# Neutron Scattering, correlation functions & linear response functions

Collin Broholm
Johns Hopkins University

# Scattering cross section & $\mathcal{S}(\kappa\omega)$

$$\mathcal{H}' = \mathcal{H} + \frac{\hat{p}^2}{2m} + \hat{V}$$

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{\lambda\lambda'} p_\lambda \left|\langle k'\lambda'|V|k\lambda\rangle\right|^2 \delta\left(E_\lambda - E_{\lambda'} + \hbar\omega\right)$$

$$Q_{-\boldsymbol{\kappa}} = \frac{1}{\sqrt{N}}\frac{1}{b}\frac{m}{2\pi\hbar^2}\int d^3\mathbf{r}\, V(\mathbf{r}) e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}$$

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k} N b^2 \mathcal{S}(\kappa\omega)$$

$$\mathcal{S}(\kappa\omega) = \sum_{\lambda\lambda'} p_\lambda \left|\langle \lambda'|Q_{-\boldsymbol{\kappa}}|\lambda\rangle\right|^2 \delta\left(E_\lambda - E_{\lambda'} + \hbar\omega\right)$$

# Detailed Balance

$$p_\lambda = \frac{e^{-\beta E_\lambda}}{Z}$$

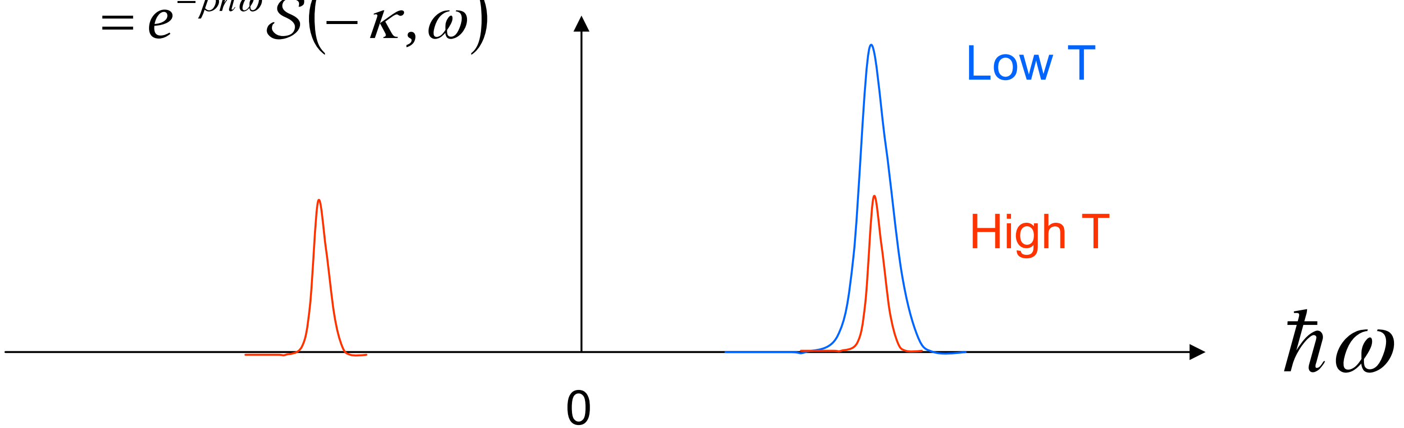
*Sample in thermal equilibrium*

$$\mathcal{S}(\kappa,-\omega) = Z^{-1}\sum_{\lambda\lambda'} e^{-\beta E_\lambda} \left|\langle\lambda'|Q_{-\boldsymbol{\kappa}}|\lambda\rangle\right|^2 \delta(E_\lambda - E_{\lambda'} - \hbar\omega)$$

$$= Z^{-1}\sum_{\lambda\lambda'} e^{-\beta E_\lambda} \left|\langle\lambda'|Q_{-\boldsymbol{\kappa}}|\lambda\rangle\right|^2 \delta(E_{\lambda'} - E_\lambda + \hbar\omega)$$

$$= Z^{-1}\sum_{\lambda\lambda'} e^{-\beta E_{\lambda'}} e^{-\beta\hbar\omega} \left|\langle\lambda'|Q_{-\boldsymbol{\kappa}}|\lambda\rangle\right|^2 \delta(E_{\lambda'} - E_\lambda + \hbar\omega)$$

$$= e^{-\beta\hbar\omega}\mathcal{S}(-\kappa,\omega)$$



# $\mathcal{S}(\kappa\omega)$ & correlation functions

$$\delta(\hbar\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} e^{-i\omega t}\,dt$$

$$\mathcal{S}(\kappa\omega) = \frac{1}{2\pi\hbar}\sum_{\lambda\lambda'} p_\lambda \left|\langle\lambda'|Q_{-\mathbf{\kappa}}|\lambda\rangle\right|^2 \int \exp\left(-i\left((E_\lambda - E_{\lambda'})\hbar^{-1} + \omega\right)t\right)$$

$$= \frac{1}{2\pi\hbar}\int dt e^{-i\omega t}\sum_{\lambda\lambda'} p_\lambda \langle\lambda|Q_{\mathbf{\kappa}}|\lambda'\rangle\langle\lambda'|e^{i\mathcal{H}t/\hbar}Q_{-\mathbf{\kappa}}e^{-i\mathcal{H}t/\hbar}|\lambda\rangle$$

$$= \frac{1}{2\pi\hbar}\int dt e^{-i\omega t}\langle Q_{\mathbf{\kappa}}Q_{-\mathbf{\kappa}}(t)\rangle$$

