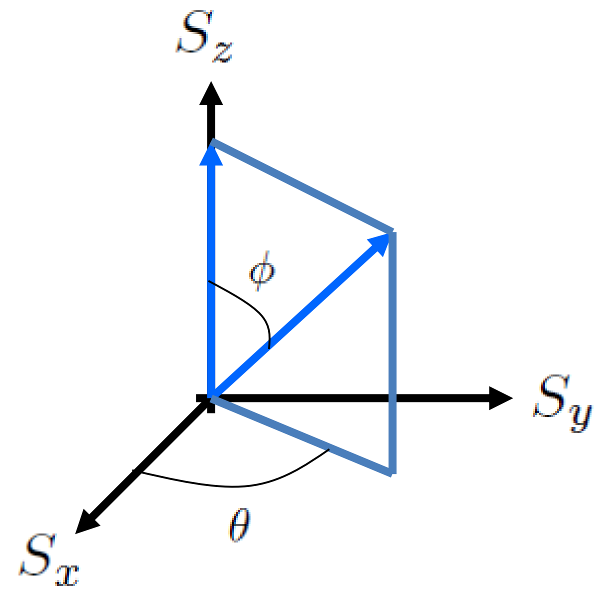
Deviation away from saturation:

$$S_z = S - D$$

$$\cos\phi = 1 - \frac{D}{S}$$

Spin components written in terms of the deviation:

$$
\begin{aligned}
S^+ &= \sqrt{2S}\sqrt{1-\frac{D}{2S}}\,\sqrt{D}\; e^{+i\theta} \\
S^- &= \sqrt{2S}\sqrt{1-\frac{D}{2S}}\,\sqrt{D}\; e^{-i\theta} \\
S_z &= S - D
\end{aligned}
$$



How to describe the « deviation » ?

The deviation is expressed using a boson operator:

$$[b, b^+] = 1$$

$$n_b = b^+ b = 0, 1, 2, ..., \infty$$

$$\begin{aligned} S^+ &= \sqrt{2S}\sqrt{1 - \frac{D}{2S}}\sqrt{D}\, e^{+i\theta} \\ S^- &= \sqrt{2S}\sqrt{1 - \frac{D}{2S}}\sqrt{D}\, e^{-i\theta} \\ S_z &= S - D \end{aligned}$$

$$\begin{aligned} S^+ &= \sqrt{2S}\sqrt{1 - \frac{n_b}{2S}}\, b \\ S^- &= \sqrt{2S}\, b^+ \sqrt{1 - \frac{n_b}{2S}} \\ S_z &= S - n_b \end{aligned}$$

« Holstein – Primakov »

$$\begin{aligned} S^+ &\approx \sqrt{2S}\, b \\ S^- &\approx \sqrt{2S}\, b^+ \\ S^z &= S - n_b \end{aligned}$$

How to describe the « deviation » ?

The deviation is expressed using a boson operator:

$$[b, b^+] = 1$$

$$n_b = b^+ b = 0, 1, 2, ..., \infty$$

May become unphysical

$$\begin{aligned} S^+ &= \sqrt{2S}\,\sqrt{1-\frac{n_b}{2S}}\;b \\ S^- &= \sqrt{2S}\;b^+\sqrt{1-\frac{n_b}{2S}} \\ S_z &= S - n_b \end{aligned}$$

$$\begin{aligned} S^+ &= \sqrt{2S}\,\sqrt{1-\frac{D}{2S}}\,\sqrt{D}\;e^{+i\theta} \\ S^- &= \sqrt{2S}\,\sqrt{1-\frac{D}{2S}}\,\sqrt{D}\;e^{-i\theta} \\ S_z &= S - D \end{aligned}$$

« Holstein – Primakov »

$$\begin{aligned} S^+ &\approx \sqrt{2S}\;b \\ S^- &\approx \sqrt{2S}\;b^+ \\ S^z &= S - n_b \end{aligned}$$

Ferromagnet

Heisenberg Hamiltonian

$$\mathcal{H} = \frac{1}{2} \sum_{m,n} \mathbf{S}_m J_{m,n} \, \mathbf{S}_n$$

Holstein-Primakov

$$\begin{aligned} S_m^x &\approx \frac{\sqrt{2S}}{2} \left(b_m + b_m^+\right) \\ S_m^y &\approx \frac{\sqrt{2S}}{2i} \left(b_m - b_m^+\right) \\ S_m^z &\approx S - b_m^+ \, b_m \end{aligned}$$

Keep 2nd order terms (small deviations)

$$\mathcal{H} \approx \frac{1}{2} \sum_{m,n} \quad \frac{S}{2}(b_m + b_m^+) \, J_{m,n} \, (b_n + b_n^+) - \frac{S}{2}(b_m - b_m^+) \, J_{m,n} \, (b_n - b_n^+)$$
$$+ \, (S - b_m^+ b_m) \; J_{m,n} \; (S - b_n^+ b_n)$$

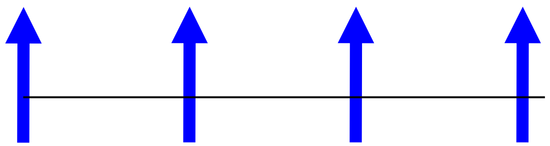
$$\mathcal{H} \approx \frac{1}{2} \sum_{m,n} \quad S \, J_{m,n} \, (b_m \, b_n^+ + b_m^+ \, b_n) - S \, J_{m,n} \, b_n^+ b_n - b_m^+ b_m \, J_{m,n} \, S$$

Fourier Transform : dispersion of the spin waves

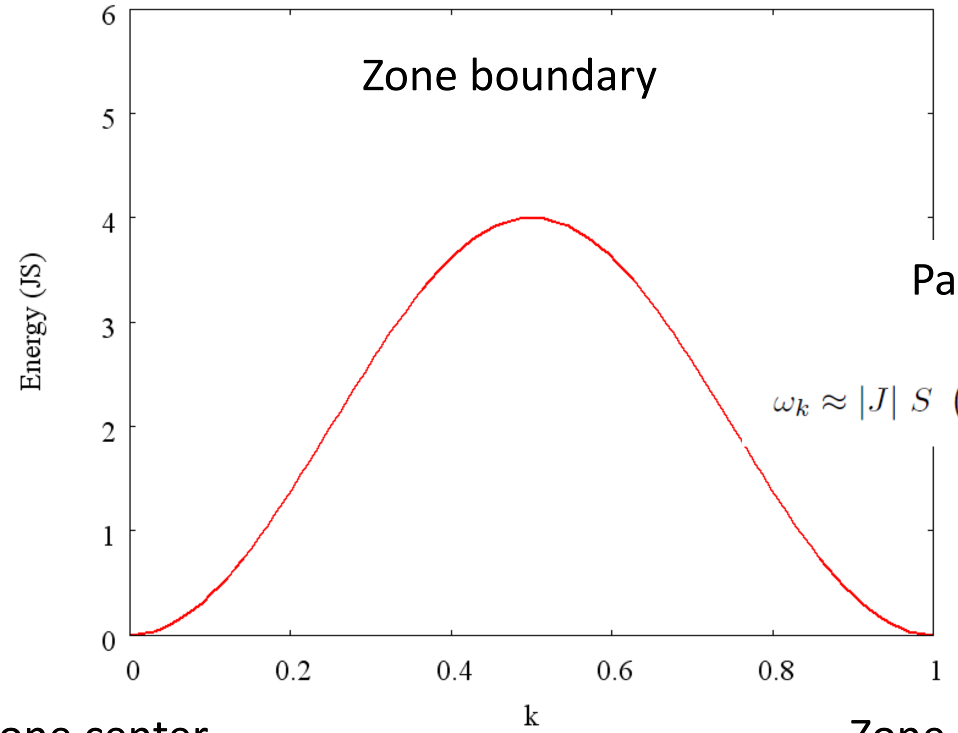
$$\mathcal{H} \approx \sum_k \quad J_k \, S \, b_k^+ \, b_k - \left(\sum_\Delta J_{m,m+\Delta} \right) b_k^+ b_k$$

$$\omega_k = (J_k - zJ) \, S = z|J| \, S \, (1 - \gamma_k) \qquad \gamma_k = \frac{1}{z} \sum_\Delta e^{ik\Delta}$$

Ferromagnet



$$\omega_k = (J_k - zJ)\,S = z|J|\;S\;(1-\gamma_k) \qquad \gamma_k = \frac{1}{z}\sum_{\Delta} e^{ik\Delta}$$



Parabolic dispersion at small k

$$\omega_k \approx |J|\;S\;\left((k_x\;a_x)^2 + (k_y\;a_y)^2 + (k_z\;a_z)^2\right)$$

Zone center

Zone center

Ferromagnet

Checking consistency of the approximation

$$\langle S^z \rangle \approx S - \sum_k \langle b_k^+ \, b_k \rangle$$
$$\approx S - \sum_k n_B(\omega_k)$$

$$\sum_k \longrightarrow \int dk^d = \int dk \, \frac{k^{d-1}}{(2\pi)^d}$$

$$n_B(E) \longrightarrow \frac{k_B T}{E}$$

$$\sum_k n_B(\omega_k) \longrightarrow \int dk \, \frac{k^{d-1}}{(2\pi)^d} \, \frac{T}{k^2}$$

The thermal fluctuations prevent long range ordering for $d \leq 2$

The Breakdown of the spin wave theory is consistent with the « Mermin and Wagner » theorem

Ferromagnet

To calculate the cross section from spin waves, we calculate the spin in terms of the spin wave bosons:

$$\sum_m e^{i\mathbf{k}\mathbf{R}_m}\ \mathbf{S}_m(t) = \mathbf{S}_k(t) = \begin{pmatrix} \frac{\sqrt{2S}}{2}(b_k e^{+i\omega_k t} + b_k^+ e^{-i\omega_k t}) \\ \frac{\sqrt{2S}}{2i}(b_k e^{+i\omega_k t} - b_k^+ e^{-i\omega_k t}) \\ S - \sum_k b_k^+ b_k \end{pmatrix}$$

The time dependency is known and directly reflects the spin wave energies:

$$\begin{aligned} \langle b_k b_k^+(t)\rangle &= (1 + n_B(\omega_k))e^{-i\omega_k t} \\ \langle b_k^+ b_k(t)\rangle &= n_B(\omega_k)e^{+i\omega_k t} \end{aligned}$$

Cross section (inelastic + elastic)

$$\begin{aligned} \mathcal{S}(Q,\omega) &= S\left(1 + \frac{Q^z\ Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\sum_\tau \\ &\times \sum_k \{n_B(\omega_k)\delta(\omega+\omega_k)\ \delta(Q+k-\tau)\ +\ (1+n_B(\omega_k))\delta(\omega-\omega_k)\ \delta(Q-k-\tau)\} \\ &+ \left(1 - \frac{Q^z\ Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\langle S\rangle^2\sum_\tau \delta(\omega)\ \delta(Q-\tau) \end{aligned}$$

Ferromagnet

$$\mathcal{S}(Q,\omega) = S\left(1+\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\sum_{\tau}$$

$$\times\sum_{k}\{n_B(\omega_k)\delta(\omega+\omega_k)\ \delta(Q+k-\tau)\ +\ (1+n_B(\omega_k))\delta(\omega-\omega_k)\ \delta(Q-k-\tau)\}$$

$$+\left(1-\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\langle S\rangle^2\sum_{\tau}\delta(\omega)\ \delta(Q-\tau)$$

Elastic term : Bragg peaks with structure factor

Note the geometric factor (is zero if Q is along z)

Ferromagnet

Inelastic term :

$$
\begin{aligned}
\mathcal{S}(Q,\omega) \quad = \quad & S\left(1+\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\sum_{\tau} \\
& \times\sum_{k}\{n_B(\omega_k)\delta(\omega+\omega_k)\;\delta(Q+k-\tau)\;+\;(1+n_B(\omega_k))\delta(\omega-\omega_k)\;\delta(Q-k-\tau)\} \\
& +\left(1-\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\langle S\rangle^2\sum_{\tau}\delta(\omega)\;\delta(Q-\tau)
\end{aligned}
$$

Elastic term : Bragg peaks with structure factor
Note the geometric factor (is zero if Q is along z)

Ferromagnet

Periodic (reciprocal lattice)

Inelastic term :

$$\begin{aligned}
\mathcal{S}(Q,\omega) \;&=\; S\left(1+\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\sum_{\tau} \\
&\times\sum_{k}\{n_B(\omega_k)\delta(\omega+\omega_k)\;\delta(Q+k-\tau)\;+\;(1+n_B(\omega_k))\delta(\omega-\omega_k)\;\delta(Q-k-\tau)\} \\
&+\left(1-\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\langle S\rangle^2\sum_{\tau}\delta(\omega)\;\delta(Q-\tau)
\end{aligned}$$

Elastic term : Bragg peaks with structure factor
Note the geometric factor (is zero if Q is along z)

Ferromagnet

Periodic (reciprocal lattice)

Creation and annihilation & Detailed Balance

Inelastic term :

$$\mathcal{S}(Q,\omega) = S\left(1+\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\sum_\tau$$

$$\times\sum_k\{n_B(\omega_k)\delta(\omega+\omega_k)\;\delta(Q+k-\tau)\;+\;(1+n_B(\omega_k))\delta(\omega-\omega_k)\;\delta(Q-k-\tau)\}$$

$$+\left(1-\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\langle S\rangle^2\sum_\tau\delta(\omega)\;\delta(Q-\tau)$$

Elastic term : Bragg peaks with structure factor

Note the geometric factor (is zero if Q is along z)

Ferromagnet

Periodic (reciprocal lattice)

Creation, annihilation & Detailed Balance

Geometric factor (maximum when Q is along z)

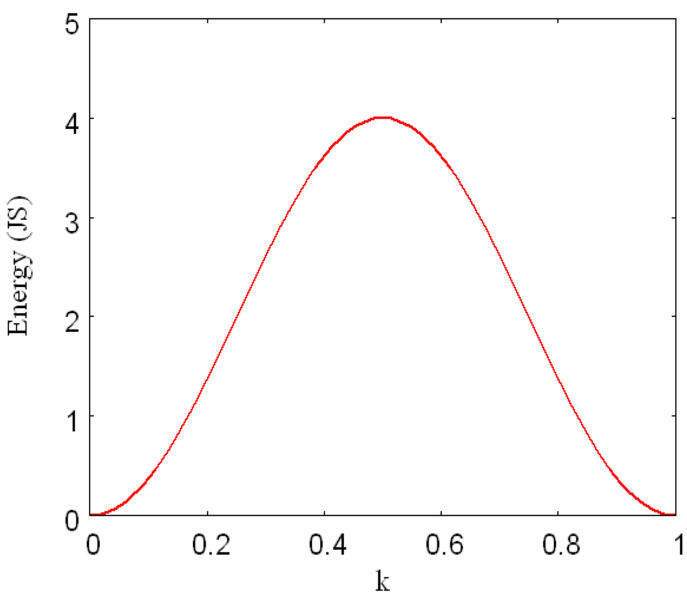
Inelastic term :

