Inelastic Neutron Scattering

Mechthild Enderle

Institute Laue-Langevin, Grenoble

October 12, 2015

Outline

- A bit of theory
- How we measure
- Local excitations
- Collective excitations

Magnetic cross section

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{\substack{\sigma_i,\sigma_f \\ n_0,n_1}} p(\sigma_i)p(n_0) \left| \langle \boldsymbol{k}_f \sigma_f n_1 | V_M | \boldsymbol{k}_i \sigma_i n_0 \rangle \right|^2 \delta(\epsilon_1 - \epsilon_0 - \hbar\omega)$$

$$= \frac{k_f}{k_i}(-\gamma r_0)^2 \sum_{\substack{\sigma_i,\sigma_f \\ n_0,n_1}} p(\sigma_i)p(n_0) \left| \langle \sigma_f n_1 | \boldsymbol{\sigma} \cdot \boldsymbol{M}_\perp(\boldsymbol{Q}) | \sigma_i n_0 \rangle \right|^2 \delta(\epsilon_1 - \epsilon_0 - \hbar\omega)$$

magnetic selection rule

$$\boldsymbol{M}_\perp(\boldsymbol{Q}) = \hat{\boldsymbol{Q}} \times (\boldsymbol{M}(\boldsymbol{Q}) \times \hat{\boldsymbol{Q}}) = \boldsymbol{M}(\boldsymbol{Q}) - (\boldsymbol{M}(\boldsymbol{Q}) \cdot \hat{\boldsymbol{Q}})\, \hat{\boldsymbol{Q}}$$

FT of the magnetization (operator) density

$$\boldsymbol{M}(\boldsymbol{Q}) = \int d^3r\; e^{i\boldsymbol{Q}\boldsymbol{r}}\; \boldsymbol{M}(\boldsymbol{r}).$$

Unpolarized neutrons

$$
\begin{aligned}
\frac{d^2\sigma}{d\Omega dE_f} &= \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{n_0,n_1} p(n_0)\,\left|\,\langle n_1|V(\boldsymbol{Q})|n_0\rangle\,\right|^2 \;\delta(\epsilon_1 - \epsilon_0 - \hbar\omega) \\
&= \frac{k_f}{k_i}\underbrace{\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{n_0} p(n_0)\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt\; e^{-i\omega t}\,\langle\, n_0|\; V^*(\boldsymbol{Q},0)V(\boldsymbol{Q},t)\;|\; n_0\rangle} \\
&= \frac{k_f}{k_i}\quad S(\boldsymbol{Q},\omega) \quad \text{dynamic scattering function}
\end{aligned}
$$

Dynamic scattering function

$$S_N(\boldsymbol{Q},\omega) = \sum_{n_0} p(n_0) \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} dt\ e^{-i\omega t} \langle\ n_0 |\ N^*(\boldsymbol{Q},0) N(\boldsymbol{Q},t)\ |\ n_0 \rangle$$

$$= \frac{1}{2\pi\hbar} \int d^3r dt\ e^{i(\boldsymbol{Q}\boldsymbol{r}-\omega t)} \langle\ N(0,0) N(\boldsymbol{r},t)\ \rangle_T$$

$$S_M(\boldsymbol{Q},\omega) = (\gamma r_0)^2 \sum_{n_0} p(n_0) \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} dt\ e^{-i\omega t} \langle\ n_0 |\ \boldsymbol{M}_\perp^*(\boldsymbol{Q},0) \boldsymbol{M}_\perp(\boldsymbol{Q},t)\ |\ n_0 \rangle$$

$$= (\gamma r_0)^2 \frac{1}{2\pi\hbar} \int d^3r dt\ e^{i(\boldsymbol{Q}\boldsymbol{r}-\omega t)} \langle\ \boldsymbol{M}_\perp(0,0) \boldsymbol{M}_\perp(\boldsymbol{r},t)\ \rangle_T$$

$S(\boldsymbol{Q},\omega)$ is the space-time FT
of the nuclear-positional resp. magnetic
time-dependent pair correlation function.

Separating phonons and magnons via Q

Coherent 1-phonon scattering function

$$S(\mathbf{Q},\omega) = \sum_{s=1}^{3r} \frac{1}{\omega_s} \left| \underbrace{\sum_{j=1}^{r} \frac{\overline{b_j}}{\sqrt{M_j}} \exp(-W_j) \exp(i\boldsymbol{Q}\boldsymbol{d}_j)}(\boldsymbol{Q}\cdot\boldsymbol{e}_{js}) \right|^2 \delta(\boldsymbol{k}_i - \boldsymbol{k}_f - \boldsymbol{Q})$$

$$\cdot \left[\langle n_s + 1\rangle_T \, \delta(\omega - \omega_s) + \langle n_s\rangle_T \, \delta(\omega + \omega_s) \right]$$

FT of a point
periodic in Q (for simple positions d_j)

increases with Q^2

Coherent 1-magnon scattering function ($T<<T_c$)

$$S(\mathbf{Q},\omega) = \sum_{s=1}^{r} \left| \sum_{j=1}^{r} \underbrace{f_j(\boldsymbol{Q})} \boldsymbol{e}_{\perp,js}(\boldsymbol{Q},\boldsymbol{q},\omega_s) \right|^2 \delta(\boldsymbol{k}_i - \boldsymbol{k}_f - \boldsymbol{Q})$$

$$\cdot \left[\langle n_s + 1\rangle_T \, \delta(\omega - \omega_s) + \langle n_s\rangle_T \, \delta(\omega + \omega_s) \right]$$

FT of unpaired-electron shell
falls off for large Q

periodic in Q (depending on d_j)

Dynamic scattering function

$$
\begin{aligned}
S^{\alpha\beta}(\boldsymbol{Q},\omega) &= \langle\ M^{\alpha *}(\boldsymbol{Q}) M^{\beta}(\boldsymbol{Q})\ \rangle_{T,\omega} \\
&= \frac{1}{2\pi} \int dt\ e^{-i\omega t} \langle M^{\alpha}(-\boldsymbol{Q},0) M^{\beta}(\boldsymbol{Q},t) \rangle_T \\
&= \frac{1}{2\pi} \int d^3r\ dt\ e^{i(\boldsymbol{Q}\boldsymbol{r}-\omega t)} \underbrace{\langle M^{\alpha}(0,0) M^{\beta}(\boldsymbol{r},t) \rangle_T}
\end{aligned}
$$

time dependent magnetic pair correlation function

$t \to \infty$ limit of pair correlations

$$
\begin{aligned}
S^{\alpha\beta}_{Bragg}(\boldsymbol{Q},\omega) &= \frac{1}{2\pi}\int dt\; e^{-i\omega t}\;\lim_{t\to\infty}\langle\; M^{\alpha}(-\boldsymbol{Q},0)M^{\beta}(\boldsymbol{Q},t)\;\rangle \\
&= \frac{1}{2\pi}\int dt\; e^{-i\omega t}\;\langle M^{\alpha}(-\boldsymbol{Q},0)M^{\beta}(\boldsymbol{Q},\infty)\rangle
\end{aligned}
$$

$t \to \infty$ limit of pair correlations

$$
\begin{aligned}
S^{\alpha\beta}_{Bragg}(\boldsymbol{Q},\omega) &= \frac{1}{2\pi}\int dt\ e^{-i\omega t}\ \lim_{t\to\infty}\langle\ M^{\alpha}(-\boldsymbol{Q},0)M^{\beta}(\boldsymbol{Q},t)\ \rangle \\
&= \frac{1}{2\pi}\int dt\ e^{-i\omega t}\ \langle M^{\alpha}(-\boldsymbol{Q},0)\rangle\ \langle M^{\beta}(\boldsymbol{Q},\infty)\rangle
\end{aligned}
$$

$t \to \infty$ limit of pair correlations

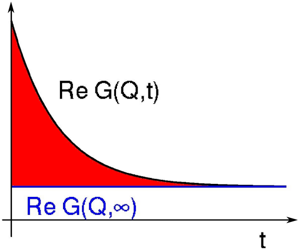
$$
\begin{aligned}
S^{\alpha\beta}_{Bragg}(\boldsymbol{Q},\omega) &= \frac{1}{2\pi}\int dt\; e^{-i\omega t}\;\lim_{t\to\infty}\langle\; M^{\alpha}(-\boldsymbol{Q},0)M^{\beta}(\boldsymbol{Q},t)\;\rangle \\
&= \frac{1}{2\pi}\int dt\; e^{-i\omega t}\;\langle M^{\alpha}(-\boldsymbol{Q},0)\rangle\;\langle M^{\beta}(\boldsymbol{Q},0)\rangle
\end{aligned}
$$