We measure a time and space fourier transform of the interaction potential

$$I(\mathbf{\kappa},t) = \langle Q_{\mathbf{\kappa}}Q_{-\mathbf{\kappa}}(t)\rangle$$

$$I(\mathbf{\kappa},\infty) = \lim_{\varepsilon\to 0^+}\int_{-\varepsilon}^{\varepsilon}\mathcal{S}(\kappa\omega)\hbar d\omega$$

No elastic scattering for a liquid!

$$I(\mathbf{\kappa},0) = \int_{-\infty}^{\infty}\mathcal{S}(\kappa\omega)\hbar d\omega$$

Equal time correlation function

# Fluctuation Dissipation Theorem

$$\Phi_{\boldsymbol{\kappa}}(t) = \frac{i}{\hbar}\left\langle \left[ Q_{\boldsymbol{\kappa}}(t), Q_{-\boldsymbol{\kappa}} \right] \right\rangle$$ Is linear response function

$$\frac{1}{2\pi i}\int e^{i\omega t}\Phi_{\boldsymbol{\kappa}}(t)dt = \frac{1}{2\pi\hbar}\int dt\left(\left\langle Q_{\boldsymbol{\kappa}}(t)Q_{-\boldsymbol{\kappa}}\right\rangle - \left\langle Q_{-\boldsymbol{\kappa}}Q_{\boldsymbol{\kappa}}(t)\right\rangle\right)e^{i\omega t}$$

$$\left\langle Q_{\boldsymbol{\kappa}}(t)Q_{-\boldsymbol{\kappa}}\right\rangle = \left\langle Q_{\boldsymbol{\kappa}}Q_{-\boldsymbol{\kappa}}(-t)\right\rangle$$ Time translational invariance

$$\left\langle Q_{-\boldsymbol{\kappa}}Q_{-\boldsymbol{\kappa}}(t)\right\rangle = \left\langle Q_{\boldsymbol{\kappa}}Q_{-\boldsymbol{\kappa}}(-t + i\beta\hbar)\right\rangle$$ Trace is invariant under cyclic permutation

$$\frac{1}{2\pi i}\int e^{i\omega t}\Phi_{\boldsymbol{\kappa}}(t)dt = \mathcal{S}(\kappa\omega)\left(1 - e^{-\beta\hbar\omega}\right)$$ Fluctuation dissipation theorem v1

$$\chi(\boldsymbol{\kappa}\omega) = -\lim_{\varepsilon\to 0^+}\int_0^\infty dt e^{-i(\omega - i\varepsilon)t}\Phi_{\boldsymbol{\kappa}}(t)$$ Generalized susceptibility

$$\mathcal{S}(\kappa\omega) = \frac{1}{1 - e^{-\beta\hbar\omega}}\frac{\chi''(\boldsymbol{\kappa}\omega)}{\pi}$$ Fluctuation dissipation theorem v2

# First Moment Sum-rule

$$\Phi_{\mathbf{\kappa}}(t) = i\int d\omega e^{-i\omega t}\mathcal{S}(\kappa\omega)\left(1-e^{-\beta\hbar\omega}\right)$$

Fourier transform

$$\partial_t \Phi_{\mathbf{\kappa}}(t)\Big|_{t=0} = \int \omega\, d\omega\, \mathcal{S}(\kappa\omega)\left(1-e^{-\beta\hbar\omega}\right)$$

Time derivative evaluated at *t*=0

$$= 2\int \omega\, d\omega\, \mathcal{S}(\kappa\omega)$$

Use detailed balance

$$\partial_t Q_{\mathbf{\kappa}}(t) = \frac{1}{i\hbar}\left[Q_{\mathbf{\kappa}}(t), \mathcal{H}\right]$$

Use this result from interaction picture to Derive $\partial_t \Phi_{\mathbf{\kappa}}(t)\Big|_{t=0}$

$$\int \hbar\omega\, \hbar d\omega\, \mathcal{S}(\kappa\omega) = -\tfrac{1}{2}\left\langle\left[Q_{\mathbf{\kappa}},\left[Q_{-\mathbf{\kappa}},\mathcal{H}\right]\right]\right\rangle$$

First moment sum-rule

To learn it’s significance look at interaction potentials: Nuclear then magnetic

# Neutron-Nuclear interaction

$$V(\mathbf{r}) = \sum_n b \frac{2\pi\hbar^2}{m} \delta\left(\mathbf{r} - \hat{\mathbf{R}}_n\right)$$

Fermi Pseudo potential

$$\hat{Q}_{\boldsymbol{\kappa}} = \frac{1}{\sqrt{N}} \sum_n e^{i\boldsymbol{\kappa}\cdot\hat{\mathbf{R}}_n}$$