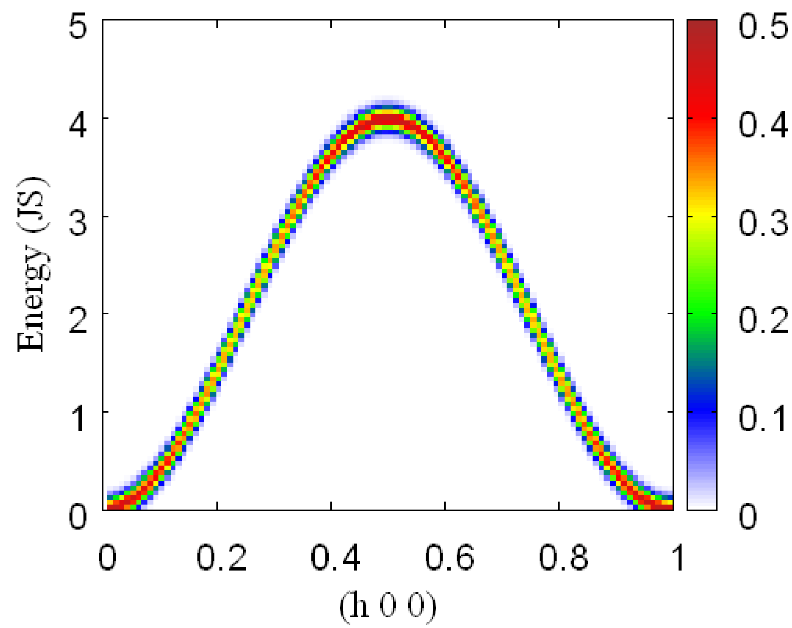
$$\begin{aligned}
\mathcal{S}(Q,\omega) \; &= \; S\left(1+\frac{Q^z\,Q^z}{Q^2}\right)\,\frac{(2\pi)^3}{v_o}\,\sum_{\tau} \\
&\times \sum_{k}\left\{n_B(\omega_k)\delta(\omega+\omega_k)\;\delta(Q+k-\tau)\;+\;(1+n_B(\omega_k))\delta(\omega-\omega_k)\;\delta(Q-k-\tau)\right\} \\
&+\left(1-\frac{Q^z\,Q^z}{Q^2}\right)\,\frac{(2\pi)^3}{v_o}\,\langle S\rangle^2\,\sum_{\tau}\,\delta(\omega)\;\delta(Q-\tau)
\end{aligned}$$

Elastic term : Bragg peaks with structure factor

Note the geometric factor (is zero if Q is along z)

$$\begin{aligned}\mathcal{S}(Q,\omega) &= S\left(1+\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\sum_\tau \\ &\times\sum_k \{n_B(\omega_k)\delta(\omega+\omega_k)\;\delta(Q+k-\tau)\;+\;(1+n_B(\omega_k))\delta(\omega-\omega_k)\;\delta(Q-k-\tau)\} \\ &+\left(1-\frac{Q^z\,Q^z}{Q^2}\right)\frac{(2\pi)^3}{v_o}\,\langle S\rangle^2\sum_\tau\,\delta(\omega)\;\delta(Q-\tau)\end{aligned}$$





Outline

- Introduction
- Cross section

- Localized excitations : Crystal field modes
- « Nearly » localized modes : excitons
- Collective excitations : spin waves

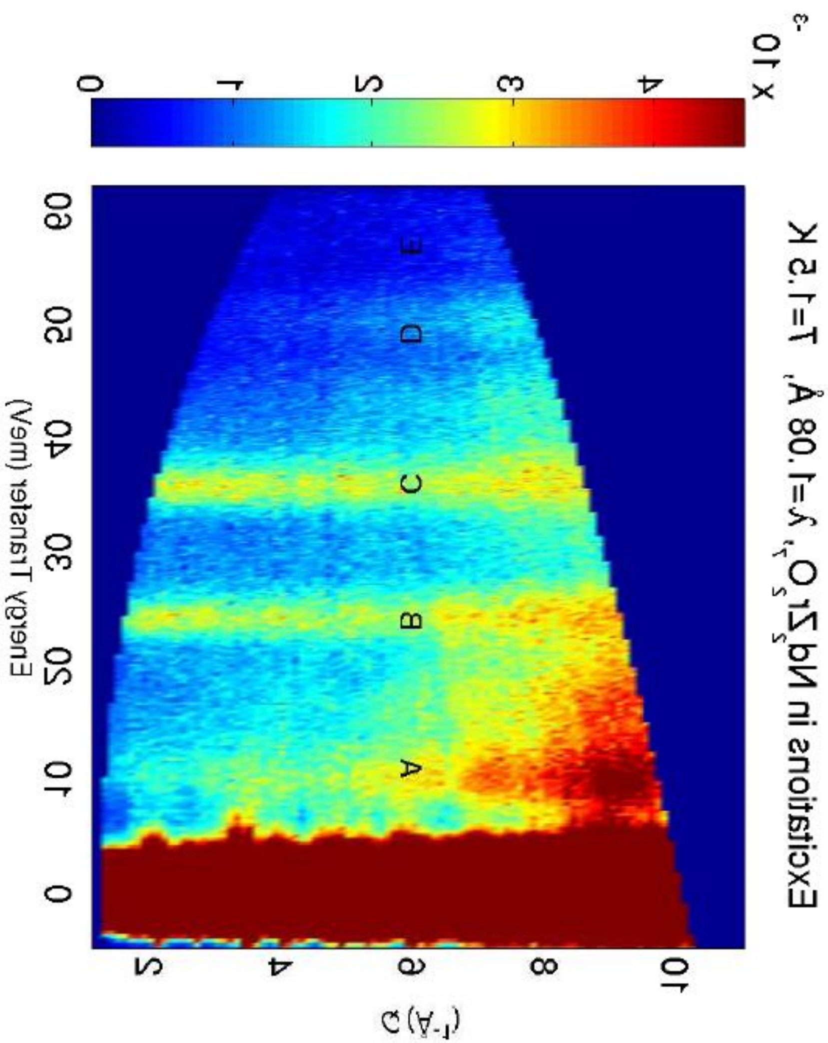
$\Delta S=1$
Collective excitations
Well defined modes

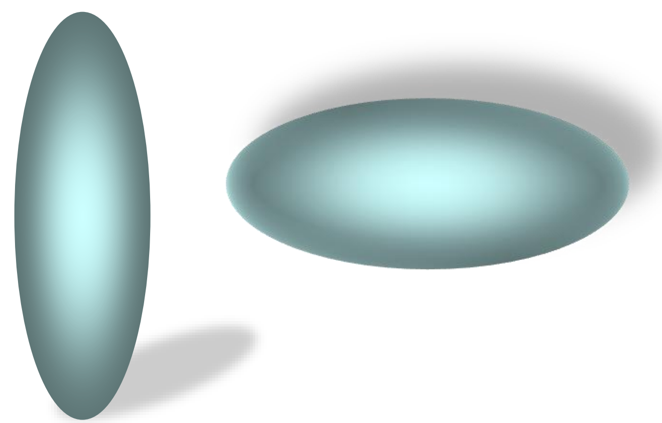
- Quantum magnets (spin ½ and spin 1)
- Metals

$\Delta S=1/2$
Collective excitations
Continuum

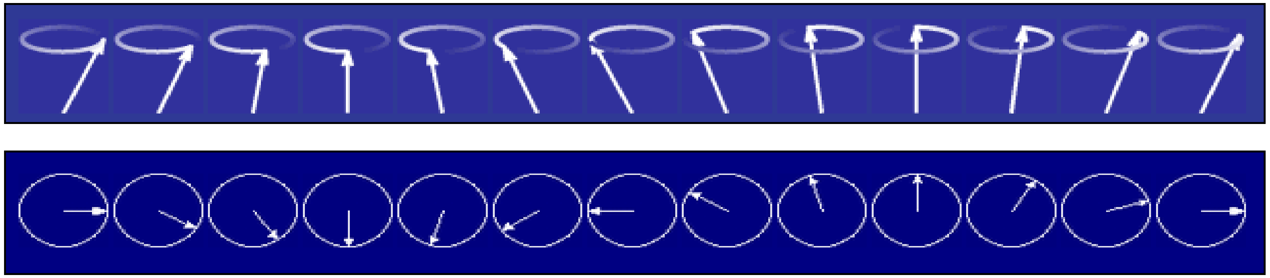
Possible emergence of bound states and of well defined modes

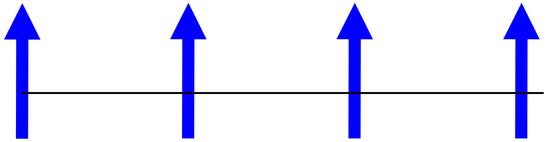
Crystal field

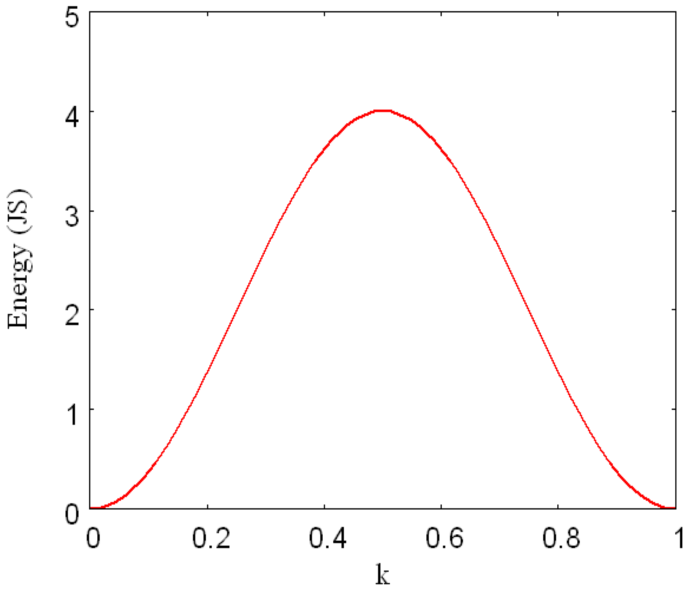


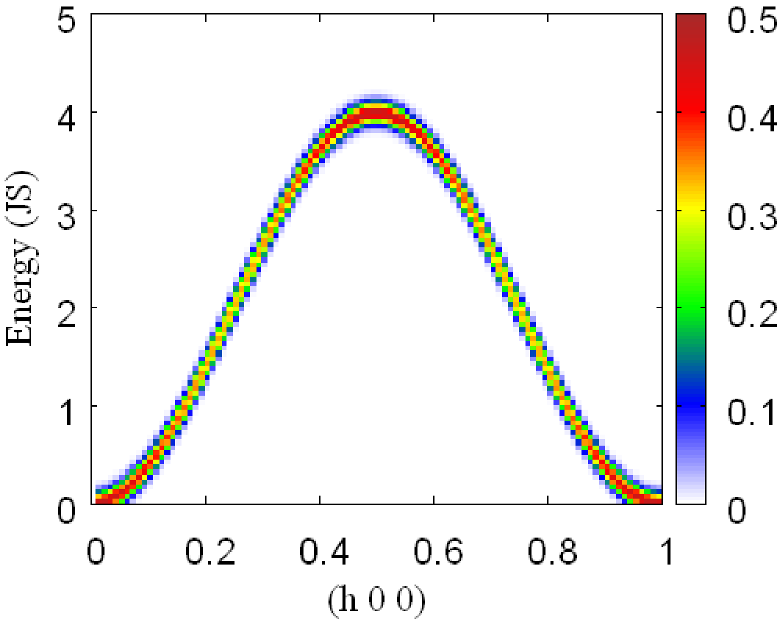


Ferromagnet



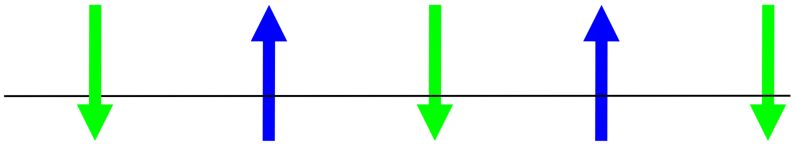


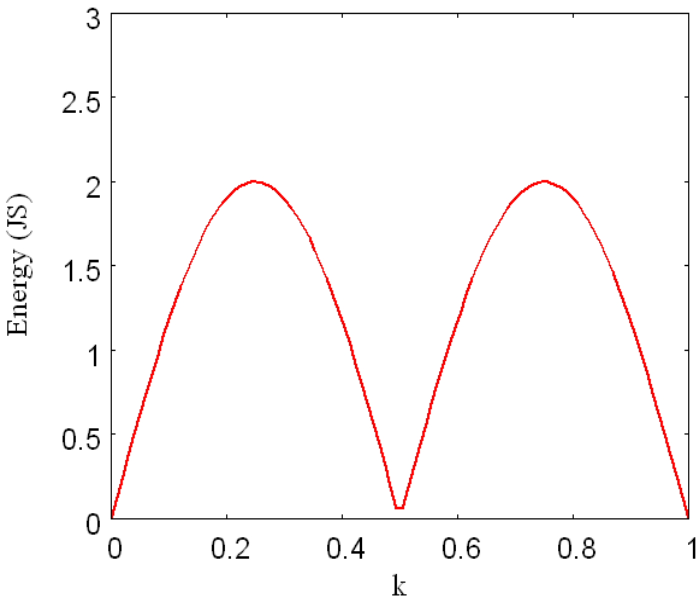


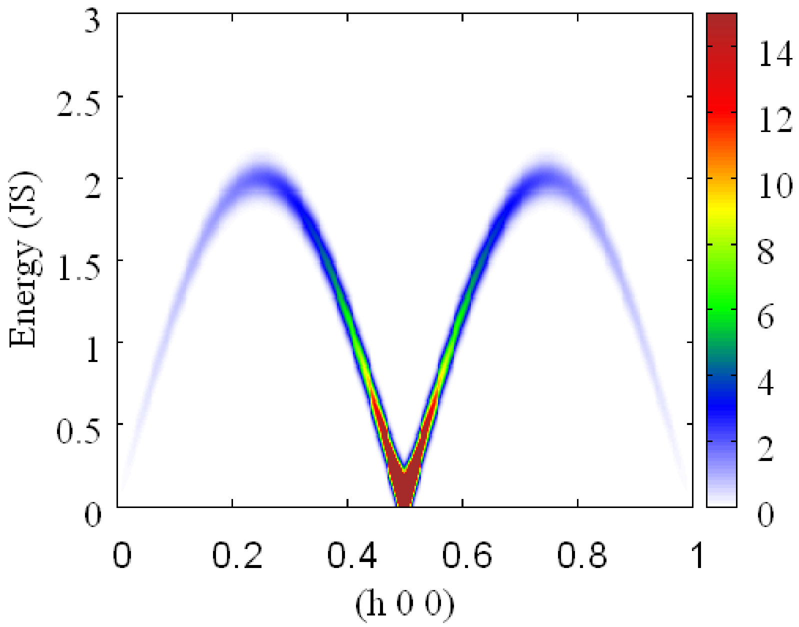


Antiferromagnet

$$
\begin{aligned}
\mathcal{S}(Q,\omega) &= I_{\text{Bragg}}\ \delta(\omega) \\
&+ S\,\frac{(2\pi)^3}{v_o}\sum_{s}\sum_{\tau,k} \\
&\quad |A_{k,s}|^2\ n_B(E_{k,s})\delta(\omega+E_{k,s})\ \delta(Q+k-\tau)\ + \\
&\quad |A_{k,s}|^2(1+n_B(E_{k,s}))\delta(\omega-E_{k,s})\ \delta(Q-k-\tau)
\end{aligned}
$$



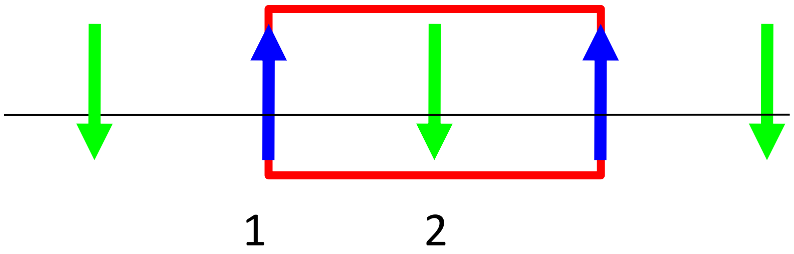




Antiferromagnet

AF Heisenberg Hamiltonian : distinguish up and down spins

$$\mathcal{H} = \frac{1}{2} \sum_{m,n} \mathbf{S}_{m,1} J \, \mathbf{S}_{n,2} \; + \; \mathbf{S}_{m,2} J \, \mathbf{S}_{n,1}$$



