Magnetic and lattice excitations characterized with neutron scattering

Mechthild Enderle

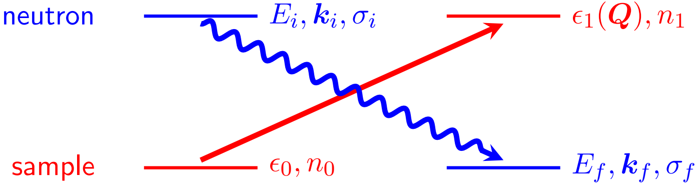
Institute Laue-Langevin, Grenoble

September 17, 2024

Outline

- Interaction neutron – matter
- Collective dynamics: Dispersion
- Collective dynamics: Intensities
- More than spin waves: Quantum many-body states

Master equation for neutron scattering



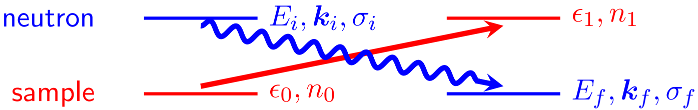
- incoming/scattered neutron plane wave
- energies far away from nuclear resonances

Energy conservation	$E_i - E_f$	$=$	$\epsilon_1 - \epsilon_0$
Momentum/wave vector conservation	$\boldsymbol{k}_i - \boldsymbol{k}_f$	$=$	$\boldsymbol{Q}$

Master equation for neutron scattering

- feable interaction V → single scattering process
- 1st order perturbation - 1st Born approximation - - Fermi's golden rule

$$\frac{d^2\sigma}{d\Omega dE_f}\Big|_{n_0,\sigma_i\rightarrow n_1,\sigma_f} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \left|\langle \boldsymbol{k}_f\sigma_f n_1|V|\boldsymbol{k}_i\sigma_i n_0\rangle\right|^2 \cdot \delta(\epsilon_1-\epsilon_0-(E_i-E_f))$$

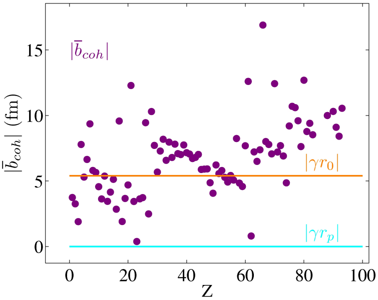


Intensities can be calculated

Interactions of neutrons with matter V

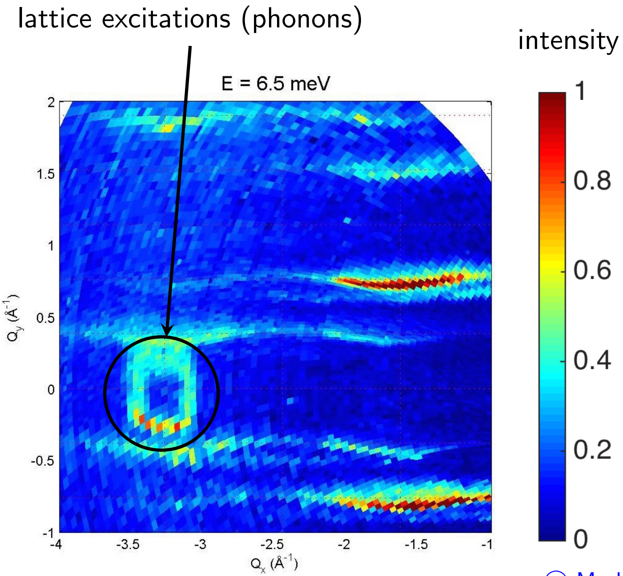
n	–	atomic nucleus	strong interaction
n	–	electronic magn. moment	dipole-dipole interaction
n	–	electric field	spin-orbit + Foldy interaction

	average (absolute) scattering length		
nucleus	+6.5	fm	
el. magn. mom.	−5.4	fm	$(\cdot S(J))$
electric field	+1.5	am	

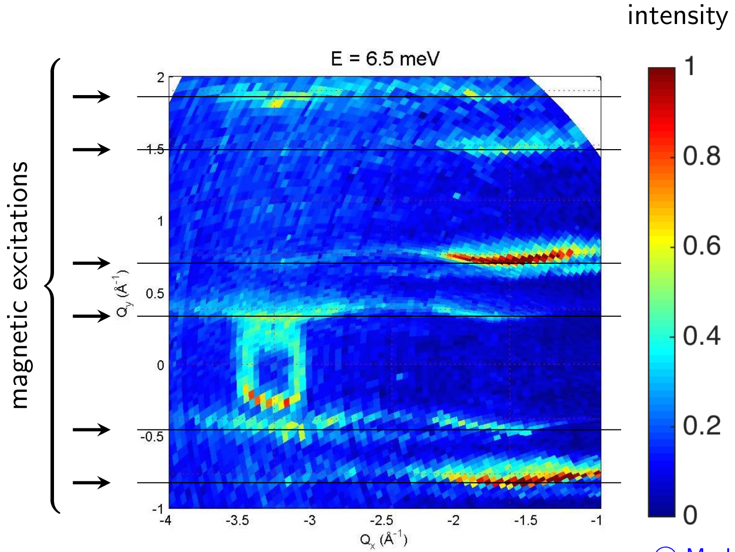


magnetic/nuclear intensities comparable
charge $<1/10^6$

Magnetic and lattice excitations: comparable intensity



Magnetic and lattice excitations: comparable intensity



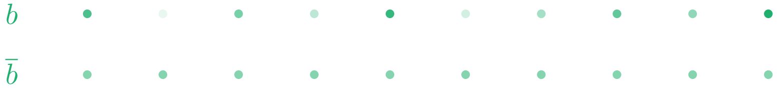
Nuclear interaction potential

Fermi pseudopotential V_N for neutron scattering from one nucleus

$$V_N(\boldsymbol{r}) \;=\; \frac{2\pi\hbar^2}{m} b\, \delta(\boldsymbol{r} - \boldsymbol{R}_j)$$

Coherent scattering length $\bar{b}$: average over $\left\{ \begin{array}{c} \text{nuclear spin states} \\ \text{isotopes} \end{array} \right.$ of an element.

entire sample: $$V_N(\boldsymbol{r}) \;=\; \frac{2\pi\hbar^2}{m} \sum_{j=1}^{10^{23}} \bar{b}_j\, \delta(\boldsymbol{r} - \boldsymbol{R}_j) \;=\; \frac{2\pi\hbar^2}{m}\, N(\boldsymbol{r})$$