Hats indicate sample space operator

$$\mathcal{S}(\kappa\omega) = \frac{1}{2\pi\hbar} \frac{1}{N} \int dt e^{-i\omega t} \sum_{nn'} \left\langle e^{i\boldsymbol{\kappa}\cdot\mathbf{R}_n} e^{-i\boldsymbol{\kappa}\cdot\mathbf{R}_{n'}(t)} \right\rangle$$

FT density-density correlations

For free atoms $\mathcal{H} = \sum_n \frac{\hat{P}_n^{\,2}}{2M}$ can readily compute double commutator

$$\tfrac{1}{2}\left\langle \left[Q_{\boldsymbol{\kappa}}, \left[Q_{-\boldsymbol{\kappa}}, \mathcal{H}\right]\right] \right\rangle = -\frac{(\hbar\kappa)^2}{2M} \quad \Longrightarrow \quad \int \hbar\omega \hbar d\omega \, \mathcal{S}(\kappa\omega) = \frac{(\hbar\kappa)^2}{2M}$$

Recoil!

$$\mathcal{S}(\kappa\omega) = \int \delta\left(\hbar\omega - \frac{(\hbar\kappa)^2}{2M} - \frac{\hbar\boldsymbol{\kappa}\cdot\mathbf{p}}{M}\right) f(\mathbf{p}) d^3\mathbf{p}$$

Impulse approximation holds generally.
Use to measure momentum distribution

# Neutron-electron spin interactions

$$\boldsymbol{\mu}_n = -\gamma \frac{e}{m}\frac{\hbar}{2}\boldsymbol{\sigma}$$

Neutron is spin-1/2 magnetic dipole $\gamma=1.913$

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

Dipole in inhomogeneous field from sample

$$B(\mathbf{r}) = -\sum_n \nabla \times \left( \frac{\mu_0}{4\pi} \frac{g\mu_B \mathbf{s}_n \times \hat{\mathbf{r}}}{r^2} \right)$$

B-Field from partially filled electron shells

$$V(\mathbf{r}) = -\gamma \frac{e}{m}\frac{\hbar}{2}\boldsymbol{\sigma} \cdot \sum_n \nabla \times \left( \frac{\mu_0}{4\pi} \frac{g\mu_B \mathbf{s}_n \times \hat{\mathbf{r}}}{r^2} \right)$$

Interaction potential

$$Q_{\boldsymbol{\kappa}} = -\boldsymbol{\sigma} \cdot \sum_n \hat{\boldsymbol{\kappa}} \times \mathbf{s}_n \times \hat{\boldsymbol{\kappa}} \exp(i\boldsymbol{\kappa} \cdot \mathbf{r}_n) \frac{1}{\sqrt{N}}$$

Dimensionless scattering operator

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

$$r_0 = \frac{1}{4\pi\varepsilon_0}\frac{e^2}{mc^2} = 2.818\ \mathrm{fm} \quad ; \qquad F(\boldsymbol{\kappa})$$

Is magnetic form factor: Fourier Transf. of spin density distribution on atom

# Magnetic correlation & response function

$$\mathcal{S}^{\alpha\beta}(\kappa\omega)=\frac{1}{2\pi\hbar}\int dt e^{-i\omega t}\left\langle s_{\boldsymbol{\kappa}}^{\alpha}s_{-\boldsymbol{\kappa}}^{\beta}(t)\right\rangle$$

time and spatial FT of 2-point correlations

$$\mathbf{s}_{\boldsymbol{\kappa}}=\frac{1}{\sqrt{N}}\sum_{n}\exp(i\boldsymbol{\kappa}\cdot\mathbf{R}_n)\mathbf{s}_n$$

$$\Phi_{\boldsymbol{\kappa}}^{\alpha\beta}(t)=\frac{i}{\hbar}\left\langle\left[s_{\boldsymbol{\kappa}}^{\alpha}(t),s_{-\boldsymbol{\kappa}}^{\beta}\right]\right\rangle$$

Linear response function

$$\chi_{\alpha\beta}(\boldsymbol{\kappa}\omega)=-(g\mu_B)^2\int_0^{\infty}dt\, e^{-i\omega t}\Phi_{\boldsymbol{\kappa}}^{\alpha\beta}(t)$$

Generalized susceptibility

$$\mathcal{S}^{\alpha\beta}(\kappa\omega)=\frac{1}{1-e^{-\beta\hbar\omega}}\frac{\chi''_{\alpha\beta}(\boldsymbol{\kappa}\omega)}{(g\mu_B)^2\pi}$$

