INSTITUTE FOR QUANTUM MATTER

A collaboration between
JOHNS HOPKINS UNIVERSITY
and PRINCETON UNIVERSITY

Magnetic Neutron Scattering

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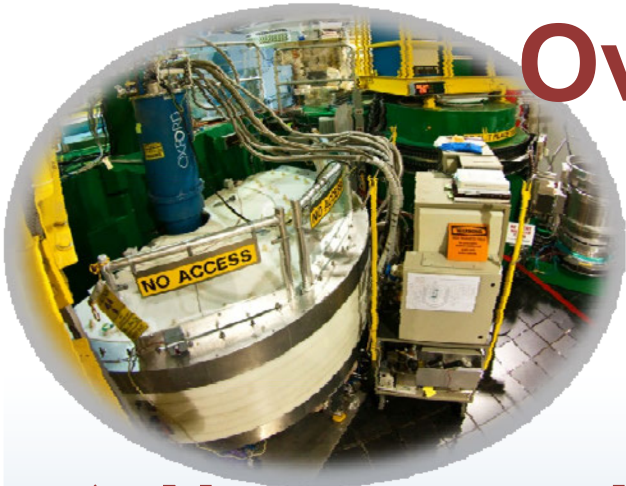
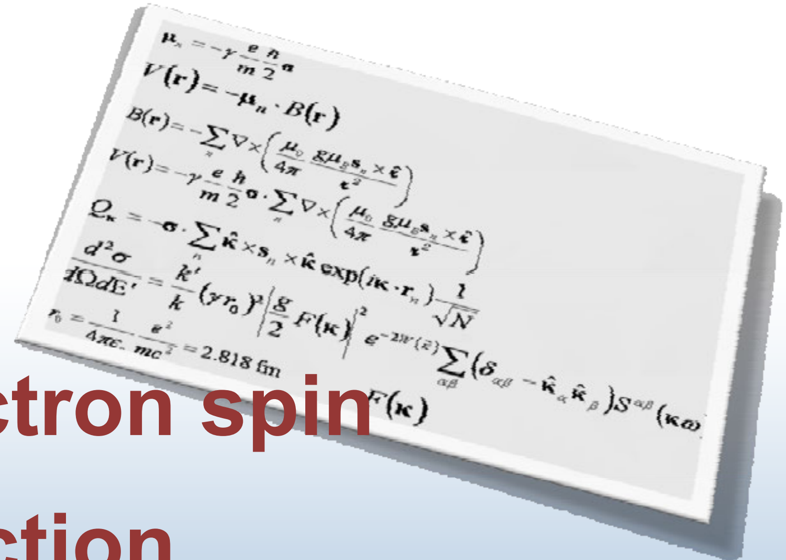
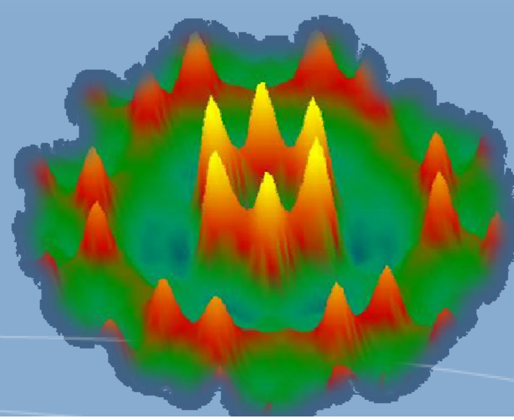
JOHNS HOPKINS UNIVERSITY

Overview of Tutorial





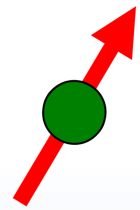
- Neutron spin meets electron spin
- Magnetic neutron diffraction
- Inelastic magnetic neutron scattering
- Polarized neutron scattering
- Summary



Magnetic properties of the neutron

The neutron has a dipole moment

$$\vec{\mu}_n = -\gamma \mu_B \frac{m_e}{m} \vec{\sigma}$$



μ_n is 960 times smaller than the electron moment

$$\frac{\mu_e}{\mu_n} = \frac{m}{m_e \gamma} = \frac{1836}{1.913} = 960$$

A dipole in a magnetic field has potential energy

$$V(\mathbf{r}) = -\vec{\mu} \cdot \mathbf{B}(\mathbf{r})$$

Correspondingly the field exerts a torque and a force

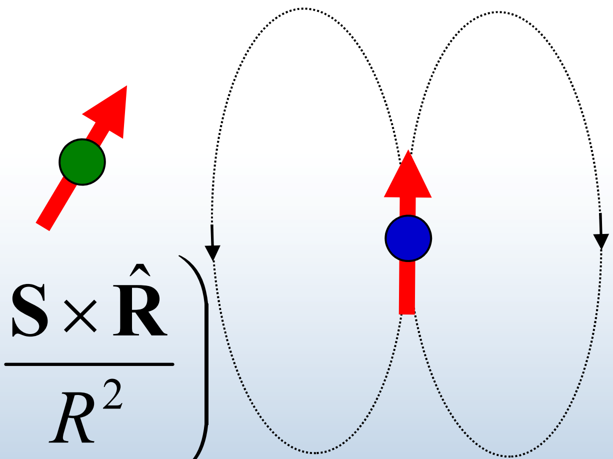
$$\vec{\tau} = \vec{\mu} \times \mathbf{B} \qquad \mathbf{F} = \nabla(\vec{\mu} \cdot \mathbf{B})$$

driving the neutron parallel to high field regions

The transition matrix element

The dipole moment of unfilled shells yield inhomog. B−field

$$\mathbf{B} = \nabla \times \left(\frac{\mu_0}{4\pi} \frac{g\mu_B \mathbf{S} \times \hat{\mathbf{R}}}{R^2} \right)$$



The magnetic neutron senses the field

$$V_m(\mathbf{r}) = -\vec{\mu} \cdot \mathbf{B}(\mathbf{r}) = -\frac{\mu_0}{4\pi} g\gamma \frac{m_e}{m} \mu_B^2 \vec{\sigma} \cdot \nabla \times \left(\frac{\mathbf{S} \times \hat{\mathbf{R}}}{R^2} \right)$$

The transition matrix element in Fermi's golden rule

$$\frac{m}{2\pi\hbar^2} \langle \mathbf{k}'\sigma'\lambda' | V_m | \mathbf{k}\sigma\lambda \rangle = -\gamma r_0 \frac{g}{2} F(\mathbf{q}) \langle \sigma'\lambda' | \vec{\sigma} \cdot \mathbf{S}_{\perp l} | \sigma\lambda \rangle \exp(i\mathbf{q} \cdot \mathbf{r}_l)$$

Magnetic scattering is similar in strength to nuclear scattering

$$\gamma r_0 = \gamma \frac{\mu_0}{4\pi} \frac{e^2}{m_e} = 0.54 \times 10^{-12} \text{ cm}$$

It is sensitive to atomic dipole moment perp. to $\mathbf{q}$

$$\mathbf{S}_{\perp l} = \mathbf{S}_l - (\mathbf{S}_l \cdot \mathbf{q})\mathbf{q}$$

The magnetic scattering cross section

Spin density spread out ➡ scattering decreases at high κ

$$F(\mathbf{q}) = \int s(\mathbf{r}) \exp(i\mathbf{q}\cdot\mathbf{r}) \mathrm{d}\mathbf{r}$$