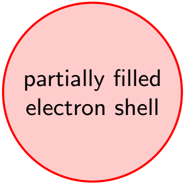
Magnetic interaction potential

magnetic potential V_M for neutron scattering from one electron

$$V_M(\boldsymbol{r}) = -\boldsymbol{\mu}_n \cdot \boldsymbol{B}_e$$

$$V_M(\boldsymbol{r}) = -\boldsymbol{\mu}_n \cdot \nabla \times \boldsymbol{A}$$

$$V_M(\boldsymbol{r}) = -\boldsymbol{\mu}_n \cdot \nabla \times \overbrace{\left(\underbrace{\nabla \times \frac{\boldsymbol{\mu}_e^S}{r}}_{\text{spin}} + \underbrace{\nabla \times \frac{\boldsymbol{\mu}_e^L}{r}}_{\text{orbital current}} \right)}^{\text{vector potential } \boldsymbol{A}}$$



total (spin+orbital)
magnetic moment $\boldsymbol{\mu}_e$

Magnetic matrix element - Fourier transform

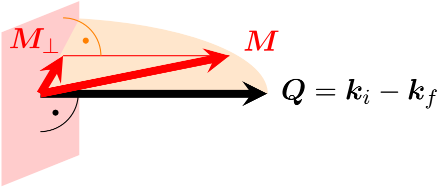
one electron

$$\langle \boldsymbol{k}_i \sigma_i n_0 | -\boldsymbol{\mu}_n \cdot \nabla \times \left(\nabla \times \frac{\boldsymbol{\mu}_e^{tot}}{r} \right) | \boldsymbol{k}_f \sigma_f n_1 \rangle$$

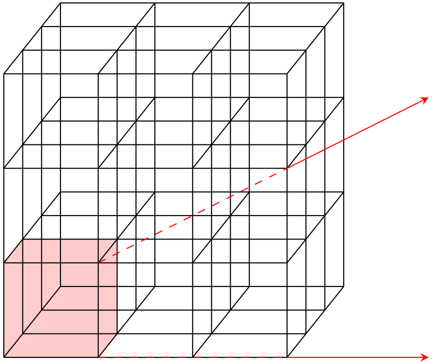
$$= \langle \sigma_i n_0 | -\boldsymbol{\mu}_n \cdot \underbrace{\left(\hat{\boldsymbol{Q}} \times (\hat{\boldsymbol{Q}} \times \boldsymbol{\mu}_e^{tot}(\boldsymbol{Q})) \right)} | \sigma_f n_1 \rangle$$

$$= \langle \sigma_i n_0 | -\boldsymbol{\mu}_n \cdot \boldsymbol{\mu}_{e\,\perp}^{tot}(\boldsymbol{Q}) | \sigma_f n_1 \rangle$$

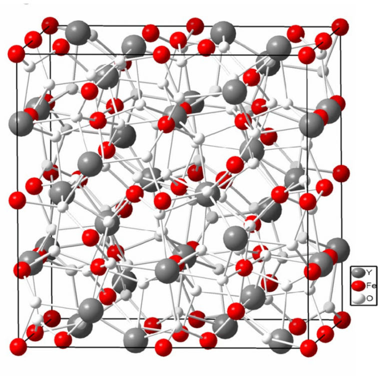
entire sample $\sum \boldsymbol{\mu}_{e\,\perp}^{tot}(\boldsymbol{Q}) = \boldsymbol{M}_\perp(\boldsymbol{Q})$

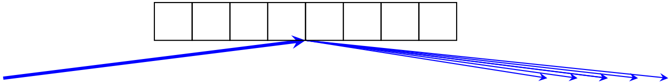


Single crystals – periodic arrays



unit cell





Coherent and incoherent scattering

Coherent scattering	Incoherent scattering
average scattering amplitude	standard deviation
equal objects	unequal objects – isotopes – nuclear spin directions – electronic spin directions
spatial and temporal interference of many objects	temporal interference of 1 object with itself
Pair correlation	Autocorrelation
Full spatial information	No spatial information

Coherent and incoherent scattering

Coherent scattering

average scattering amplitude

equal ~~objects~~ mag. moments

polarisation analysis

low temperature

Incoherent scattering

standard deviation

unequal objects

– ~~isotopes~~

– ~~nuclear spin directions~~

– ~~electronic spin directions~~

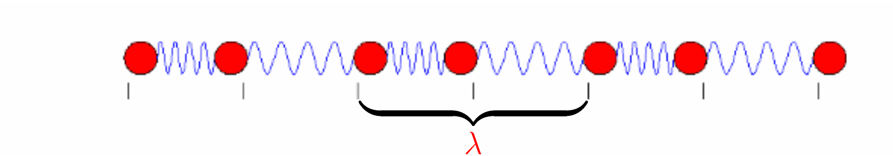
magnetic pair correlations separate from { incoherent scattering, phonons, background }

Collective motion – coherent dynamics – "ballet"

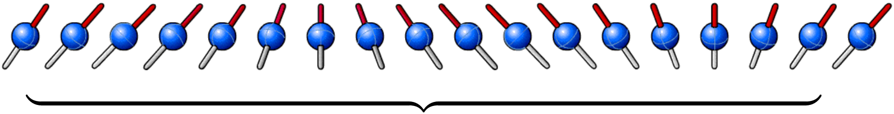
"snapshot" interference patterns

taken with the stroboscopic frequency $\hbar\omega = E_i - E_f$

phonons



magnons



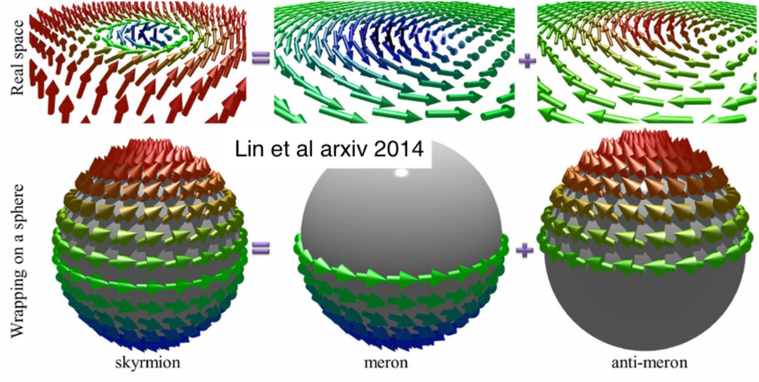
wavelength λ

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

Collective motion – coherent dynamics – "ballet"

topological magnetic excitations:

solitons – skyrmions – merons – antimerons

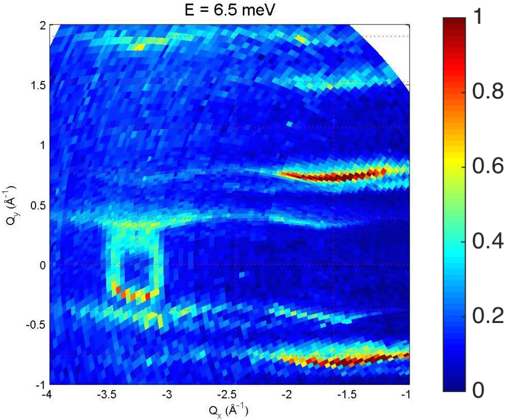


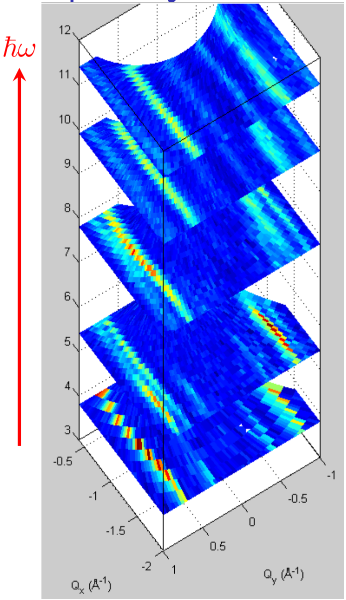
quantum mechanical analogues:

spinons, vector bosons . . .