Fluctuation dissipation theorem

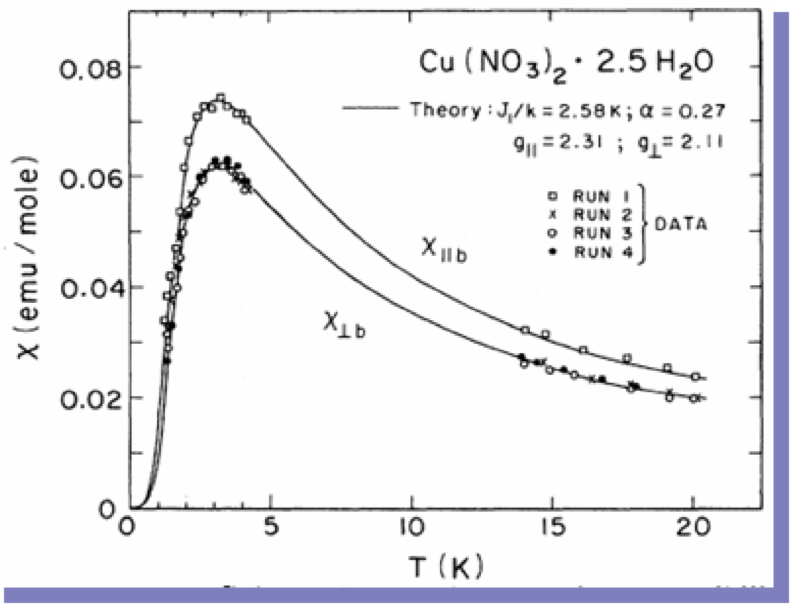
$$\hbar^2\int\omega\; d\omega\,\mathcal{S}(\vec{\kappa},\omega)=-\frac{1}{3}\frac{1}{N}\sum_{nn'}J_{nn'}<\mathbf{s}_n\cdot\mathbf{s}_{n'}>\left(1-\cos\boldsymbol{\kappa}\cdot(\mathbf{R}_n-\mathbf{R}_{n'})\right)$$

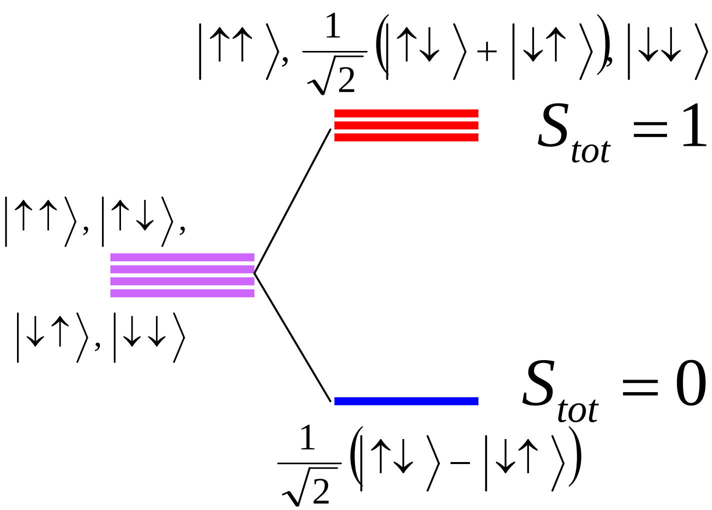
1st moment

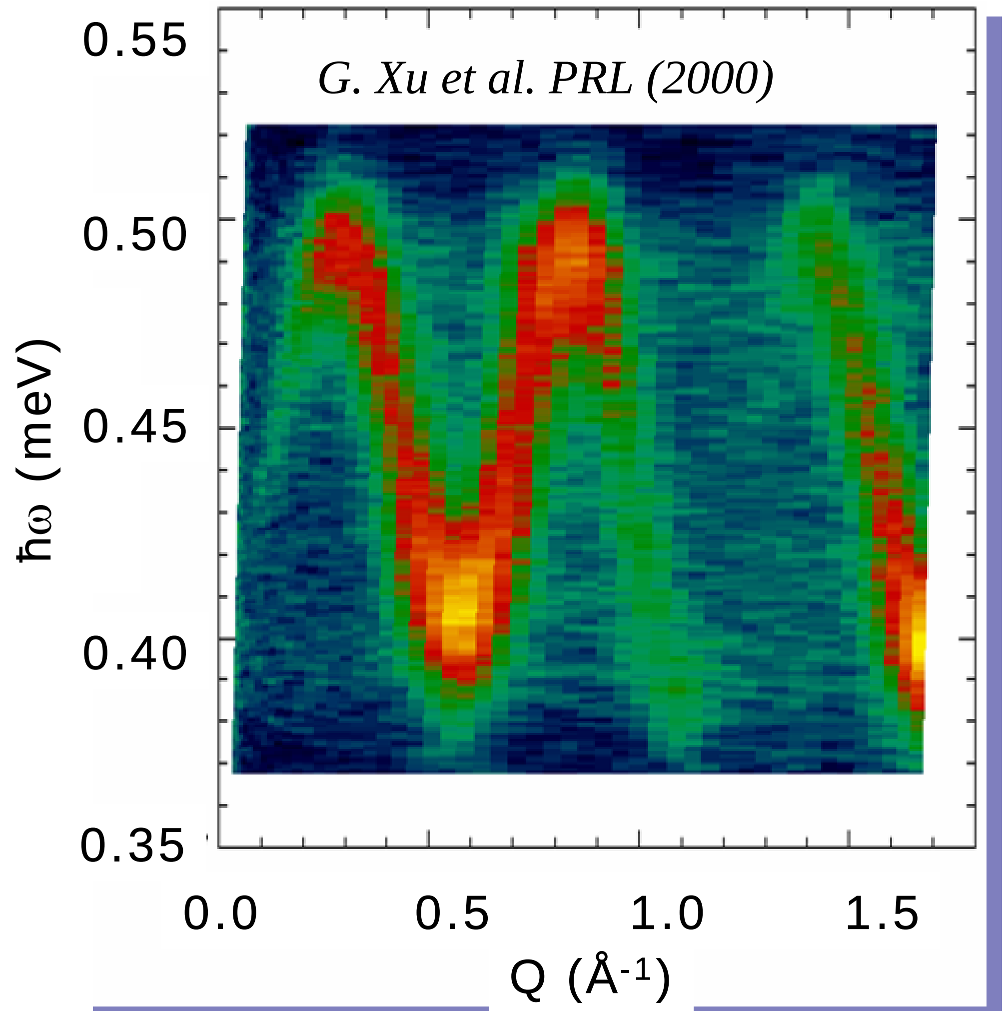
$$\frac{1}{\int d^3\boldsymbol{\kappa}}\sum_{\alpha}\int d^3\boldsymbol{\kappa}\int\hbar\mathrm{d}\omega\,\mathcal{S}^{\alpha\alpha}(\boldsymbol{\kappa}\omega)=s(s+1)$$

