Linear Response
&
RPA Dispersive Magnetic Excitations

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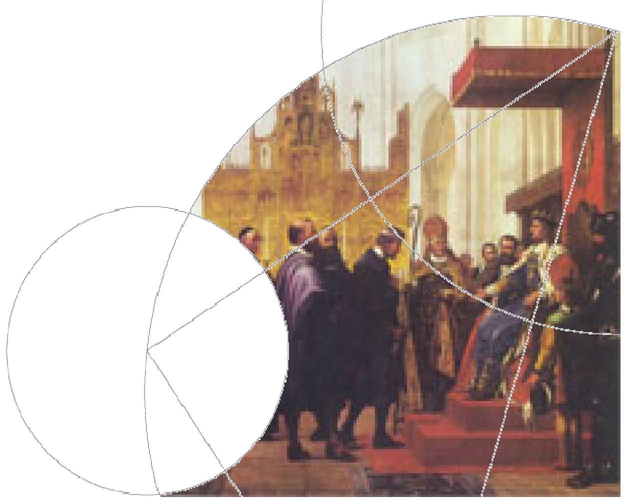


Linear response theory

Non-interacting spin system: $\mathcal{H}(i) = \mathcal{H}_{\mathrm{CF}} - (\vec{h}_0 + \delta\vec{h}\, e^{-i\omega t}) \cdot \vec{J}, \qquad \vec{h} \equiv g\mu_B \vec{H}$

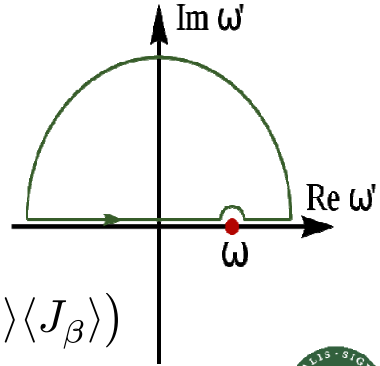
Thermal average: $\langle \hat{A} \rangle = \frac{1}{Z} \mathrm{Tr}\left(\hat{A} e^{-\mathcal{H}/k_B T} \right), \quad Z = \mathrm{Tr}\left(e^{-\mathcal{H}/k_B T} \right)$

Linear response to a time-~~in~~dependent field: $\delta\langle \vec{J} \rangle = \overline{\overline{\chi}}(\vec{H}_0, T, \omega) \cdot \delta\vec{h}\, e^{-i\omega t}$

$$\chi_{\alpha\beta}(\omega) = \sum_{ab}^{E_a \neq E_b} \frac{\langle a \,|\, J_\alpha \,|\, b \rangle \langle b \,|\, J_\beta \,|\, a \rangle}{E_b - E_a - \hbar(\omega + i\epsilon)} (n_a - n_b) + \chi^{\mathrm{el}}_{\alpha\beta} = \chi'_{\alpha\beta}(\omega) + i\,\chi''_{\alpha\beta}(\omega)$$

$$\chi^{\mathrm{el}}_{\alpha\beta} = \left(\frac{i\epsilon}{\omega + i\epsilon} \right)^2 \frac{1}{k_B T} \left\{ \sum_{ab}^{E_a = E_b} \langle a \,|\, J_\alpha \,|\, b \rangle \langle b \,|\, J_\beta \,|\, a \rangle\, n_a - \langle J_\alpha \rangle \langle J_\beta \rangle \right\}$$

Dirac's formula: $\lim_{\epsilon \to 0^+} \frac{1}{\omega_0 - \omega - i\epsilon} = \mathcal{P} \frac{1}{\omega_0 - \omega} + i\pi\delta(\omega_0 - \omega)$

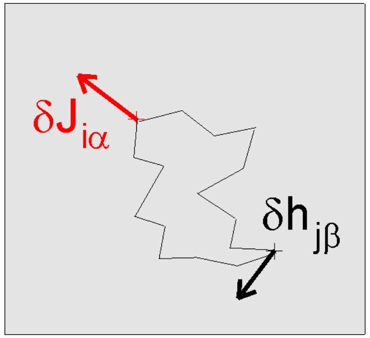


Kramers-Kronig relation: $\chi_{\alpha\beta}(\omega) = \frac{1}{i\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi_{\alpha\beta}(\omega')}{\omega' - \omega} d\omega'$