The magnetic neutron scattering cross section

$$\left.\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'}\right|_{\sigma\to\sigma'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{\lambda\lambda'} p_\lambda \left|\langle \mathbf{k}'\sigma'\lambda' | V_m | \mathbf{k}\sigma\lambda\rangle\right|^2 \delta\left(E_\lambda - E_{\lambda'} - \hbar\omega\right)$$

$$= \frac{k'}{k}(\gamma r_0)^2 \left|\frac{g}{2} F(\mathbf{q})\right|^2 e^{-2W(\vec{\kappa})} \int dt\, e^{-i\omega t} \sum_{ll'} e^{i\mathbf{q}\cdot(\mathbf{R}_l - \mathbf{R}_{l'})}$$

$$\times \left\langle \langle \sigma | \vec{\sigma}\cdot\mathbf{S}_{\perp l}(0) | \sigma' \rangle \langle \sigma' | \vec{\sigma}\cdot\mathbf{S}_{\perp l'}(t) | \sigma \rangle \right\rangle$$

For unspecified incident & final neutron spin states

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'} = \tfrac{1}{2}\sum_{\sigma\sigma'} \left.\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'}\right|_{\sigma\to\sigma'}$$

Un-polarized magnetic scattering

Squared form factor

DW factor

Polarization factor

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}(\gamma r_0)^2 \left|\frac{g}{2}F(\mathbf{q})\right|^2 e^{-2W(\mathbf{q})} \sum_{\alpha\beta}\left(\delta_{\alpha\beta} - \hat{q}_\alpha \hat{q}_\beta\right)$$

$$\times \int dt\, e^{-i\omega t} \sum_{ll'} e^{i\mathbf{q}\cdot(\mathbf{r}_l - \mathbf{r}_{l'})} \left\langle \mathbf{S}_l^\alpha(0)\mathbf{S}_{l'}^\beta(t)\right\rangle$$

Fourier transform

Spin correlation function

Magnetic neutron diffraction

Time independent spin correlations ➡ elastic scattering

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \left| \frac{g}{2} F(\mathbf{q}) \right|^2 e^{-2W(\mathbf{q})} \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \hat{q}_\alpha \hat{q}_\beta \right) \sum_{ll'} e^{i\mathbf{q}\cdot(\mathbf{r}_l - \mathbf{r}_{l'})} \left\langle \mathbf{S}_l^\alpha \right\rangle \left\langle \mathbf{S}_{l'}^\beta \right\rangle$$

Periodic magnetic structures ➡ Magnetic Bragg peaks

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 N_m \frac{(2\pi)^3}{v_m} \sum_{\vec{\tau}_m} \left(\left| \vec{\mathcal{F}}(\mathbf{q}) \right|^2 - \left| \hat{\mathbf{q}} \cdot \vec{\mathcal{F}}(\mathbf{q}) \right|^2 \right) \delta(\mathbf{q} - \vec{\tau}_m)$$

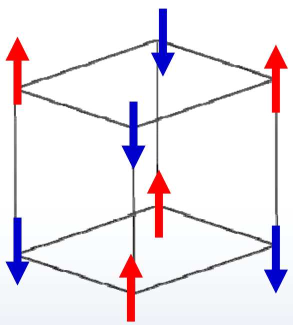
Magnetic primitive unit cell greater than chemical P.U.C.

➡ Magnetic Brillouin zone smaller than chemical B.Z.

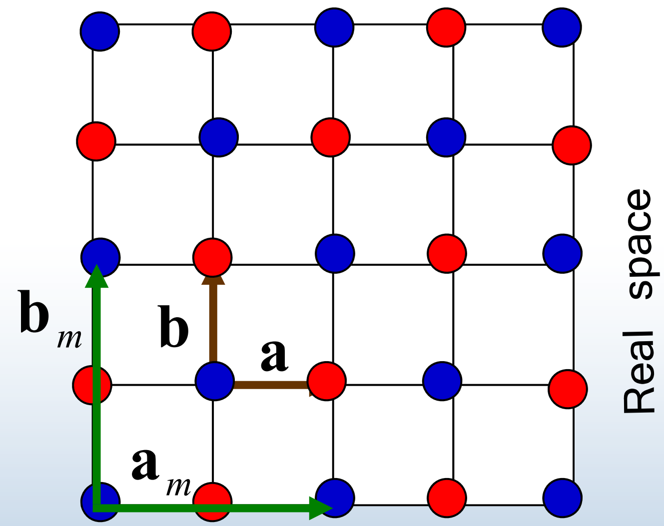
The magnetic vector structure factor is

$$\vec{\mathcal{F}}(\mathbf{q}) = \sum_{\mathrm{d}} \frac{g_{\mathbf{d}}}{2} F_{\mathbf{d}}(\mathbf{q}) e^{-2W_{\mathbf{d}}(\mathbf{q})} \left\langle \mathbf{S}_{\mathbf{d}} \right\rangle e^{i\mathbf{q}\cdot\mathbf{d}}$$

Simple cubic antiferromagnet



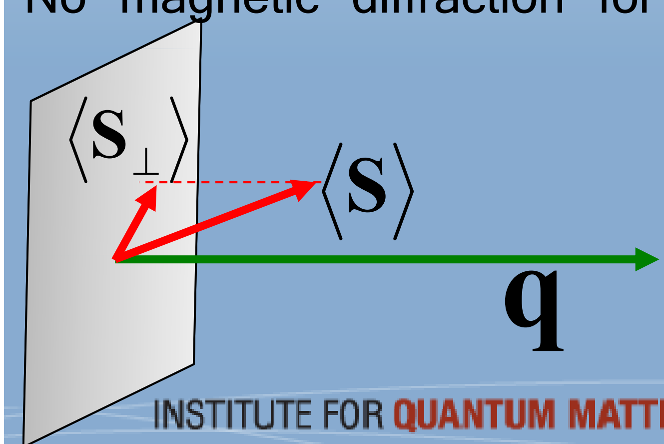
$$g\langle \mathbf{S} \rangle \equiv \frac{m_s}{\mu_B}\hat{\mathbf{z}}$$

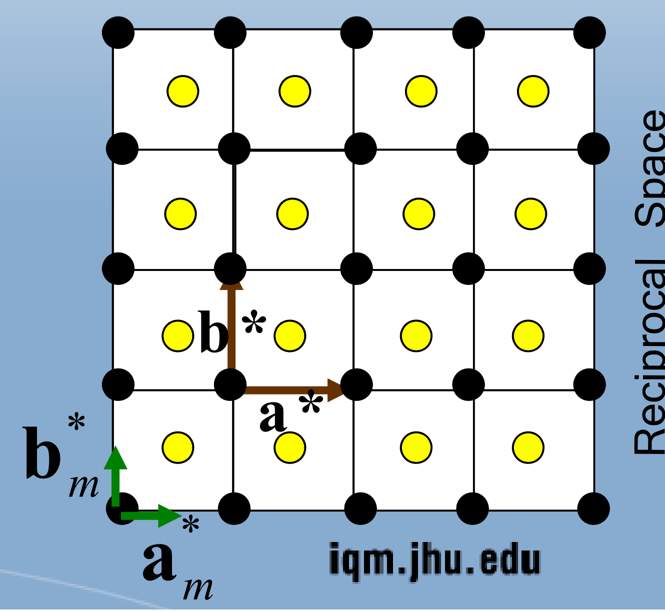


$$\vec{\mathcal{F}}(\vec{\kappa}) = \frac{m_s\hat{\mathbf{z}}}{2\mu_B}F(\mathbf{q})e^{-2W(\mathbf{q})}8\sin\pi h\sin\pi k\sin\pi l$$