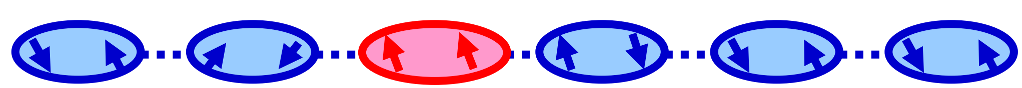
0st moment

# Weakly interacting dimers in $Cu(NO_3)_2 \cdot 2.5\ H_2O$



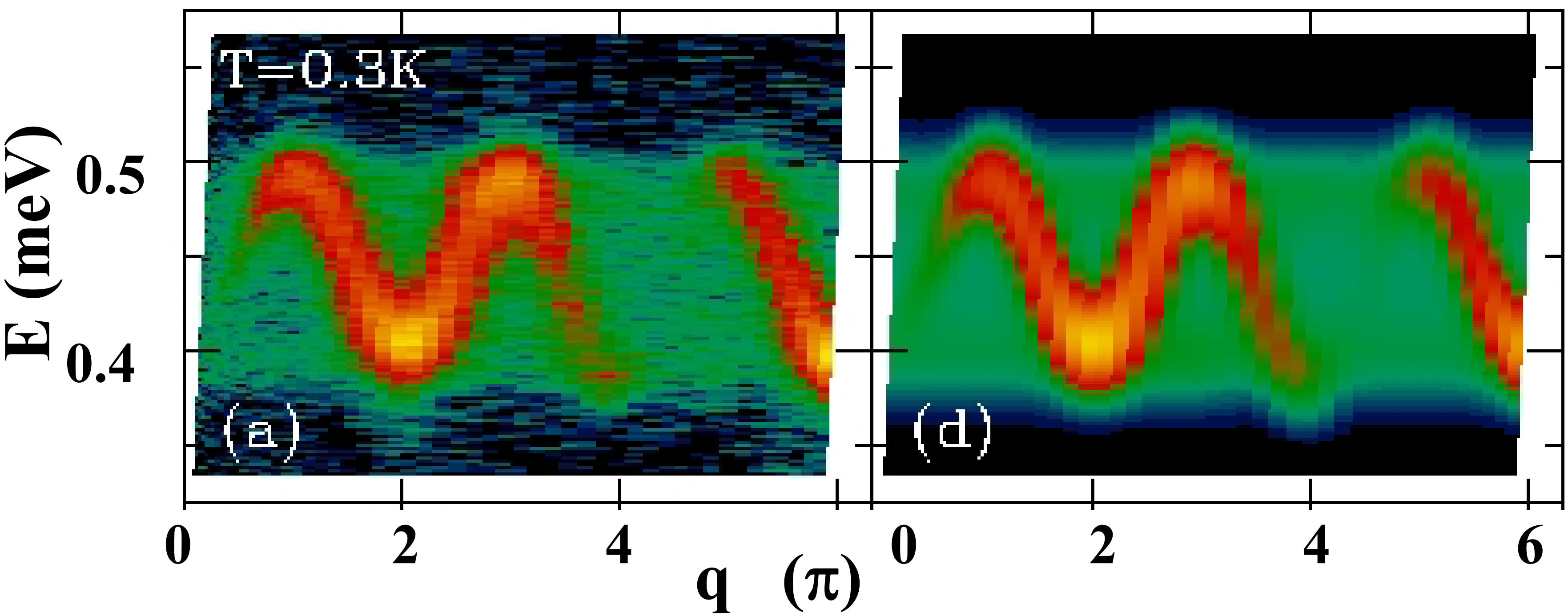
# "Guessing" S(qω)

When a coherent mode dominates the spectrum:

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

First moment Sum-rule links S(q) and $\varepsilon$(q)

