Inelastic Neutron Scattering

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Outline

- Diffuse and collective motion
- Lattice dynamics: phonons
- Magnetic dynamics: spin waves and more
- Key features as seen in powder samples
- Choice of instrument

Collective and diffuse motion

Collective dynamics

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

Snapshot:
periodic pattern – wave

Diffuse (Brownian) motion

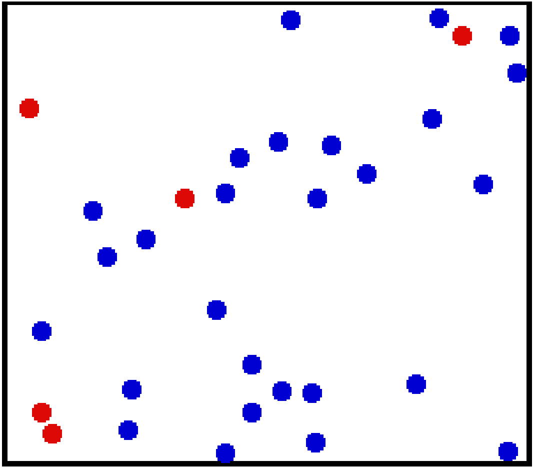
"random walk"
uncorrelated, diffuse motion

disordered

Diffuse (Brownian) motion

Disordered materials
(non-periodic materials)

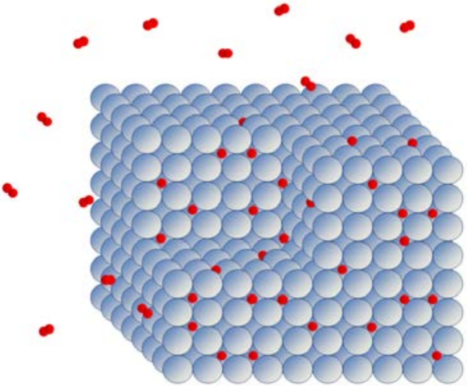
- polymers
- liquids
- macromolecules
- biological cells



Diffuse motion: examples

Disordered phenomena in crystals

- hydrogen diffusion
- spin diffusion, e.g. critical scattering near a phase transition



Signature of diffuse motion: Quasielastic scattering

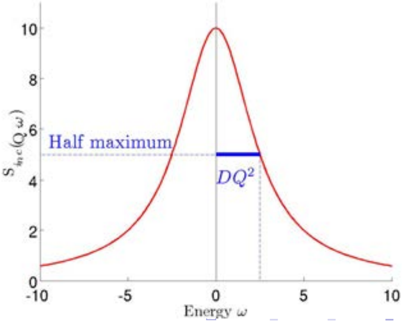
$$\left.\frac{d^2\sigma}{d\omega d\Omega}\right|_{\text{inc}} = \frac{\sigma_{\text{inc}}}{4\pi}\frac{k_f}{k_i} N S_{\text{inc}}(\boldsymbol{Q},\omega)$$

$$S_{\text{inc}}(\boldsymbol{Q},\omega) = \frac{1}{2\pi\hbar}\int dt\, d\boldsymbol{r}\; e^{i(\boldsymbol{Q}\boldsymbol{r}-\omega t)}\; G_{\text{self}}(\boldsymbol{r},t)$$

$$G_{\text{self}}(\boldsymbol{r},t) = \frac{1}{N}\sum_j \int d\boldsymbol{r}'\langle\; \delta(\boldsymbol{r}'-\boldsymbol{R}_j(0))\; \delta(\boldsymbol{r}'+\boldsymbol{r}-\boldsymbol{R}_j(t))\;\rangle_T$$

Incoherent cross section $\sim$ autocorrelation

correlation of a particle/spin at $\boldsymbol{r}=0, t=0$ with the same particle/spin at $\boldsymbol{r}, t$



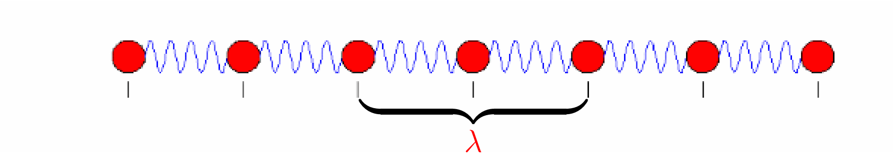
Instruments for Quasielastic scattering

Slow dynamics $\Rightarrow$ high energy resolution

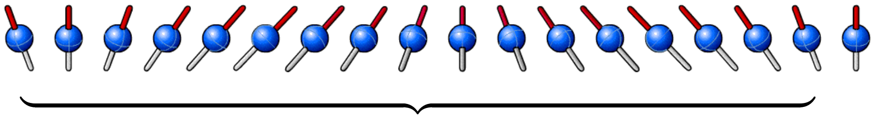
- cold Time Of Flight
- backscattering
- spin echo spectroscopy

Collective motion – coherent dynamics – "ballet"

phonons



magnons



wavelength λ

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

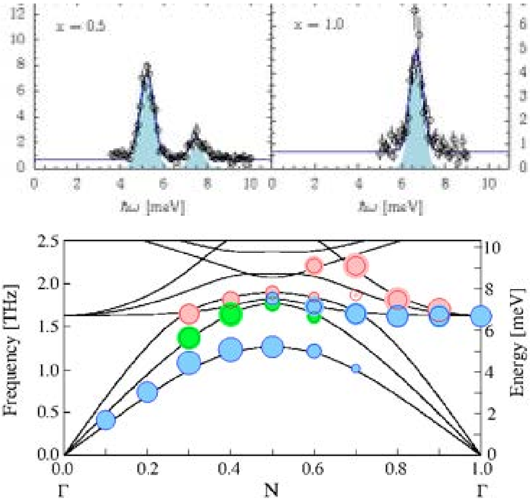
Collective dynamics: Signature dispersion

phonons (IN8)

- in periodic arrays/ crystals
- in liquids (sound wave)

Dispersion:

discrete $\hbar\omega$ at each $\boldsymbol{Q}$



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

Collective motion – wave – interference pattern

Coherent cross section

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right|_{\text{coh}} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{n_0,n_1} p(n_0) \left| \langle n_1 | V(\boldsymbol{Q}) | n_0 \rangle \right|^2 \delta(\varepsilon_1 - \varepsilon_0 - \hbar\omega)$$

$$= \frac{k_f}{k_i} \underbrace{\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{n_0} p(n_0) \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} dt\; e^{-i\omega t} \langle n_0 \,|\, V^*(\boldsymbol{Q},0) V(\boldsymbol{Q},t) \,|\, n_0 \rangle}$$

$$= \frac{k_f}{k_i} \quad S(\boldsymbol{Q},\omega)$$ coherent dynamic scattering function