« Holstein-Primakov » representation for the two sites (two quantification axes)

$$\begin{aligned} S^x_{m,1} &\approx \frac{\sqrt{2S}}{2} \left(b_{m,1} + b^+_{m,1}\right) \\ S^y_{m,1} &\approx \frac{\sqrt{2S}}{2i} \left(b_{m,1} - b^+_{m,1}\right) \\ S^z_{m,1} &\approx S - b^+_{m,1} \, b_{m,1} \end{aligned}$$

$$\begin{aligned} S^x_{m,2} &\approx + \frac{\sqrt{2S}}{2} \left(b_{m,2} + b^+_{m,2}\right) \\ S^y_{m,2} &\approx - \frac{\sqrt{2S}}{2i} \left(b_{m,2} - b^+_{m,2}\right) \\ S^z_{m,2} &\approx - \left(S - b^+_{m,2} \, b_{m,2}\right) \end{aligned}$$

Antiferromagnet

Keep 2nd order terms (small deviations)

$$
\begin{aligned}
\mathcal{H} \;\approx\; & \frac{1}{2}\sum_{m,n}\frac{S}{2}(b_{m,1} + b^{+}_{m,1})\; J\; (b_{n,2} + b^{+}_{n,2}) + \frac{S}{2}(b_{m,1} - b^{+}_{m,1})\; J\; (b_{n,2} - b^{+}_{n,2}) \\
& - \;(S - b^{+}_{m,1}b_{m,1})\; J\; (S - b^{+}_{n,2}b_{n,2}) \\
& + \;1 \leftrightarrow 2
\end{aligned}
$$

$$
\begin{aligned}
\mathcal{H} \;\approx\; & \frac{1}{2}\sum_{m,n} JS(b_{m,1}\, b_{n,2} + b^{+}_{m,1}\, b^{+}_{n,2}) + J\, S\, \left(b^{+}_{m,1}b_{m,1} + b^{+}_{n,2}b_{n,2}\right) \\
& + \;1 \leftrightarrow 2
\end{aligned}
$$

Fourier Transform :

$$
\begin{aligned}
\mathcal{H} \;\approx\; & \frac{1}{2}\sum_{k} J\, S\, (\gamma_{-k}\, b_{k,1}\, b_{-k,2} + \gamma_k\, b^{+}_{-k,1}\, b^{+}_{k,2}) + J\, S\, \left(b^{+}_{k,1}b_{k,1} + b^{+}_{k,2}b_{k,2}\right) \\
& + \; J\, S\, (\gamma_{-k}\, b_{k,2}\, b_{-k,1} + \gamma_k\, b^{+}_{-k,2}\, b^{+}_{k,1}) + J\, S\, \left(b^{+}_{k,2}b_{k,2} + b^{+}_{k,1}b_{k,1}\right)
\end{aligned}
$$

$$
\mathcal{H} \approx \frac{1}{2}\sum_{k} \left(b^{+}_{k,1}\; b^{+}_{k,2}\; b_{-k,1}\; b_{-k,2}\right)
\begin{pmatrix}
z\,J\,S & 0 & 0 & z\,J\,S\,\gamma_{-k} \\
0 & z\,J\,S & z\,J\,S\,\gamma_{-k} & 0 \\
0 & z\,J\,S\,\gamma_k & z\,J\,S & 0 \\
z\,J\,S\,\gamma_k & 0 & 0 & z\,J\,S
\end{pmatrix}
\begin{pmatrix}
b_{k,1} \\ b_{k,2} \\ b^{+}_{-k,1} \\ b^{+}_{-k,2}
\end{pmatrix}
$$

Antiferromagnet: Bogolubov transform

We need a final transform to get a free bosons Hamiltonian.

$$\begin{pmatrix} b_{k,1} \\ b_{k,2} \\ b^+_{-k,1} \\ b^+_{-k,2} \end{pmatrix} = P \begin{pmatrix} y_{k,1} \\ y_{k,2} \\ y^+_{-k,1} \\ y^+_{-k,2} \end{pmatrix}$$

$$\mathcal{H} = \sum_k E_{k,1}\; y^+_{k,1} y_{k,1} \;+\; E_{k,2}\; y^+_{k,2} y_{k,2}$$

This transformation (due to Bogolubov) is a « rotation » defined as:

$$B \;=\; P\, Y \qquad\qquad P^{-1} = P^+$$

We impose that the Hamiltonian describes free independent bosons:

$$B^+ \; h \; B = Y^+ \; E \; Y \qquad\qquad P^+ \; h \; P = E$$

Since the Y are bosons :

$$[B, B^+] = g$$

$$[Y, Y^+] = g$$

$$g = \begin{pmatrix} 1 & & & & & \\ & .. & & & & \\ & & 1 & & & \\ & & & -1 & & \\ & & & & .. & \\ & & & & & -1 \end{pmatrix}$$

$$g = P \; g \; P^+$$

Antiferromagnet: Bogolubov transform

$$P^{-1} = P^{+}$$
$$P^{+}\ h\ P = E$$
$$g = P\ g\ P^{+}$$

$$(g\ h)\ P \quad = \quad (P\ g\ P^{+})\ h\ P = P\ (g\ E)$$

1. The spin wave energies are eigenvalues of the (g h) matrix (and not of the h matrix)
2. Because of the Bogolubov transform, the number of deviations at low T is **not** zero !

$$g\ h = \frac{z\ J\ S}{2} \begin{pmatrix} 1 & 0 & 0 & \gamma_{-k} \\ 0 & 1 & \gamma_{-k} & 0 \\ 0 & -\gamma_k & -1 & 0 \\ -\gamma_k & 0 & 0 & -1 \end{pmatrix}$$

$$E_k = \pm \frac{zJS}{2}\sqrt{1 - |\gamma_k|^2}$$

Antiferromagnet: Bogolubov transform

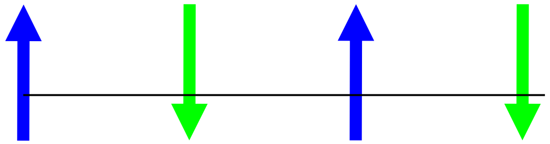
$$P = \begin{pmatrix} u & & & v^* \\ & u & v^* & \\ & v & u^* & \\ v & & & u^* \end{pmatrix}$$

$$u_k^2 - v_k^2 = 1$$

$$\begin{aligned} u_k^2 &= \frac{1}{2}\left(+1 + \frac{z\ J\ S}{E_k}\right) \\ v_k^2 &= \frac{1}{2}\left(-1 + \frac{z\ J\ S}{E_k}\right) \\ u_k\ v_k &= \frac{z\ J\ S\ \gamma_k}{2\ E_k} \end{aligned}$$

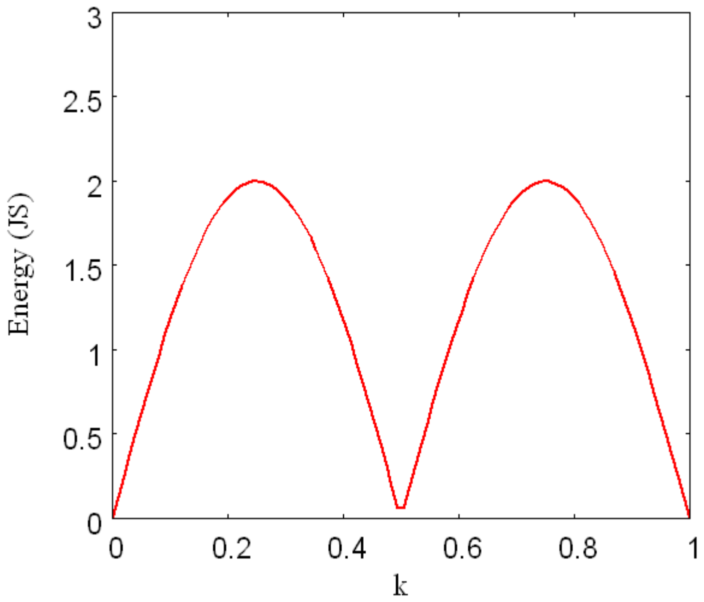
Antiferromagnet

1. Spin wave energies



$$E_k = \pm \frac{zJS}{2}\sqrt{1 - |\gamma_k|^2}$$

$$\gamma_k = \frac{1}{z}\sum_{\Delta} e^{ik\Delta}$$



Zone center Zone boundary Zone center

Linear dispersion at small k

$$E_k \sim |k|$$

2. Average deviation away from equilibrium direction (at low T): reduction of the average moment

Bogolubov transform

$$b_{k,1} = u\ y_{k,1}\ +\ v\ y^{+}_{-k,2}$$

Average deviation

$$\langle b^{+}_{k,1} b_{k,1} \rangle = u^2\ \langle y^{+}_{k,1} y_{k,1} \rangle\ +\ v^2\ \langle y_{-k,2} y^{+}_{-k,2} \rangle$$

Quantum effect

$$\langle b^{+}_{k,1} b_{k,1} \rangle = u^2\ n_B(E_{k,1})\ +\ v^2\ [1\ +\ n_B(E_{k,2})]$$

2. Average deviation away from equilibrium direction (at low T): reduction of the average moment

$$\langle S^z \rangle \approx S - \sum_k \langle b_k^+ \ b_k \rangle$$

$$\langle S \rangle \approx S - \sum_k v_k^2 + (u_k^2 + v_k^2) \ n_B(E_k)$$

$$\langle S \rangle \approx S + \frac{1}{2} - \sum_k \frac{z \ J \ S}{E_k} \left(n_B(E_k) + \frac{1}{2} \right)$$

Thermal fluctuations

Quantum fluctuations

The vacuum of the Bogolubov bosons is not « the vaccum of deviations »

Antiferromagnet

$$\sum_k \frac{z\ J\ S}{E_k}\left(n_B(E_k)+\frac{1}{2}\right)\longrightarrow\int dk\ \frac{k^{d-1}}{(2\pi)^d}\frac{1}{k}\left(\frac{T}{k}+\frac{1}{2}\right)$$

The thermal fluctuations prevent long range ordering for $d \leq 2$

The Quantum fluctuations already destroy LRO in $d \leq 1$

Breakdown of the spin wave theory is consistent with Mermin and Wagner theorem

Antiferromagnet

To calculate the cross section from spin waves, we calculte the spin in terms of the Bogolubov bosons:

$$\sum_m e^{i\mathbf{k}\mathbf{R}_m}\,\mathbf{S}_{m,1}(t) = \mathbf{S}_{k,1} = \begin{pmatrix} \frac{\sqrt{2S}}{2}(b_{k,1} + b^{+}_{-k,1}) \\ \frac{\sqrt{2S}}{2i}(b_{k,1} - b^{+}_{-k,1}) \\ S - \sum_k b^{+}_{k,1} b_{k,1} \end{pmatrix}$$

$$\sum_m e^{i\mathbf{k}\mathbf{R}_m}\,\mathbf{S}_{m,2}(t) = \mathbf{S}_{k,2} = \begin{pmatrix} + \frac{\sqrt{2S}}{2}(b_{k,2} + b^{+}_{-k,2}) \\ - \frac{\sqrt{2S}}{2i}(b_{k,2} - b^{+}_{-k,2}) \\ - \left(S - \sum_k b^{+}_{k,2} b_{k,2}\right) \end{pmatrix}$$

The time dependency is known and directly reflects the spin wave energies:

$$\begin{aligned} \langle y_k y_k^{+}(t)\rangle &= (1 + n_B(E_k))e^{-iE_k t} \\ \langle y_k^{+} y_k(t)\rangle &= n_B(E_k)e^{+iE_k t} \end{aligned}$$

Cross section

$$\begin{aligned} \mathcal{S}(Q,\omega) &= I_{\text{Bragg}}\,\delta(\omega) \\ &+ S\,\frac{(2\pi)^3}{v_o}\sum_s\sum_{\tau,k} \\ &\quad |A_{k,s}|^2\, n_B(E_{k,s})\delta(\omega + E_{k,s})\,\delta(Q + k - \tau)\; + \\ &\quad |A_{k,s}|^2(1 + n_B(E_{k,s}))\delta(\omega - E_{k,s})\,\delta(Q - k - \tau) \end{aligned}$$

Antiferromagnet

Elastic term : Bragg peaks with structure factor
Geometric factor (is zero if Q is along z)

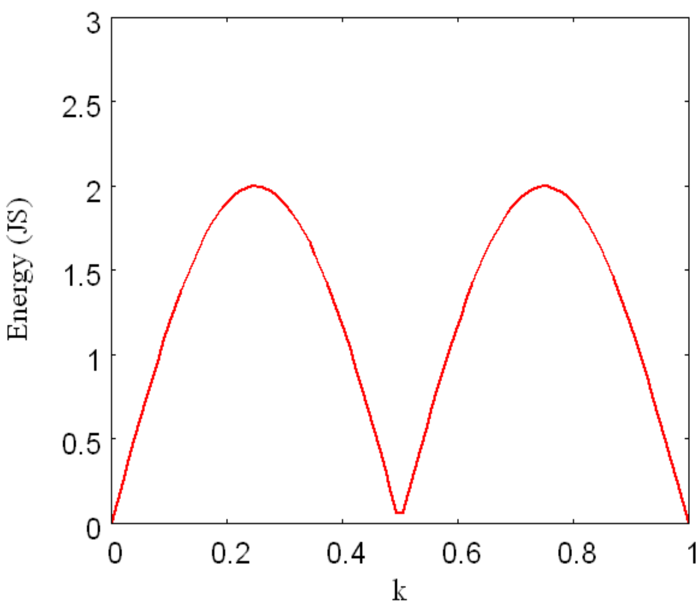
$$
\begin{aligned}
\mathcal{S}(Q,\omega) \quad &= \quad I_{\text{Bragg}}\ \delta(\omega) \\
&+ \quad S\ \frac{(2\pi)^3}{v_o} \sum_s \sum_{\tau,k} \\
&\quad |A_{k,s}|^2\ n_B(E_{k,s})\delta(\omega + E_{k,s})\ \delta(Q + k - \tau)\ + \\
&\quad |A_{k,s}|^2 (1 + n_B(E_{k,s}))\delta(\omega - E_{k,s})\ \delta(Q - k - \tau)
\end{aligned}
$$

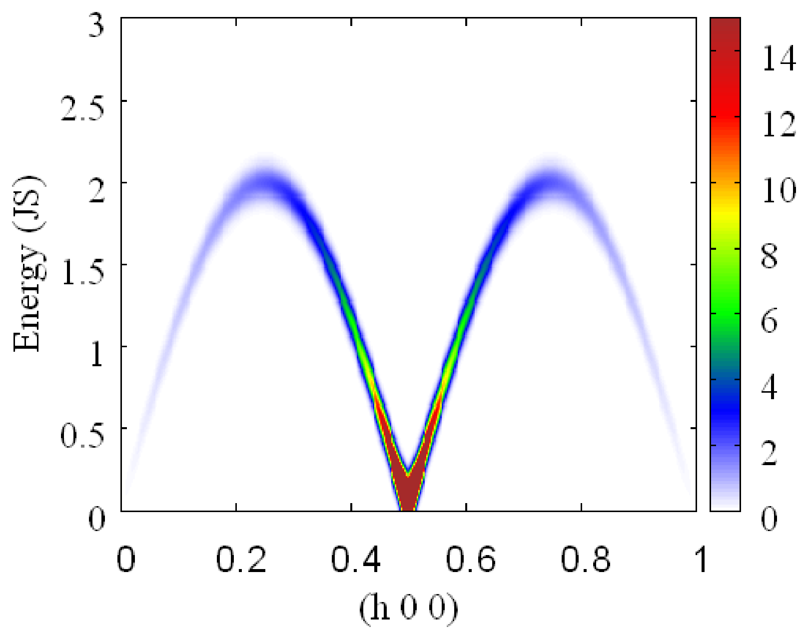
Inelastic term :

Periodic (reciprocal lattice)
Creation, annihilation & Detailed Balance
Geometric factor (maximum when Q is along z)

Antiferromagnet

$$\mathcal{S}(Q,\omega) = I_{\text{Bragg}}\ \delta(\omega)$$
$$+ \quad S\ \frac{(2\pi)^3}{v_o} \sum_s \sum_{\tau,k}$$
$$|A_{k,s}|^2\ n_B(E_{k,s})\delta(\omega + E_{k,s})\ \delta(Q + k - \tau)\ +$$
$$|A_{k,s}|^2(1 + n_B(E_{k,s}))\delta(\omega - E_{k,s})\ \delta(Q - k - \tau)$$