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

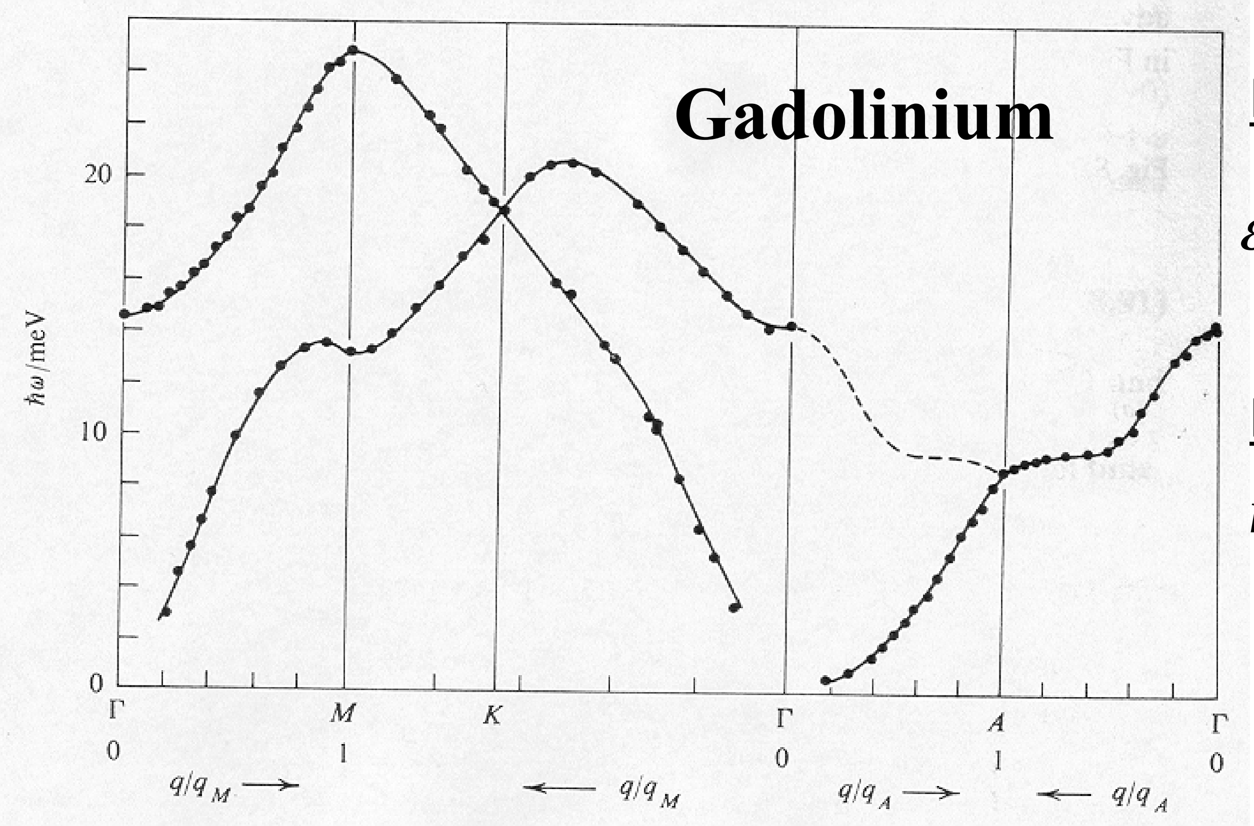


# Spin waves in ferromagnet

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

Magnon creation

Magnon destruction




Dispersion relation

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

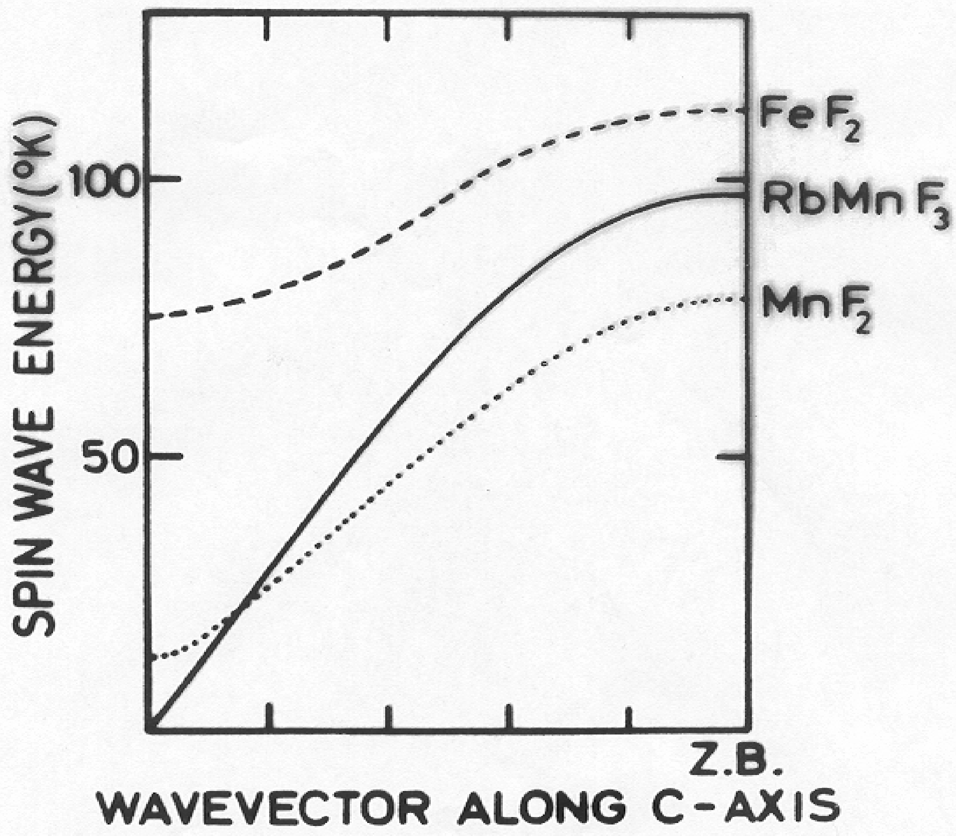
Magnon occupation prob.