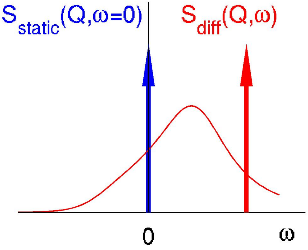
$t \to \infty$ limit of pair correlations

$$S^{\alpha\beta}_{Bragg}(\boldsymbol{Q},\omega) = \frac{1}{2\pi}\int dt\ e^{-i\omega t} \lim_{t\to\infty} \langle\ M^{\alpha}(-\boldsymbol{Q},0)M^{\beta}(\boldsymbol{Q},t)\ \rangle$$

$$= \delta(\omega)\ \langle M^{\alpha\,*}(\boldsymbol{Q},0)\rangle\ \langle M^{\beta}(\boldsymbol{Q},0)\rangle$$

Bragg scattering



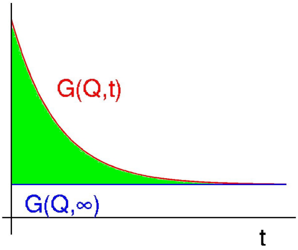


Bragg and diffuse/inelastic scattering

$$\langle M^{\alpha *}(\boldsymbol{Q},0) M^{\beta}(\boldsymbol{Q},t)\rangle_T$$

$$= \langle M^{\alpha *}(\boldsymbol{Q},0)\rangle \, \langle M^{\beta}(\boldsymbol{Q},0)\rangle \ldots \quad \} \rightarrow \text{Bragg, time independent}$$

$$\ldots + \underbrace{\langle \, (M^{\alpha *}(\boldsymbol{Q},0) - \langle M^{\alpha *}(\boldsymbol{Q},0)\rangle)(M^{\beta}(\boldsymbol{Q},t) - \langle M^{\beta}(\boldsymbol{Q},0)\rangle) \, \rangle_T}_{\text{correlation of fluctuations}}$$



$$S^{\alpha\beta}(\boldsymbol{Q},\omega) = S^{\alpha\beta}_{Bragg}(\boldsymbol{Q},\omega) + S^{\alpha\beta}_{diff}(\boldsymbol{Q},\omega)$$

Dynamic susceptibility

Linear response to a space and time varying magnetic field:

$$\begin{aligned}\langle M^\alpha(\boldsymbol{r},t)\rangle &= \langle M^\alpha(\boldsymbol{r},t)\rangle\big|_{H=0} \\ &+ \sum_\beta \int d^3r'\, dt'\, \chi^{\alpha\beta}(\boldsymbol{r}-\boldsymbol{r}', t-t') H^\beta(\boldsymbol{r}', t')\end{aligned}$$

$$\langle M^\alpha(\boldsymbol{Q},\omega)\rangle = \langle M^\alpha(\boldsymbol{Q},\omega)\rangle\big|_{H=0} + \sum_\beta \chi^{\alpha\beta}(\boldsymbol{Q},\omega) H^\beta(\boldsymbol{Q},\omega)$$

Dynamic susceptibility

Linear response to a space and time varying magnetic field:

$$\langle M^\alpha(\boldsymbol{r},t)\rangle = \langle M^\alpha(\boldsymbol{r},t)\rangle\big|_{H=0} + \sum_\beta \int d^3r'\ dt'\ \chi^{\alpha\beta}(\boldsymbol{r}-\boldsymbol{r'},t-t')H^\beta(\boldsymbol{r'},t')$$

$$\langle M^\alpha(\boldsymbol{Q},\omega)\rangle = \langle M^\alpha(\boldsymbol{Q},\omega)\rangle\big|_{H=0} + \sum_\beta \chi^{\alpha\beta}(\boldsymbol{Q},\omega)H^\beta(\boldsymbol{Q},\omega)$$

"Squid-Susceptibility" - $\omega = 0$, integrated over space

$$\begin{aligned}\int d^3r\ dt\ \langle M^\alpha(\boldsymbol{r},t)\rangle &= \langle M^\alpha(\boldsymbol{Q}=0,\omega=0)\rangle \\ &= \chi'^{\,\alpha\alpha}(\boldsymbol{Q}=0,\omega=0)H^\alpha \\ &= \chi^{\alpha\alpha}(\boldsymbol{Q}=0,\omega=0)H^\alpha\end{aligned}$$

Diffuse/inel. scattering function and dynam. susceptibility

$$
\begin{aligned}
S^{\alpha\beta}_{diff}(\boldsymbol{Q},\omega) &= S^{\alpha\beta}(\boldsymbol{Q},\omega) - S^{\alpha\beta}_{Bragg}(\boldsymbol{Q},\omega) \\
&= -\frac{\hbar}{\pi}\frac{1}{1-e^{-\frac{\hbar\omega}{k_B T}}}\ \frac{1}{2i}\left(\ \chi^{\alpha\beta}(\boldsymbol{Q},\omega) - \chi^{\beta\alpha\,*}(\boldsymbol{Q},\omega)\ \right) \\
S^{\alpha\alpha}_{diff}(\boldsymbol{Q},\omega) &= -\frac{\hbar}{\pi}\quad\frac{1}{1-e^{-\frac{\hbar\omega}{k_B T}}}\quad \Im(\ \chi^{\alpha\alpha}(\boldsymbol{Q},\omega)\ )
\end{aligned}
$$

Diffuse/inel. scattering function and dynam. susceptibility

$$S^{\alpha\beta}_{diff}(\boldsymbol{Q},\omega) = S^{\alpha\beta}(\boldsymbol{Q},\omega) - S^{\alpha\beta}_{Bragg}(\boldsymbol{Q},\omega)$$

$$= -\frac{\hbar}{\pi}\frac{1}{1-e^{-\frac{\hbar\omega}{k_B T}}}\ \frac{1}{2i}\left(\ \chi^{\alpha\beta}(\boldsymbol{Q},\omega) - \chi^{\beta\alpha\,*}(\boldsymbol{Q},\omega)\ \right)$$

$$S^{\alpha\alpha}_{diff}(\boldsymbol{Q},\omega) = -\frac{\hbar}{\pi}\quad\frac{1}{1-e^{-\frac{\hbar\omega}{k_B T}}}\quad \chi''^{\,\alpha\alpha}(\boldsymbol{Q},\omega)$$

$$S_{diff}(\boldsymbol{Q},\omega) = -\frac{\hbar}{\pi}\frac{1}{1-e^{-\frac{\hbar\omega}{k_B T}}}\cdot$$

$$\cdot\sum_{\alpha\beta}\left(\ \delta_{\alpha\beta} - \hat{Q}^{\alpha}\hat{Q}^{\beta}\ \right)\ \frac{1}{2}\left(\ \chi''^{\,\alpha\beta}(\boldsymbol{Q},\omega) + \chi''^{\,\beta\alpha}(\boldsymbol{Q},\omega)\ \right)$$

Neutron response and Squid-susceptibility

Kramers-Kronig relation

$$\chi'^{\,\alpha\alpha}(\boldsymbol{Q},\omega) \;=\; \frac{1}{\pi}\int d\omega' \;\frac{\chi''^{\,\alpha\alpha}(\boldsymbol{Q},\omega')}{\omega'-\omega}$$

$$\chi'^{\,\alpha\alpha}(\boldsymbol{Q},0) \;=\; \frac{1}{\pi}\int d\omega \;\frac{\chi''^{\alpha\alpha}(\boldsymbol{Q},\omega)}{\omega-0}$$

Neutron response and Squid-susceptibility

Kramers-Kronig relation

$$\chi'^{\,\alpha\alpha}(\boldsymbol{Q},\omega) = \frac{1}{\pi}\int d\omega' \, \frac{\chi''^{\,\alpha\alpha}(\boldsymbol{Q},\omega')}{\omega'-\omega}$$

$$\chi'^{\,\alpha\alpha}(\boldsymbol{Q},0) = \frac{1}{\pi}\int d\omega \, \frac{\chi''^{\alpha\alpha}(\boldsymbol{Q},\omega)}{\omega-0}$$

$$\chi'^{\,\alpha\alpha}(\boldsymbol{Q},\omega=0) = -\int_{-\infty}^{\infty} d\omega \, \frac{1-e^{-\frac{\hbar\omega}{k_BT}}}{\hbar\omega} S^{\alpha\alpha}_{diff}(\boldsymbol{Q},\omega)$$

$$\chi'^{\,\alpha\alpha}(\boldsymbol{Q}=0,\omega=0) = -\int_{-\infty}^{\infty} d\omega \, \frac{1-e^{-\frac{\hbar\omega}{k_BT}}}{\hbar\omega} S^{\alpha\alpha}_{diff}(\boldsymbol{Q}=0,\omega)$$