General case

Cross section

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

« detailed balance » Creation Annihilation

$$n(\omega_{Q,s}) = \frac{1}{e^{\frac{\hbar\omega_{Q,s}}{k_B T}}-1}$$

- Conventional magnets (molecular field)
- S=1 bosonic excitations
- L spins in the magnetic unit cell : L branches

- The theory is wrong at high T
- The theory is badly false for d=1,2
- Notice the « detailed balance »

Antiferromagnet

$$A_s \sim \sum_{\ell} P_{\ell,s}\ e^{\ i\mathbf{Q}\mathbf{r}_\ell}$$

Cross section of the s^{th} mode
=
Interference effect between
« partial » spin fluctuations of
the spin ℓ in mode s

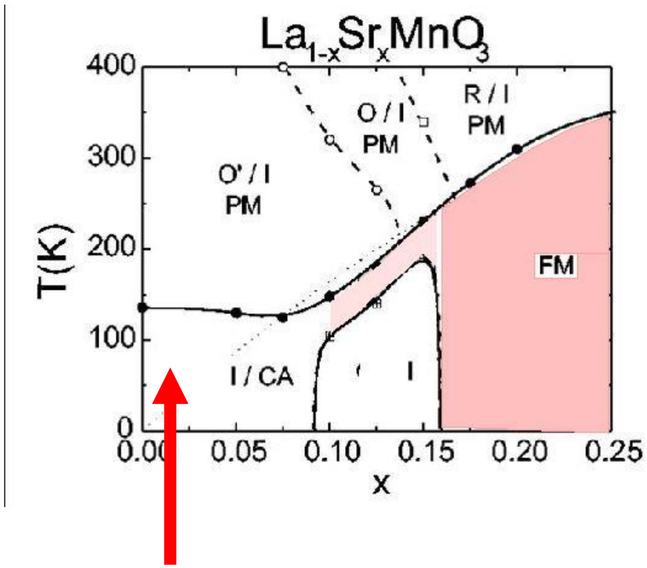
$$F_s \sim \sum_{\ell} b_\ell \frac{\hbar}{\sqrt{M_\ell \omega_{k,s}}}\ (\mathbf{Q} \cdot \mathbf{e}_{k,\ell,s})\ e^{\ i\mathbf{Q}\mathbf{r}_\ell}$$

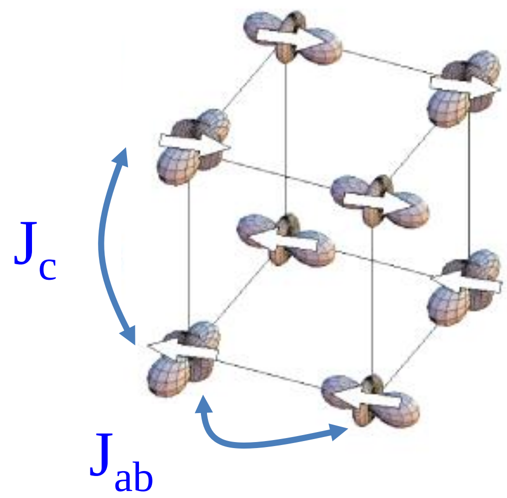
Cross section (phonons) of the
s^{th} mode =
Interference effect between
« partial » atomic motion of
the atom ℓ in mode s

3. Collective excitations : spin waves

EXAMPLES

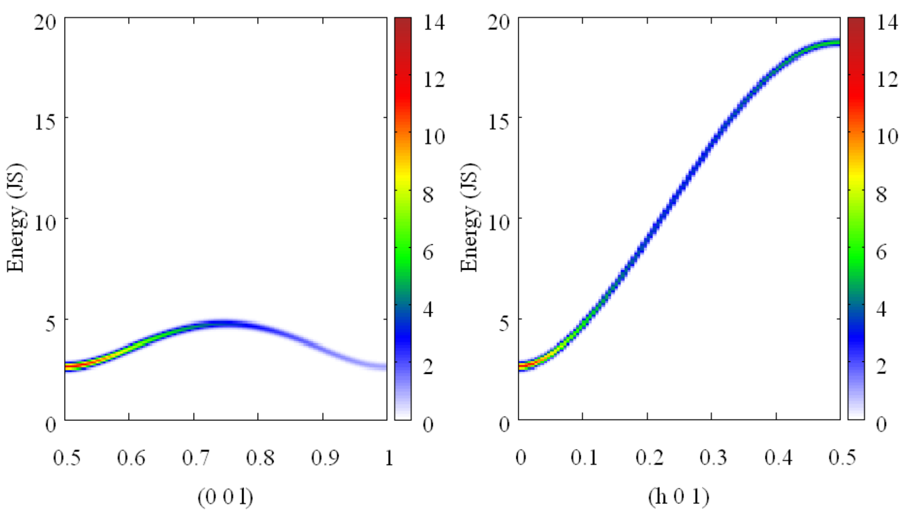
Manganites

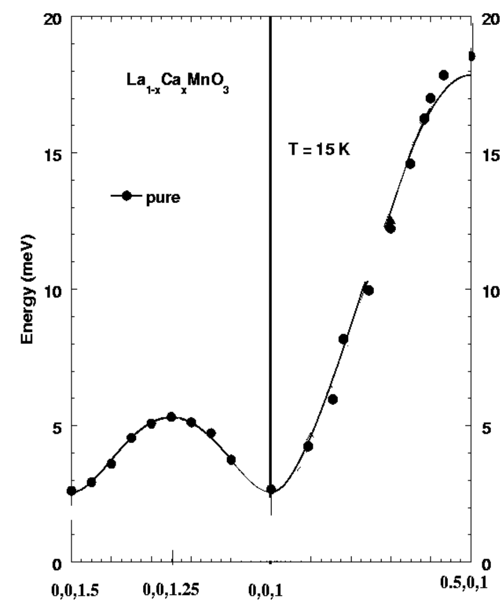




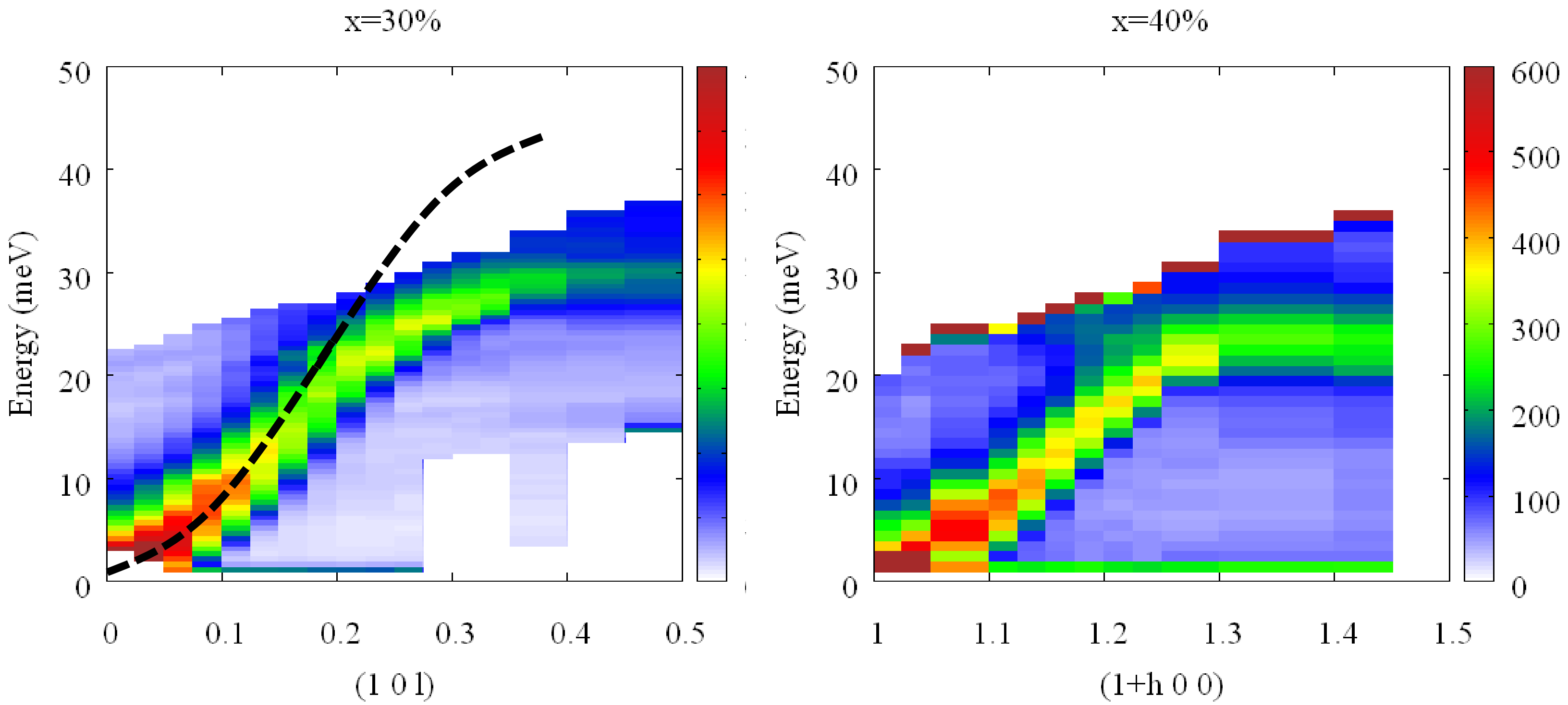
J_c ~ 0.5 meV

J_{ab} ~ -0.8 meV

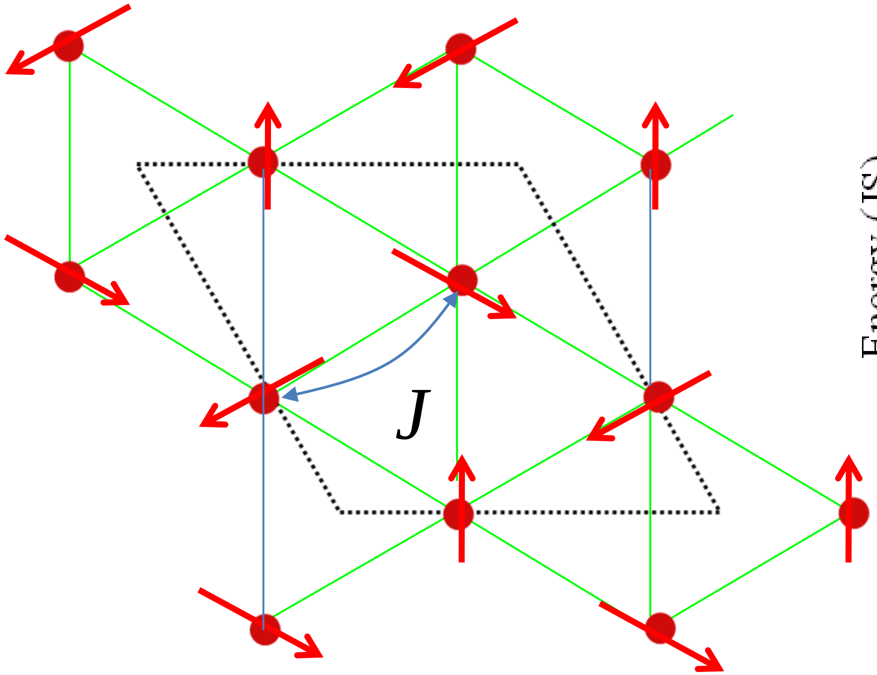




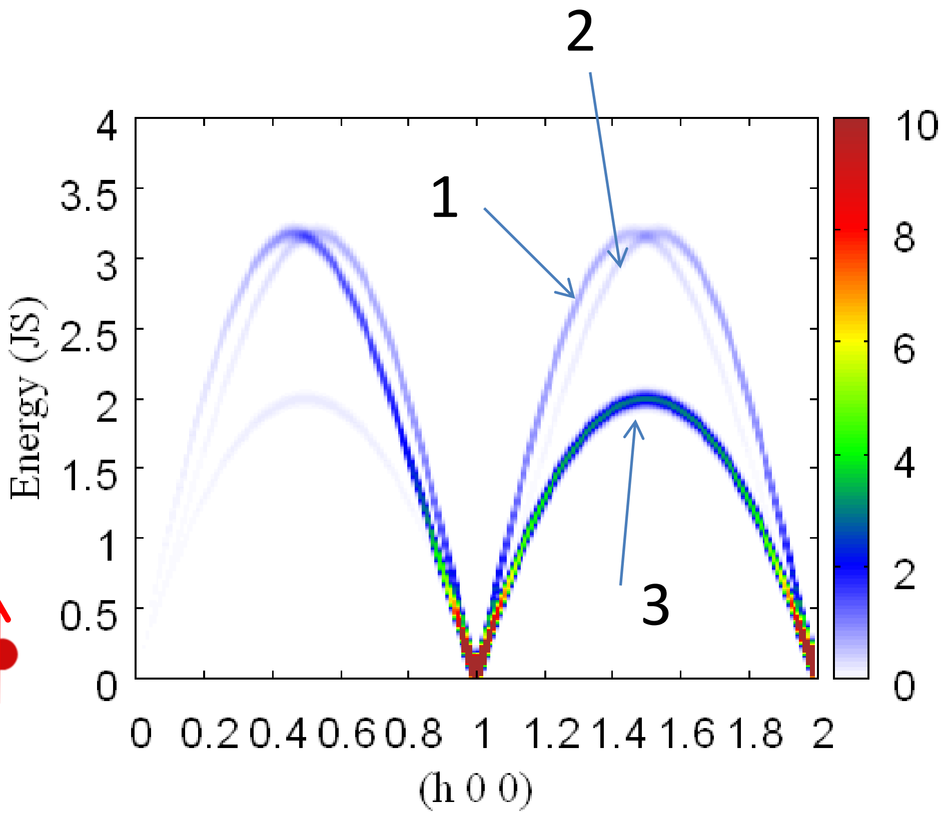
Spin waves also in the metallic state of (doped) manganites