Coherent dynamic scattering function

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

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

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

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

$S(\boldsymbol{Q},\omega)$ is the space-time Fourier transform of the

nuclear-positional / magnetic — density-density pair correlation function

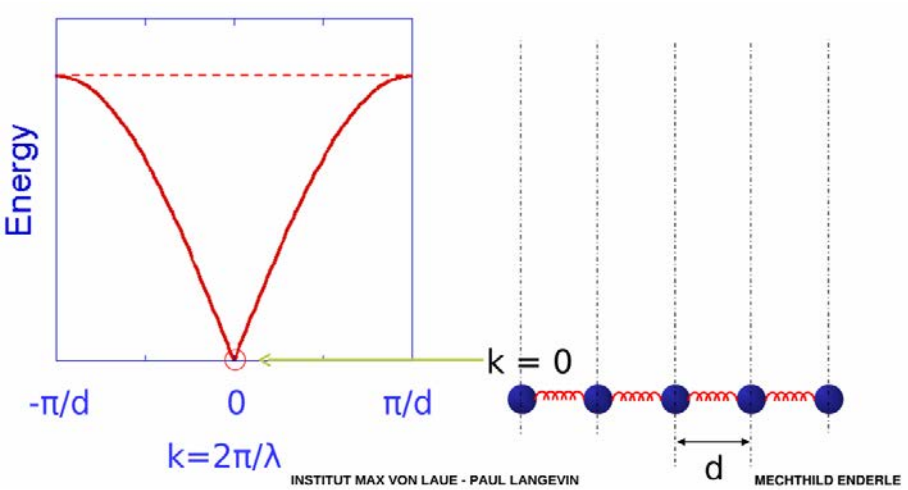
Collective and diffuse motion

Collective dynamics	Brownian motion
atoms/magnetic moments move "correlated" ("ballet")	"random walk" uncorrelated, diffuse motion
Snapshot: periodic pattern – wave	disordered
coherent cross section $\sim$ pair correlation function:	incoherent cross section $\sim$ 1-particle autocorrelation:
$\rightarrow$ dispersion relation $\rightarrow$ structural pattern	$\rightarrow$ diffusion coefficient no structural information

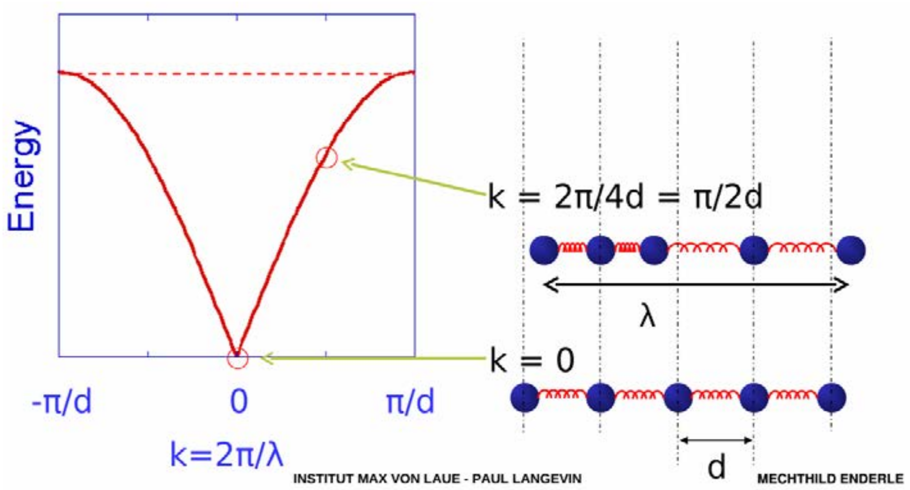
Collective dynamics in the incoherent cross section:

$\sim$ density of states $Z(\omega)$ – loss of all structural information

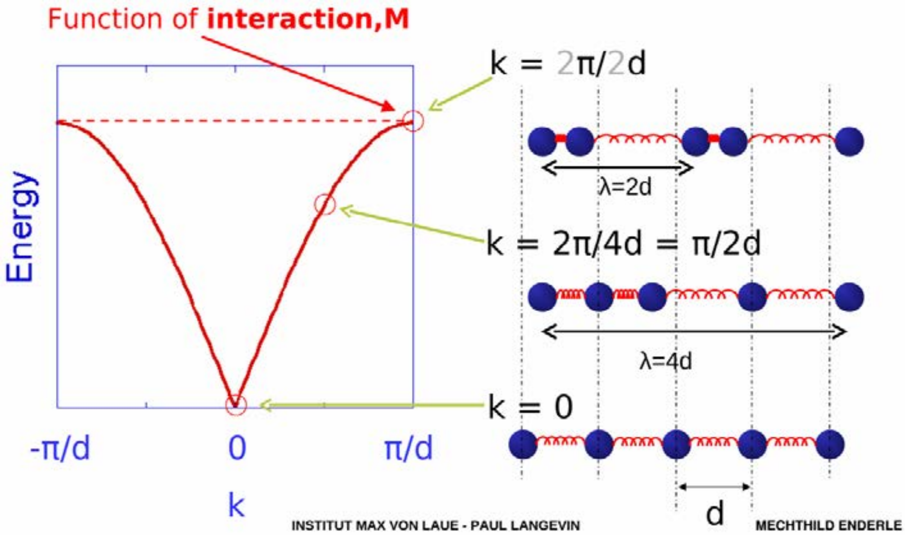
Collective excitations of the lattice: phonons



Collective excitations of the lattice: phonons



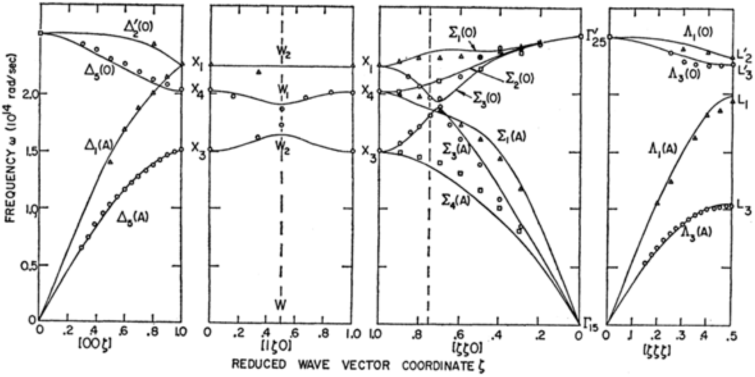
Collective excitations of the lattice: phonons



Phonons in diamond

diamond:  covalent bonds

1000 meV

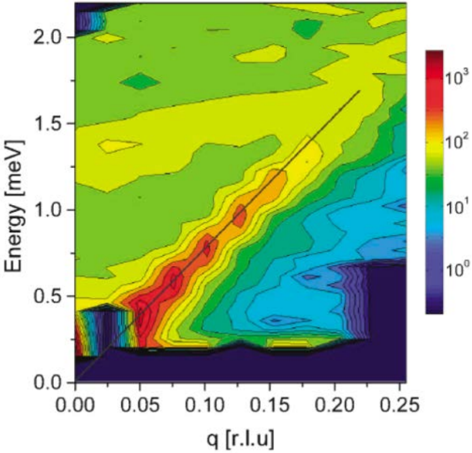


J.L. Warren *et al.* Phys.Rev. **158** 805 (1967)