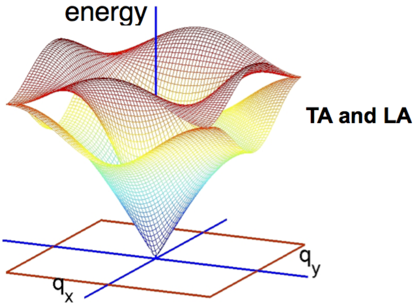
Interference pattern in the wave vector space Q

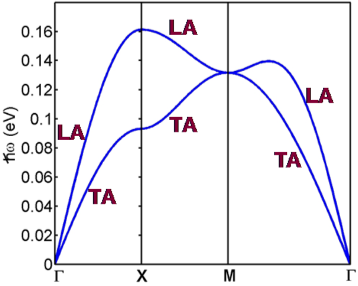
pattern changes
with the "stroboscopic" frequency
$\hbar\omega = E_i - E_f$





Dispersion surface – energy $\hbar\omega$ as function of Q_x, Q_y, Q_z





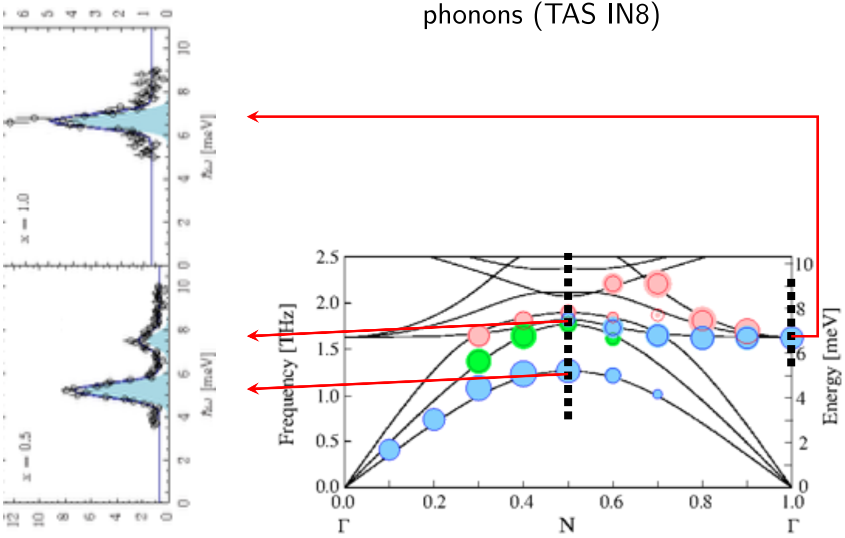
F Rana 2009

spatial interference pattern: discrete $\boldsymbol{Q}$-pattern at each $\hbar\omega$

$\Leftrightarrow$ discrete ω at given $\boldsymbol{Q}$

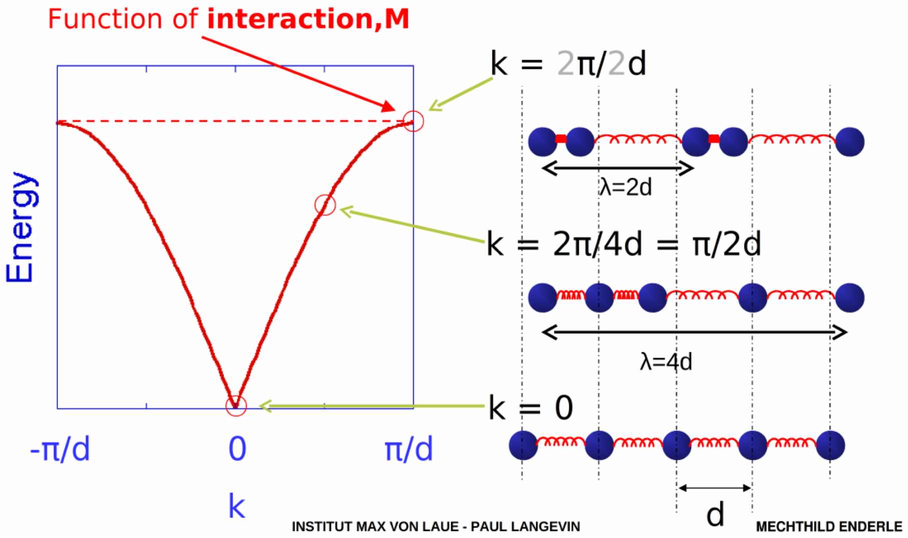
Collective dynamics: signature dispersion $\hbar\omega(\boldsymbol{Q})$

phonons (TAS IN8)



M.M. Koza *et al.* PRB **91** 014305 (2015)

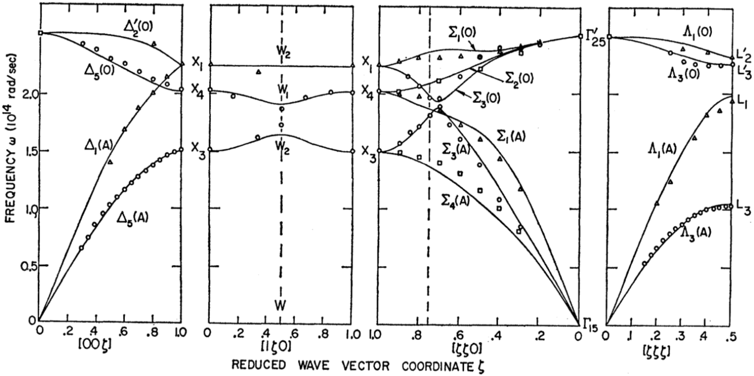
Collective lattice excitations: phonons (LA)



Phonons in diamond

diamond:  covalent bonds

1000 meV

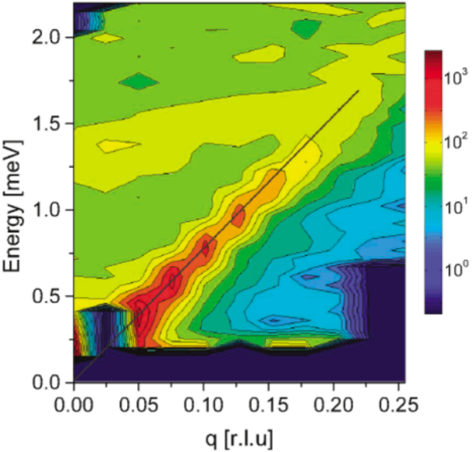


J.L. Warren *et al.* Phys.Rev. **158** 805 (1967)