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



120° Néel order

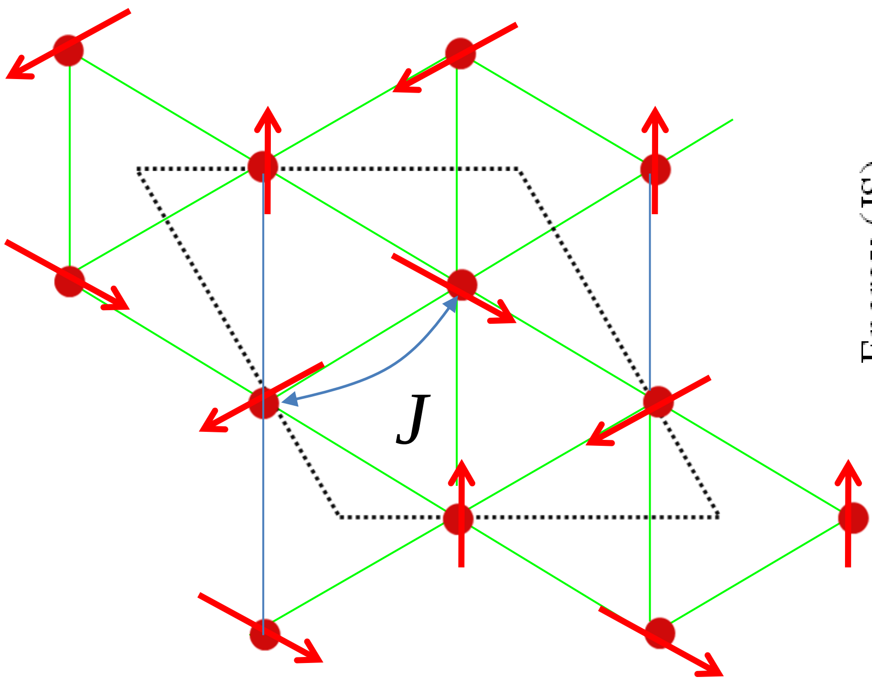


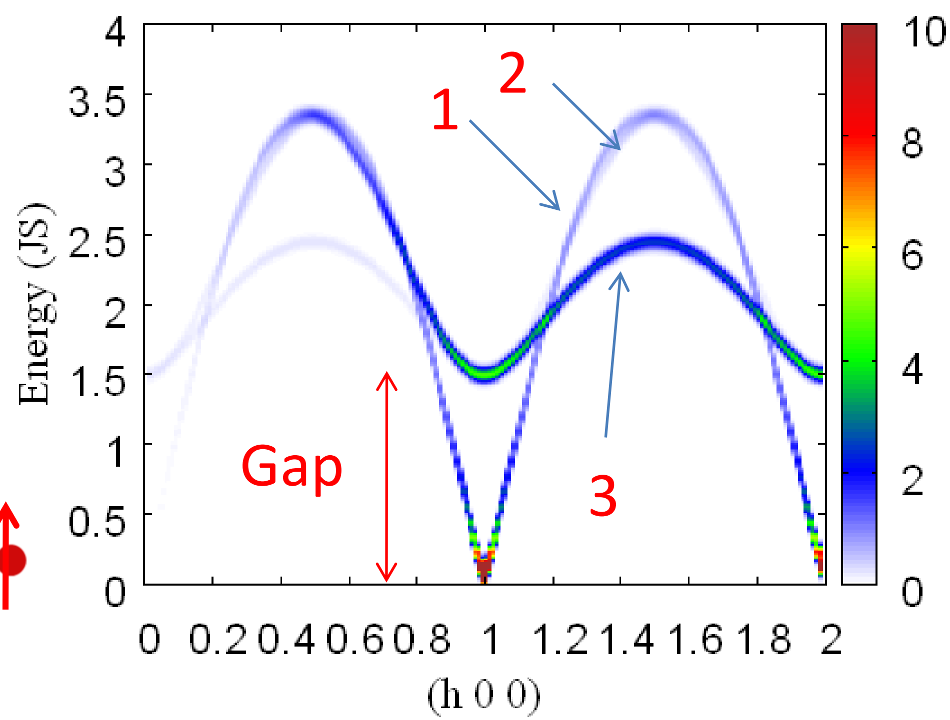
3 spins per unit cell : 3 branches

1, 2 : correlations between in plane spin components

3 : correlations between out of plane spin components

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





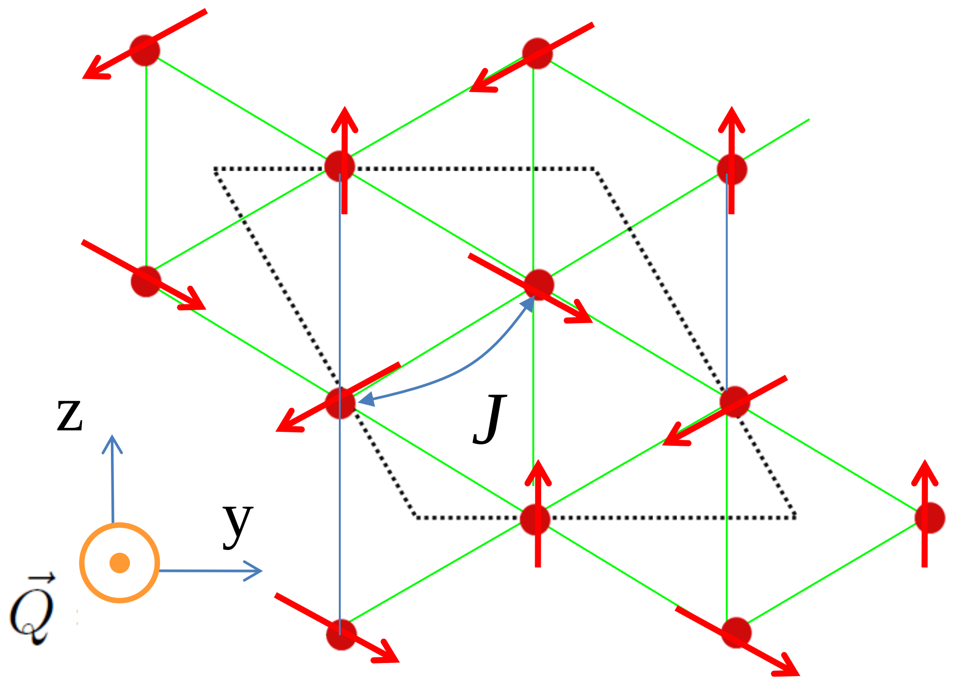
3 spins per unit cell : 3 branches

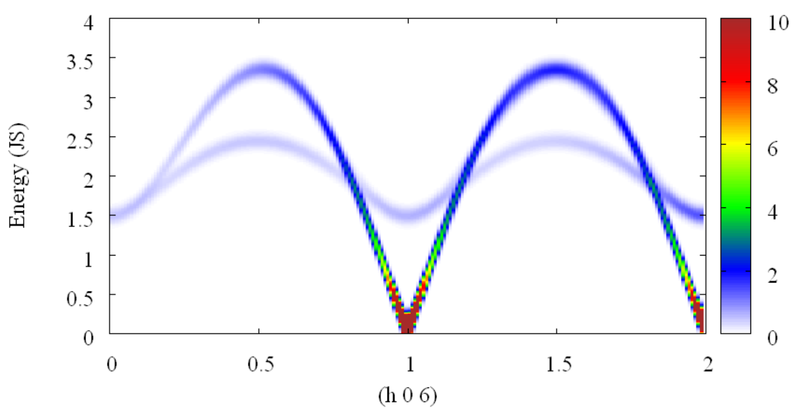
1, 2 : correlations between in plane spin components

3 : correlations between out of plane spin components

Q perpendicular to the hexagonal (yz) plane

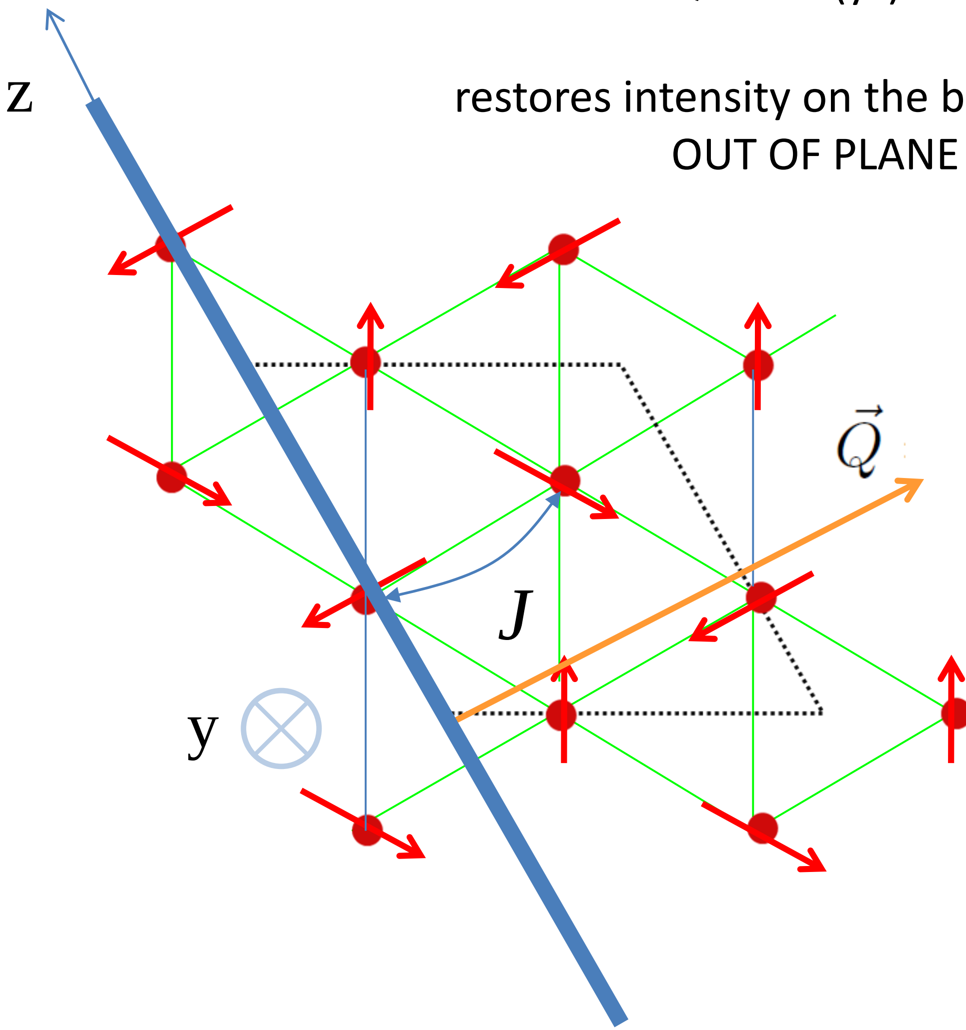
Enlightens the branches corresponding to IN PLANE correlations

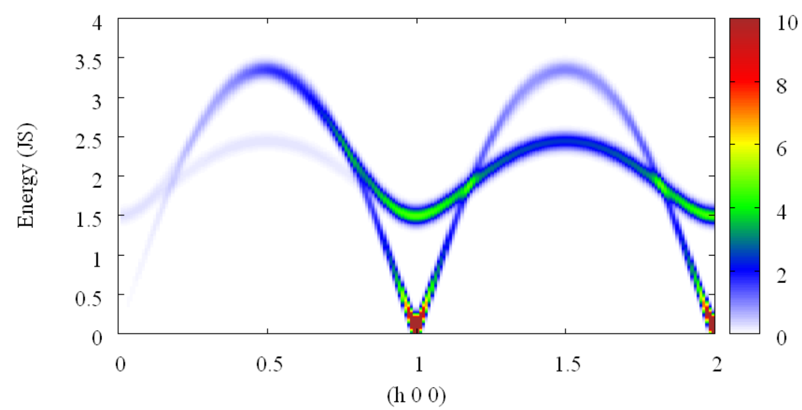




Q in the (yz) hexagonal case

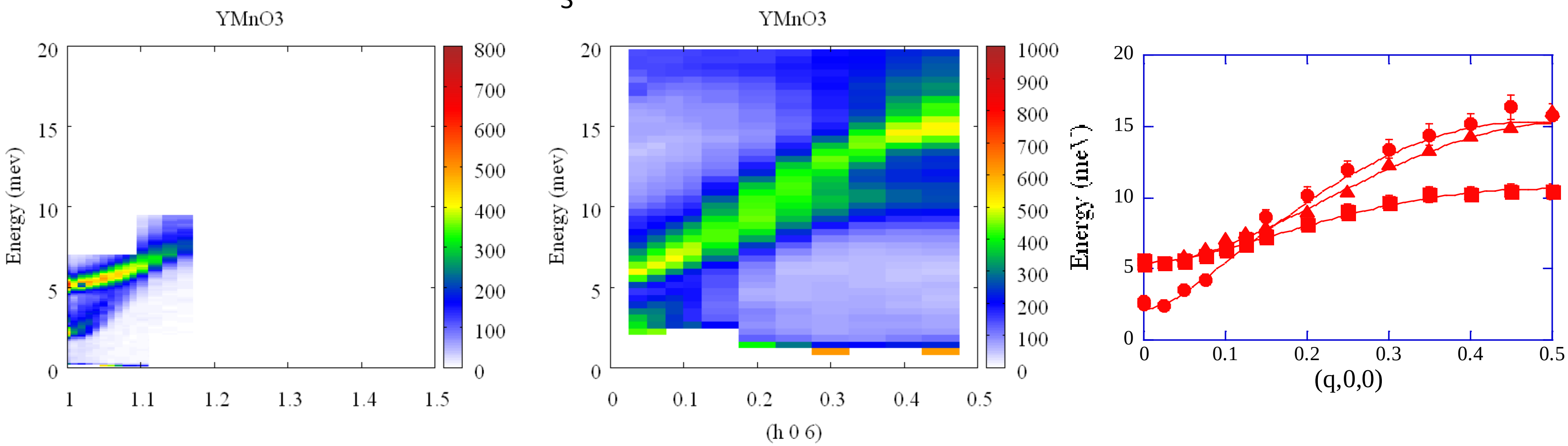
restores intensity on the branch corresponding to
OUT OF PLANE correlations

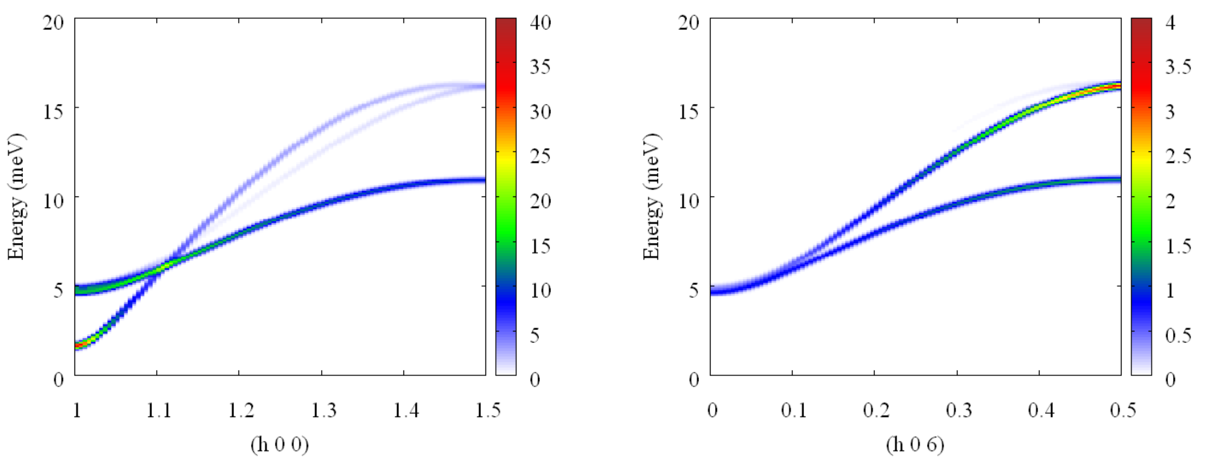




Triangular lattice

YMnO$_3$



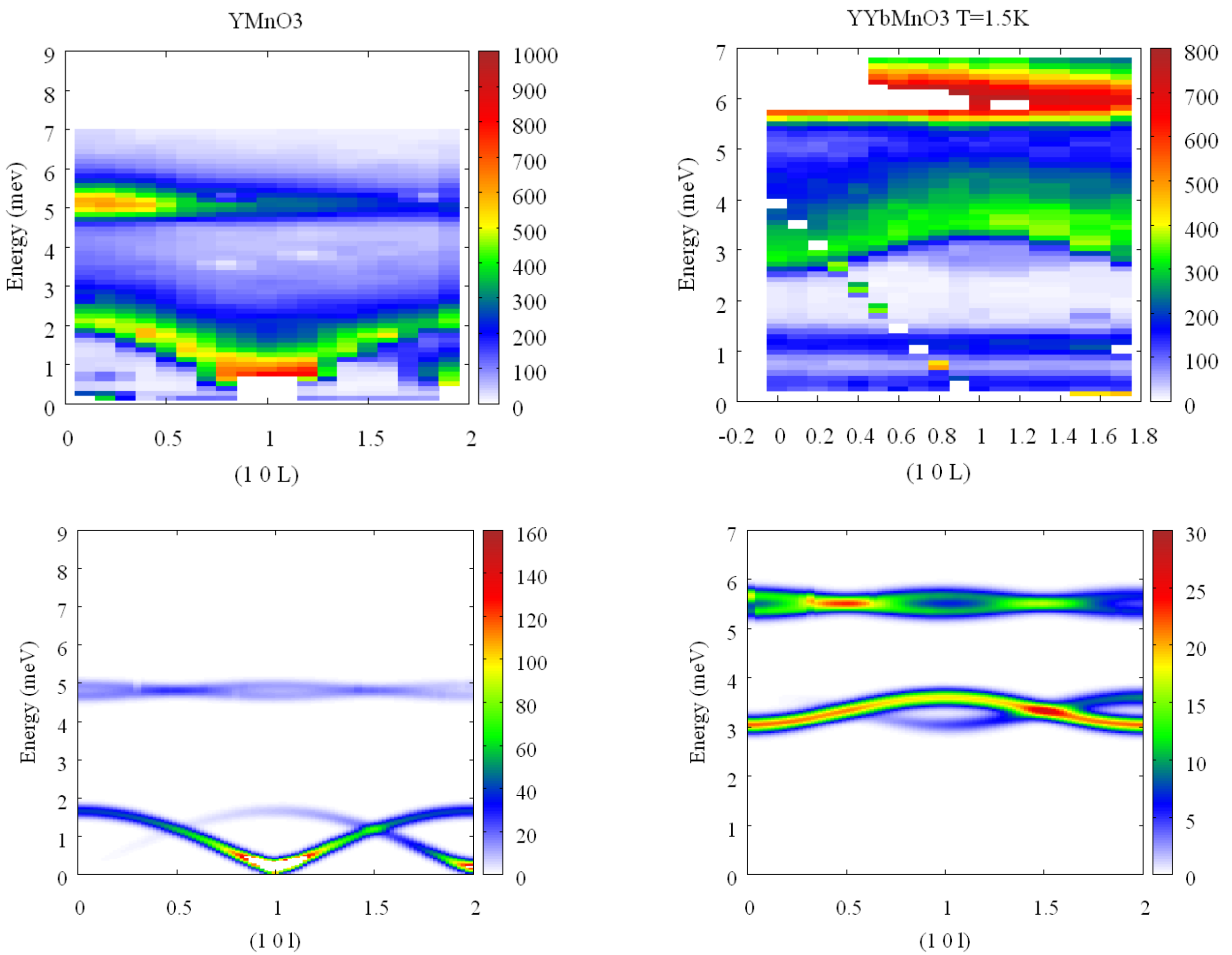


Deduce microscopic parameters from experiments

J = 2.5 meV
D = 0.5 meV

Triangular lattice

YMnO$_3$



YbMnO$_3$

Pyrochlore ferromagnet $Yb_2Ti_2O_7$

QUANTUM EXCITATIONS IN QUANTUM SPIN ICE PHYS. REV. X **1**, 021002 (2011)