Phonons in bcc ^{4}He

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

1 meV



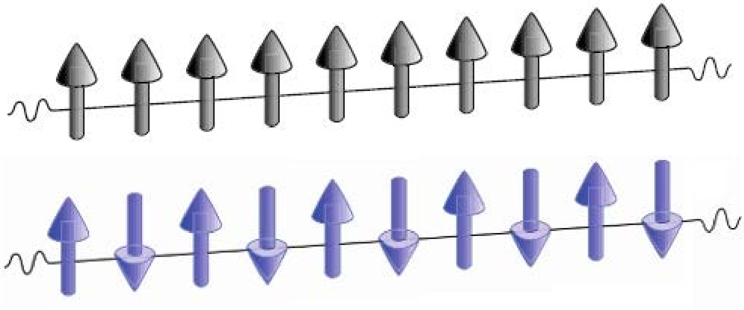
$T = 1.6400(1)$K

IN12

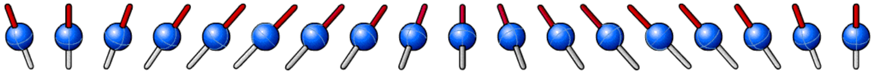
M. Markovich *et al.* PRL **88** 195301 (2002)

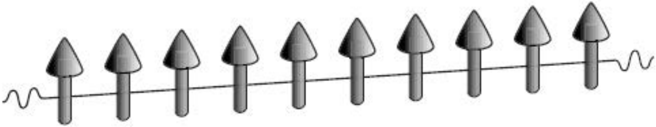
"Magnetic springs"

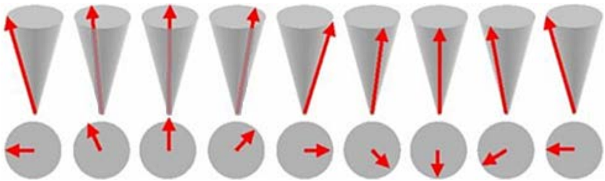
may favor parallel / antiparallel magnetic moments: Ferromagnet / Antiferromagnet