Neutron response and Squid-susceptibility

Kramers-Kronig relation

$$\chi'^{\alpha\alpha}(\boldsymbol{Q},\omega) = \frac{1}{\pi}\int d\omega' \, \frac{\chi''^{\alpha\alpha}(\boldsymbol{Q},\omega')}{\omega'-\omega}$$

$$\chi'^{\alpha\alpha}(\boldsymbol{Q},0) = \frac{1}{\pi}\int d\omega \, \frac{\chi''^{\alpha\alpha}(\boldsymbol{Q},\omega)}{\omega-0}$$

$$\chi'^{\alpha\alpha}(\boldsymbol{Q},\omega=0) = -\int_{-k_BT}^{\omega_{\text{cutoff}}} d\omega \, \frac{1-e^{-\frac{\hbar\omega}{k_BT}}}{\hbar\omega} S^{\alpha\alpha}_{diff}(\boldsymbol{Q},\omega)$$

$$\chi'^{\alpha\alpha}(\boldsymbol{Q}=0,\omega=0) = -\int_{-k_BT}^{\omega_{\text{cutoff}}} d\omega \, \frac{1-e^{-\frac{\hbar\omega}{k_BT}}}{\hbar\omega} S^{\alpha\alpha}_{diff}(\boldsymbol{Q}=0,\omega)$$

Detailed balance

Generally valid

$$\chi^{\alpha\beta}(\boldsymbol{Q},\omega) = \chi^{\alpha\beta\,*}(-\boldsymbol{Q},-\omega)$$

$$S^{\alpha\beta}_{diff}(\boldsymbol{Q},\omega) = S^{\beta\alpha\,*}_{diff}(\boldsymbol{Q},\omega)$$

Detailed balance

Generally valid

$$\chi^{\alpha\beta}(\boldsymbol{Q},\omega) \quad = \quad \chi^{\alpha\beta\,*}(-\boldsymbol{Q},-\omega)$$

$$S^{\alpha\beta}_{diff}(\boldsymbol{Q},\omega) \quad = \quad S^{\beta\alpha\,*}_{diff}(\boldsymbol{Q},\omega)$$

$$S^{\alpha\beta}_{diff}(\boldsymbol{Q},\omega) \quad = \quad e^{\frac{\hbar\omega}{k_B T}} \; S^{\beta\alpha}_{diff}(-\boldsymbol{Q},-\omega)$$

$$S_{diff}(\boldsymbol{Q},\omega) \quad = \quad e^{\frac{\hbar\omega}{k_B T}} \; S_{diff}(-\boldsymbol{Q},-\omega)$$

Detailed balance

Detailed balance for centrosymmetry

Centrosymmetric crystals and isotropic media

$$\chi'^{\alpha\beta}(\boldsymbol{Q},\omega) = \chi'^{\alpha\beta}(\boldsymbol{Q},-\omega)$$

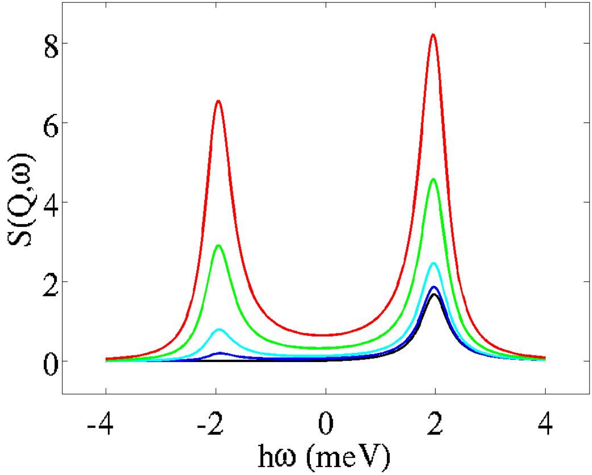
$$\chi''^{\alpha\beta}(\boldsymbol{Q},\omega) = -\chi''^{\alpha\beta}(\boldsymbol{Q},-\omega)$$

$$S^{\alpha\beta}_{diff}(\boldsymbol{Q},\omega) = e^{\frac{\hbar\omega}{k_B T}}\; S^{\beta\alpha}_{diff}(+\boldsymbol{Q},-\omega)$$

$$\underbrace{S_{diff}(\boldsymbol{Q},\omega) = e^{\frac{\hbar\omega}{k_B T}}\; S_{diff}(\boldsymbol{Q},-\omega)}_{\text{Detailed balance for centrosymmetry}}$$

Example detailed balance

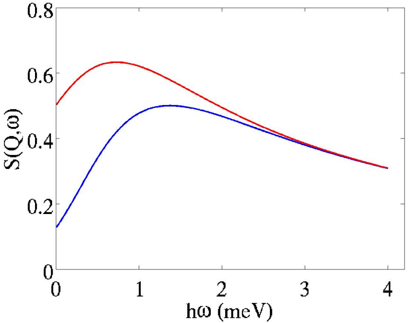
$S(\boldsymbol{Q}, \omega)$ at different T



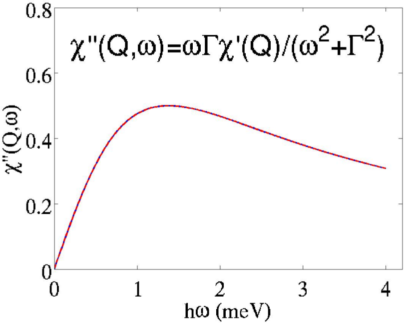
Example dynamic susceptibility

Gap or no gap ?

$S(\boldsymbol{Q}, \omega)$



$\chi''(\boldsymbol{Q}, \omega)$



Sum Rule for magnetic scattering

total magnetic scattering

$$\int d^3Q\ d\omega\ \tilde{S}_{Bragg}(\boldsymbol{Q},\omega) + \sum_{\alpha\beta} \tilde{S}^{\alpha\beta}_{diff}(\boldsymbol{Q},\omega) \quad = \quad N \sum_{\alpha} \langle \left(\ S^{\alpha} \ \right)^2 \rangle_T$$

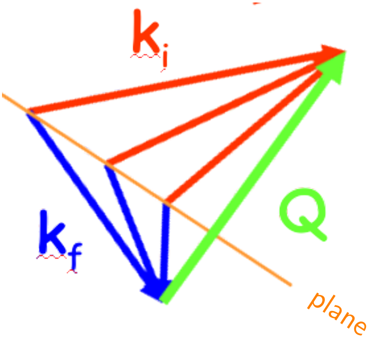
generally valid $\quad = \quad NS(S+1)$

classical complete order, for $T \to 0$ $\quad = \quad NS^2 + NS$

Scattering a neutron wave

Conservation of

	incident/ final neutron wave		sample
Momentum	$\boldsymbol{k}_i - \boldsymbol{k}_f$	$=$	$\boldsymbol{Q}$
Energy	$E_i - E_f$	$=$	$\hbar\omega$
	$\frac{\hbar^2 k_i^2}{2m_n} - \frac{\hbar^2 k_f^2}{2m_n}$	$=$	$\hbar\omega$



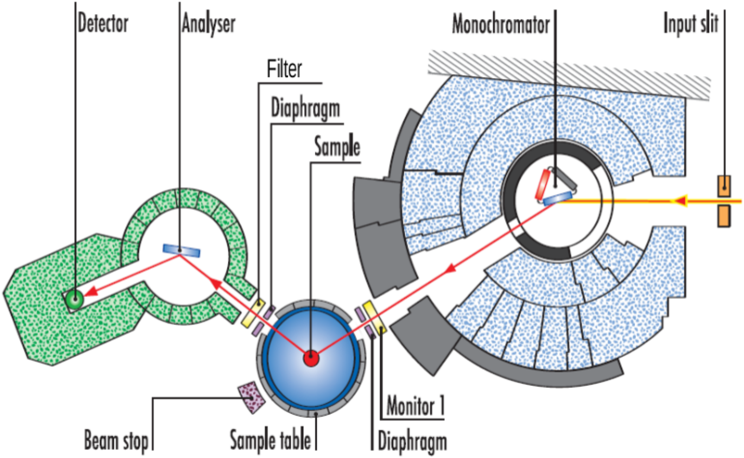
$\boldsymbol{Q}$, $\hbar\omega$

fixes $\boldsymbol{k}_i \cdot \boldsymbol{Q}$ and $\boldsymbol{k}_f \cdot \boldsymbol{Q}$