Phonons in bcc ^{4}He

bcc ^{4}He  van der Waals (+quantum effects)

1 meV



$T = 1.6400(1)$K

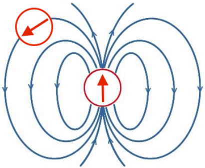
IN12

M. Markovich *et al.* PRL **88** 195301 (2002)

"Magnetic springs" - mostly super-exchange

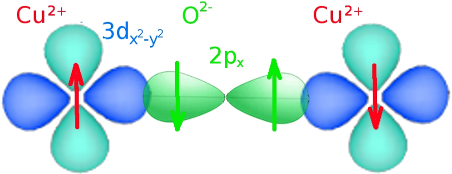
dipole-dipole $\sim$ μeV

$$E = -\boldsymbol{m}_1 \cdot \boldsymbol{B}_2$$



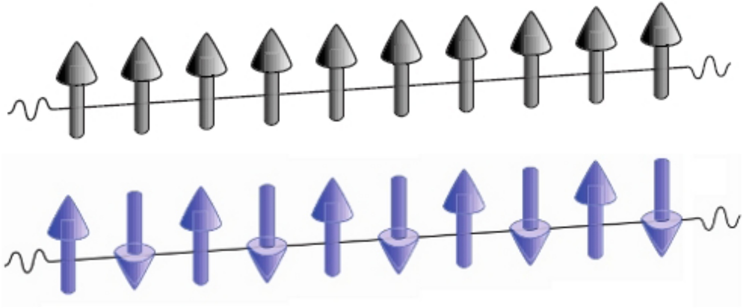
super-exchange $\sim$ meV – 0.5eV

overlapping orbitals
$+$ Pauli principle
$+$ Coulomb interaction

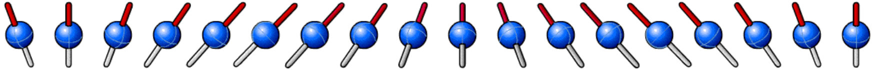


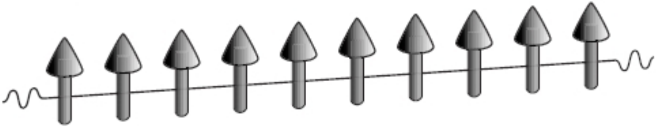
Magnetic interactions – magnetic long-range order

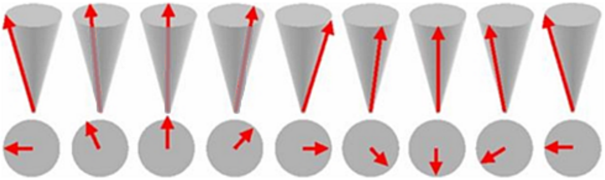
may favor parallel / antiparallel magnetic moments: Ferromagnet / Antiferromagnet