The correlation function: $S_{\alpha\beta}(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \left(\langle J_\alpha(t) J_\beta(0) \rangle - \langle J_\alpha \rangle \langle J_\beta \rangle \right)$

Fluctuation-dissipation theorem: $S_{\alpha\beta}(\omega) = \frac{2\hbar}{1 - e^{-\hbar\omega/k_B T}} \chi''_{\alpha\beta}(\omega)$

Interacting spin system



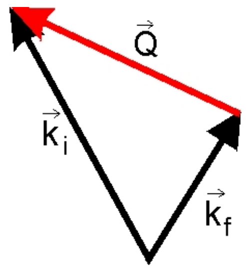
Non-local susceptibility: $\Delta\mathcal{H} = -J_{j\beta}\,\left[\delta h_{j\beta}\, e^{-i\omega t}\right]$

$$\Rightarrow \qquad \delta\langle J_{i\alpha}\rangle = \chi_{\alpha\beta}(ij,\omega)\,\left[\delta h_{j\beta}\, e^{-i\omega t}\right]$$

Differential neutron-scattering cross section:

$$\frac{d^2\sigma}{dE d\Omega} = N\,\frac{k_f}{k_i}\left(\frac{\hbar\gamma e^2}{mc^2}\right)^2 e^{-2W(\vec{Q})}|\tfrac{1}{2}gF(\vec{Q})|^2 \sum_{\alpha\beta}(\delta_{\alpha\beta} - \hat{Q}_\alpha\hat{Q}_\beta)\,\mathcal{S}^{\alpha\beta}(\vec{Q},\omega)$$

$$\vec{Q} = \vec{k}_i - \vec{k}_f, \qquad \hbar\omega = \frac{(\hbar k_i)^2}{2M} - \frac{(\hbar k_f)^2}{2M}$$



Van Hove scattering function:

$$\mathcal{S}^{\alpha\beta}(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt\, e^{i\omega t}\,\frac{1}{N}\sum_{jj'} e^{-i\vec{Q}\cdot(\vec{R}_j - \vec{R}_{j'})}\langle J_{j\alpha}(t)\, J_{j'\beta}(0)\rangle$$

$$= \delta(\hbar\omega)\sum_{j'}\langle J_{j\alpha}\rangle\,\langle J_{j'\beta}\rangle e^{-i\vec{Q}\cdot(\vec{R}_j - \vec{R}_{j'})} + \frac{1}{\pi}\,\frac{1}{1 - e^{-\hbar\omega/k_B T}}\,\chi''_{\alpha\beta}(\vec{Q},\omega)$$

Random-phase approximation (RPA)

$$\mathcal{H} = \sum_i \mathcal{H}_{\mathrm{CF}}(i) - \frac{1}{2}\sum_{i\neq j} \mathcal{J}(ij)\, \vec{J}_i \cdot \vec{J}_j = \sum_i \mathcal{H}_{\mathrm{MF}}(i) - \frac{1}{2}\sum_{i\neq j} \mathcal{J}(ij)\, (\vec{J}_i - \langle \vec{J}_i\rangle_0) \cdot (\vec{J}_j - \langle \vec{J}_j\rangle_0)$$

MF-Hamiltonian: $\mathcal{H}_{\mathrm{MF}}(i) = \mathcal{H}_{\mathrm{CF}}(i) - (\vec{J}_i - \frac{1}{2}\langle \vec{J}_i\rangle_0) \cdot \sum_j \mathcal{J}(ij)\langle \vec{J}_j\rangle_0$

"Non-interacting" MF-susceptibility: $\delta\langle \vec{J}_i\rangle = \langle \vec{J}_i(t)\rangle - \langle \vec{J}_i\rangle_0 = \overline{\overline{\chi}}{}^0_i(\omega)\left[\delta\vec{h}_i e^{-i\omega t}\right]$