Triple Axis Spectrometer

select $k_i = \frac{2\pi}{\lambda_i}$ and $k_f = \frac{2\pi}{\lambda_f}$ via Bragg reflection

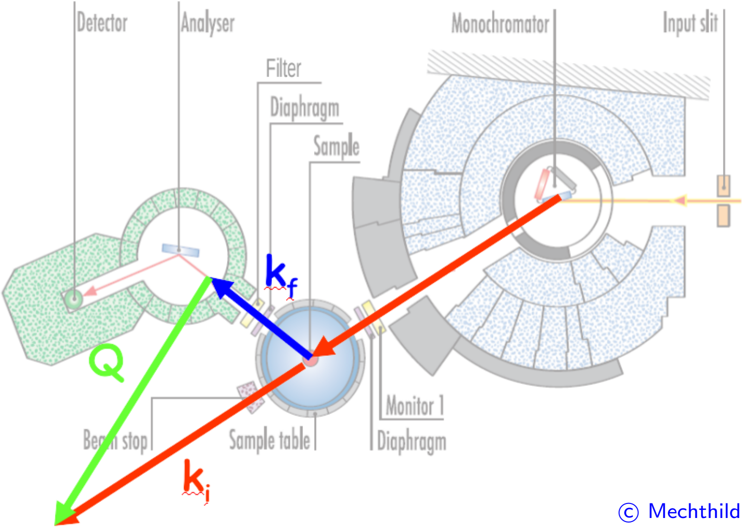
$n\lambda_f = 2d\sin\theta_f$ $\quad$ $n\lambda_i = 2d\sin\theta_i$



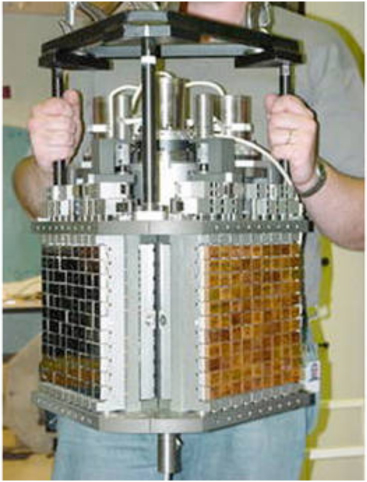
Triple Axis Spectrometer

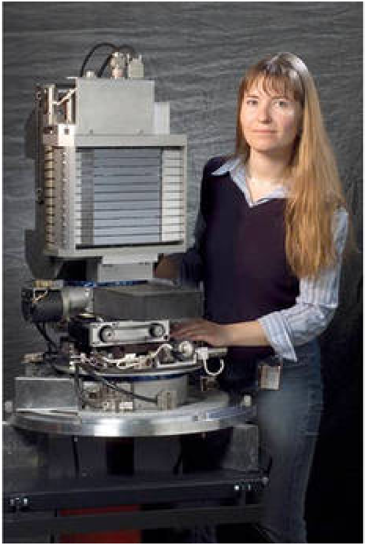
select $k_i = \frac{2\pi}{\lambda_i}$ and $k_f = \frac{2\pi}{\lambda_f}$ via Bragg reflection

$n\lambda_f = 2d\sin\theta_f$ $\qquad$ $n\lambda_i = 2d\sin\theta_i$



Monochromator/Analyser





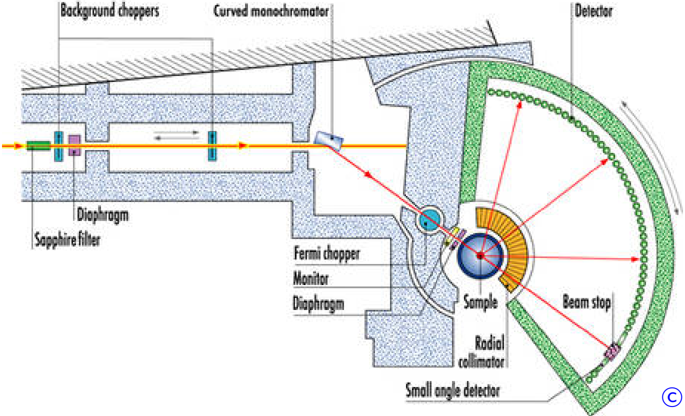
Time Of Flight Spectrometer – IN4/IN6

monochromator $n\lambda_i = 2d \sin\theta_i$ $\Rightarrow$ $k_i = \frac{2\pi}{\lambda_i}$, $E_i = \frac{\hbar^2}{2m_n} k_i^2$

Fermi chopper $\Rightarrow$ **pulse** structure of the neutron beam

Time of flight $t = t_1 + t_2$ $\Leftarrow$ $t_1 = \frac{L_1}{v_i} = \frac{L_1 m_n}{\hbar k_i}$

$k_f = \frac{1}{\hbar} m_n v_2 = \frac{m_n}{\hbar} \frac{L_2}{t_2}$ $E_f = \frac{\hbar^2}{2m_n} k_f^2$



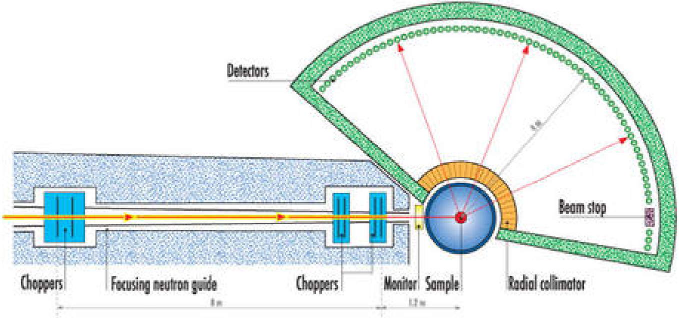
Time Of Flight Spectrometer – IN5

2 Disk choppers, relative phase ϕ and velocity Ω define $t_i = \frac{\phi}{\Omega}$

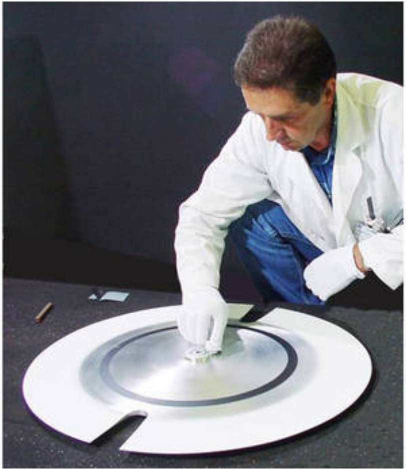
$v_i = \frac{L}{t_i} \quad \Rightarrow \quad \hbar k_i = m_n v_i, \quad E_i = \frac{\hbar^2}{2m_n} k_i^2$

Time of flight $t = t_1 + t_2 \quad \Leftarrow \quad t_1 = \frac{L_1}{v_i} = \frac{L_1 m_n}{\hbar k_i}$

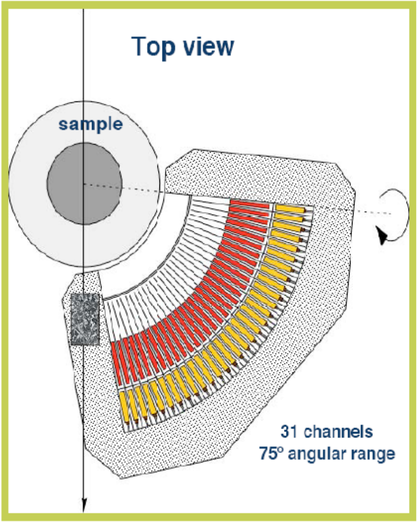
$k_f = \frac{1}{\hbar} m_n v_2 = \frac{m_n}{\hbar} \frac{L_2}{t_2} \quad E_f = \frac{\hbar^2}{2m_n} k_f^2$