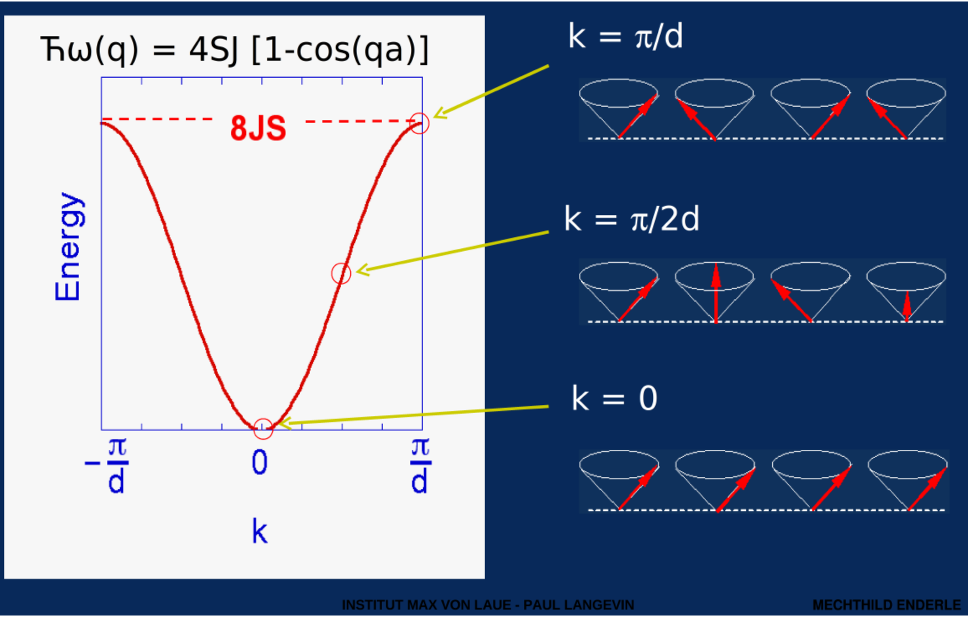
Spin waves in a ferromagnet



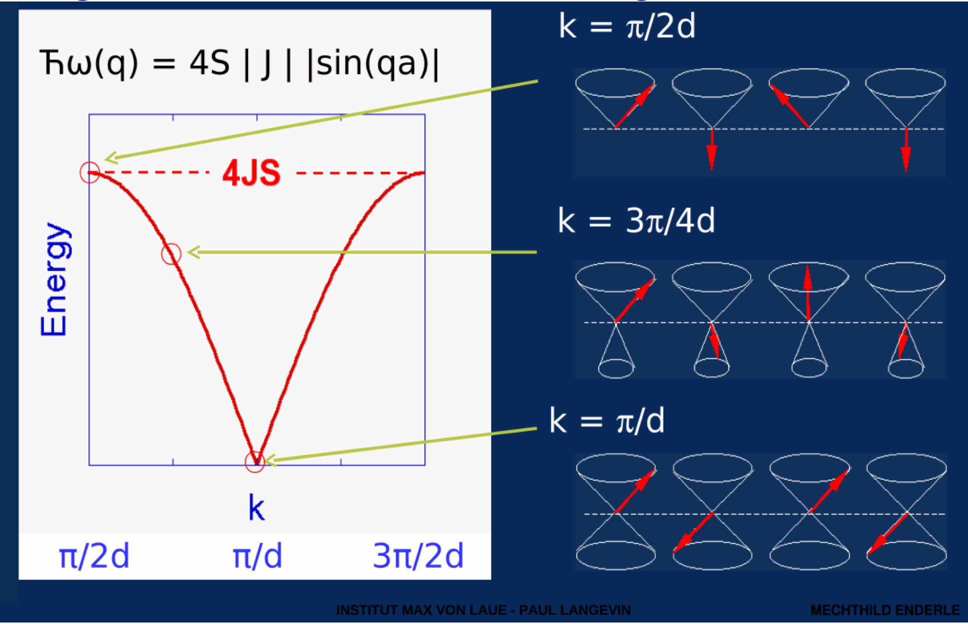




Collective excitations of the ferromagnet: magnons

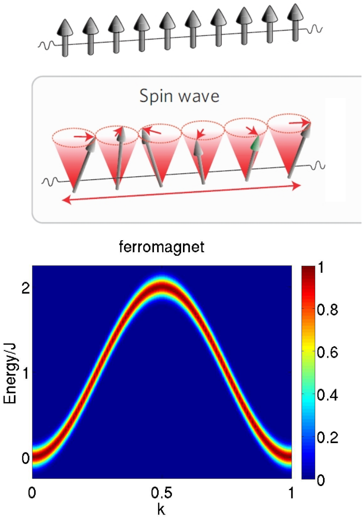


Magnons in the "classical" antiferromagnet

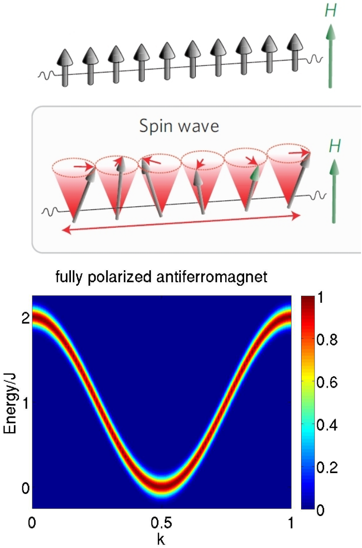


Magnon dispersion reveals microscopic interactions

Ferromagnet

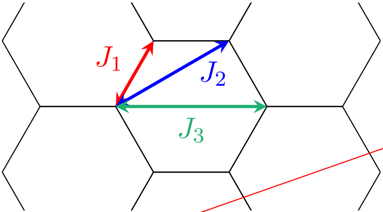


Saturated antiferromagnet $H > H_{\mathsf{sat}}$

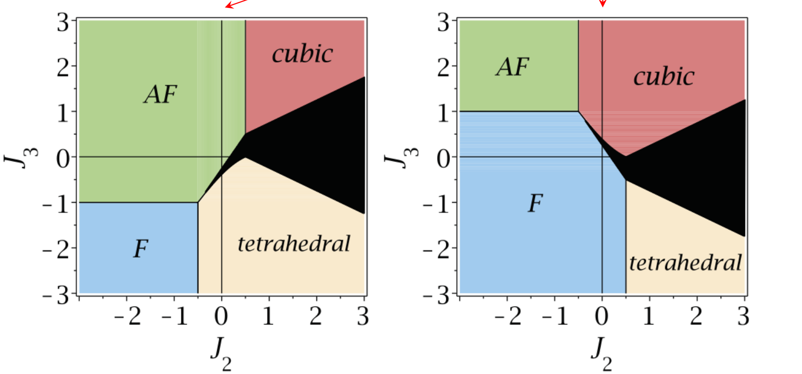


© Mechthild Enderle

Same magnetic structure for large variety of interactions



Messio et al PRB 2011

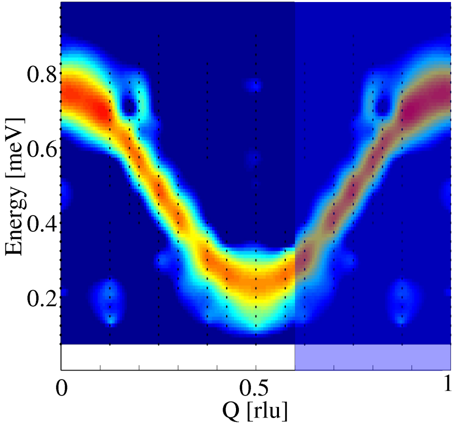


Magnon dispersion reveals microscopic interactions

$CuSO_4.5D_2O$

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

no long range order >0.1K



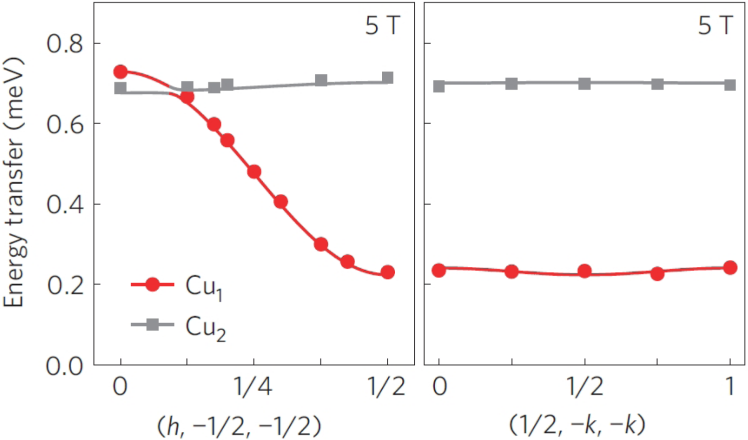
⇑

antiferromagnetic exchange

Magnon dispersion reveals microscopic interactions

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

magnetic springs only in one direction



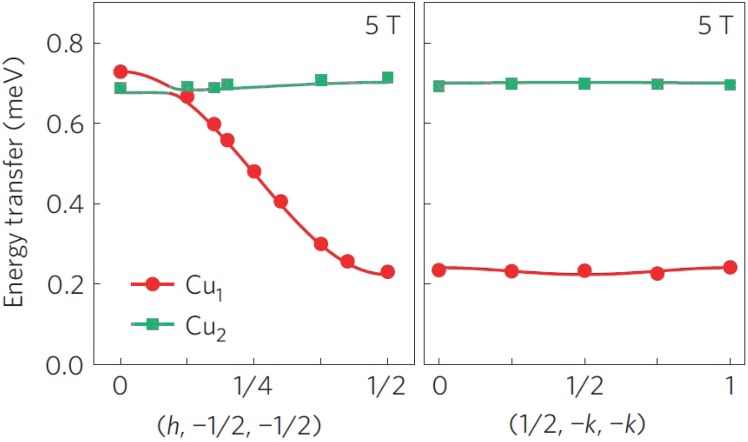
magnetically 1D !