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

# Spin waves in antiferromagnet

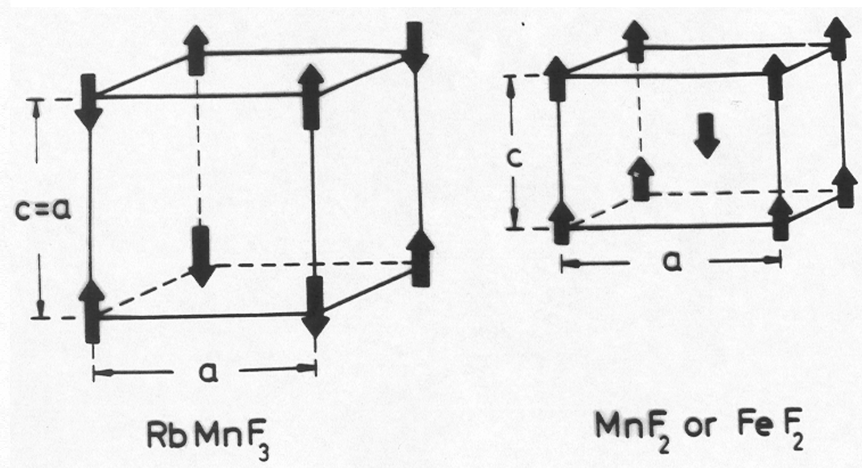
$$\mathrm{S}^{\perp}(\vec{\kappa},\omega)=\frac{S}{2}\frac{J\left(1-\frac{1}{z}\sum_{\mathbf{d}}e^{i\vec{\kappa}\cdot\mathbf{d}}\right)}{\varepsilon(\vec{\kappa})}$$