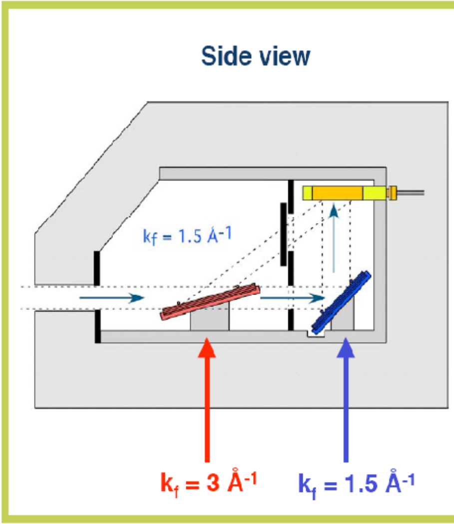


Time Of Flight Spectrometer

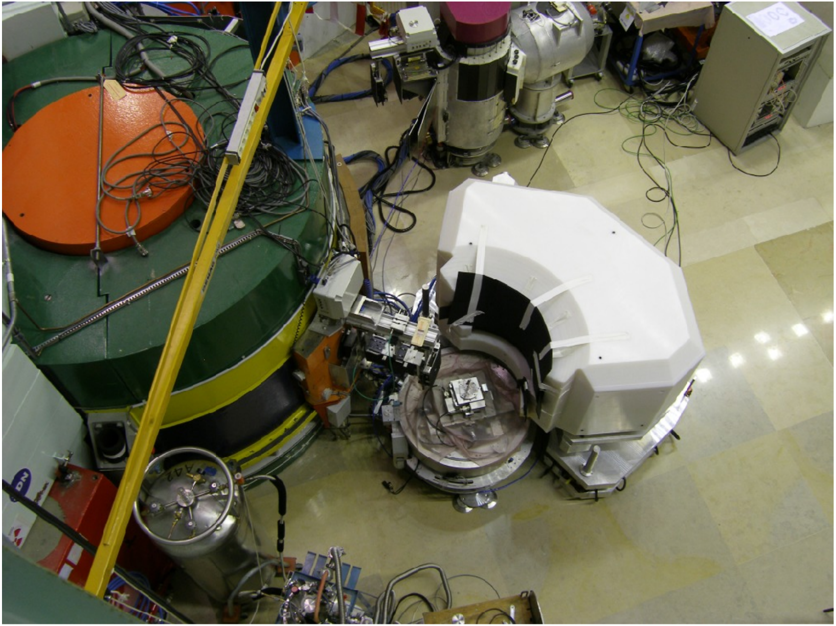


TAS multidetectors: FlatCone





TAS multidetectors: FlatCone



TAS versus TOF

TAS	TOF
highest continuous flux at sample	pulsed structure
single analyser-detector $\mathbf{1}$ E_f	highest detected solid angle continuous coverage of E_f
single crystal $>$ 5mm^3	single crystal $>$ 1cm^3

TAS versus TOF

TAS	TOF
highest continuous flux at sample	pulsed structure
single analyser-detector $\mathbf{1}\ E_f$	highest detected solid angle continuous coverage of E_f
Flatcone - multianalyser/detector Camea - several E_f	focused guide/monochromator bispectral TOF
single crystal $>$ 5mm^3	single crystal $>$ 1cm^3

Coherent dynamics

Crystal – periodic array of atoms / magnetic moments

Coherent dynamics

atoms/magnetic moments
move "correlated" ("ballet")

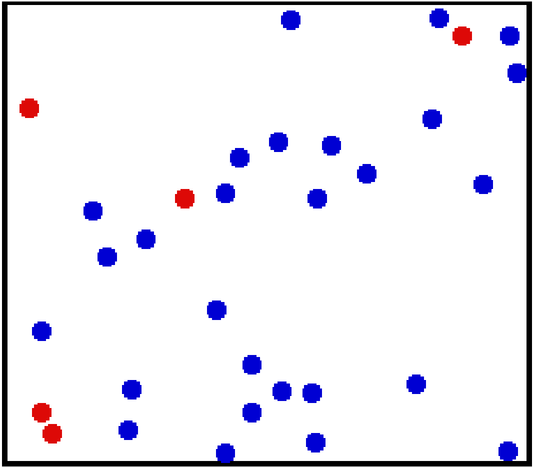
Snapshot:
periodic pattern – wave

Brownian motion

uncorrelated, diffusive motion

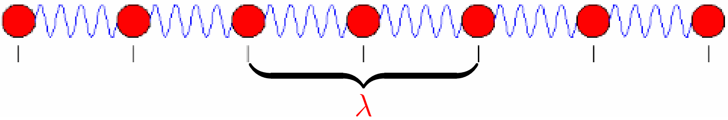
"random walk"
disordered

Brownian motion – diffuse dynamics

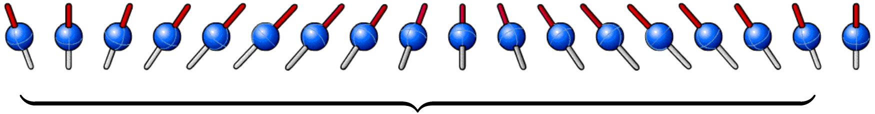


Collective motion – coherent dynamics – "ballet"

phonons



magnons



wavelength λ

wave vector $Q = \frac{2\pi}{\lambda}$

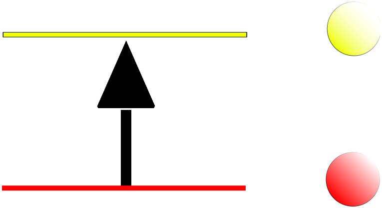
Local Excitations – infinitely weak "springs"

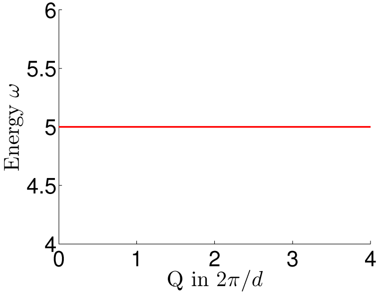
Coherent excitation ⇒ isolated/local excitation

Spectroscopy of an individual

atom/ion/molecule

spin / small group of spins





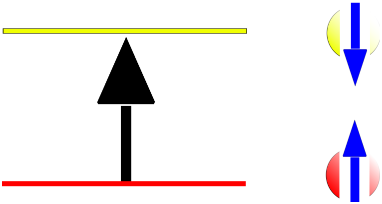
Local Excitations – infinitely weak "springs"

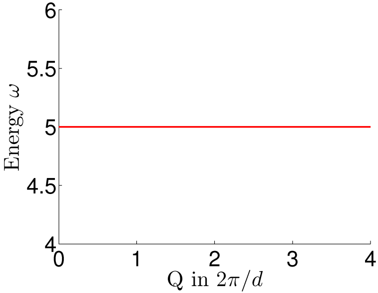
Coherent excitation ⇒ isolated/local excitation

Spectroscopy of an individual

atom/ion/molecule

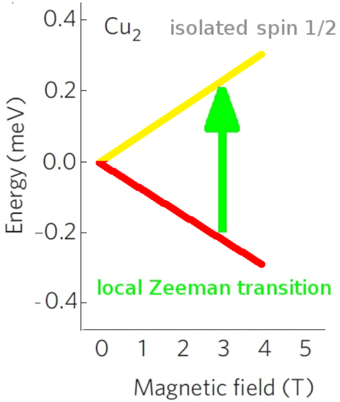
spin / small group of spins

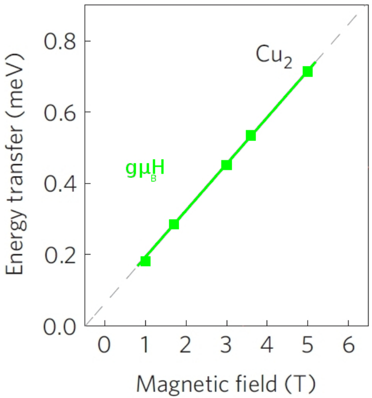




Local spin flip between Zeeman-split states

$CuSO_4.5D_2O$





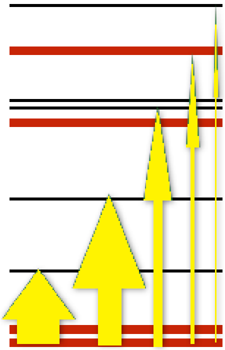
M. Mourigal, M.E. *et al.* Nat. Phys. **9** 435 (2013).