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

Magnon dispersion reveals microscopic interactions

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

no springs/ no interaction: local transition

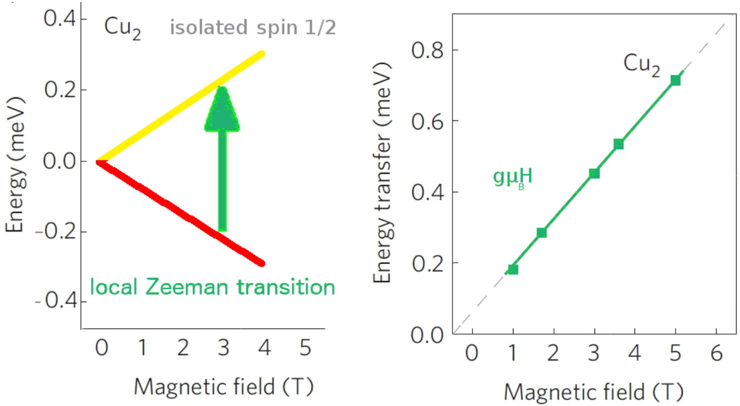


no springs

Energy independent of $\boldsymbol{Q}$ for all directions of $\boldsymbol{Q}$

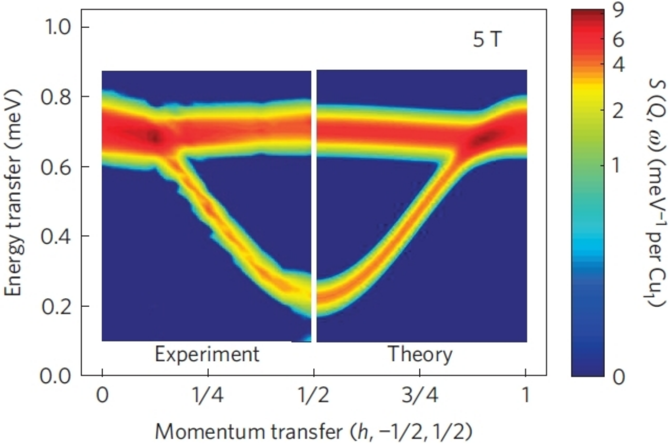
Local spin flip between Zeeman-split states

$CuSO_4.5D_2O$



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

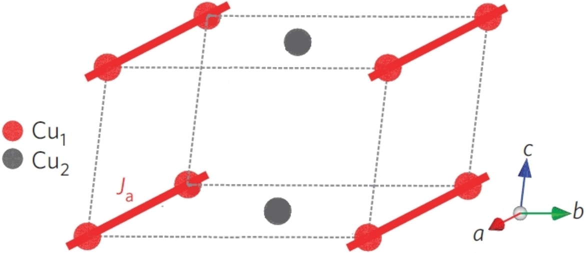
Fully saturated $CuSO_4.5D_2O$



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

Spin waves in fully saturated $CuSO_4.5D_2O$

→ microscopic scheme of magnetic interactions



Cu_1: one-dimensional arrays with antiferromagnetic interaction
Cu_2: not coupled by any interaction

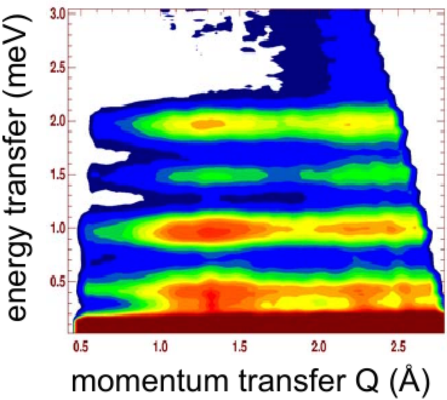
M. Mourigal, M.E. *et al.* Nat. Phys. **9** 435 (2013).

Local excitations: infinitely weak "springs"

Signature: flat dispersion

- Molecular magnets
- Crystal field excitations (Rare Earth)

$CsFe_8$ IN5



O. Waldmann, APS lecture 2006

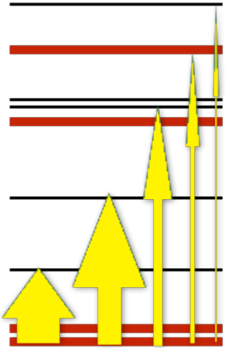
Local transitions: Crystal Electric Field Splitting

$Tb_2Ti_2O_7$

Tb^{3+}:

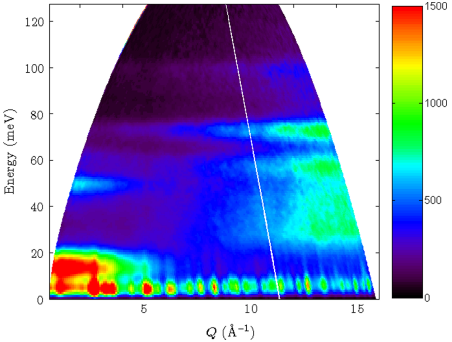
$$^7F_6 \quad \left\{ \begin{array}{c} S = 3 \\ L = 3 \end{array} \right\} \quad J = 6$$

Stark effect



Merlin $E_i = 150$meV

powder, $T = 7$K



CF phonons: $I(Q) \sim Q^2$

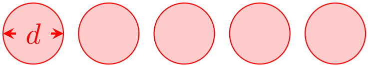
A. J. Princep *et al.* PRB **91** 224430 (2015).

Distinction between lattice and magnetic excitations

real space

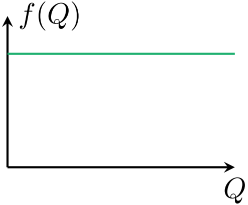
nucleus = point

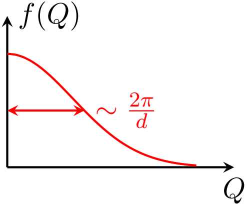
extended magnetic electron shell



Interference pattern in reciprocal $\boldsymbol{Q}$ space:

$\underbrace{\text{Fourier-transformed scattering object}}_{\text{form factor } f(Q)} \times$ reciprocal lattice



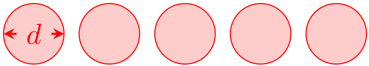


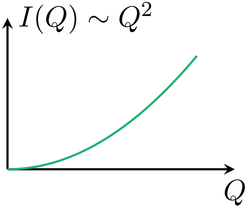
Distinction between lattice and magnetic excitations

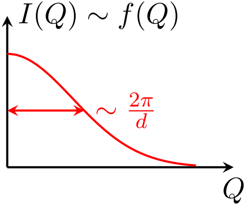
real space

nucleus = point

extended magnetic electron shell

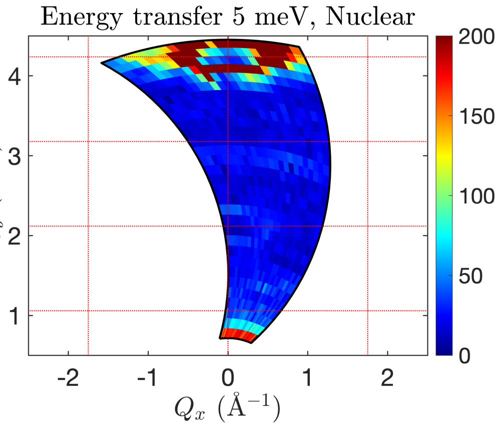


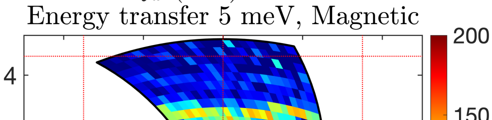




Isolation of electronic magnetic excitations

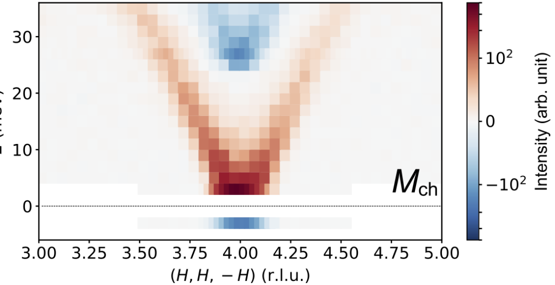
Polarized neutrons - polarization analysis of scattered neutrons