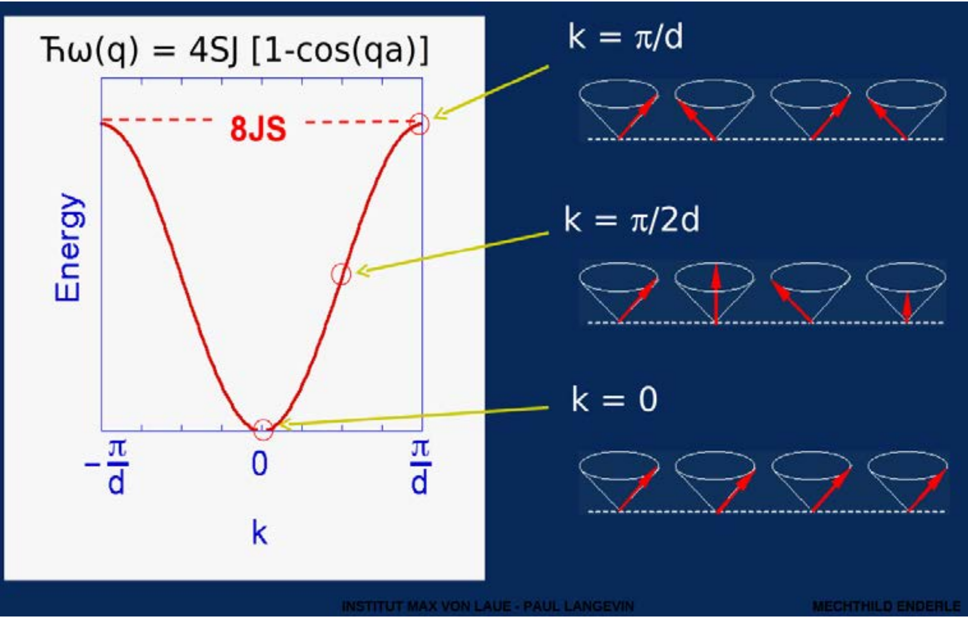
Spin waves in a ferromagnet



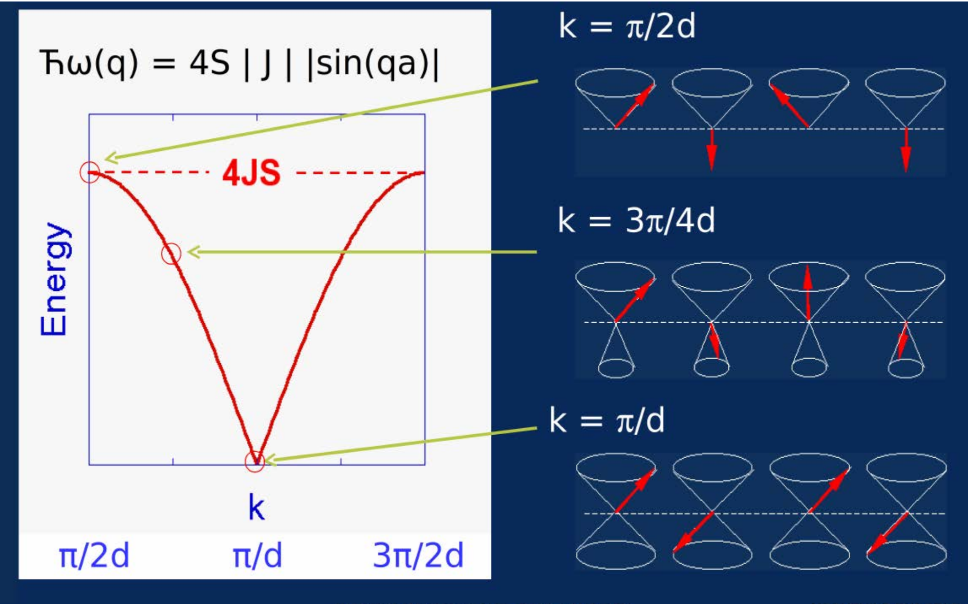




Collective excitations of the ferromagnet: magnons

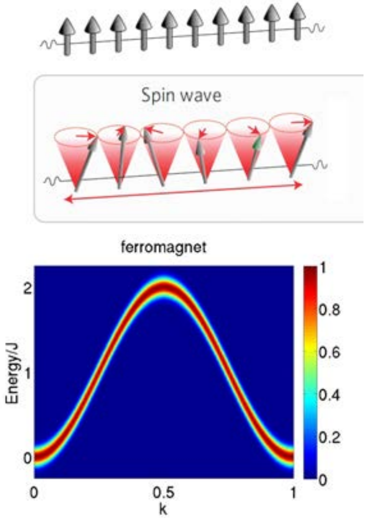


Magnons in the "classical" antiferromagnet

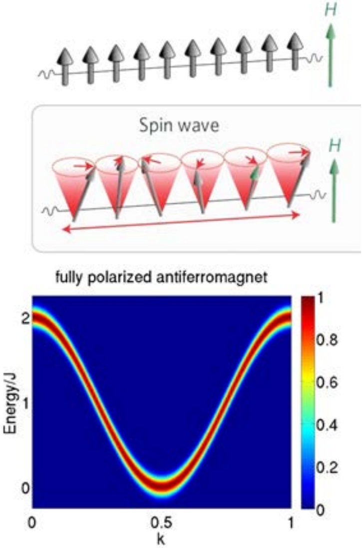


Magnon dispersion reveals microscopic interactions

Ferromagnet

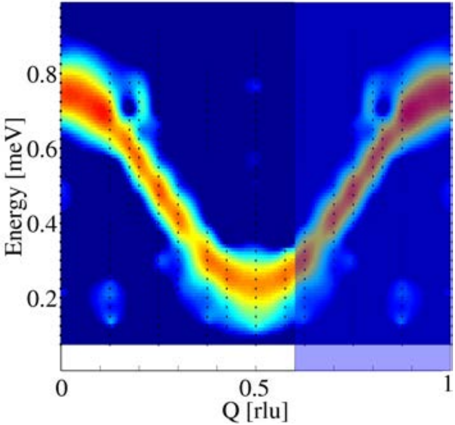


Saturated antiferromagnet $H > H_{\text{sat}}$



Magnon dispersion reveals microscopic interactions

$CuSO_4.5D_2O$



$H > H_{sat}$

no long range order >0.1K

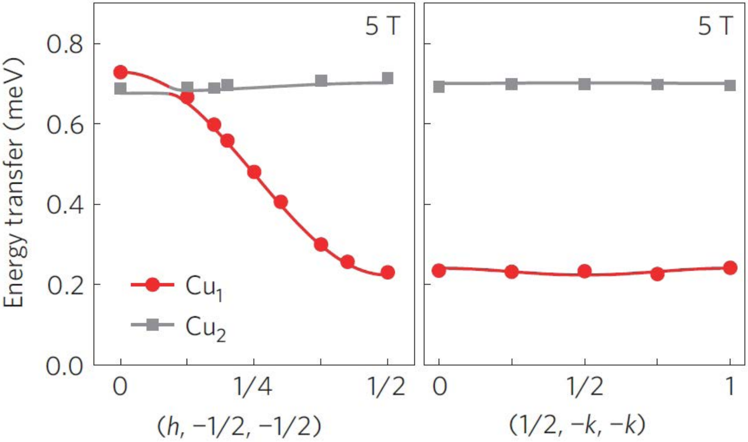
⇑

antiferromagnetic exchange

Magnon dispersion reveals microscopic interactions

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

magnetic springs only in one direction



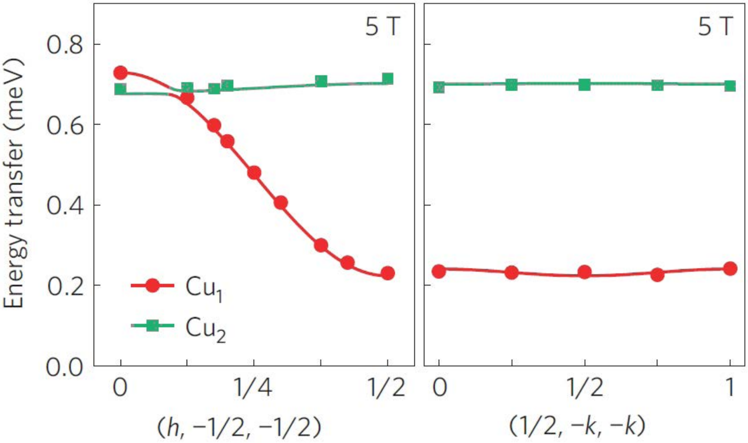
magnetically 1D !

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

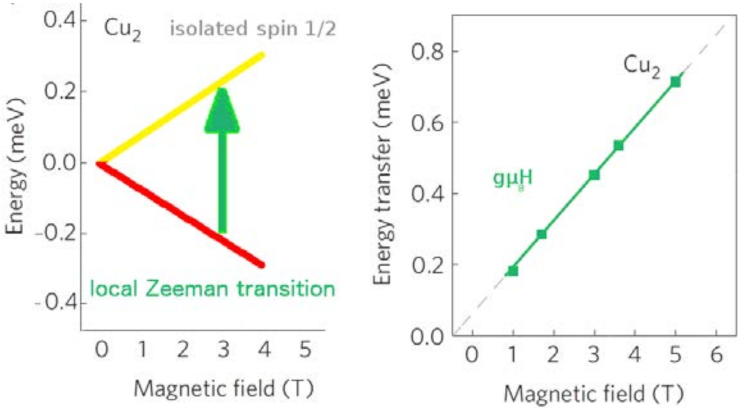


no springs

Energy independent of Q for all directions of Q

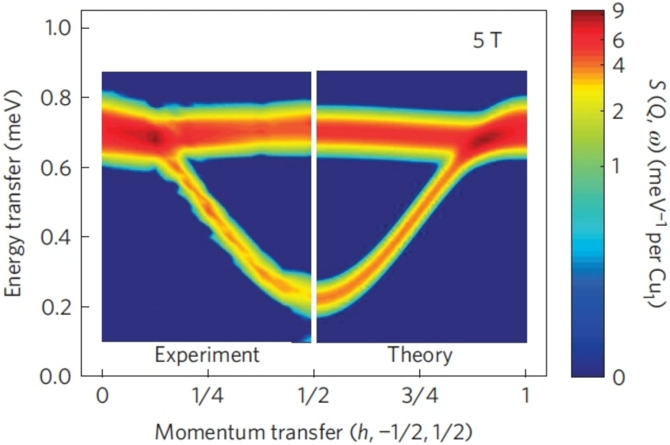
Local spin flip between Zeeman-split states

$CuSO_4.5D_2O$



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

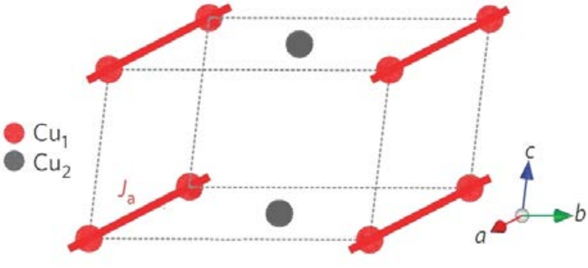
Fully saturated $CuSO_4.5D_2O$



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



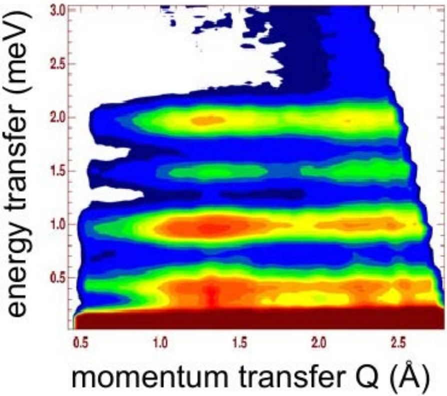
Cu_1: one-dimensional arrays with antiferromagnetic interaction
Cu_2: not coupled by any interaction