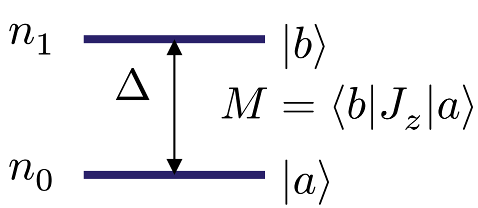
$$\delta\langle \vec{J}_i\rangle = \overline{\overline{\chi}}(ij,\omega)\left[\delta\vec{h}_j e^{-i\omega t}\right] \qquad = \overline{\overline{\chi}}{}^0_i(\omega)\left[\delta\vec{h}_j e^{-i\omega t}\right]_{\mathrm{eff}}$$

$$\left[\delta\vec{h}_j e^{-i\omega t}\right]_{\mathrm{eff}} = \left[\delta\vec{h}_j e^{-i\omega t}\right]\delta_{ij} + \sum_{j'} \mathcal{J}(ij')\left(\vec{J}_{j'}(t) - \langle \vec{J}_{j'}\rangle_0\right) \overset{\mathrm{RPA}}{=}$$

$$\left[\delta\vec{h}_j e^{-i\omega t}\right]\delta_{ij} + \sum_{j'} \mathcal{J}(ij')\left(\langle\vec{J}_{j'}(t)\rangle - \langle \vec{J}_{j'}\rangle_0\right) = \left\{\delta_{ij} + \sum_{j'} \mathcal{J}(ij')\overline{\overline{\chi}}(j'j,\omega)\right\}\left[\delta\vec{h}_j e^{-i\omega t}\right]$$

$$\boxed{\text{RPA-susceptibility:} \quad \overline{\overline{\chi}}(ij,\omega) = \overline{\overline{\chi}}{}^0_i(\omega)\left\{\delta_{ij} + \sum_{j'} \overline{\overline{\mathcal{J}}}(ij')\overline{\overline{\chi}}(j'j,\omega)\right\}}$$

Magnetic excitations (simple example)



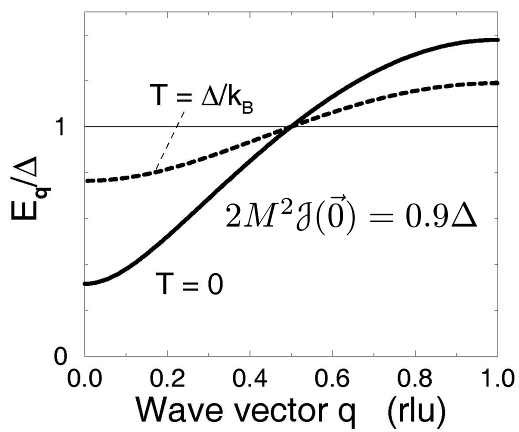
$$\chi^0_{zz}(\omega) = \frac{2n_{01}M^2\Delta}{\Delta^2 - (\hbar\omega)^2}, \qquad n_{01} = n_0 - n_1$$

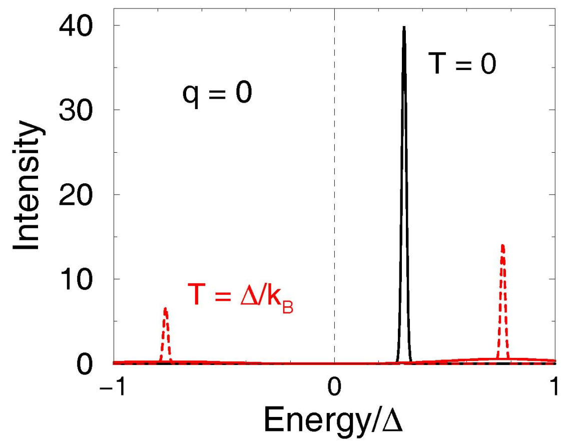
$$\chi_{zz}(\vec{q},\omega) = \chi^0_{zz}(\omega)\{1 + \mathcal{J}(\vec{q})\chi_{zz}(\vec{q},\omega)\}$$