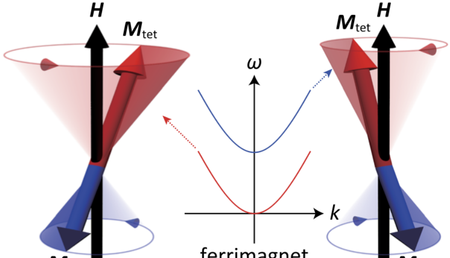




"Chirality" – precession sense of magnetic excitations

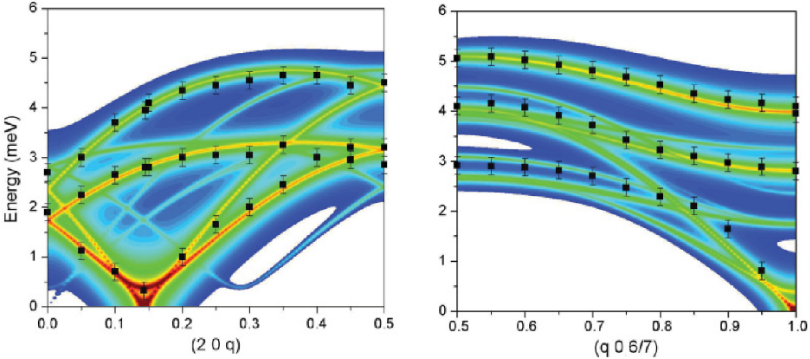
Polarized neutrons - polarization analysis of scattered neutrons





Reality is not always simple . . . – Intensities !

J. Jensen (2011) PRB 84, 104405



Coherent intensities – 4D interference pattern

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \underbrace{\sum_{n_0,\sigma_{i,f}} p(n_0) \left| \langle \boldsymbol{k}_f \sigma_f n_1 | V | \boldsymbol{k}_i \sigma_i n_0 \rangle \right|^2 \cdot \delta(\epsilon_1 - \epsilon_0 - (E_i - E_f))}_{S(\boldsymbol{Q},\omega)}$$

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

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

Coherent intensities – 4D interference pattern

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \underbrace{\sum_{n_0,\sigma_{i,f}} p(n_0)\,|\,\langle \boldsymbol{k}_f\sigma_f n_1 | V | \boldsymbol{k}_i\sigma_i n_0\rangle\,|^2 \cdot \delta(\epsilon_1 - \epsilon_0 - (E_i - E_f))}_{S(\boldsymbol{Q},\omega)}$$

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

$$S_M^{\alpha\beta}(\boldsymbol{Q},\omega) = \frac{(\gamma r_0)^2}{2\pi\hbar}\int d\boldsymbol{r}dt\; e^{i(\boldsymbol{Q}\boldsymbol{r}-\omega t)}\,\langle\, M_\perp^\alpha(\boldsymbol{0},0)\; M_\perp^\beta(\boldsymbol{r},t)\,\rangle_T$$

Coherent intensities – 4D interference pattern

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \underbrace{\sum_{n_0,\sigma_{i,f}} p(n_0)\,\left|\,\langle \boldsymbol{k}_f\sigma_f n_1|V|\boldsymbol{k}_i\sigma_i n_0\rangle\,\right|^2 \cdot \delta(\epsilon_1-\epsilon_0-(E_i-E_f))}_{S(\boldsymbol{Q},\omega)}$$

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

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

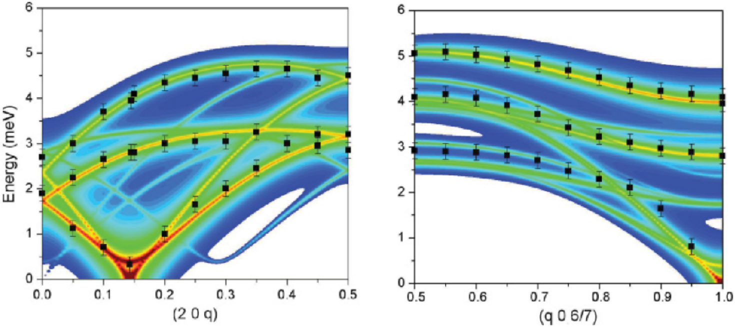
intensity — nuclear-positional, magnetic } density pair correlation function

Magnon intensities

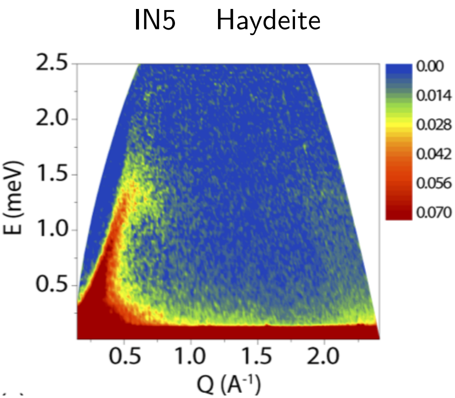
Long-range ordered structures
length of ordered moment identical at equivalent sites
transverse excitations

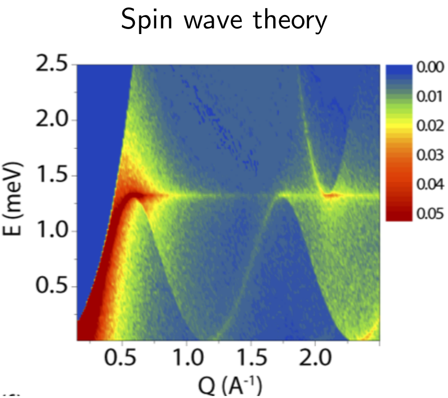
"Classical" Spin Wave Theory

J. Jensen (2011) PRB 84, 104405



Magnon intensities – essential for powder experiments !





D. Boldrin, B. Fåk, M.E., *et al.* PRB 2015

Magnetic excitations: more than spin waves

So far:

Ground state: periodically ordered atoms or magnetic moments

Collective excitations:

phonons spin waves	small oscillations around the	structural magnetic	order

Now:

periodically ordered magnetic sites with a local magnetic moment

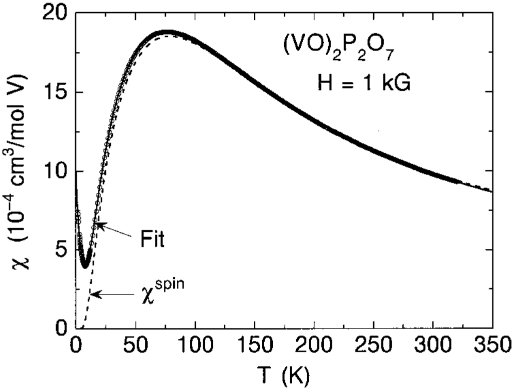
interaction between the spins (e.g. visible in $\chi(T)$)

no long-range ordered magnetic moment

Collective excitations ?