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

Local excitations: infinitely weak "springs"

Signature: flat dispersion

$CsFe_8$ IN5



- Molecular magnets
- Crystal field excitations (Rare Earth)

O. Waldmann, APS lecture 2006

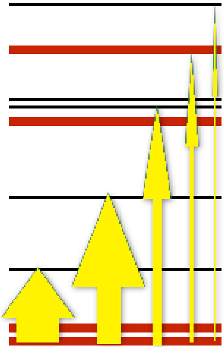
Local transitions: Crystal Electric Field Splitting

$Tb_2Ti_2O_7$

Tb^{3+}:

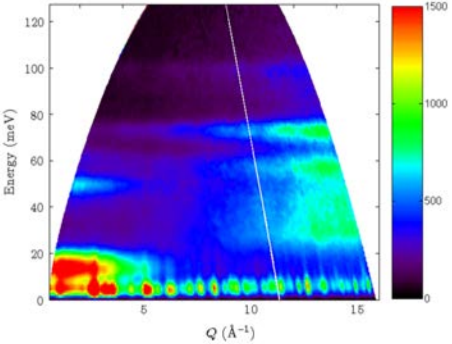
$^7F_6 \quad \left\{ \begin{array}{l} S = 3 \\ L = 3 \end{array} \right\} \; J = 6$

Stark effect



Merlin $E_i = 150\text{meV}$

powder, $T = 7\text{K}$



CF

phonons

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

Separating phonons and magnons via Q

NEUTRONS FOR SCIENCE

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 ($T \ll T_c$)

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

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

FT of unpaired-electron shell
falls off for large Q

periodic in Q (depending on d_j)

Collective dynamics reveals interactions

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 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 ?

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 $\quad \frac{1}{\sqrt{2}} \left[\, |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \,\right]$

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

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

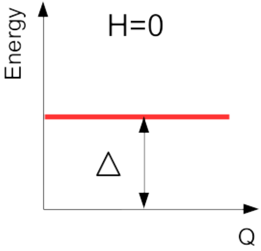
Triplons – Signature Zeeman splitting

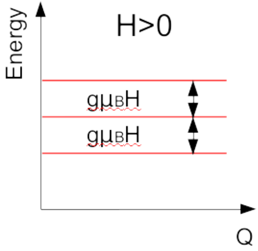
$S = \frac{1}{2}$ at each site

strong antiferromagnetic coupling between next-neighbours

no coupling between pairs

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





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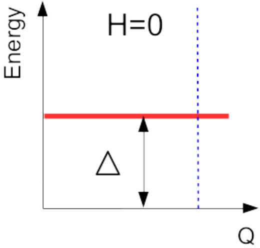
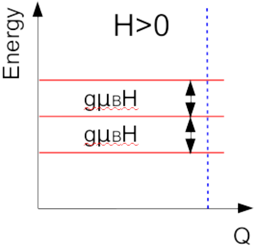
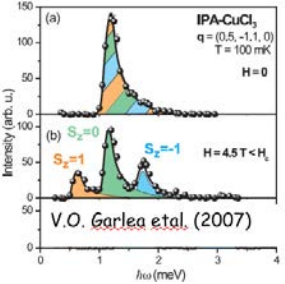




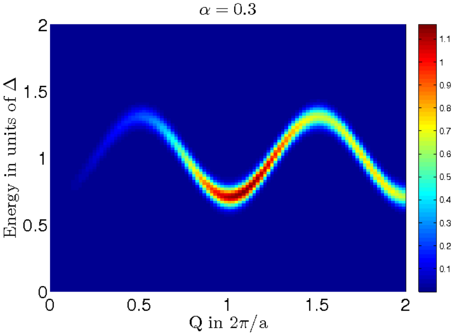


Interacting triplons – propagation – dispersion

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

strong antiferromagnetic coupling between next-neighbours

increasing coupling between pairs



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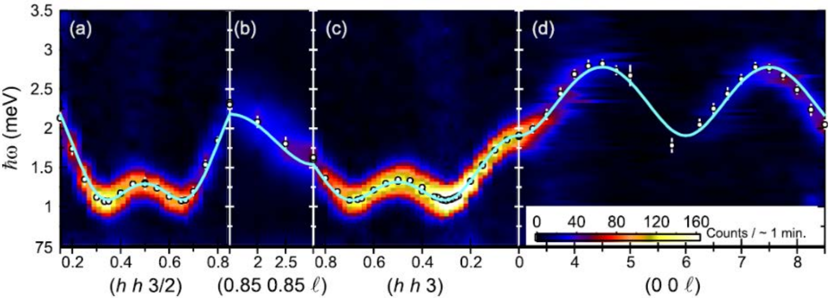
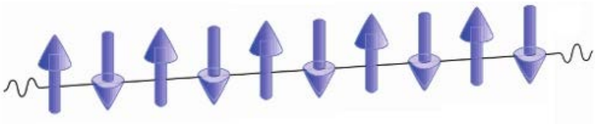
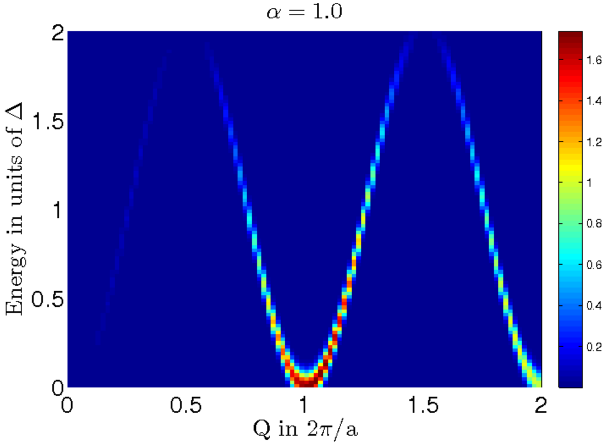


M.B. Stone *et al.* PRL **100** 237201 (2008)