Local transitions: Crystal Field Splitting

$Tb_2Ti_2O_7$

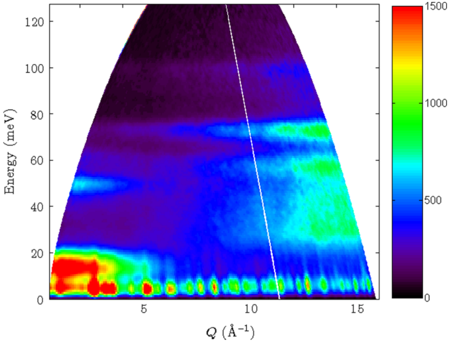
Tb^{3+}:

$^7F_6 \quad \left\{ \begin{array}{c} S=3 \\ L=3 \end{array} \right\} \quad J=6$



Merlin $E_i = 150$meV

powder, $T = 7$K

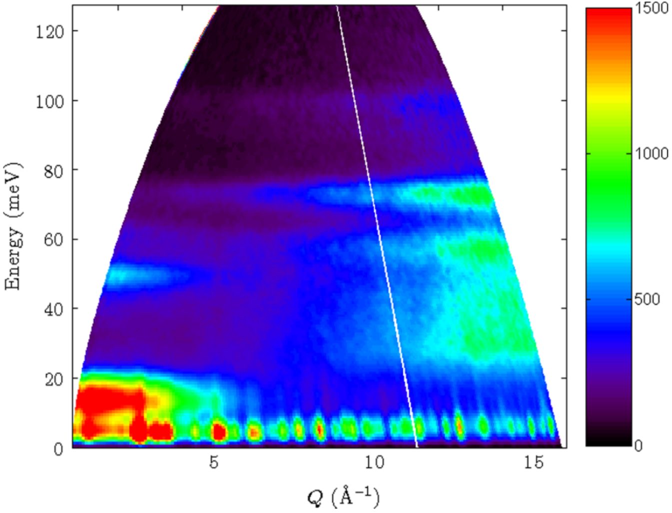


A. J. Princep *et al.* PRB **91** 224430 (2015).

Local transitions: Crystal Field Splitting

powder, $T = 7$K

$Tb_2Ti_2O_7$



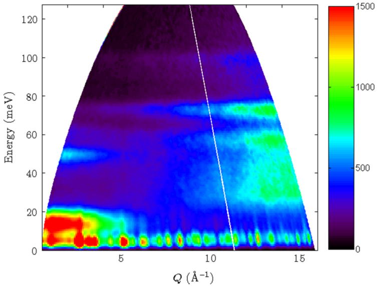
A. J. Princep *et al.* PRB **91** 224430 (2015).

Local transitions: Crystal Field Splitting

$$\mathcal{H} = \sum_{q,k} B_q^k C_q^k$$

$Tb_2Ti_2O_7$: 6 B_q^k to be determined: use energies and intensities !

$$\frac{d^2\sigma}{d\omega\, d\Omega} = N\,(\gamma r_0)^2\,\frac{k_f}{k_i}\,|f(Q)|^2\,e^{-2W}\sum_{i,f} p_i |\langle f|\boldsymbol{J}_\perp|i\rangle|^2\,\delta(\omega_f - \omega_i - \omega)$$



A. J. Princep *et al.* PRB **91** 224430 (2015).

Local transitions: Crystal Field Splitting

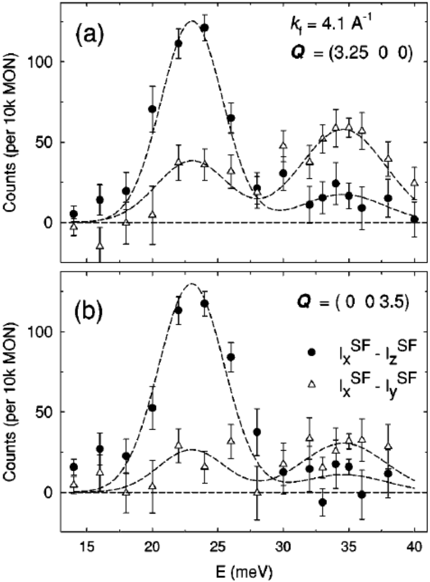
CePtSn: 2 CF-levels, 8 B_q^k.

Polarized neutrons (TAS)

+ single crystal

2 orthogonal $\boldsymbol{Q}$

$$\left.\begin{array}{l} I_x^{SF} - I_z^{SF} \\ I_x^{SF} - I_y^{SF} \end{array}\right\} \Rightarrow \left\{\begin{array}{l} |\langle f|\boldsymbol{J}_a|i\rangle|^2 \\ |\langle f|\boldsymbol{J}_b|i\rangle|^2 \\ |\langle f|\boldsymbol{J}_c|i\rangle|^2 \end{array}\right.$$



B. Janousova *et al.* PRB **69** 220412 (2004).

Polarized neutrons - Simple dipole rule

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}(-\gamma r_0)^2 \sum_{\substack{\sigma_i,\sigma_f \\ n_0,n_1}} p(\sigma_i)p(n_0) \left| \langle \sigma_f n_1 | \underbrace{\boldsymbol{\sigma} \cdot \boldsymbol{M}_\perp(\boldsymbol{Q})}_{\sigma_x M_{\perp x} + \sigma_y M_{\perp y} + \sigma_z M_{\perp z}} | \sigma_i n_0 \rangle \right|^2 \delta(\epsilon_1 - \epsilon_0 - \hbar\omega)$$

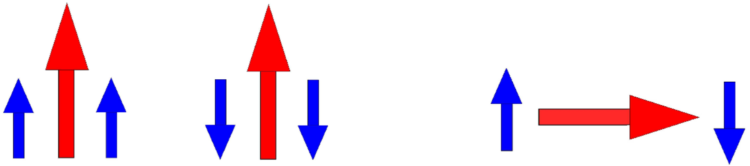
Pauli-matrices

$$\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

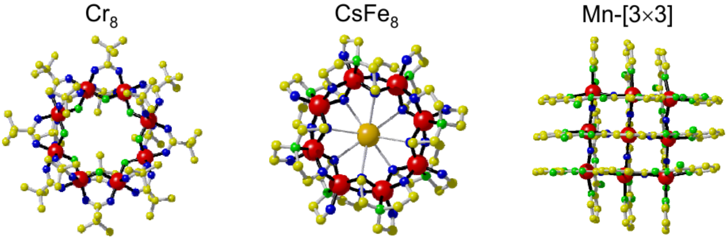
$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$$

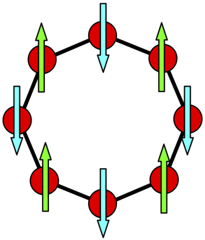
$$\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$$

- σ_z leaves its own eigenstates $\begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}$ unchanged
- $\sigma_{x,y}$ flip the state $\begin{pmatrix} 1 \\ 0 \end{pmatrix}$ $(\sigma_z = +1)$ to $\begin{pmatrix} 0 \\ 1 \end{pmatrix}$ $(\sigma_z = -1)$ and vice versa.