$$\chi_{zz}(\vec{q},\omega) = \frac{\chi^0_{zz}(\omega)}{1 - \chi^0_{zz}(\omega)\mathcal{J}(\vec{q})} = \frac{2n_{01}M^2\Delta}{E^2_{\vec{q}} - (\hbar\omega)^2}, \qquad E^2_{\vec{q}} = \Delta\left[\Delta - 2n_{01}M^2\mathcal{J}(\vec{q})\right]$$

$$\chi_{zz}(\vec{q},\omega) = n_{01}M^2\frac{\Delta}{E_{\vec{q}}}\left(\frac{1}{E_{\vec{q}} - \hbar(\omega + i\epsilon)} + \frac{1}{E_{\vec{q}} + \hbar(\omega + i\epsilon)}\right), \qquad n_{\vec{q}} = \frac{1}{e^{E_{\vec{q}}/k_BT} - 1}$$

$$\mathcal{S}^{zz}(\vec{q},\omega) = \frac{1}{\pi}\frac{1}{1 - e^{-\hbar\omega/k_BT}}\chi''_{zz}(\vec{q},\omega) = n_{01}M^2\frac{\Delta}{E_{\vec{q}}}\left[(1 + n_{\vec{q}})\,\delta(E_{\vec{q}} - \hbar\omega) + n_{\vec{q}}\,\delta(E_{\vec{q}} + \hbar\omega)\right]$$





$|\text{MF-ground state}\rangle = \prod_i |a\rangle_i$

$\langle\, |a\rangle\langle b|\,\rangle \neq 0, \quad \mathrm{P}(\delta E = 2\Delta) \neq 0$

Diagrammatic $1/z$ expansion:

$\epsilon \propto e^{-\Delta/k_BT}$

RPA - Summary

RPA-susceptibility: $\overline{\overline{\chi}}(ij,\omega) = \overline{\overline{\chi}}{}_i^0(\omega)\Big\{\delta_{ij} + \sum_{j'} \overline{\overline{\mathcal{J}}}(ij')\overline{\overline{\chi}}(j'j,\omega)\Big\}$

•The RPA-susceptibilty may be calculated numerically for any kind of crystal with atomic (localized) moments - itinerant spin systems require other methods.

•The only exception is that of a "truely" incommensurate ordered magnetic system, but even in that case useful results may be obtained by assuming a commensurate ordering with a wave vector close to the incommensurate one .

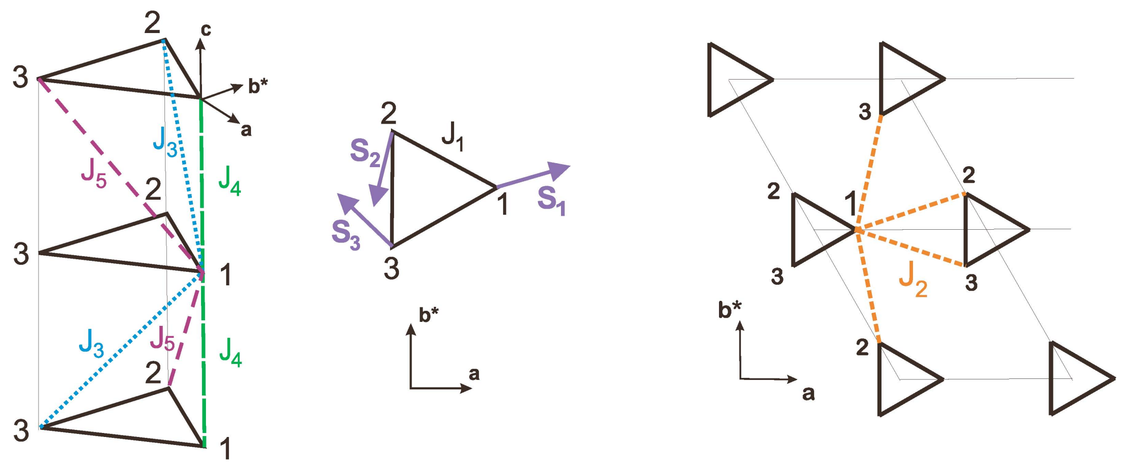
•The RPA, like the MF approximation, is most trustworthy for 3D-systems with long range interactions.