1D array – Limit of uniform coupling between $S = \frac{1}{2}$





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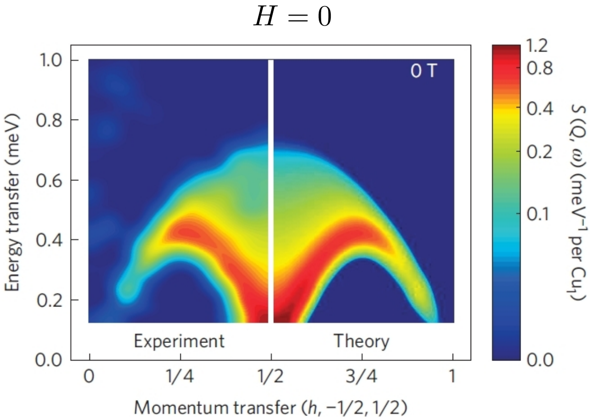
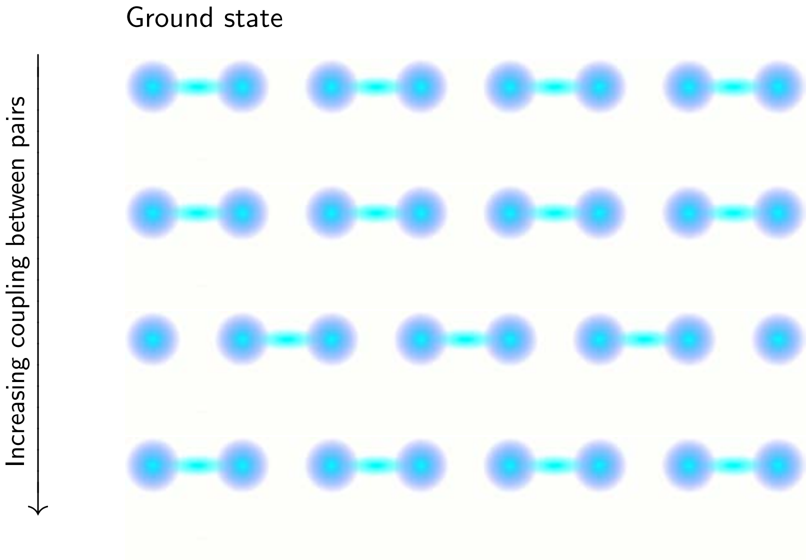


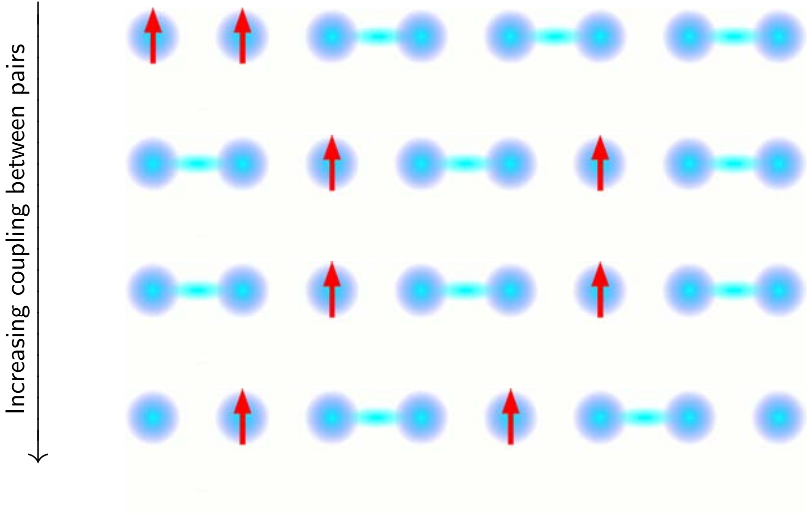
M. Mourigal, M.E. *et al.* Nat.Phys. **9** 435 (2013)

1D array – Limit of uniform coupling between $S = \frac{1}{2}$



1D array – Limit of uniform coupling between $S = \frac{1}{2}$

Triplon

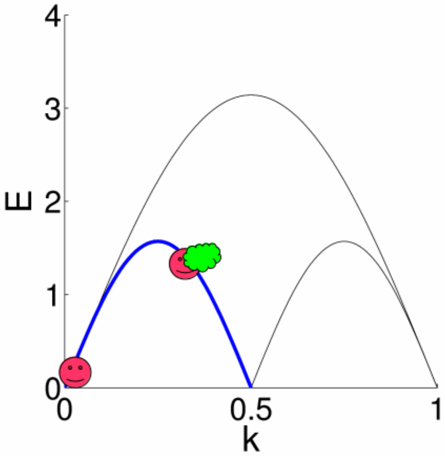


1D array – Limit of uniform coupling between $S = \frac{1}{2}$

freely propagating spin $\frac{1}{2}$ particles: spinons

Two-particle excitation: Signature continuous scattering

Neutron excites **pairs** of freely propagating spin $\frac{1}{2}$ particles



1-particle dispersion

 $E(k)$

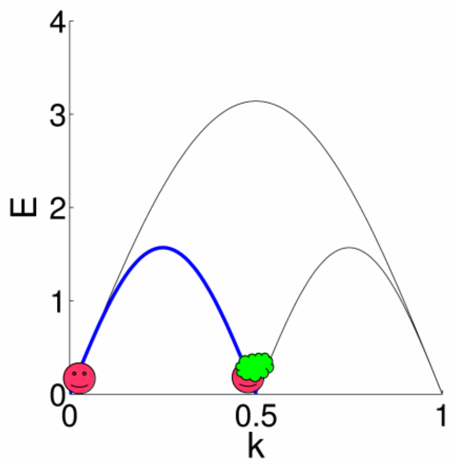
neutron excites pair

 =  + 

$$Q = k_1 + k_2$$
$$E = E(k_1) + E(k_2)$$

Two-particle excitation: Signature continuous scattering

Neutron excites **pairs** of freely propagating spin $\frac{1}{2}$ particles



1-particle dispersion

☺ $E(k)$

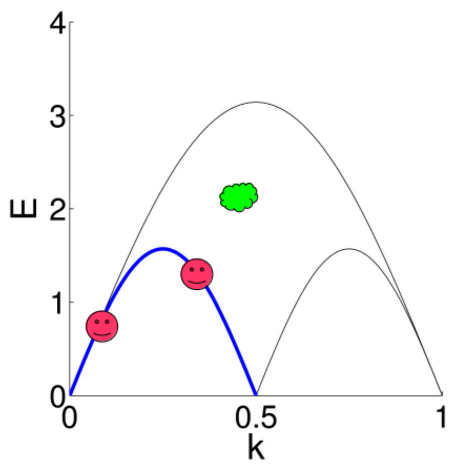
neutron excites pair

$$\text{(green blob)} = ☺ + ☺$$

$$Q = k_1 + k_2$$
$$E = E(k_1) + E(k_2)$$

Two-particle excitation: Signature continuous scattering

Neutron excites **pairs** of freely propagating spin $\frac{1}{2}$ particles



1-particle dispersion

 $E(k)$

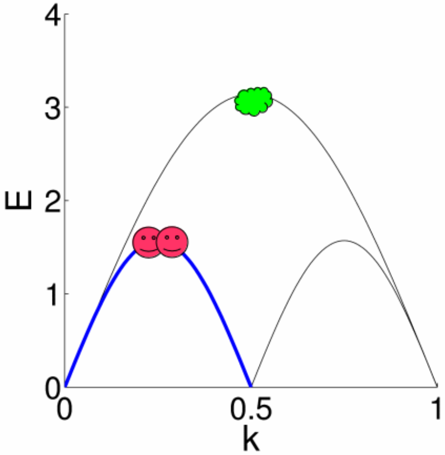
neutron excites pair

 =  + 

$$\begin{aligned} \boldsymbol{Q} &= k_1 + k_2 \\ E &= E(k_1) + E(k_2) \end{aligned}$$