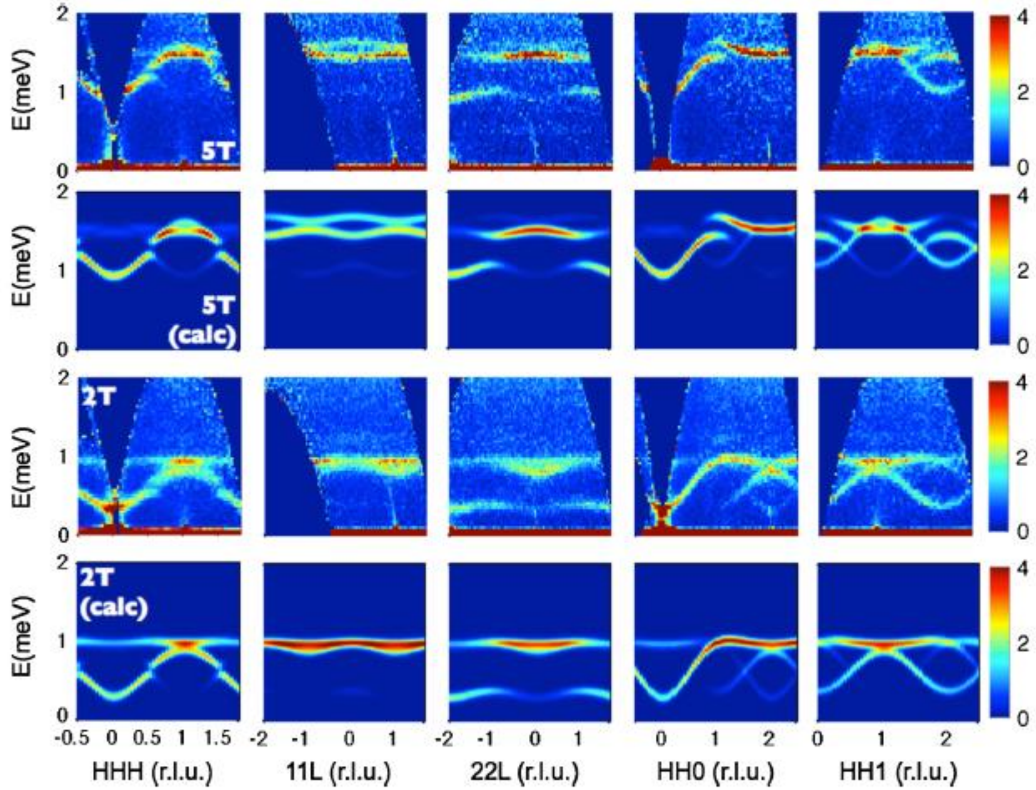


FIG. 1. The measured $S(\mathbf{Q}, \omega)$ at $T = 30$ mK, sliced along various directions in the HHL plane, for both $H = 5$ T (first row) and $H = 2$ T (third row). The second and fourth rows show the calculated spectrum for these two field strengths, based on an anisotropic exchange model with five 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 $Er_2Ti_2O_7$

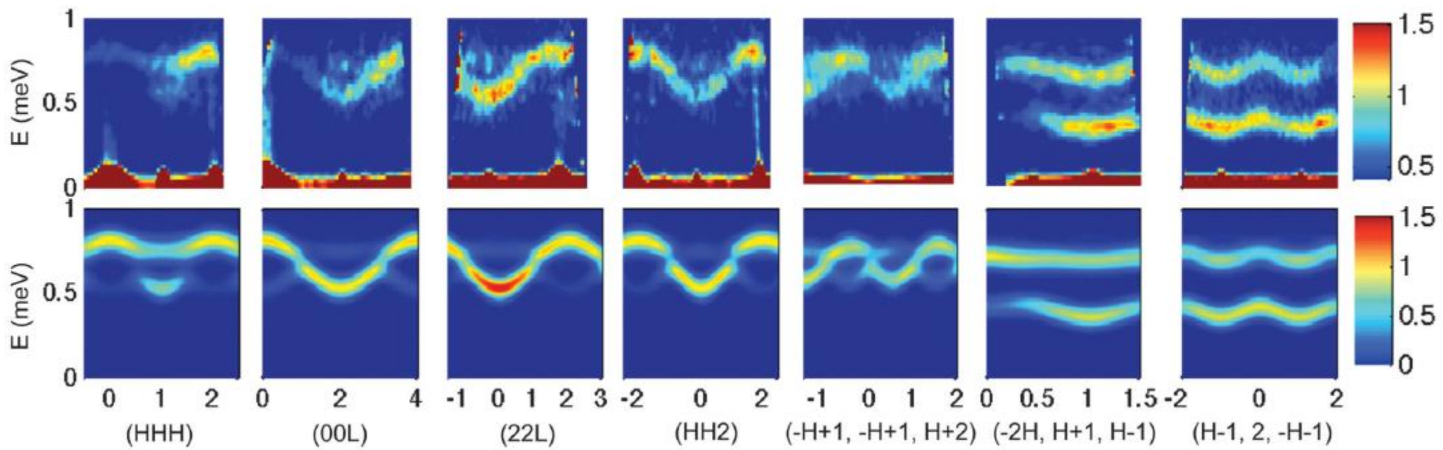
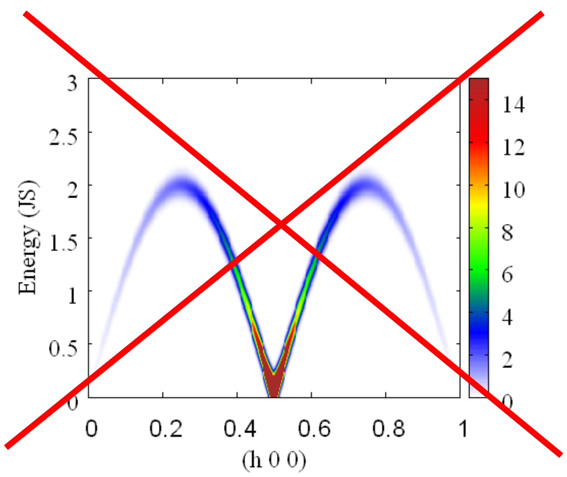


FIG. 1 (color). The measured $S(\mathbf{Q}, \omega)$ at $T = 30$ mK, $H = 3$ T sliced along several directions. The first five columns show $S(\mathbf{Q}, \omega)$ in the *HHL* plane, with the field applied along $[1\bar{1}0]$, while the last two columns show $S(\mathbf{Q}, \omega)$ for the field along [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.

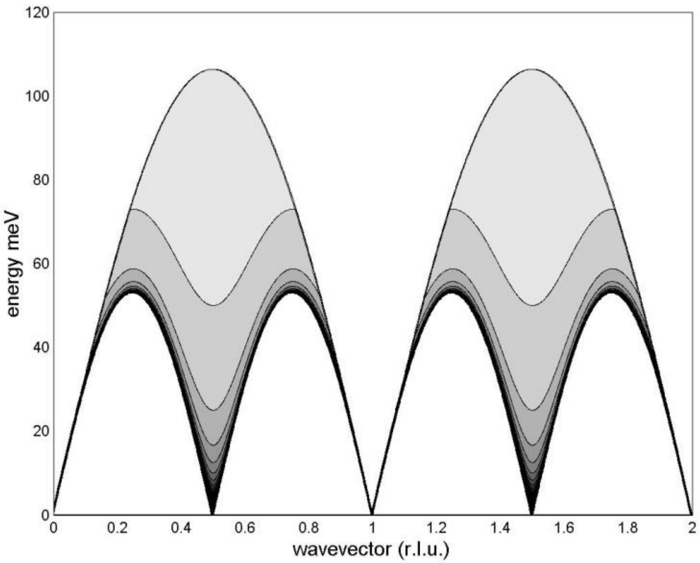
4. Quantum magnets

- Have no long range order (quantum fluctuations melt the classical Néel order).
- Power law decaying spin correlations
- No spin wave excitations…



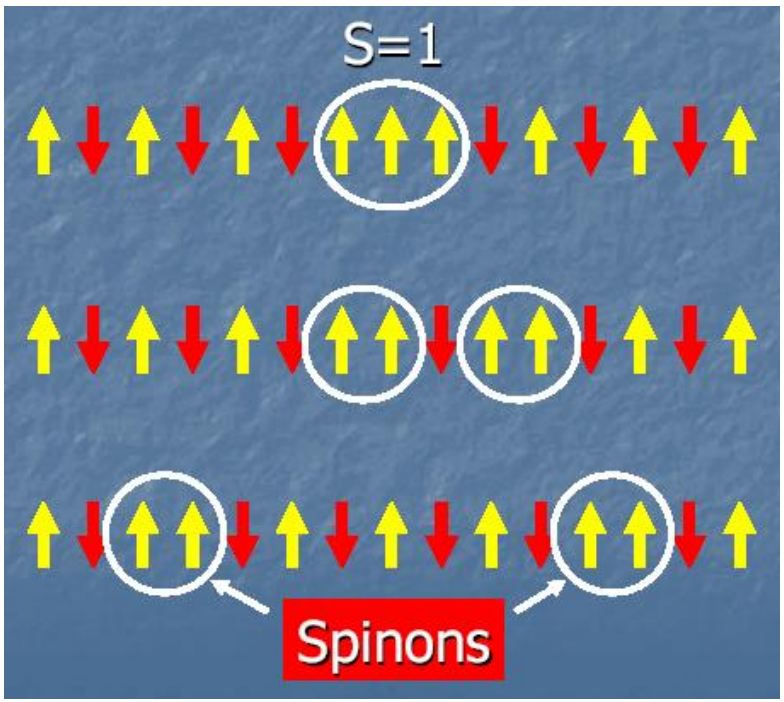
but dispersing spin ½ quasiparticles called « spinons ». To match ΔS=1, INS see a 2-particles continuum :

$$\begin{aligned} \omega &= E_q + E_{q'} \\ \mathbf{k} &= \mathbf{q} + \mathbf{q}' \end{aligned}$$

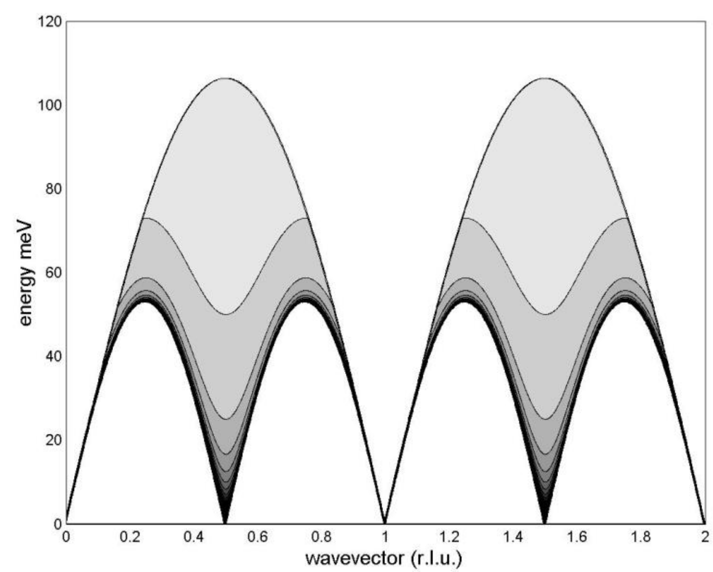


Muller *et al* PRB **24**, 1429 (1981).
Des Cloizeaux & Pearson,
Phys. Rev. 128 (1962), 2131

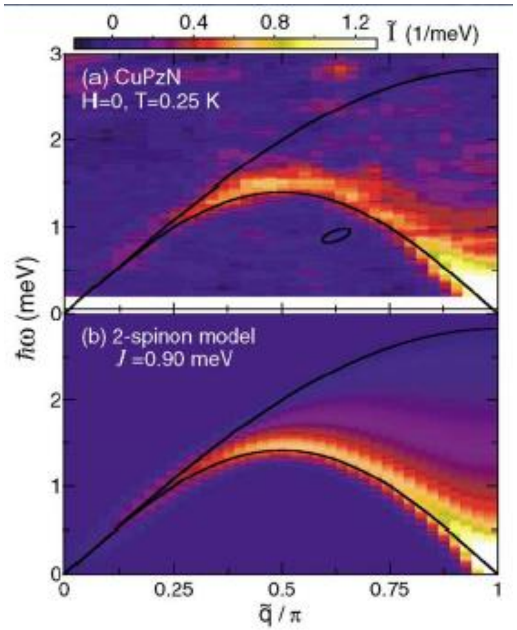
And best understood in the Ising case, where they correspond to domain walls that separate Néel configurations