•Unlike the MF approximation, the RPA does not apply at high temperatures, where the lifetimes of the magnetic excitations become short due to uncorrelated thermal fluctuations [the linewidths of quasielastic paramagnetic excitations at high temperatures may be determined by the use of certain "sum rules"].

•Special care should be shown in cases, where one interaction between a few neighbors is the dominating one, as in the case of dimer, trimer, tetramer, or weakly coupled chain systems.

Chiral spin-wave excitations of the spin-5/2 trimers in the langasite compound $Ba_3NbFe_3Si_2O_{14}$

Hund's rules applied on Fe^{3+} ion with five $3d$ electrons $\quad \Rightarrow \quad S = \frac{5}{2},\ L = 0$



Trigonal space group $(P321)$. Structural chirality $\epsilon_T = \pm 1$

Different super-superexchange paths for J_5 and J_3.

$T < T_N = 27$ K: Helix with wave vector $\vec{\tau} \simeq \frac{1}{7}\vec{c}^*$, i.e. $\phi = \epsilon_H \frac{2\pi}{7}$ $\sim$ helicity $\epsilon_H = \pm 1$

The angle between $\langle \vec{S}_1 \rangle$ and $\langle \vec{S}_2 \rangle$ is $\gamma = \epsilon_\gamma \frac{2\pi}{3}$ $\sim$ spin triangle orientation: $\epsilon_\gamma = \pm 1$

$$\tan\phi = R\sin\gamma, \quad R = \frac{2(J_5 - J_3)}{2J_4 - J_3 - J_5} \quad \Rightarrow \quad \epsilon_H = \mathrm{sign}(R)\,\epsilon_\gamma$$

Cluster MF/RPA model

$$\mathcal{H} = \sum_i \mathcal{H}_T(i) + \frac{1}{2}\sum_{i,\xi}\sum_{j,\eta} J_{\xi\eta}(ij)\,\vec{S}_\xi(i)\cdot\vec{S}_\eta(j)$$

$$\mathcal{H}_T = J_1\left(\vec{S}_1\cdot\vec{S}_2 + \vec{S}_2\cdot\vec{S}_3 + \vec{S}_3\cdot\vec{S}_1\right) + D_c\left(\vec{S}_1\times\vec{S}_2 + \vec{S}_2\times\vec{S}_3 + \vec{S}_3\times\vec{S}_1\right)\cdot\hat{c}$$

$J_1 = 1.25,\ J_2 = 0.2,\ J_3 = 0.1,\ J_4 = 0.064,\ J_5 = 0.29$

$D_c = 0.0038$ (in units of meV)

for a crystal with the structural chirality $\epsilon_T = -1$

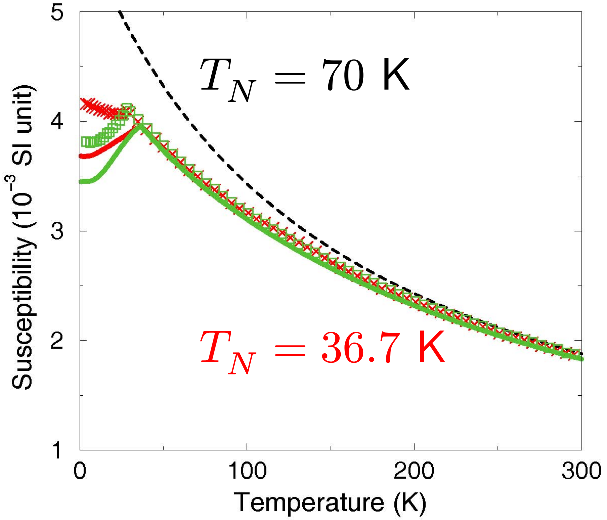
Technicalities:

$\vec{S}_1 \otimes \vec{S}_2 \otimes \vec{S}_3 \quad \Rightarrow \quad 6^3 = 216$ states/trimer