Two-particle excitation: Signature continuous scattering

Neutron excites **pairs** of freely propagating spin $\frac{1}{2}$ particles



1-particle dispersion

$E(k)$

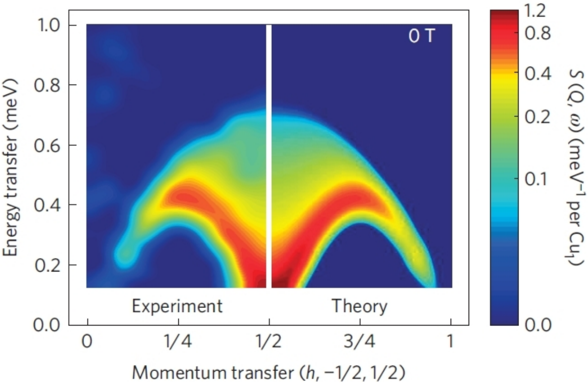
neutron excites pair

$= \quad + $

$$Q = k_1 + k_2$$
$$E = E(k_1) + E(k_2)$$

Spinon continuum in $CuSO_4.5D_2O$

$H = 0$



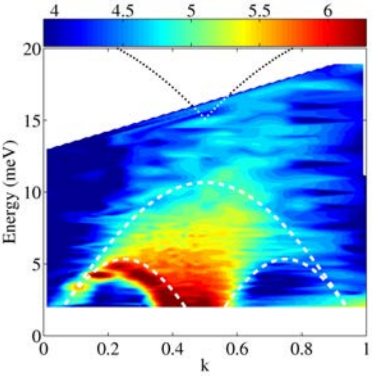
M. Mourigal, M.E. *et al.* Nat.Phys. **9** 435 (2013)

New many-particle states and excitations

2 zig-zag coupled 1D spin $\frac{1}{2}$ arrays

Continuum:
pairs of free particles

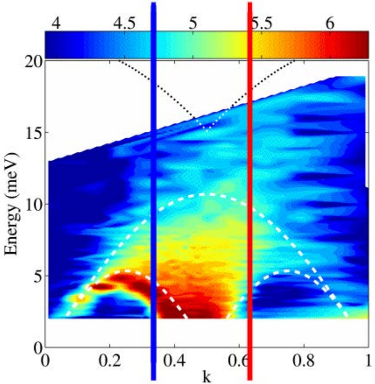
discrete branch:
bound particle-pairs

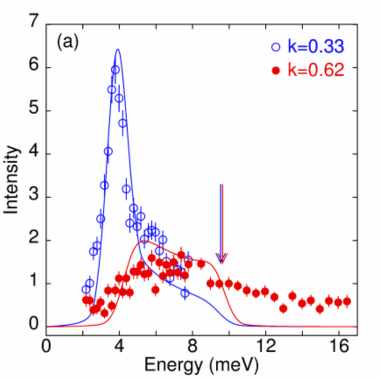


M.E. *et al.* PRL **104** 237207 (2010)

New many-particle states and excitations

2 zig-zag coupled 1D spin $\frac{1}{2}$ arrays

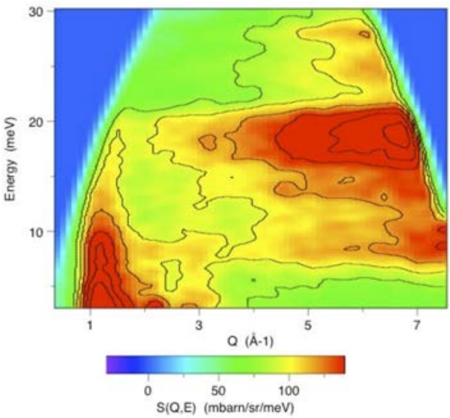




M.E. *et al.* PRL **104** 237207 (2010)

Collective excitations – powder samples

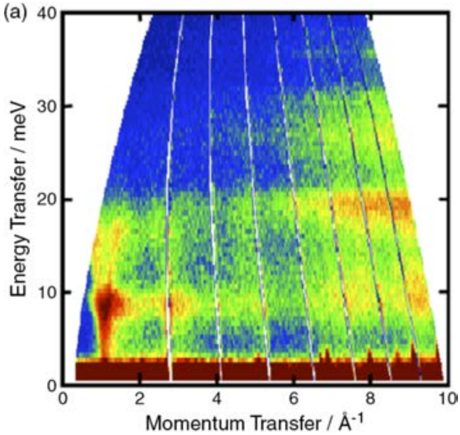
Continuum



B. Fåk *et al.* EPL **81** 17006 (2008)

Deuteronium jarosite

magnons

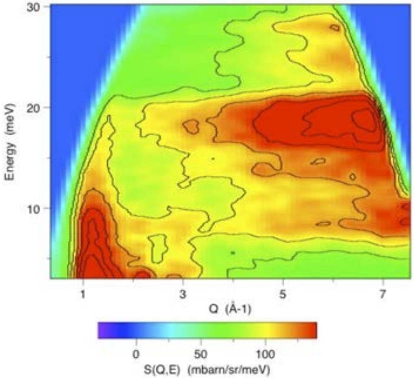


F. Coomer *et al.* JPCM **18** 8847 (2006)

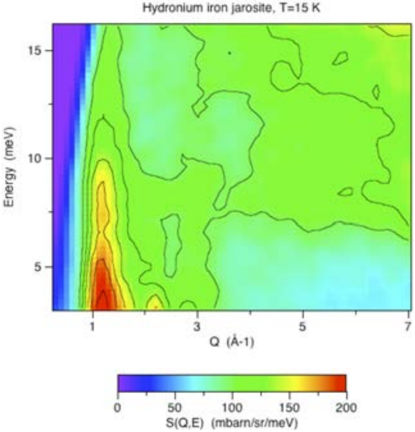
K-jarosite

Collective excitations – powder samples

IN5: deuteronium jarosite powder



magnetic only

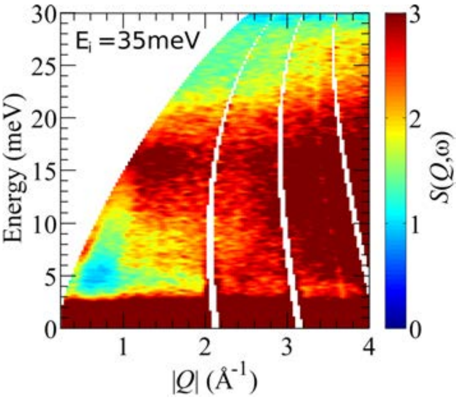


B. Fåk *et al.* EPL **81** 17006 (2008)

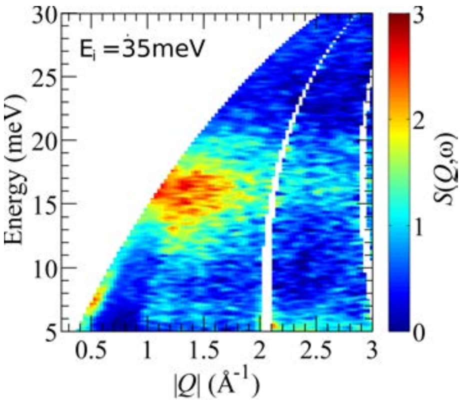
Malachite Inelastic neutron scattering on deuterated powder