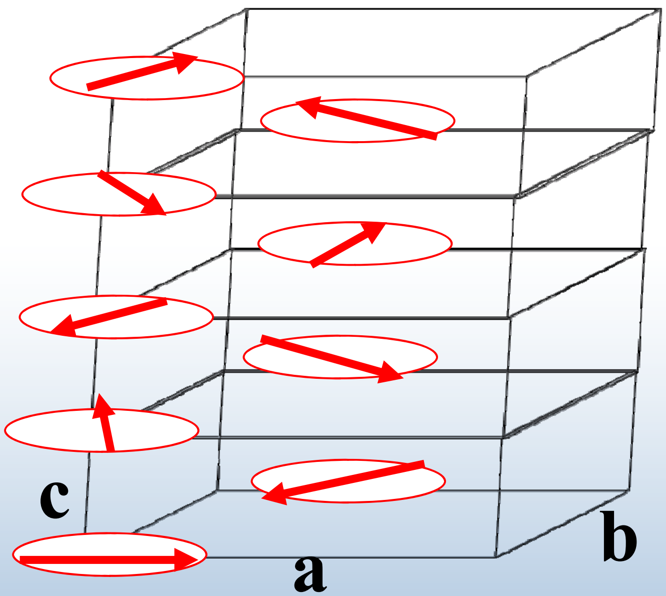
$$\frac{d\sigma}{d\Omega} = N\left(\gamma r_0\frac{m_s}{2\mu_B}\right)^2 e^{-2W(\mathbf{q})}\left|F(\mathbf{q})\right|^2\left(1-\hat{q}_z^2\right)\frac{(2\pi)^3}{v}\sum_{\vec{\tau}_m}\delta(\mathbf{q}-\vec{\tau}_m)$$

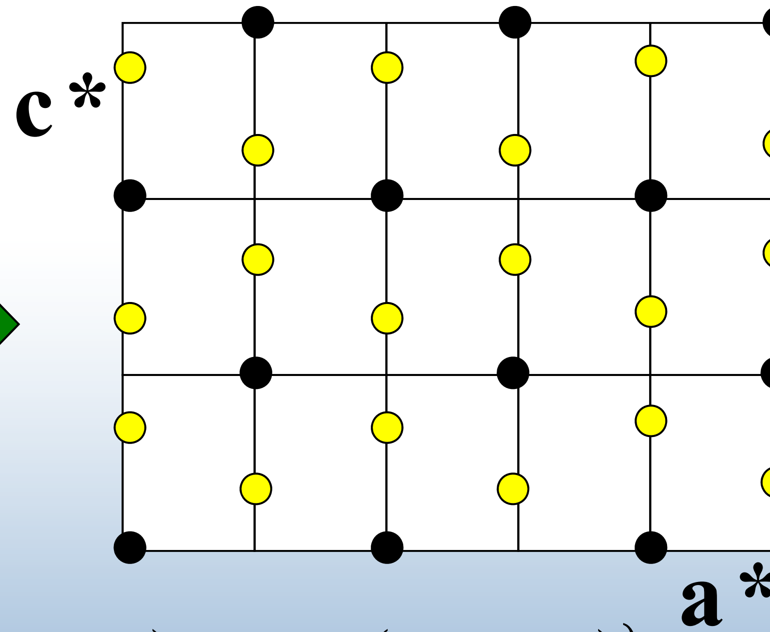
No magnetic diffraction for $\mathbf{q} \parallel \langle \mathbf{S} \rangle$





Not so simple Heli-magnet : MnO_2





$$\langle \mathbf{S}_l \rangle = S \exp(i\mathbf{w} \cdot \mathbf{R}_l)\{\hat{\mathbf{x}} \cos(\mathbf{Q}_m \cdot \mathbf{R}_l) + \hat{\mathbf{y}} \sin(\mathbf{Q}_m \cdot \mathbf{R}_l)\}$$

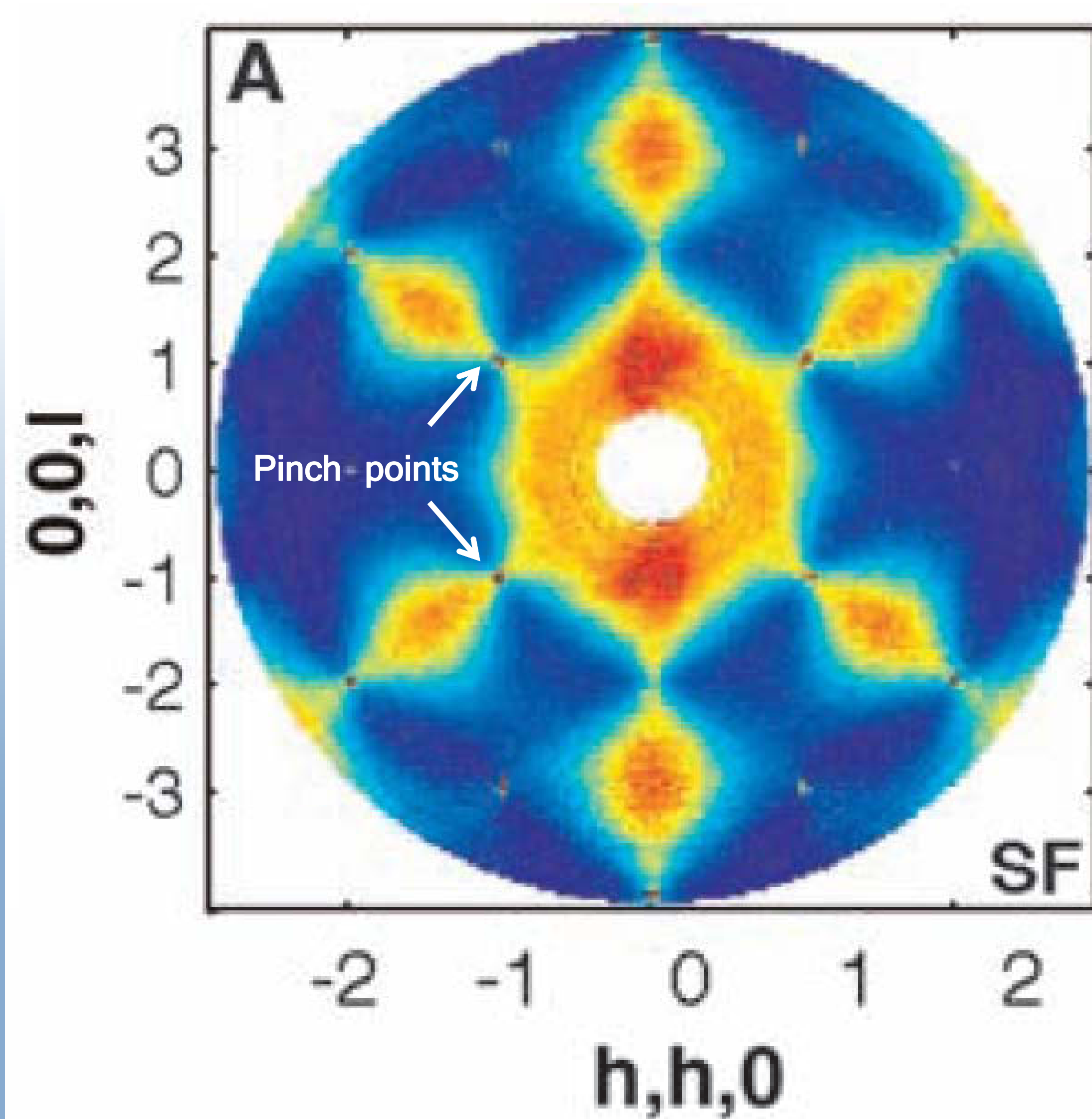
$\mathbf{w} = (111)$ and $\mathbf{Q}_m \approx (00\tfrac{2}{7})$ characterize structure