MF-susceptibility (9×9 matrices): $\left(\overline{\overline{\chi}}^{\,0}\right)^{\xi\eta}(\omega)$

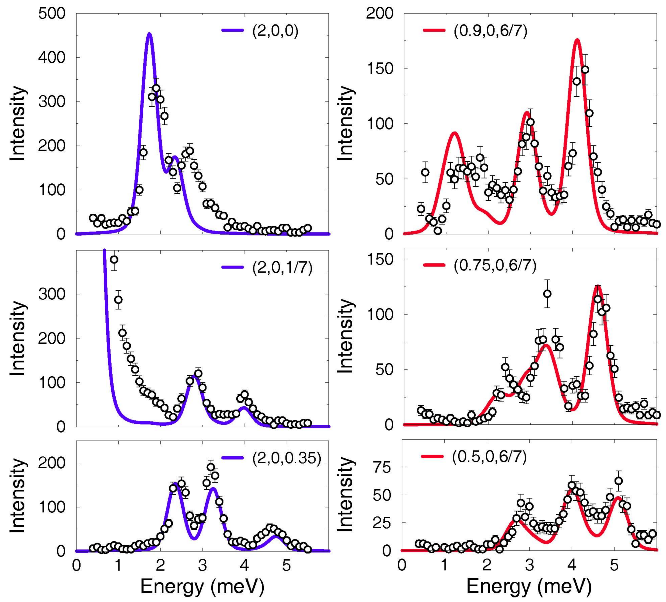
7 trimer sublattices.

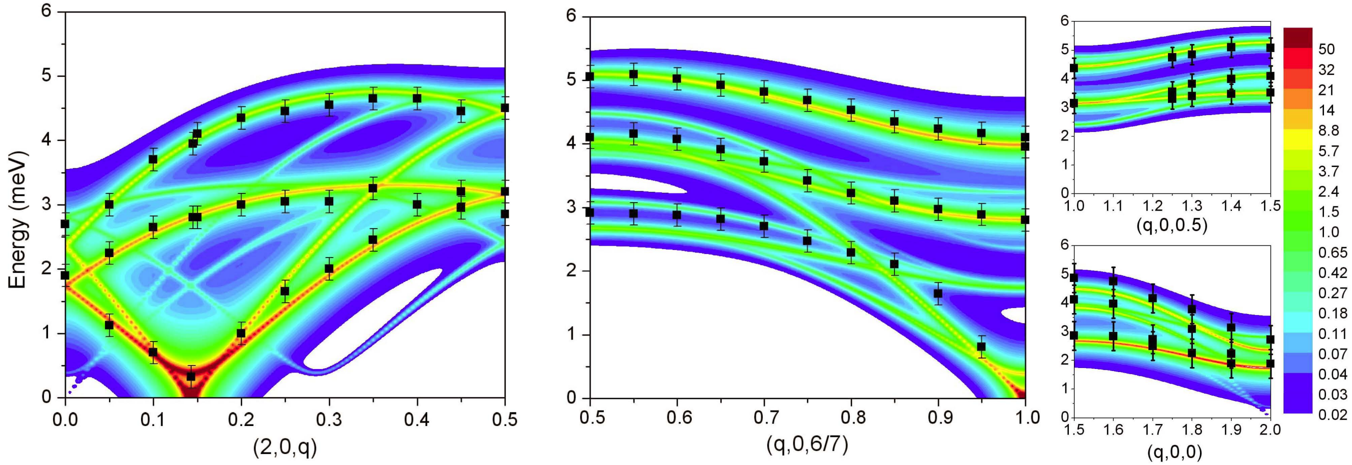
$$I(\vec{q},\omega) = \sum_{\alpha\beta}\frac{\delta_{\alpha\beta} - q_\alpha q_\beta/q^2}{3\pi\left(1 - e^{-\hbar\omega/k_B T}\right)} \times \sum_{\xi\eta}\mathrm{Im}\left[\chi^{\xi\eta}_{\alpha\beta}(\vec{q},\omega)e^{-i\,\vec{q}\cdot(\vec{R}_\xi - \vec{R}_\eta)}\right]$$



Unpolarized neutron scattering

Experiments at 2.5 K performed by
C. Stock, L. C. Chapon, A. Schneidewind, Y. Su,
P.G. Radaelli, D. F. McMorrow, A. Bombardi,
N. Lee, and S.-W. Cheong,
Phys. Rev. B **83**, 104426 (2011).