Local transitions: Molecular magnets

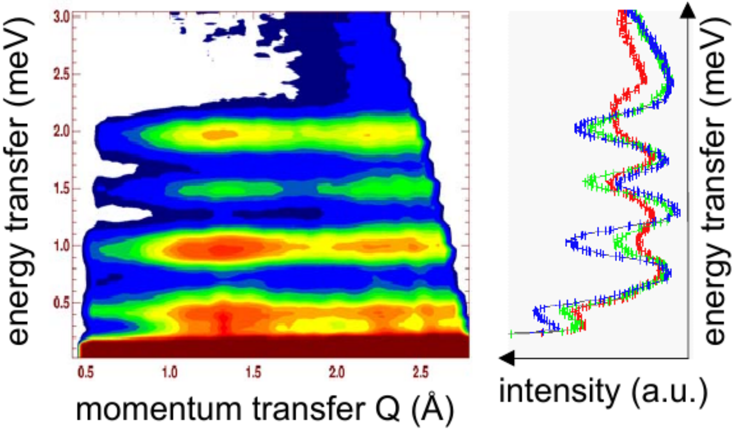




O. Waldmann APS-lecture 2006.

Local transitions: Molecular magnets

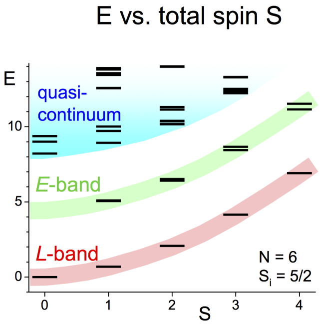
$CsFe_8$

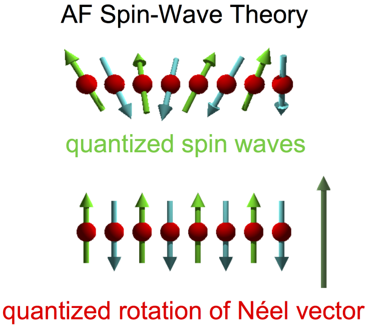


O. Waldmann *et al.* PRB 03, PRB 05.

Local transitions: Molecular magnets

Néel vector rotation and spin waves

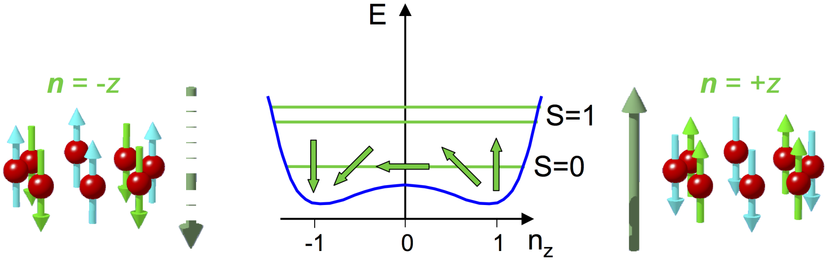




O. Waldmann APS-lecture 2006.

Local transitions: Molecular magnets

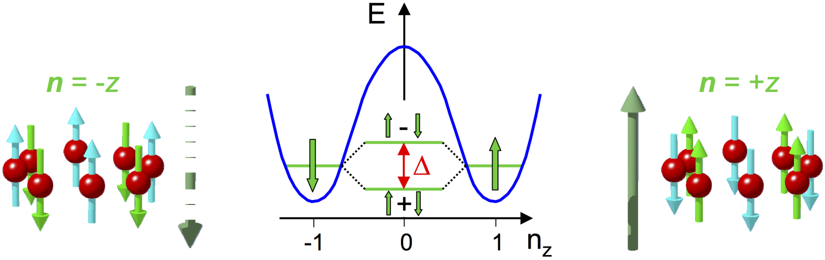
Néel vector rotation



O. Waldmann APS-lecture 2006.

Local transitions: Molecular magnets

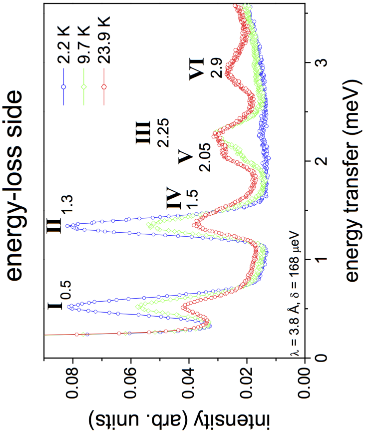
Néel vector tunneling

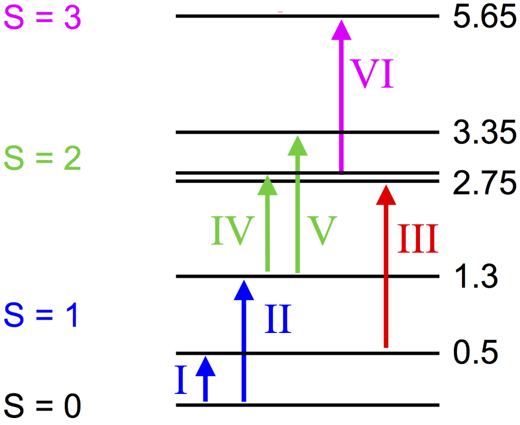


O. Waldmann APS-lecture 2006.

Local transitions: Molecular magnets

Néel vector rotation





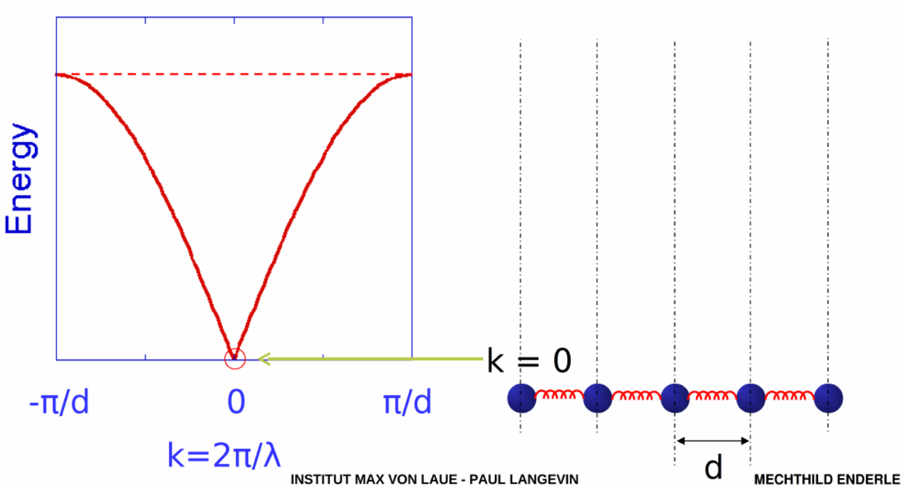
O. Waldmann APS-lecture 2006.

Local transitions

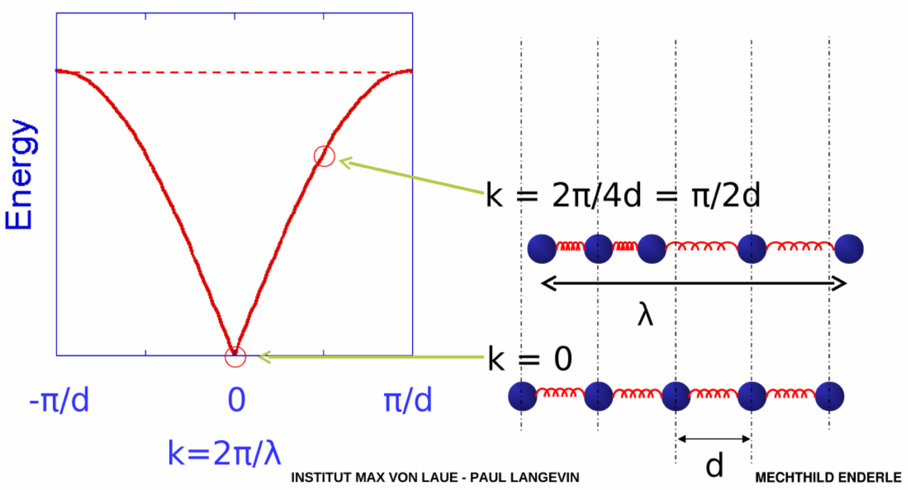
- energy independent of Q
- intensity: form factor $|f(Q)|^2$
  magnetic structure factor of local cluster (MM)
- powder measurement often sufficient
- integrate large Q-areas
- subtract phonons via nonmagnetic "blank"
- details (intensity pattern, transition matrix element):
  single crystal, polarized neutrons !

Ideal with TOF

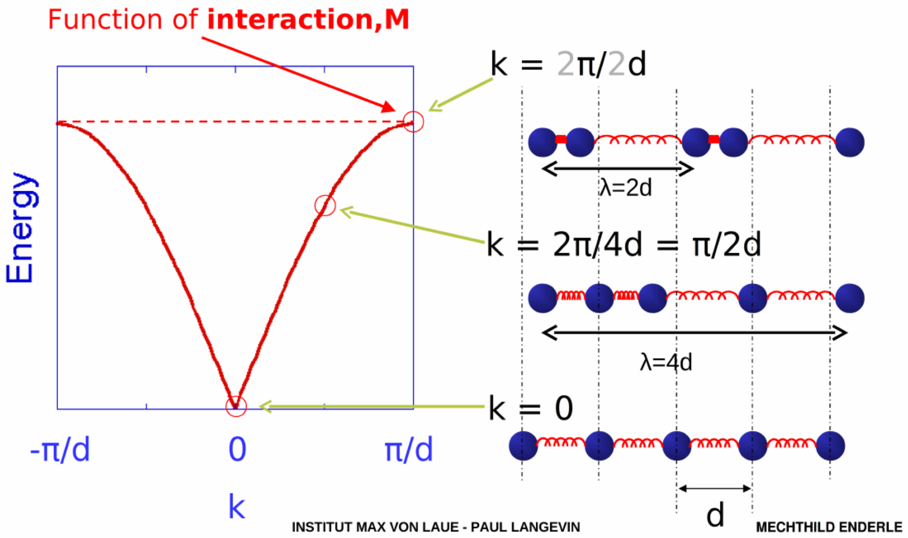
Collective excitations of the lattice: phonons