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

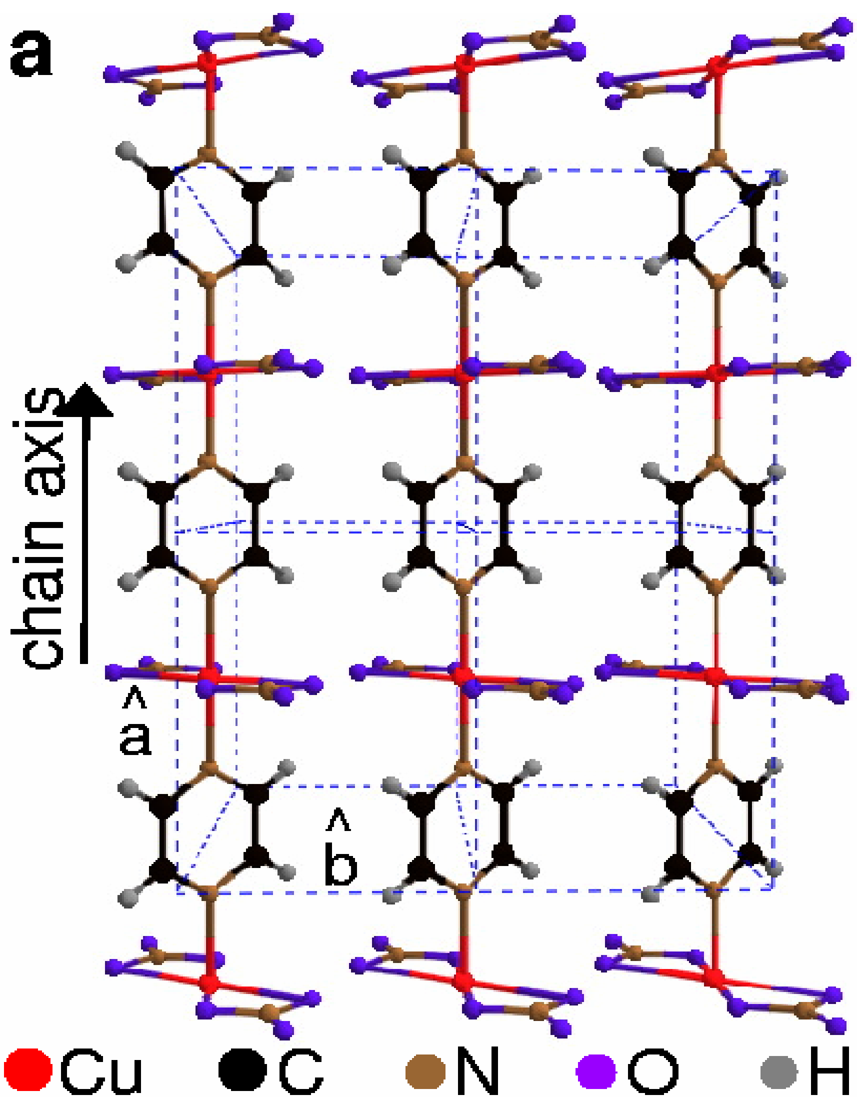



Dispersion relation

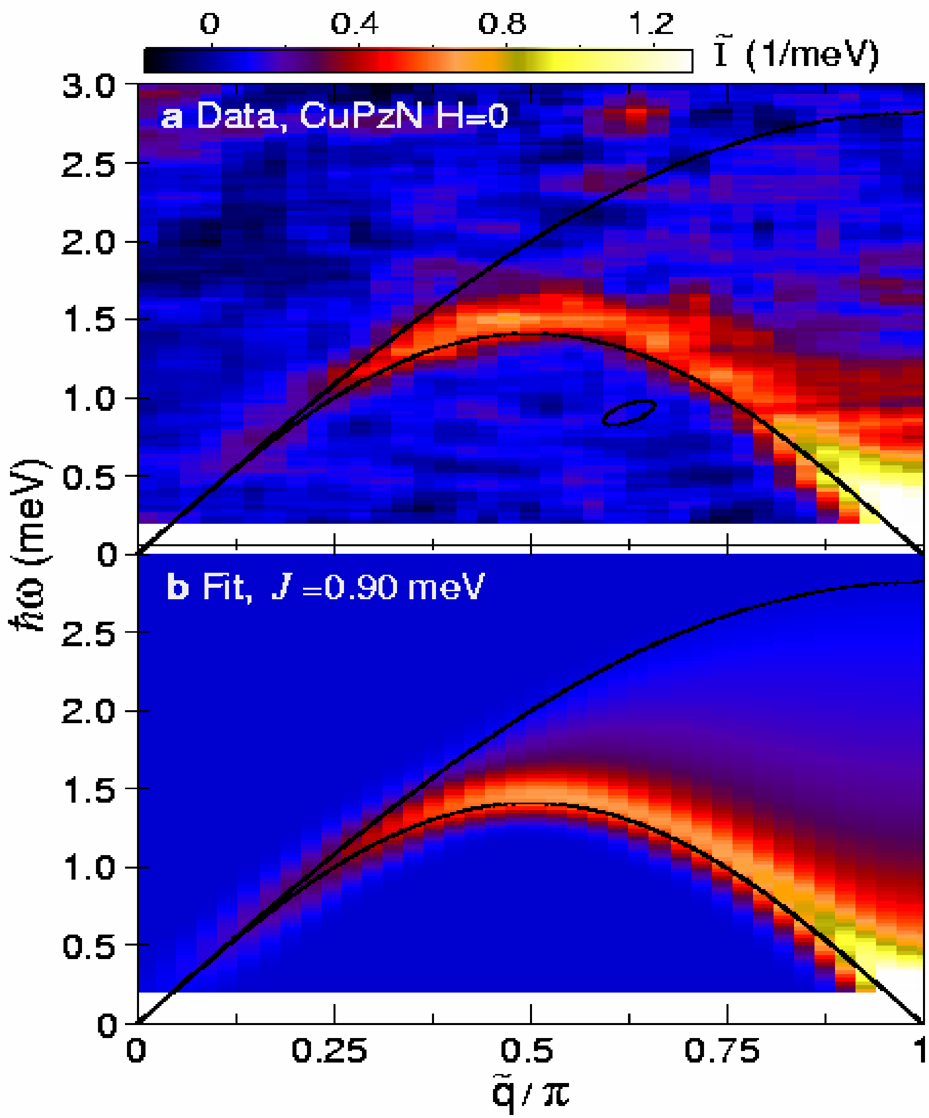
$$\varepsilon(\vec{\kappa})=2S\sqrt{\mathrm{J}(0)^2-\mathrm{J}(\vec{\kappa})^2}$$



# Two-particle continuum scattering




$Cu(C_4H_4N_2)(NO_3)_2$




*Stone et al. (2003).*

# Summary

- Nucl. scattering $$\mathcal{S}(\kappa\omega)=\frac{1}{2\pi\hbar}\frac{1}{N}\int dte^{-i\omega t}\sum_{nn'}\left\langle e^{i\boldsymbol{\kappa}\cdot\mathbf{R}_n}e^{-i\boldsymbol{\kappa}\cdot\mathbf{R}_{n'}(t)}\right\rangle$$
- Magnetic Scattering $$\mathcal{S}^{\alpha\beta}(\kappa\omega)=\frac{1}{2\pi\hbar}\int dte^{-i\omega t}\left\langle s_{\boldsymbol{\kappa}}^{\alpha}s_{-\boldsymbol{\kappa}}^{\beta}(t)\right\rangle$$
- Detailed balance $$\mathcal{S}(\kappa,-\omega)=e^{-\beta\hbar\omega}\mathcal{S}(-\kappa,\omega)$$
- Fluctuation dissipation $$\mathcal{S}(\kappa\omega)=\frac{1}{1-e^{-\beta\hbar\omega}}\frac{\chi''(\boldsymbol{\kappa}\omega)}{\pi}$$
- Know your sum-rules and happy scattering!