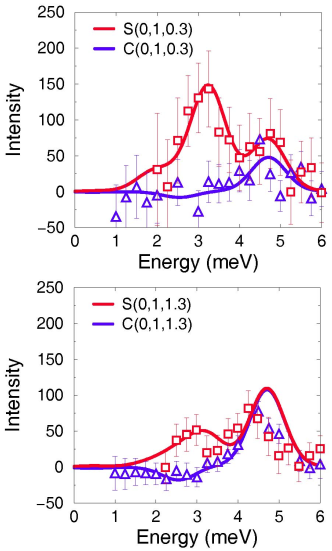
Polarized neutron scattering

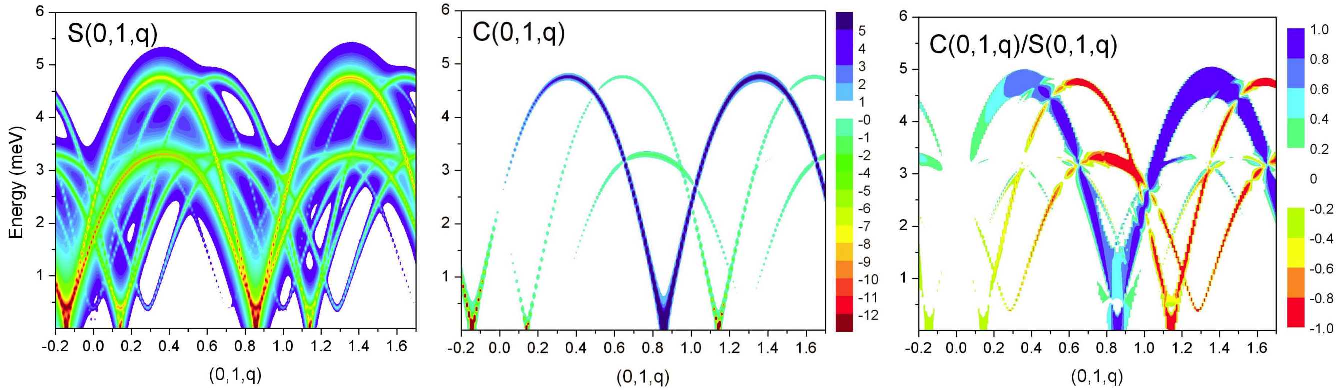
Experiments at 1.5 K performed by
M. Loire, V. Simonet, S. Petit, K. Marty, P. Bordet, P. Lejay, J. Ollivier,
M. Enderle, P. Steffens, E. Ressouche, A. Zorko, and R. Ballou, arXiv:1010.2008.

R. M. Moon, T. Riste, and W. C. Koehler, Phys. Rev. **181**, 920 (1969)

$$\frac{d\sigma^{\pm/\mp}}{d\Omega} = \sum_{ij} e^{i\vec{k}\cdot\vec{r}_{ij}}\, p_i p_j^* \left[\vec{S}_{\perp i}\cdot\vec{S}_{\perp j} \mp i\hat{z}\cdot(\vec{S}_{\perp i}\times\vec{S}_{\perp j})\right]$$

$$S(\vec{Q},\omega) = \frac{I^{\pm}(\vec{Q},\omega) + I^{\mp}(\vec{Q},\omega)}{2}, \quad C(\vec{Q},\omega) = \frac{I^{\pm}(\vec{Q},\omega) - I^{\mp}(\vec{Q},\omega)}{2}, \quad \hat{z} = \hat{Q}$$





sign$(R) = -1$
or $\epsilon_H = -\epsilon_\gamma$

$D_c > 0$

$\epsilon_H = +1$ for a crystal with $\epsilon_T = -1$