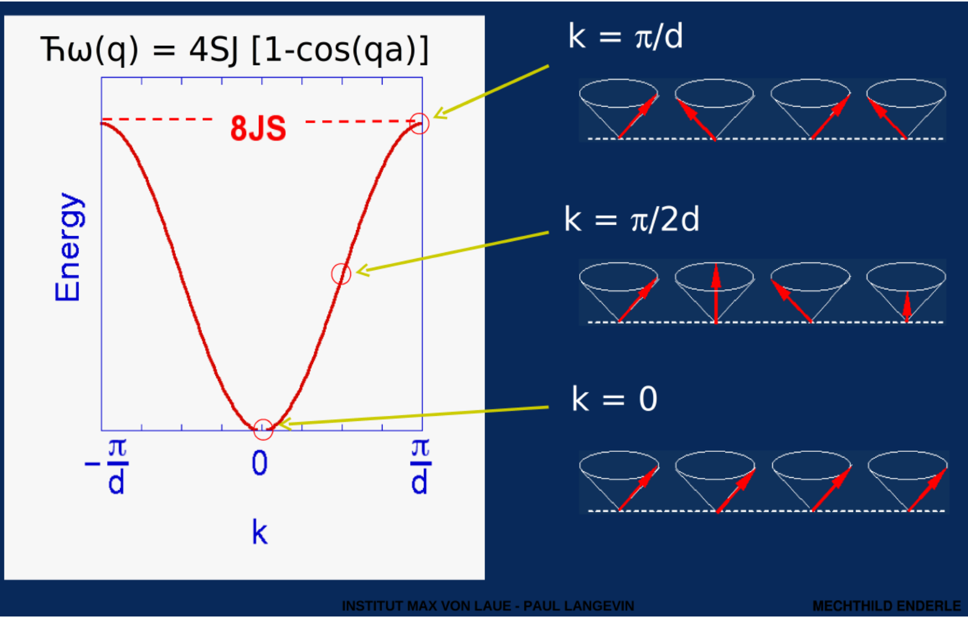
Collective excitations of the lattice: phonons



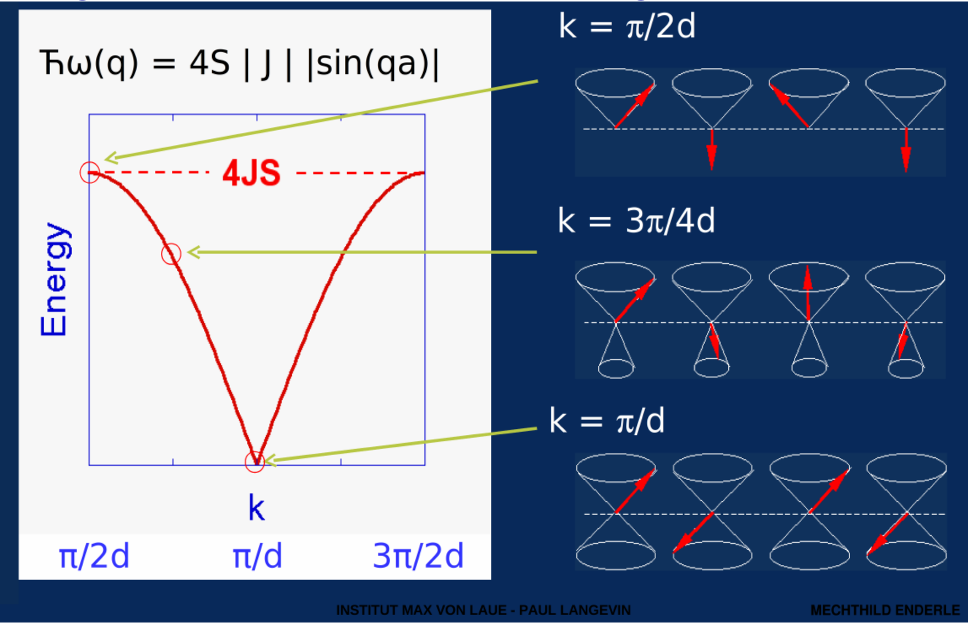
Collective excitations of the lattice: phonons



Collective excitations of the ferromagnet: magnons

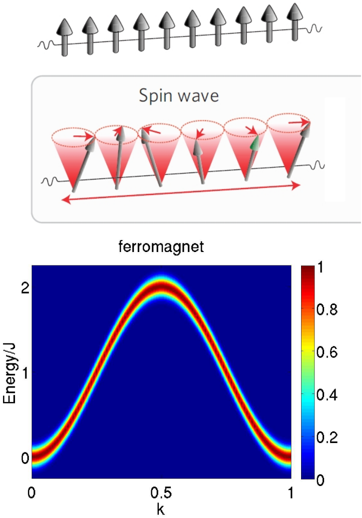


Magnons in the "classical" antiferromagnet



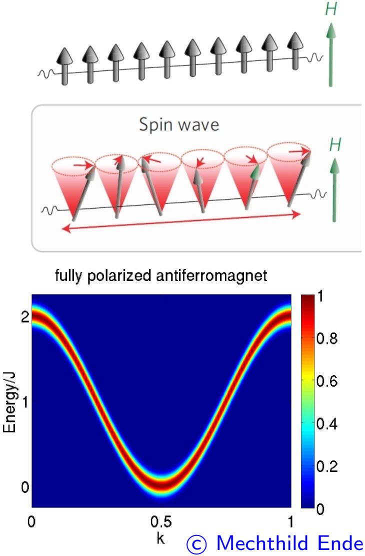
Magnon dispersion reveals microscopic interactions

Ferromagnet



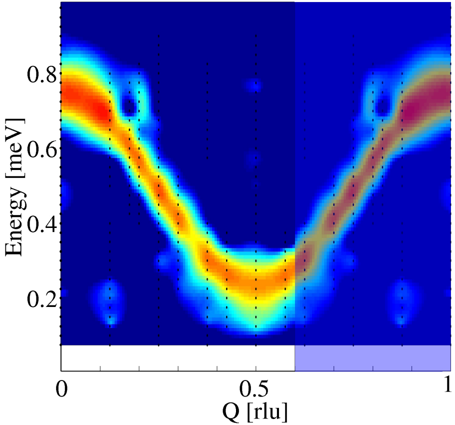
Fully saturated antiferromagnet

$H > H_{\text{sat}}$



Magnon dispersion reveals microscopic interactions

$CuSO_4.5D_2O$



$\Uparrow$

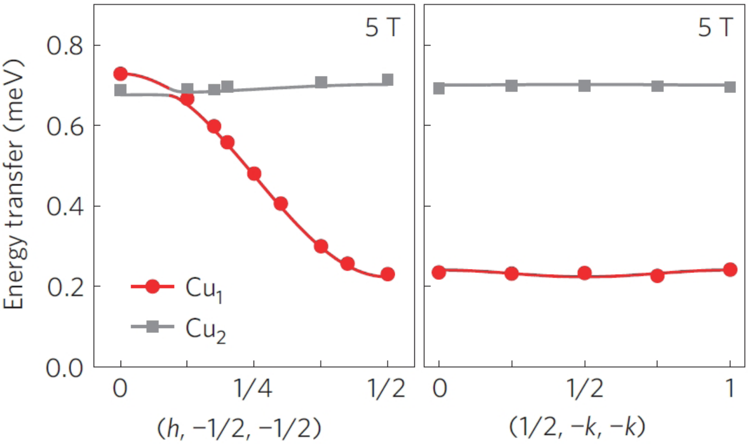
antiferromagnetic exchange

$H > H_{\mathrm{sat}}$

no long range order >0.1K

Magnon dispersion reveals microscopic interactions

$CuSO_4.5D_2O$ fully saturated $H > H_{sat}$



magnetically 1D !

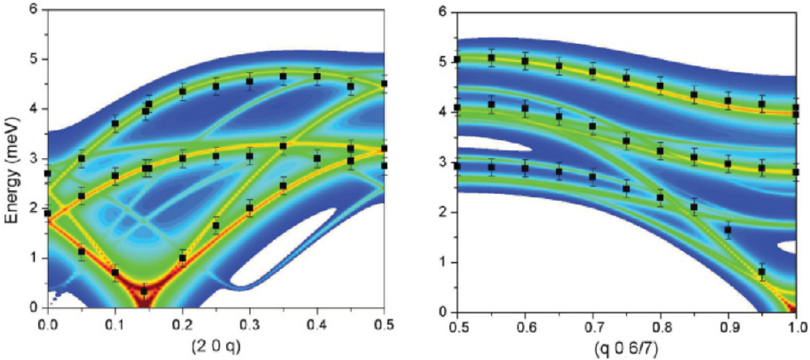
M. Mourigal, M.E. *et al.* Nat. Phys. **9** 435 (2013).

Magnetic dispersion reveals microscopic interactions

...but we need a theory !

Long-range ordered structures: "Classical" Spin Wave Theory

J. Jensen (2011) PRB 84, 104405



Magnetic dispersion reveals microscopic interactions

periodically ordered spin sites with a local magnetic moment

interaction between the spins (e.g. visible in $\chi(T)$)

no long-range ordered magnetic moment

Collective excitations ?