Insert into diffraction cross section to obtain

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = N(\gamma r_0 S)^2 e^{-2W(\mathbf{q})} \left|\frac{g}{2} F(\mathbf{q})\right|^2 (1+\hat{q}_z^2)\frac{(2\pi)^3}{v}$$

$$\times \sum_{\vec{\tau}} \{\delta(\mathbf{q} + \mathbf{w} - \mathbf{Q}_\mathbf{m} - \boldsymbol{\tau}) + \delta(\mathbf{q} + \mathbf{w} + \mathbf{Q}_\mathbf{m} - \boldsymbol{\tau})\}$$

Diffuse Elastic Magnetic Scattering



Magnetic Coulomb Phase in the Spin Ice $Ho_2Ti_2O_7$

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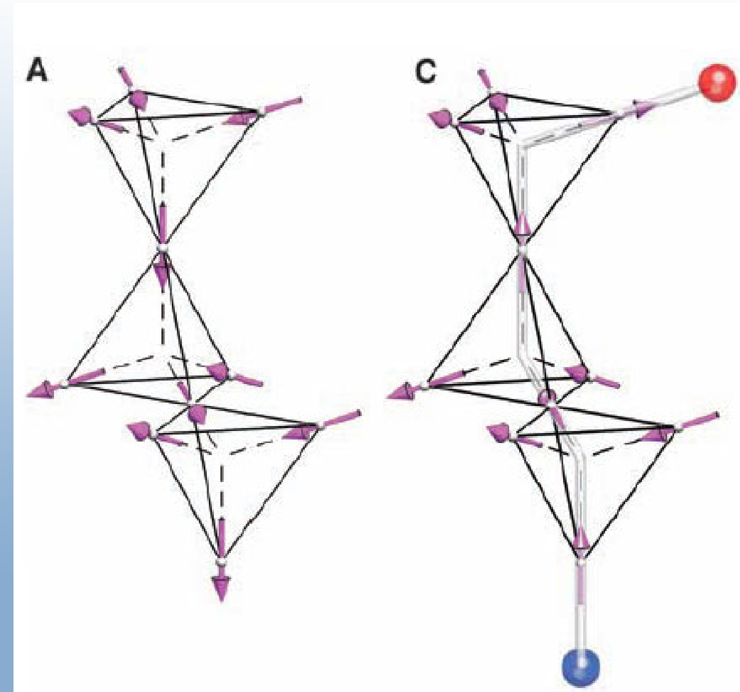

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Understanding Inelastic Magnetic Scattering:

Isolate the "interesting part" of the cross section

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k} N(\gamma r_0)^2 \left|\frac{g}{2}F(\mathbf{q})\right|^2 e^{-2W(\mathbf{q})} \sum_{\alpha\beta}\left(\delta_{\alpha\beta} - \hat{q}_\alpha \hat{q}_\beta\right) \mathcal{S}^{\alpha\beta}(\mathbf{q},\omega)$$

The "scattering law" is defined as

$$\mathcal{S}^{\alpha\beta}(\mathbf{q},\omega) = \int dt e^{-i\omega t} \tfrac{1}{N} \sum_{ll'} e^{i\mathbf{q}(\mathbf{r}_l - \mathbf{r}_{l'})} \left\langle S_l^\alpha(0) S_{l'}^\beta(t)\right\rangle$$

For systems in thermodynamic equilibrium $\mathcal{S}^{\alpha\beta}(\vec{\kappa},\omega)$ satisfies sum−rules

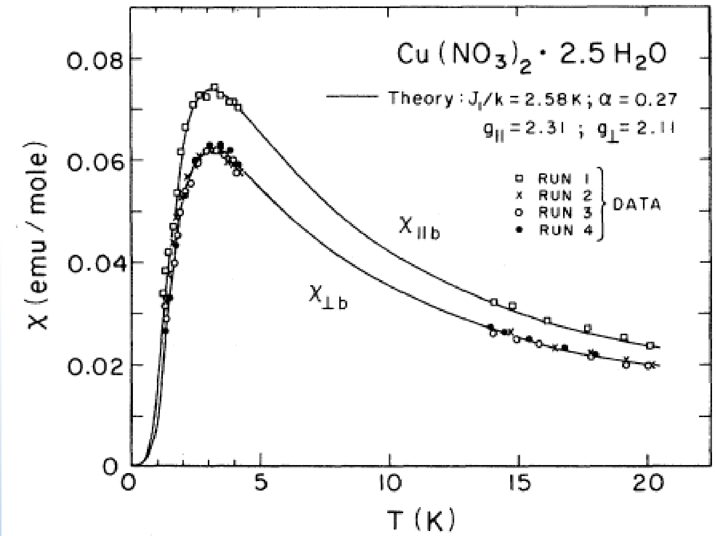
Detailed balance $\mathcal{S}(\mathbf{q},\omega) = \exp(\beta\hbar\omega)\mathcal{S}(-\mathbf{q},-\omega)$