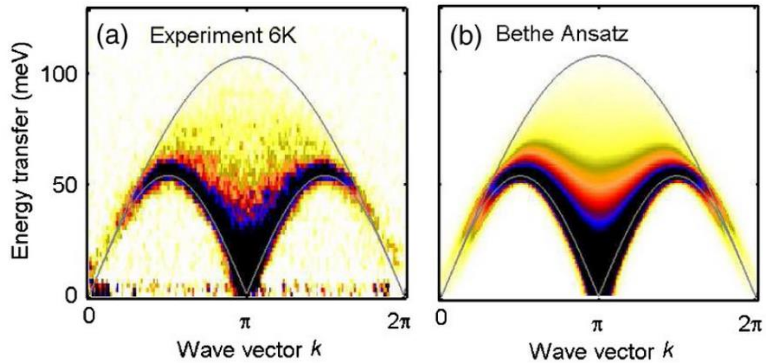
Cartoon by F. Mila



4. Quantum magnets



CuPzN, Stone et al, PRL 2003



$KCuF_3$, Lake et al, Nature Materials 2005

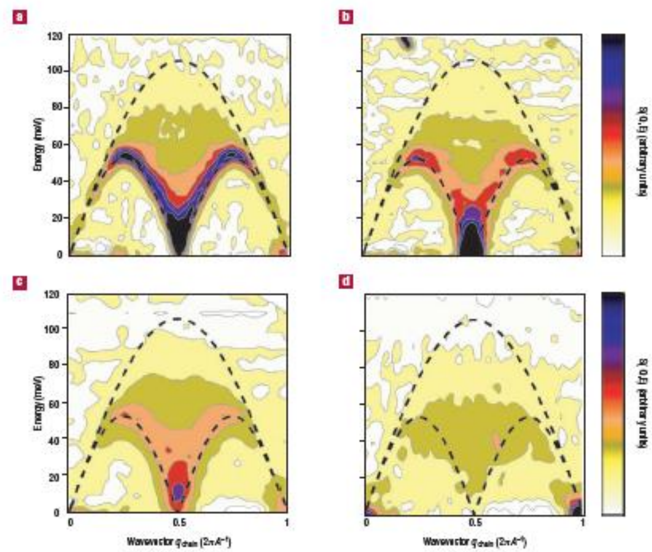
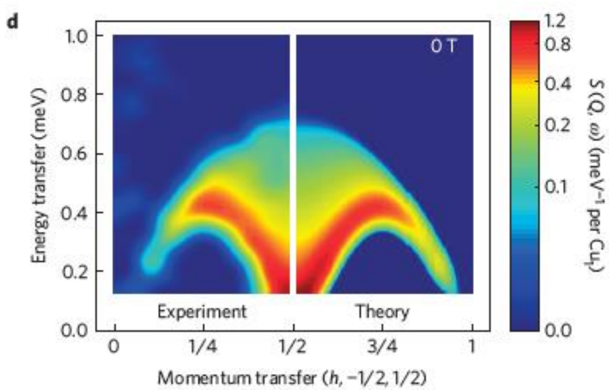


Figure 1 Inelastic neutron scattering data for $KCuF_3$. The data is plotted as a function of E and q parallel to the chains for the temperatures a, $T = 6$ K, b, $T = 50$ K, c, $T = 150$ K and d, $T = 300$ K. The colours indicate the size of the neutron scattering cross-section $S(q,E)$ and the superimposed 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

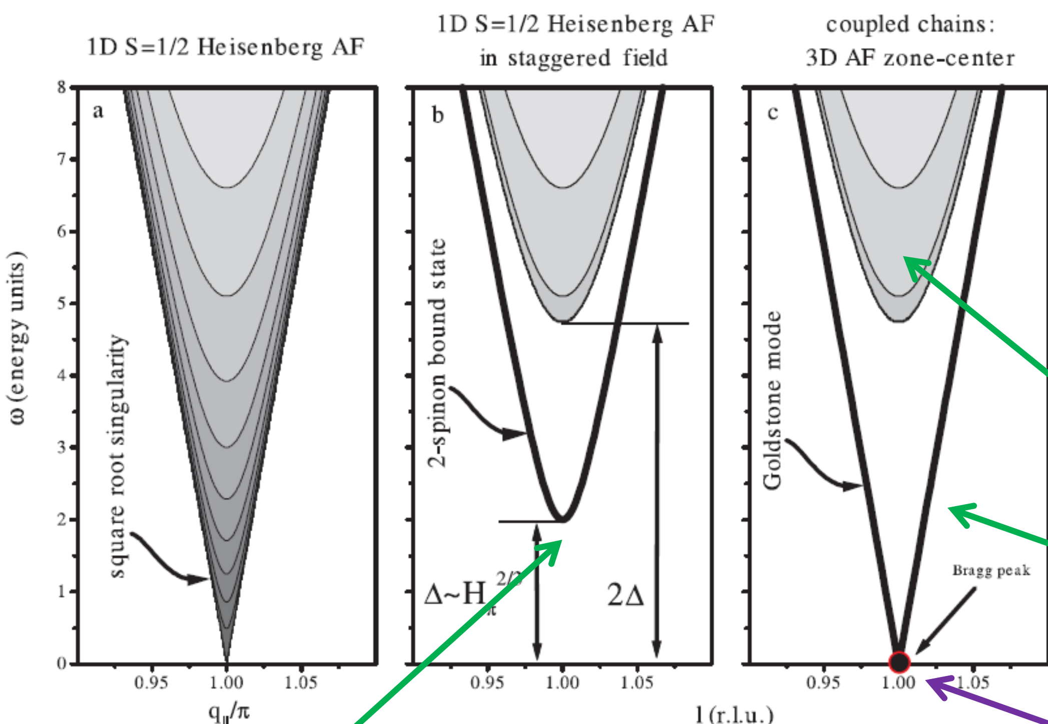
4. Quantum magnets

Physical realizations of 1D systems eventually order at low temperature, owing to small inter-chain couplings

Nature of the cross-over from 1D to 3D ?

4. Quantum magnets

H creates bound states between spinons … the 3 flavours transform into 2 transverse modes



Single chain in a staggered field H
RPA approximation

$\Delta \sim H^{2/3}$

Continuum

Bound state
Goldstone
Transverse modes

Bragg peak

with a gap at zone boundaries

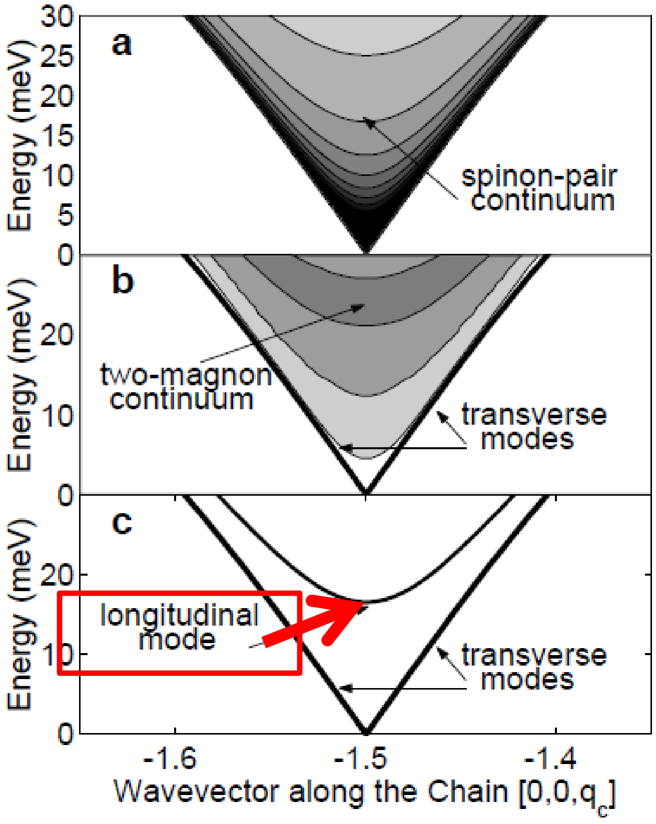
Cartoon from A. Zheludev

Appl. Phys. A 74 [Suppl.], S1–S5 (2002)

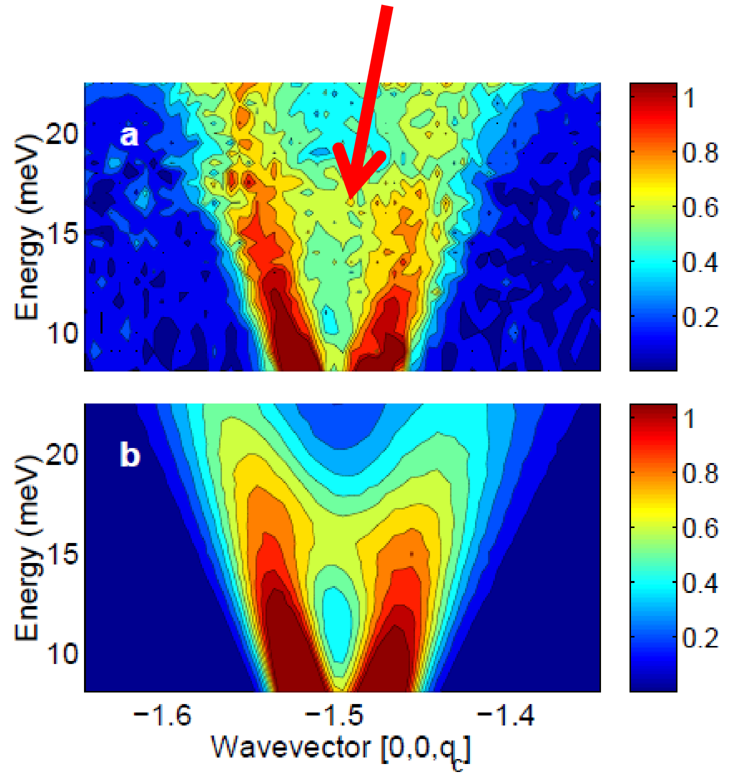
H. L. Essler, A.M. Tsvelik, G. Delfino,
Phys. Rev. B **56**, 11 001 (1997).
H. J. Schulz
Phys. Rev. Lett. **77**, 2790 (1996).

Bound states of spinons

+ an additional Amplitude (Higgs) mode; it signals the cross-over from 1D to 3D behaviour and indicates a reduction in the ordered spin moment.



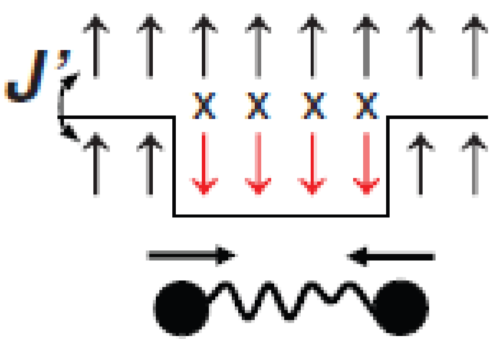
Overdamped L mode

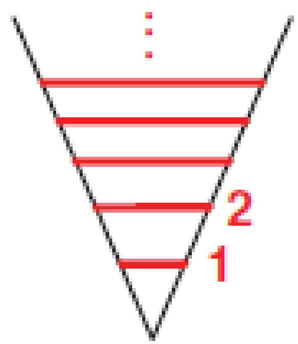


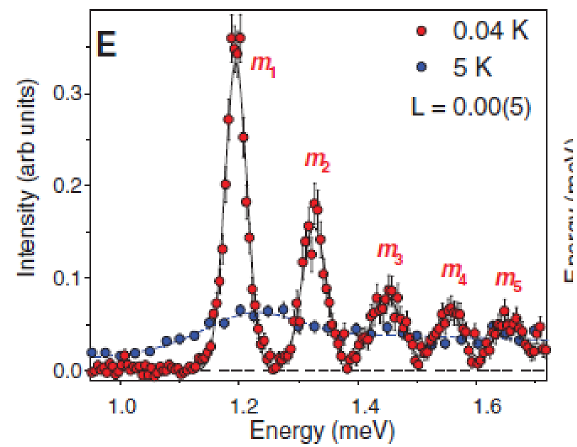
$KCuF_3$, Lake et al, Nature

Bound states of spinons

The neighboring chains induce a linear potential giving rise to bound states at specific values (negative zeroes of the Airy function). Ex $CoNb_2O_6$

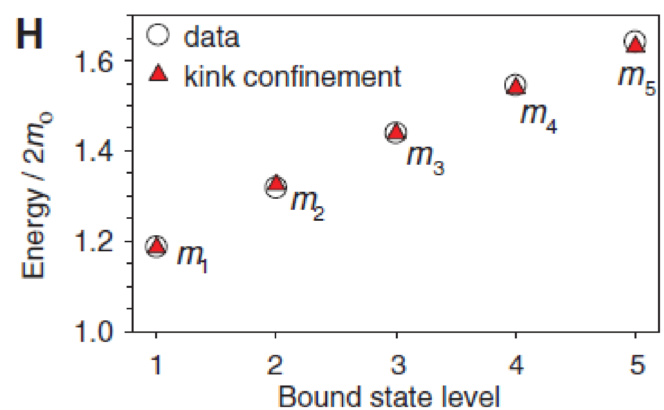






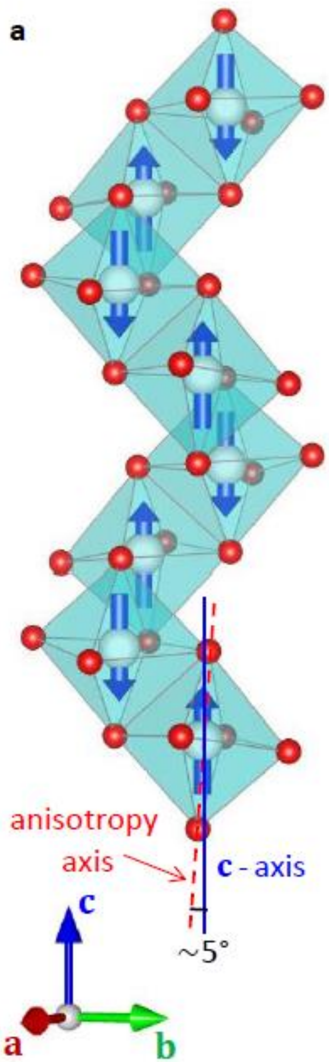
$$m_j = 2m_o + z_j \lambda^{2/3} \left(\frac{\hbar^2}{\mu}\right)^{1/3}$$