Collective phenomena without magnetic long-range order

χ displays interactions – but no phase transition



Two spins $\frac{1}{2}$ and an antiferromagnetic spring

$S = \frac{1}{2}$ at each site

strong antiferromagnetic	coupling between	next-neighbours
no	coupling between	pairs

Dimer: Pair spin 0 $\qquad \frac{1}{\sqrt{2}} \left[\, | \uparrow\downarrow\rangle - | \downarrow\uparrow\rangle \, \right]$

Local singlet-triplet excitations

$S = \frac{1}{2}$ at each site

strong antiferromagnetic	coupling between	next-neighbours
no	coupling between	pairs

Triplon: Pair spin 1

$$\begin{cases} \quad |\uparrow\uparrow\rangle \\ \frac{1}{\sqrt{2}} \left[\, |\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \, \right] \\ \quad |\downarrow\downarrow\rangle \end{cases}$$

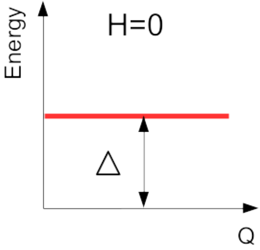
Triplons – Signature Zeeman splitting

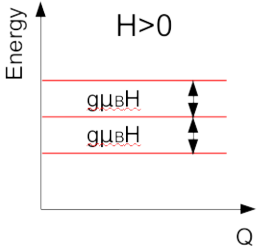
$S = \frac{1}{2}$ at each site

strong antiferromagnetic coupling between next-neighbours

no coupling between pairs

$$\begin{matrix} & |\uparrow\uparrow\rangle & \\ \frac{1}{\sqrt{2}} & \left[\, |\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \,\right] & \\ & |\downarrow\downarrow\rangle & \end{matrix} \Bigg\}$$



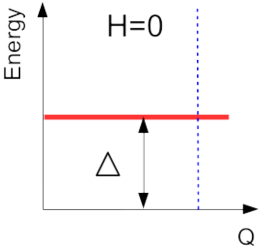


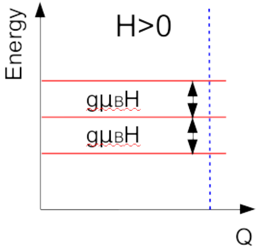
Triplons – Signature Zeeman splitting

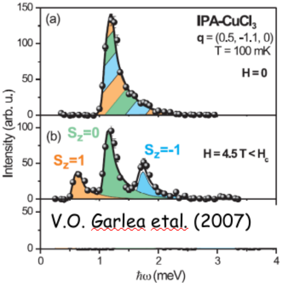
$S = \frac{1}{2}$ at each site

strong antiferromagnetic coupling between next-neighbours

no coupling between pairs



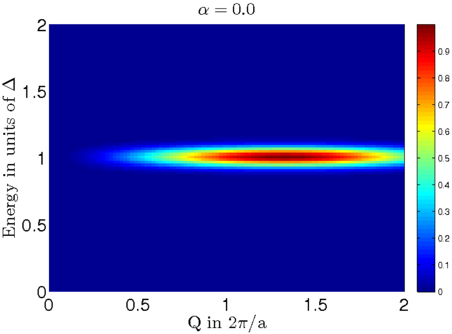




Non-Interacting triplons – intensity signature

$S = \frac{1}{2}$ at each site

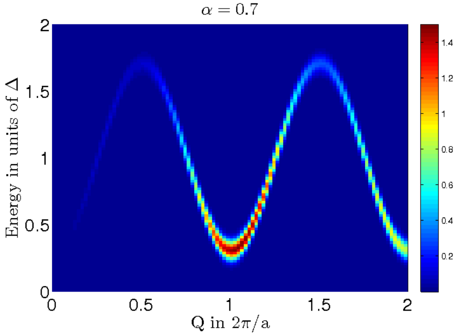
strong antiferromagnetic	coupling between	next-neighbours
no	coupling between	pairs



Interacting triplons – propagation – dispersion

$S = \frac{1}{2}$ at each site

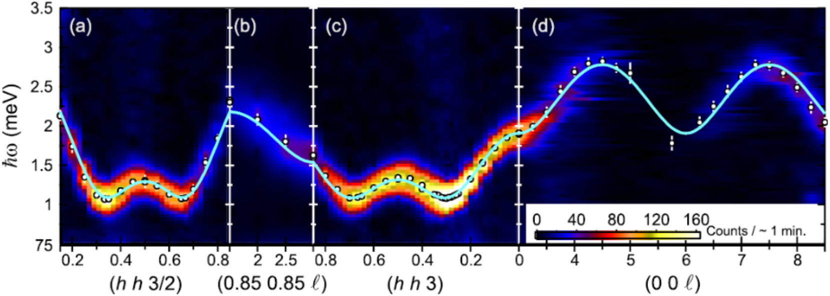
strong antiferromagnetic	coupling between	next-neighbours
increasing	coupling between	pairs



Interacting triplons – propagation – dispersion

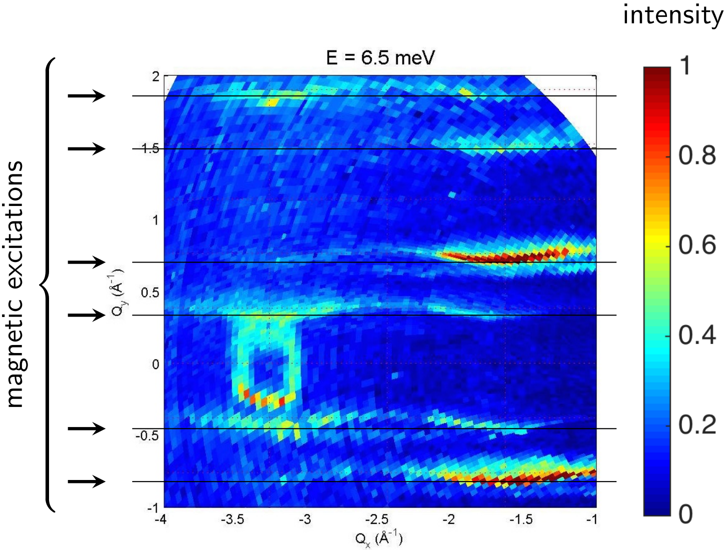
$S = \frac{1}{2}$ at each site

strong antiferromagnetic	coupling between	next-neighbours
increasing	coupling between	pairs



M.B. Stone *et al.* PRL **100** 237201 (2008)

Triplons in nearly-1D coupled dimers: $(VO)_2P_2O_7$



M.E., U. Hansen, P. Steffens, IN20

Coherent excitations – How do we measure them ?

- powder on TOF – valuable info
- single crystal TOF – large overview of Q-E-space
- single crystal TAS – large overview of Q-space at selected E
- questions at specific $\boldsymbol{Q}$, specific H,p,T: TAS
- small single crystal: TAS
- inelastic polarized: TAS (today !)