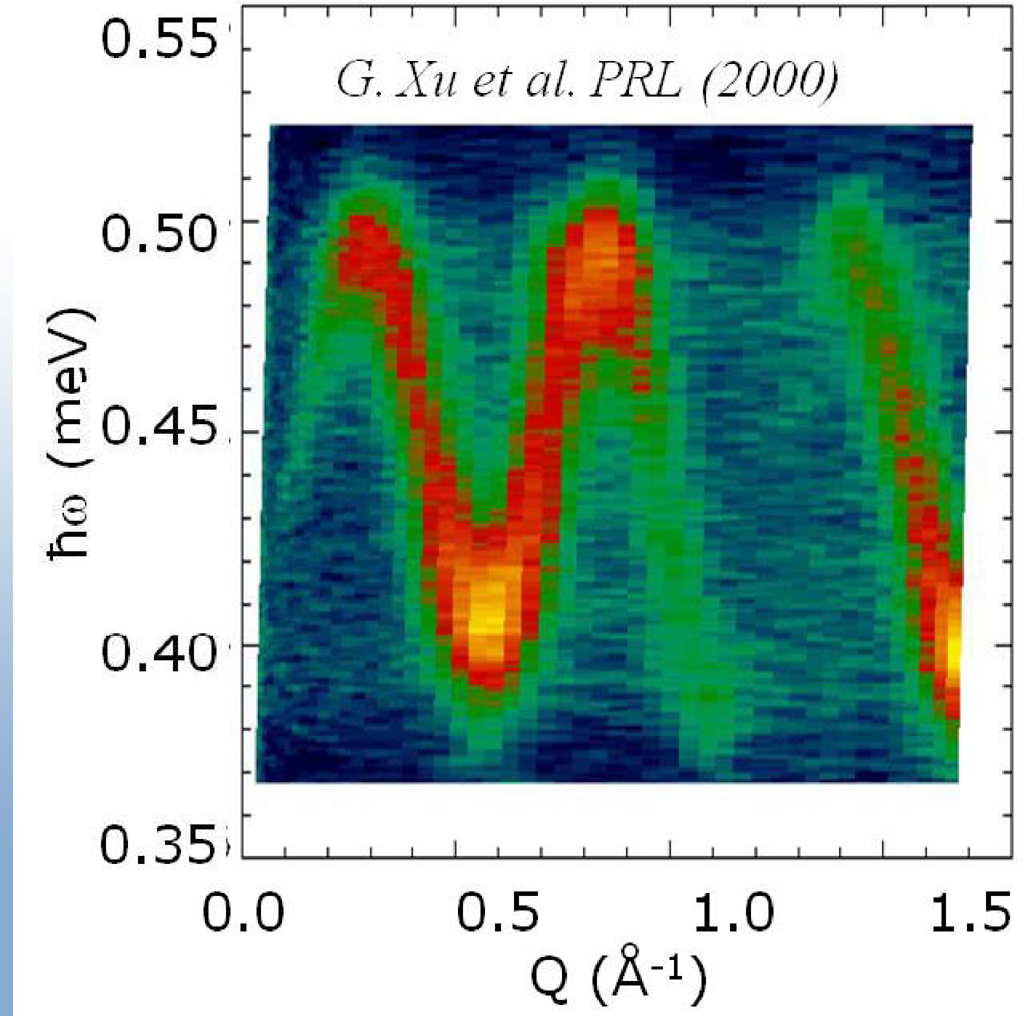
Total moment & 1st moment

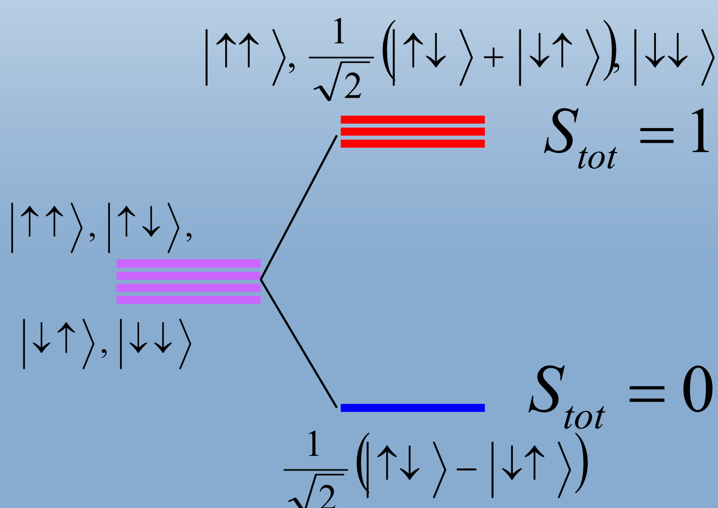
$$\hbar \frac{1}{\int d^3\mathbf{q}} \sum_\alpha \int d^3\mathbf{q} \int d\omega\, \mathcal{S}^{\alpha\alpha}(\mathbf{q},\omega) = S(S+1)$$

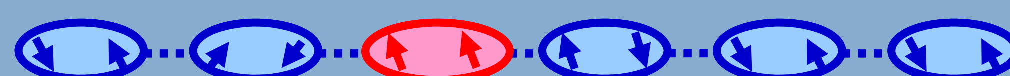
$$\hbar^2 \int \omega \; d\omega \, \mathcal{S}(\mathbf{q},\omega) = -\frac{1}{3}\frac{1}{N}\sum_{ll'} J_{ll'} \left\langle \mathrm{S}_l \cdot \mathrm{S}_{l'}\right\rangle \left(1 - \cos \mathbf{q}\cdot(\mathbf{r}_l - \mathbf{r}_{l'})\right)$$

Weakly Interacting spin-1/2 pairs in Cu-nitrate









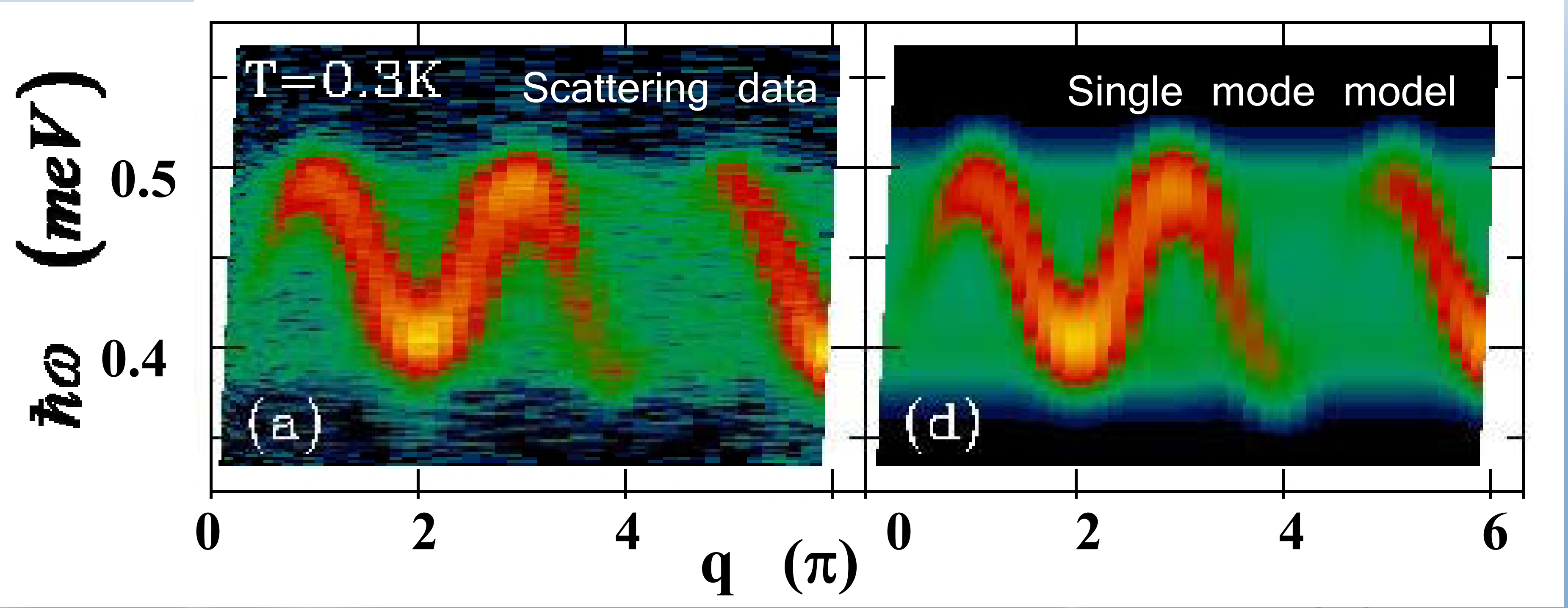
Sum rules and the single mode approximation