MARI/ISIS

all data



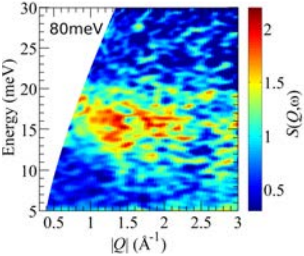
magnetic part



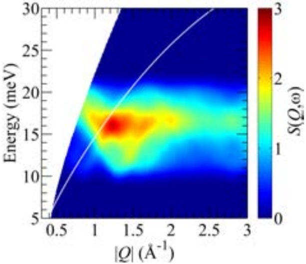
Malachite Inelastic neutron scattering on D-powder

Extracting information from powder samples: Triplons

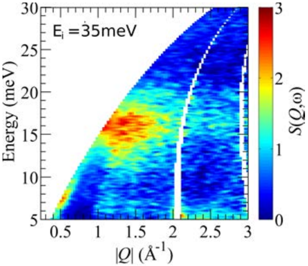
magnetic $E_i = 35$meV



model



magnetic $E_i = 80$meV

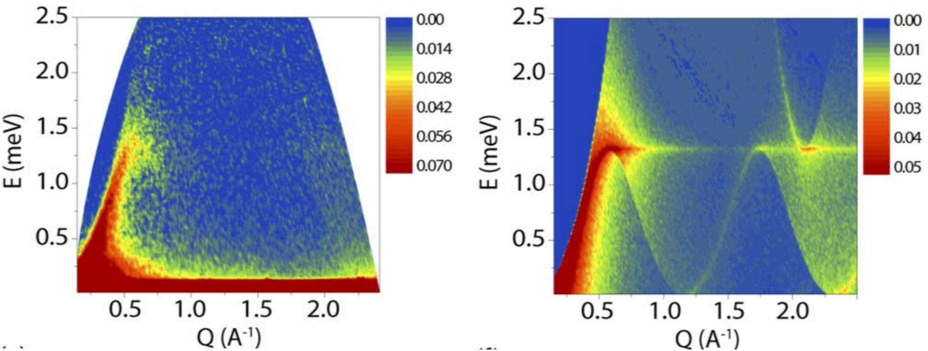


Collective excitations – powder samples: magnons

Extracting information from powder samples: Spinwaves/Magnons

IN5 Haydeite

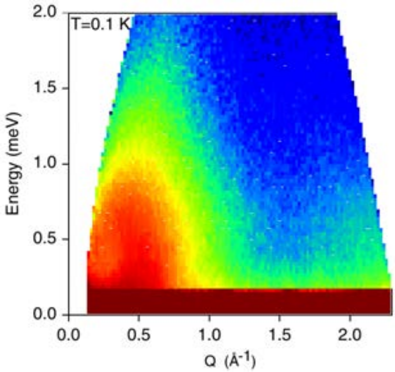
Spin wave theory



D. Boldrin, B. Fåk, M.E., *et al.* PRB **91** 220408 (2015)

Collective excitations – powder samples

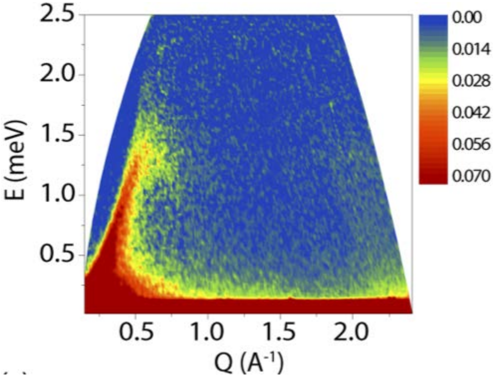
Continuum



B. Fåk *et al.*
PRL **109** 037208 (2012)

IN5 Kapellasite

Magnons

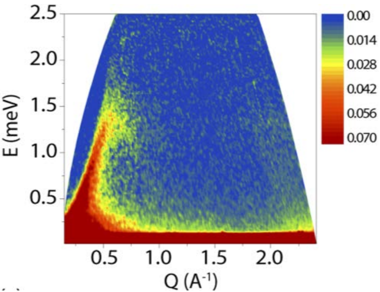


D. Boldrin, B. Fåk, M.E. *et al.*
PRB **91** 220408 (2015)

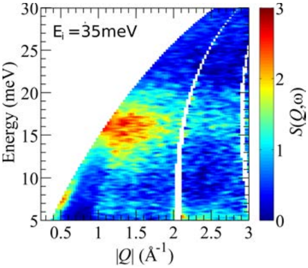
IN5 Haydeite

Collective excitations – powder samples

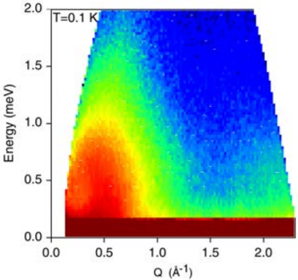
Magnons



Triplons

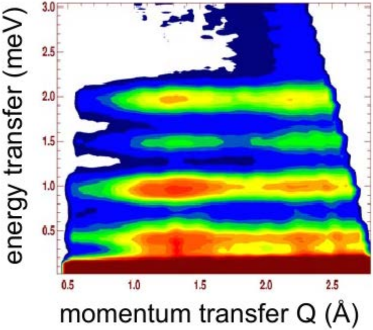


Continuum

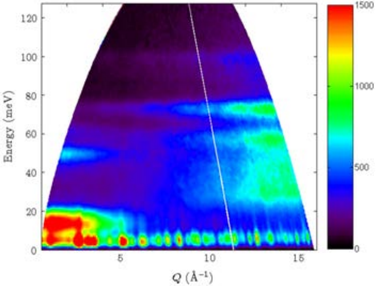


Collective excitations – powder samples

local excitations
(molecule-spin states)



local (Crystal field)
and phonons



Correlated Excitations – How do we measure them ?

- powder on TOF – valuable info
- single crystal TOF – large overview of Q-E-space
- Questions at specific Q/H,p,T: TAS
- Small single crystal: TAS
- inelastic polarized: TAS (today !)

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 $> 5\text{mm}^3$	single crystal $> 1\text{cm}^3$

TAS versus TOF

TAS	TOF
highest continuous flux at sample	pulsed structure
single analyser-detector **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 $> 5\text{mm}^3$	single crystal $> 1\text{cm}^3$