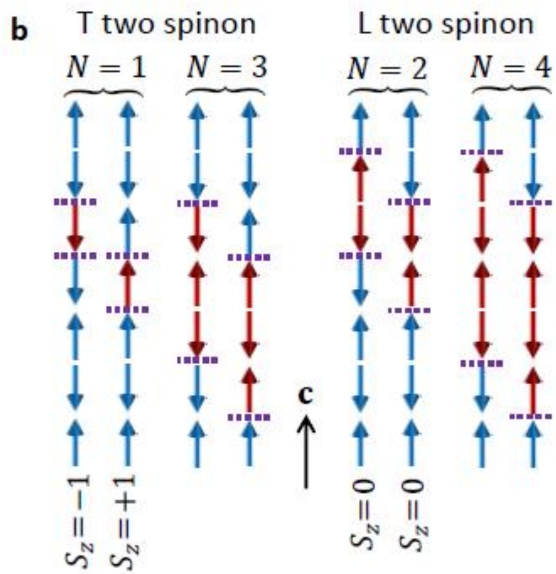
$$j = 1, 2, 3, \ldots$$

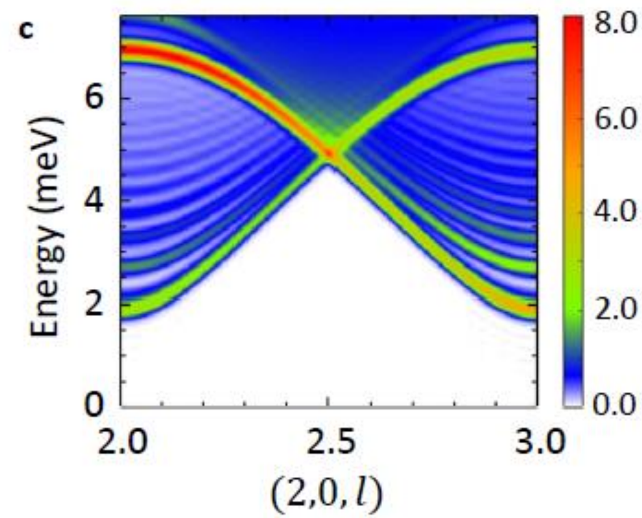


R. Coldea *et al.*
Science **327**, 177 (2010);
DOI: 10.1126/science.1180085

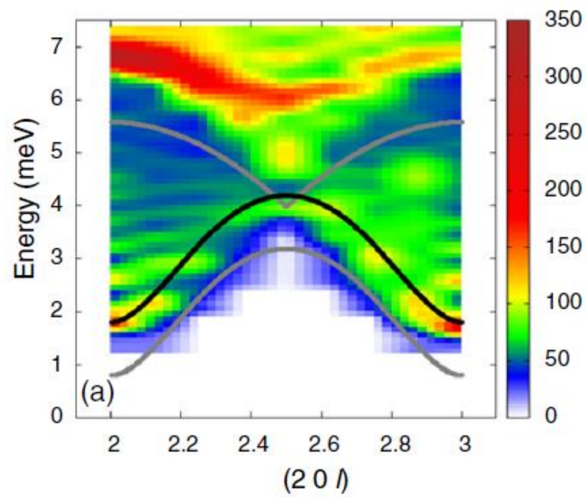
Bound states of spinons



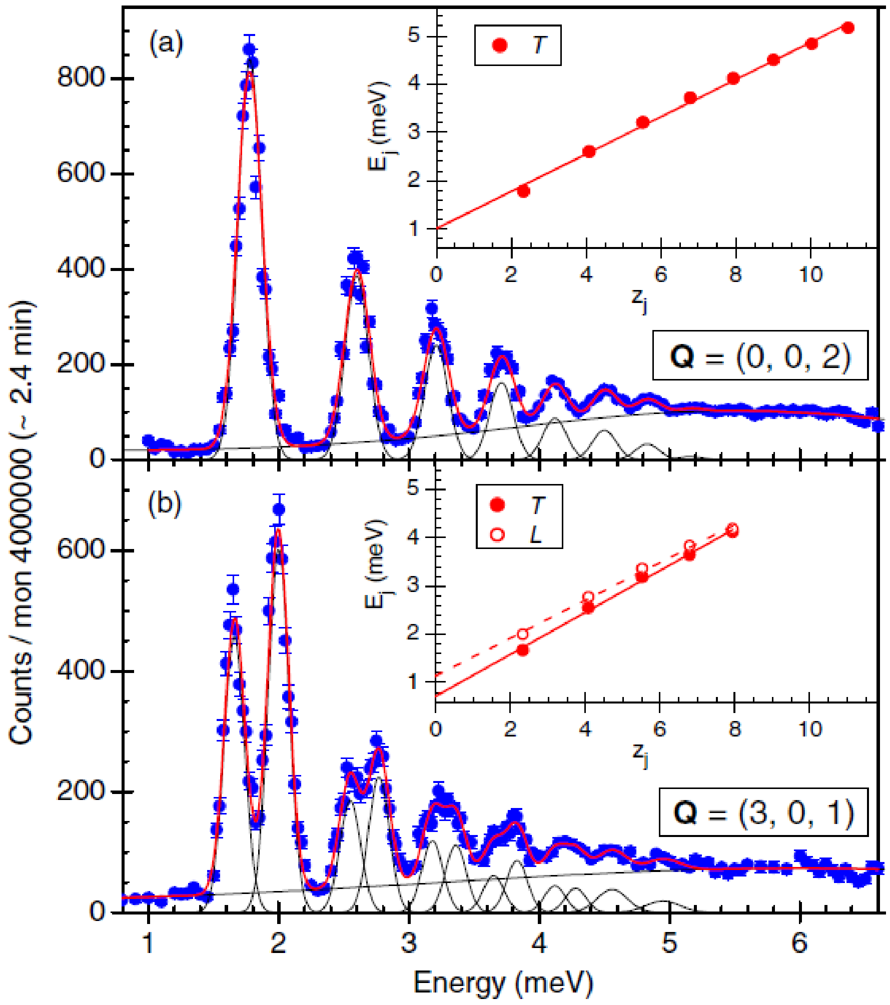




Example :
Antiferromagnet
$BaCo_2V_2O_8$

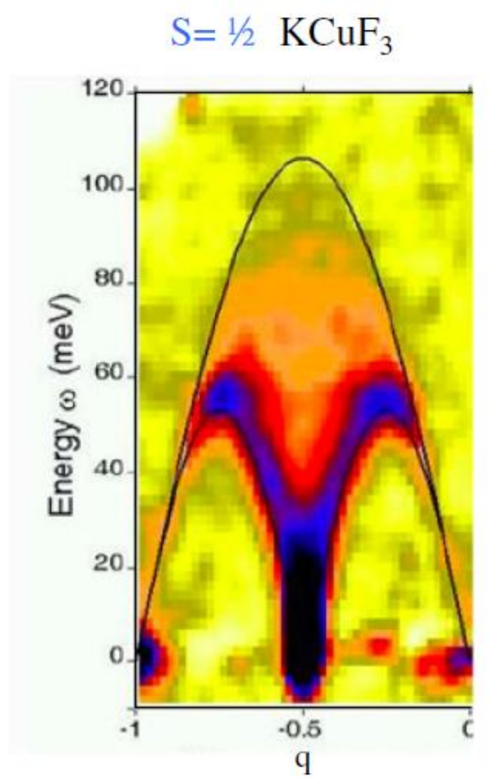


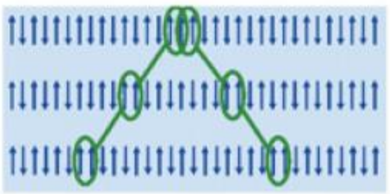
Bound states of spinons



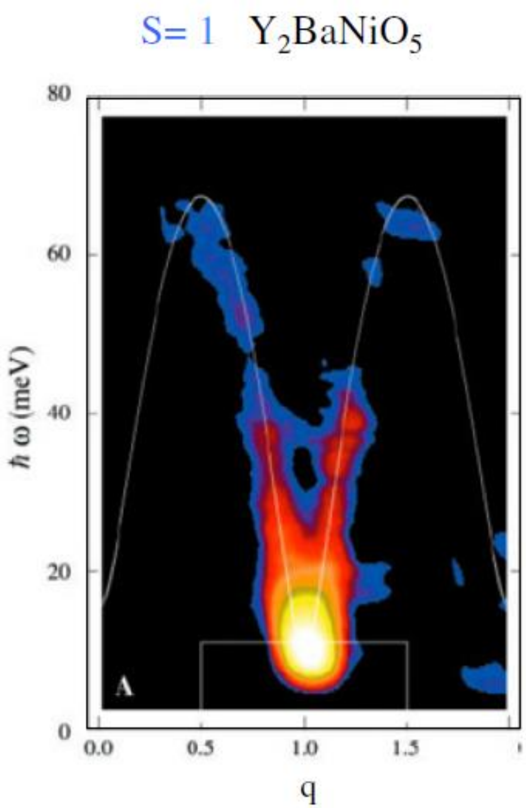
$$E_j^{T,L} = 2E_0^{T,L} + \alpha z_j \quad \text{with} \quad j = 1, 2, 3, \ldots,$$

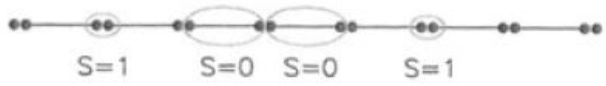
4. Quantum magnets





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





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

Electrons are dispersing spin ½ quasiparticles !!
To match ΔS=1, INS see a 2-particles continuum :

$$\omega = E_q + E_{q'}$$
$$\mathbf{k} = \mathbf{q} + \mathbf{q}'$$

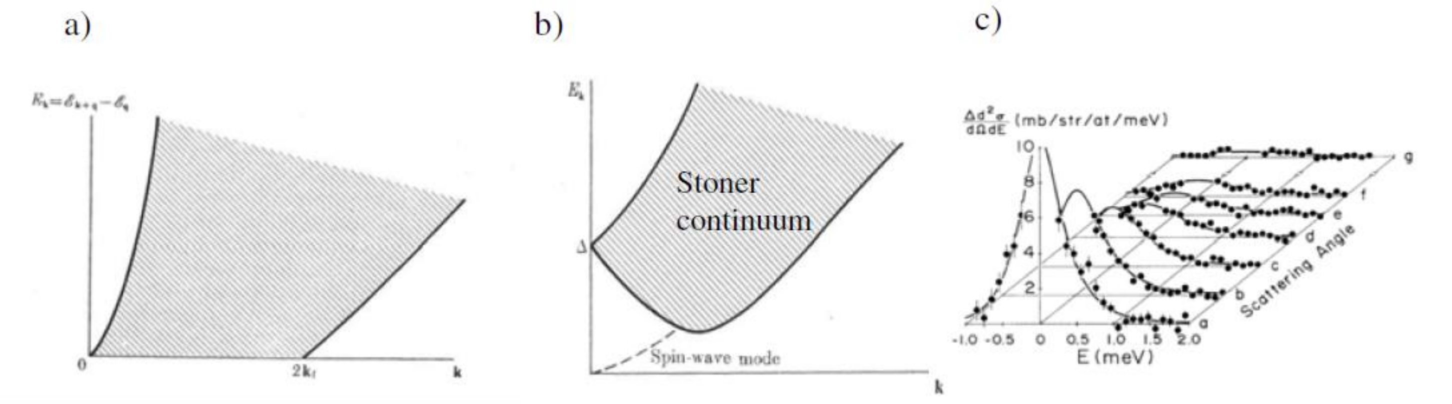


Figure 9. a) Energy spectrum of electron-hole excitation in a metal. b) Energy spectrum for interband transition in a ferromagnetic itinerant metal for $U > \epsilon_F$. c) Fluctuation spectrum of Ni_3Ga [20].

$$\chi_0(\mathbf{q},\omega) = -2\mu_B^2 \sum_{\mathbf{k}} \frac{f_{\mathbf{k}+\mathbf{q},\sigma'} - f_{\mathbf{k},\sigma}}{\epsilon_{\mathbf{k}+\mathbf{q},\sigma'} - \epsilon_{\mathbf{k},\sigma} - \hbar\omega + i\epsilon}$$

The susceptibility (hence the correlations) are sensitive to nesting properties of the Fermi surface

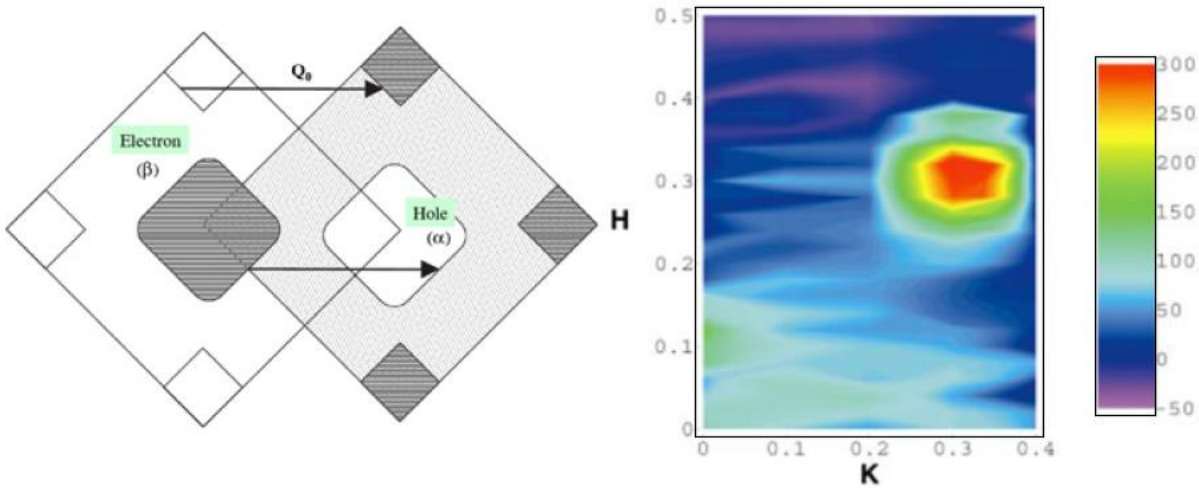
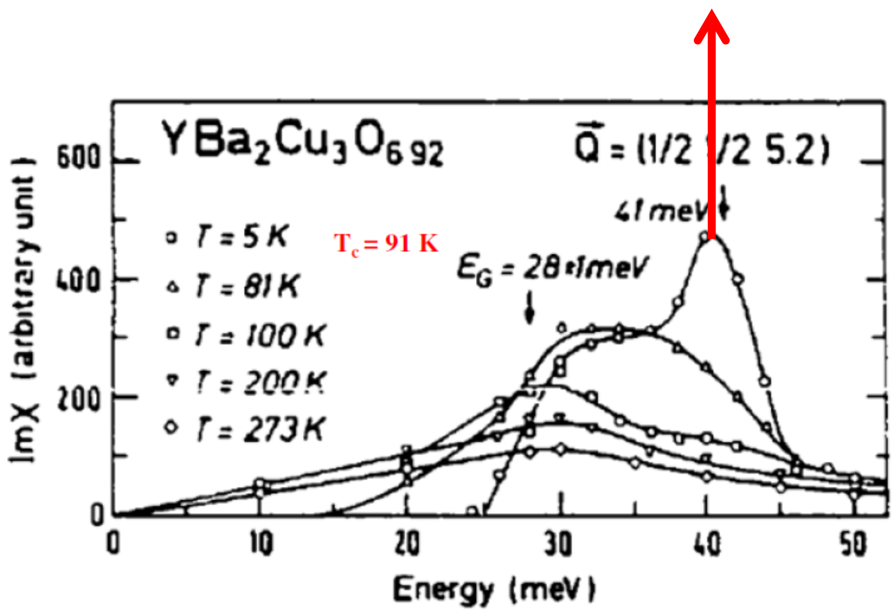


Figure 10. Left: sketch of the nesting feature between α and β bands of Sr_2RuO_4. Right: (H, K) map of the magnetic excitations in Sr_2RuO_4 at 10 K for an energy transfer of 4.1 meV [21].

Interactions may create a bound state (spin exciton)
Resonance peak in High Tc superconductors ?

$$\chi_0(\mathbf{q},\omega) = -2\mu_B^2 \sum_{\mathbf{k}} \frac{f_{\mathbf{k+q},\sigma'} - f_{\mathbf{k},\sigma}}{\epsilon_{\mathbf{k+q},\sigma'} - \epsilon_{\mathbf{k},\sigma} - \hbar\omega + i\epsilon}$$

$$\chi_0(\mathbf{q},\omega) = -2\mu_B^2 \sum_{\mathbf{k}} \left(1 - \frac{\Delta_{\mathbf{k}}\Delta_{\mathbf{k+q}} + \varepsilon_{\mathbf{k}}\varepsilon_{\mathbf{k+q}}}{E_{\mathbf{k}} E_{\mathbf{k+q}}}\right) \frac{f_{\mathbf{k+q},\sigma'} + f_{\mathbf{k},\sigma} - 1}{E_{\mathbf{k+q},\sigma'} - E_{\mathbf{k},\sigma} - \hbar\omega + i\epsilon} \,.$$



THANK YOU FOR YOUR ATTENTION

Local frames

The long range ordering defines local frames (attached to the direction of the local magnetization)

$$S^{a=x,y,z} = \mathcal{R}\ S^{a=\xi,\zeta,\eta}$$

$$\mathcal{R} = \begin{pmatrix} R_{11} & R_{12} & R_{13} \\ R_{21} & R_{22} & R_{23} \\ R_{31} & R_{32} & R_{33} \end{pmatrix} \qquad \mathbf{z} = \begin{pmatrix} R_{11} + iR_{12} \\ R_{21} + iR_{22} \\ R_{31} + iR_{32} \end{pmatrix} \quad \eta = \begin{pmatrix} R_{13} \\ R_{22} \\ R_{32} \end{pmatrix}$$

$$S_\ell^{a=x,y,z} = \frac{\sqrt{2S}}{2} \bar{z}_\ell^a\ b_\ell\ +\ \frac{\sqrt{2S}}{2}\ z_\ell^a b_\ell^+\ +\ \eta_\ell^a\ \left(S - b_\ell^+ b_\ell\right)$$

A spin in a field

Spin operator $\mathbf{S} = (S_x, S_y, S_z)$

Hamiltonian $\mathcal{H} = -\mathbf{S}\cdot\mathbf{h} = -h\ S_z$

Energies $E_n = h\ n \qquad n = -S, ..., S$

Manetization $S_z|n\rangle = n\ |n\rangle$

Raising and lowering operators

$$
\begin{aligned}
S^+|n\rangle &= \sqrt{S(S+1) - n(n+1)}\ |n+1\rangle \\
S^-|n\rangle &= \sqrt{S(S+1) - n(n-1)}\ |n-1\rangle
\end{aligned}
$$