When a coherent mode dominates the spectrum:

$$\mathcal{S}(\mathbf{q},\omega) \approx \mathcal{S}(\mathbf{q})\delta\left(\hbar\omega - \varepsilon(\mathbf{q})\right)$$

Sum-rules link $\mathcal{S}(\mathbf{q})$ and $\varepsilon(\mathbf{q})$:

$$\mathcal{S}(\mathbf{q}) \approx \frac{\hbar^2 \int \omega \; d\omega \, \mathcal{S}(\mathbf{q},\omega)}{\varepsilon\,(\mathbf{q})} = -\frac{1}{3}\frac{\frac{1}{N}\sum_{ll'} J_{ll'} \langle \mathrm{S}_l \cdot \mathrm{S}_{l'} \rangle \left(1 - \cos \mathbf{q} \cdot (\mathrm{r}_l - \mathrm{r}_{l'})\right)}{\varepsilon\,(\mathbf{q})}$$

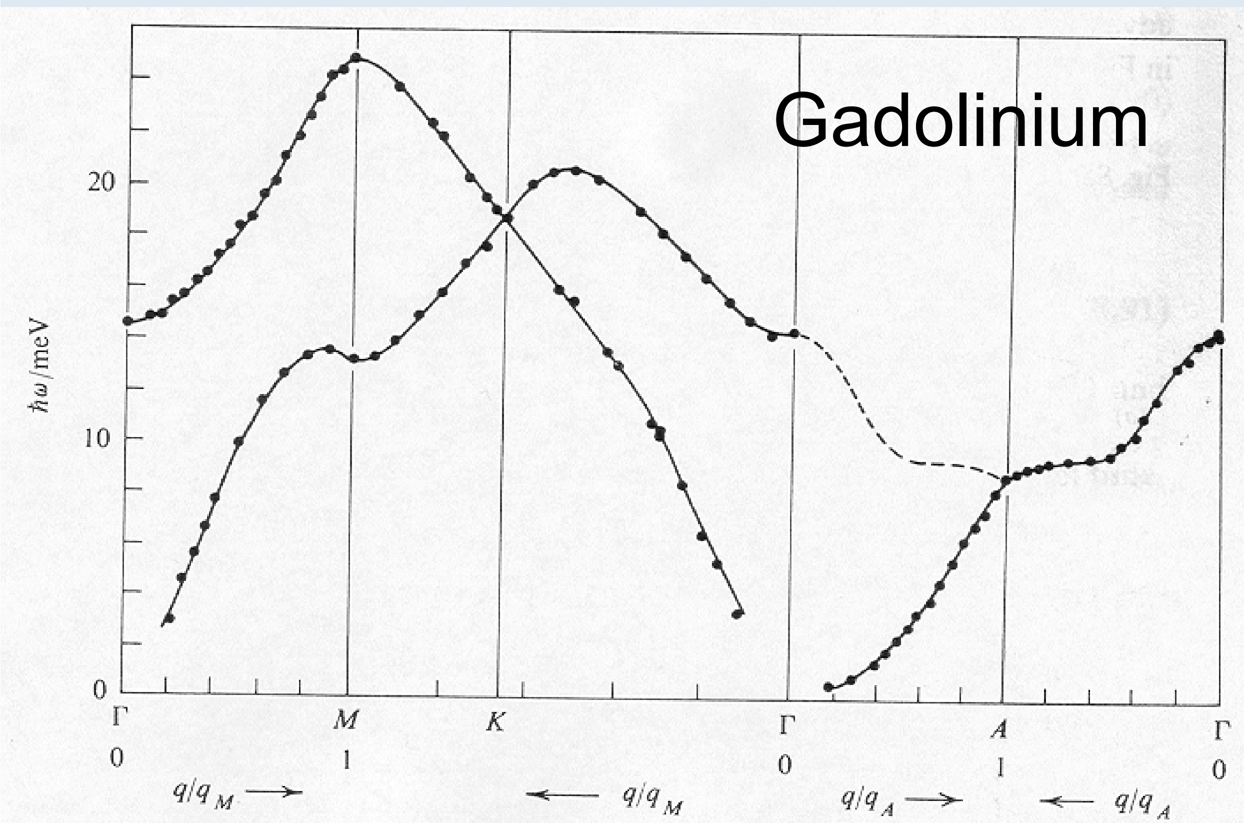


Spin waves in a ferromagnet

$$S^{\perp}(\vec{\kappa},\omega)=\frac{S}{2}\{\delta(\varepsilon(\vec{\kappa})-\hbar\omega)(n(\hbar\omega)+1)+\delta(\varepsilon(\vec{\kappa})+\hbar\omega)n(\hbar\omega)\}$$

Magnon creation

Magnon destruction



Dispersion relation

$$\varepsilon(\vec{\kappa})=2S\left(\mathrm{J}(0)-\mathrm{J}(\vec{\kappa})\right)$$

Magnon occupation number

$$n(\omega)=\frac{1}{\exp\left(\hbar\omega/k_BT\right)-1}$$

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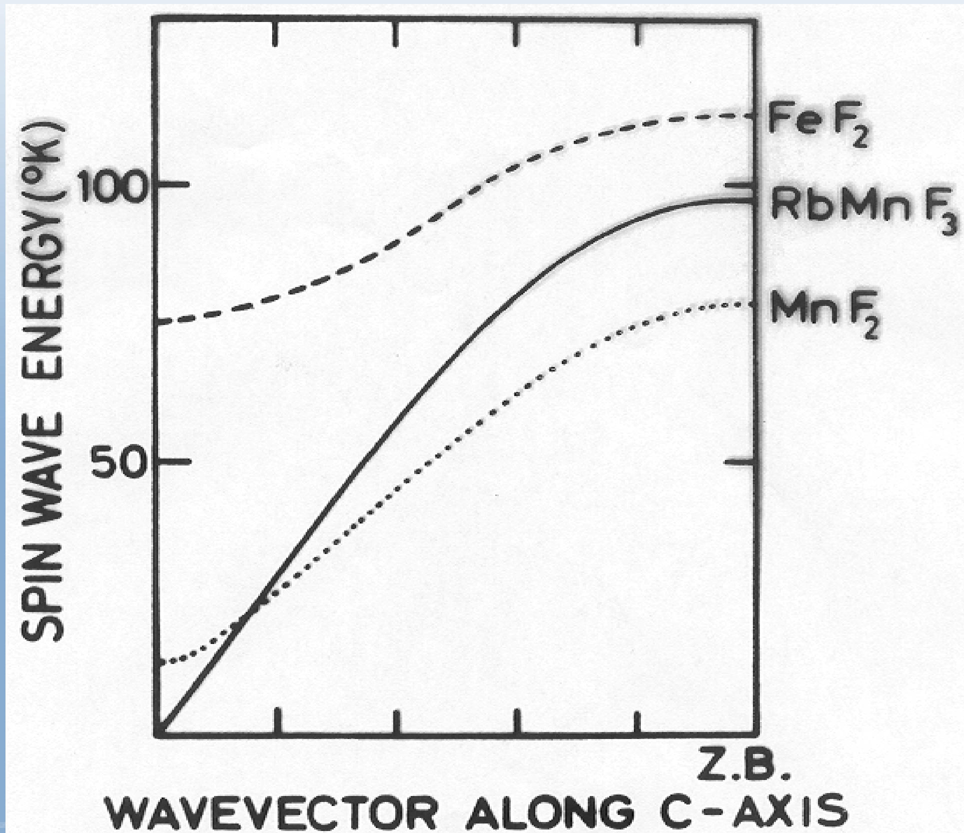
Spin waves in an antiferromagnet

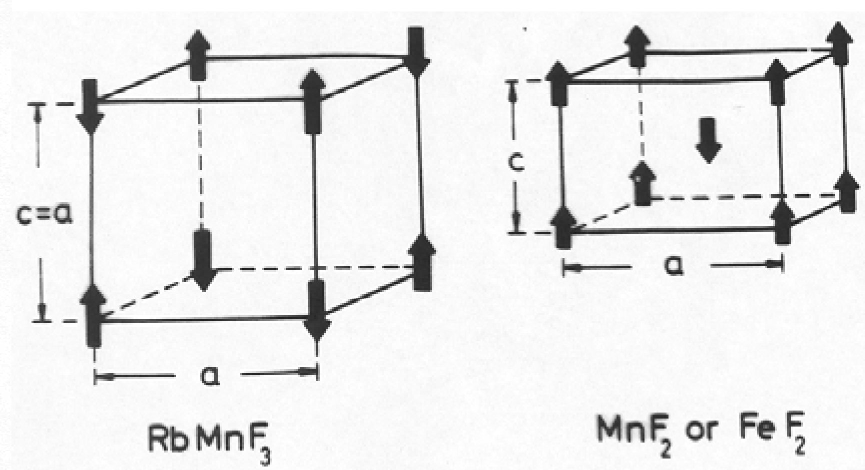
$$S^{\perp}(\vec{\kappa},\omega)=\frac{S}{2}\,\frac{J\left(1-\frac{1}{z}\sum_{\mathbf{d}}e^{i\vec{\kappa}\cdot\mathbf{d}}\right)}{\varepsilon(\vec{\kappa})}$$

$$\times\left\{\delta\left(\varepsilon(\vec{\kappa})-\hbar\omega\right)\left(n(\hbar\omega)+1\right)+\delta\left(\varepsilon(\vec{\kappa})+\hbar\omega\right)n(\hbar\omega)\right\}$$



Dispersion relation

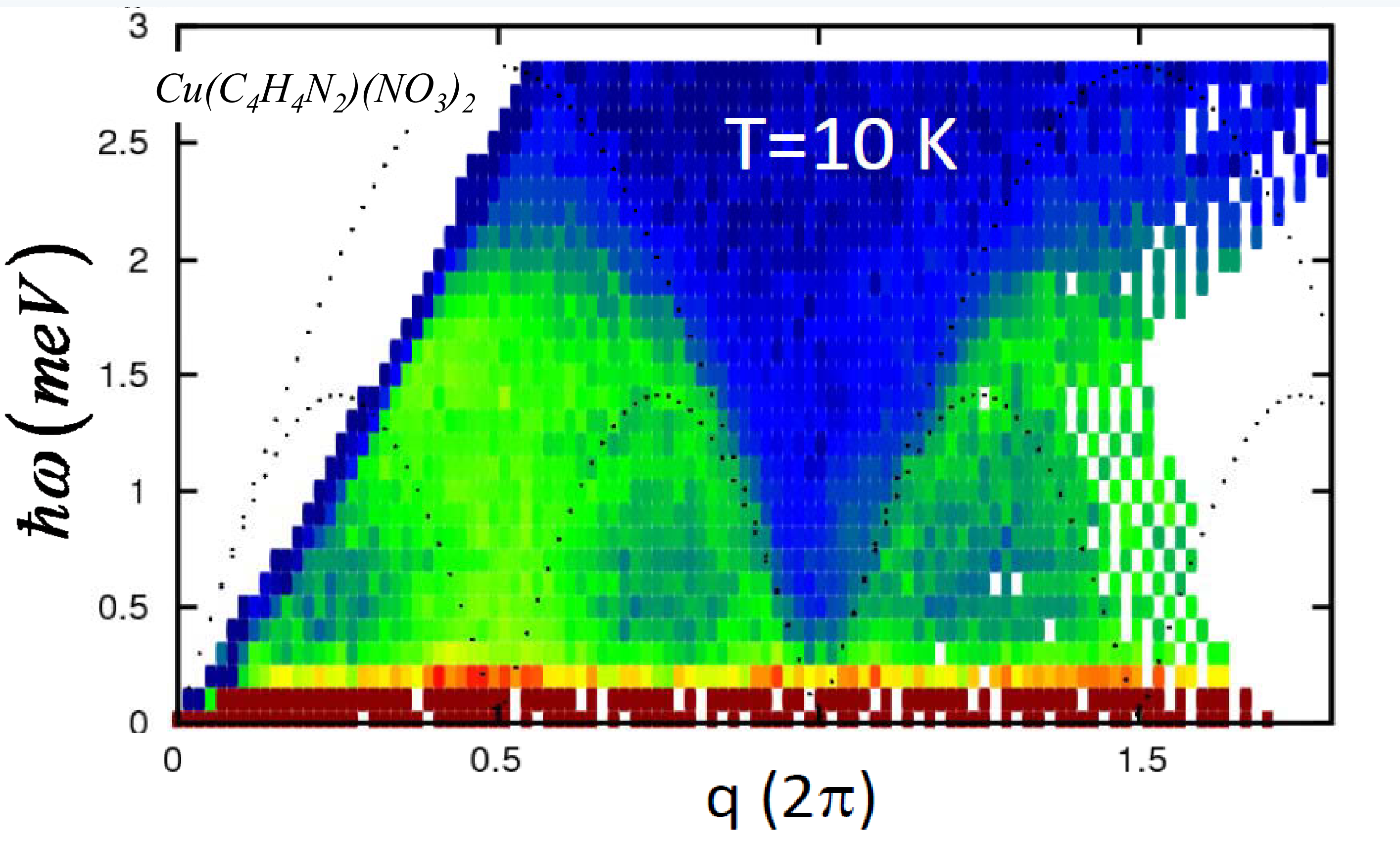
$$\varepsilon(\vec{\kappa})=2S\sqrt{J(0)^2-J(\vec{\kappa})^2}$$

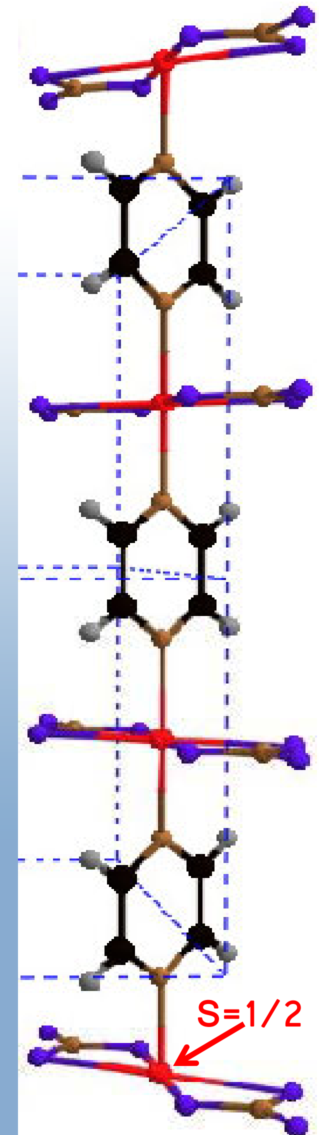


Continuum magnetic inelastic scattering

Inelastic scattering is not confined to disp. relations when

- ✓ a thermal ensemble of excitations is present
- ✓ $\mathbf{q}$ and $\hbar\omega$ do not uniquely specify excited state





$\mathcal{S}^{\alpha\beta}(\vec{\kappa},\omega)$ & the generalized susceptibility

$$\mathcal{S}^{\alpha\beta}(\mathbf{q},\omega)=\int dt\, e^{-i\omega t}\,\tfrac{1}{N}\sum_{ll'} e^{i\vec{\kappa}(\mathbf{r}_l-\mathbf{r}_{l'})}\left\langle S_l^{\alpha}(0)\,S_{l'}^{\beta}(t)\right\rangle$$

Compare to the generalized susceptibility

$$\chi^{\alpha\beta}(\mathbf{q},\omega)=\frac{(g\mu_B)^2}{N}\int dt\, e^{-i\omega t}\sum_{ll'} e^{i\mathbf{q}(\mathbf{r}_l-\mathbf{r}_{l'})}\left\langle\left[S_l^{\alpha}(t),S_{l'}^{\beta}(0)\right]\right\rangle$$

They are related by a **fluctuation–dissipation theorem**

$$\mathcal{S}^{\alpha\beta}(\mathbf{q},\omega)=\frac{\mathrm{Im}\left\{\chi^{\alpha\beta}(\mathbf{q}\omega)\right\}}{\pi(g\mu_B)^2}\frac{1}{1-e^{-\beta\hbar\omega}}$$

We convert inelastic scattering data to $\chi^{\alpha\beta}(\mathbf{q}\omega)$ to

- Compare with bulk susceptibility data
- Expose non–trivial temperature dependence
- Compare with theories

Polarized magnetic neutron scattering

Specify the incident and final neutron spin state

$$\left\langle \left\langle \sigma \middle| \vec{\sigma} \cdot \mathbf{S}_{\perp l}(0) \middle| \sigma' \right\rangle \left\langle \sigma' \middle| \vec{\sigma} \cdot \mathbf{S}_{\perp l'}(t) \middle| \sigma \right\rangle \right\rangle =$$

$$\begin{cases} + \rightarrow + & \left\langle S^{z}_{\perp l}(0) S^{z}_{\perp l}(t) \right\rangle \\ - \rightarrow - & \left\langle S^{z}_{\perp l}(0) S^{z}_{\perp l}(t) \right\rangle \\ + \rightarrow - & \left\langle S^{-}_{\perp l}(0) S^{+}_{\perp l}(t) \right\rangle \\ - \rightarrow - & \left\langle S^{+}_{\perp l}(0) S^{-}_{\perp l}(t) \right\rangle \end{cases}$$

Non spin flip:

$\mathbf{S} \parallel \mathbf{H} \quad \mathbf{S} \perp \mathbf{q}$

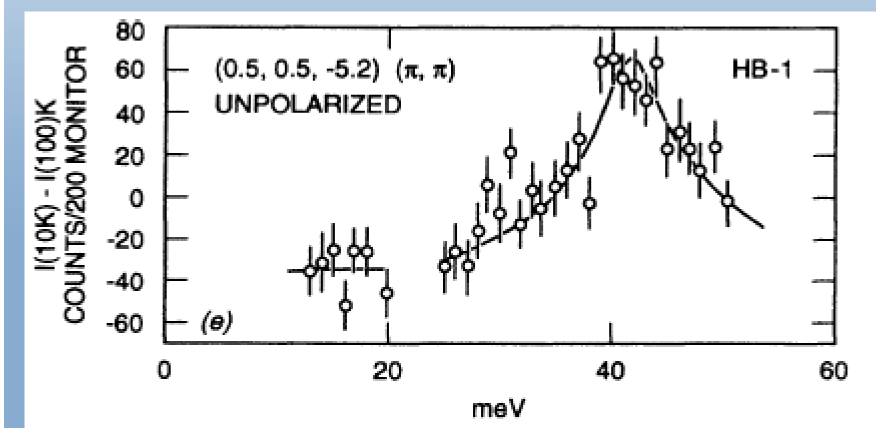
Spin flip:

$\mathbf{S} \perp \mathbf{H} \quad \mathbf{S} \perp \mathbf{q}$

Polarized neutron scattering

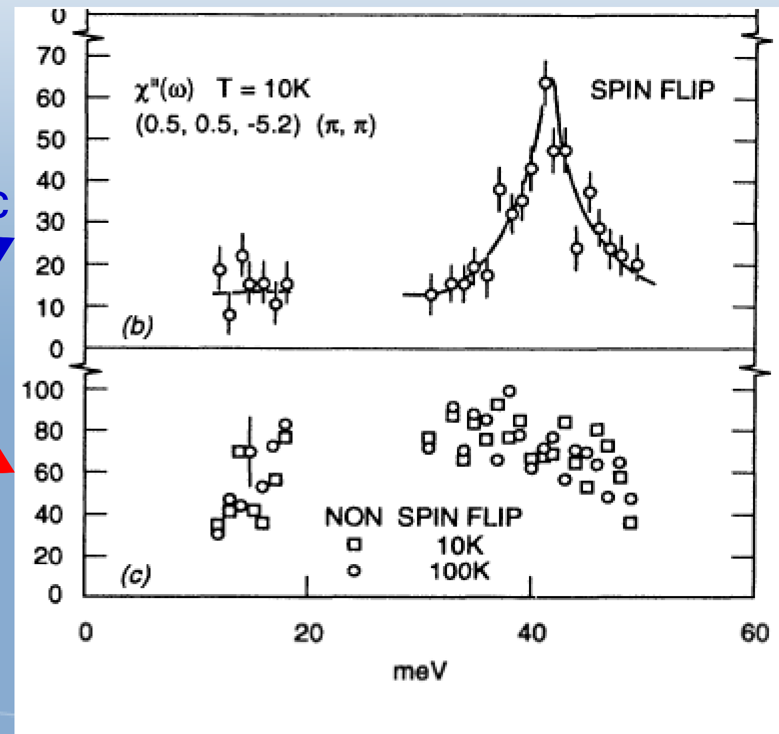
Type of scattering	H//κ		H perp κ	
	SF	NSF	SF	NSF
Nuclear coherent	0	1	0	1
Nuclear isotope incoherent	0	1	0	1
Nuclear spin incoherent	2/3	1/3	2/3	1/3
Magnetic	$S^{xx}+S^{yy}$	0	S^{xx}	S^{yy}

$YBa_2Cu_3O_7$ by Mook *et al.* PRL (1993)



magnetic

Nuclear



Summary

- The neutron has a small dipole moment causing scattering from electrons
- Magnetic scattering is similar in magnitude to nuclear scattering
- Elastic magnetic scattering probes static magnetic structure
- Inelastic magnetic scattering probes dynamics correlations through $\mathcal{S}^{\alpha\beta}(\mathbf{q},\omega)$
- Spin resolved scattering distinguishes magnetic and nuclear processes and spin components