Linear Response Theory and the Random Phase Approximation

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J. Jensen and A. R. Mackintosh, "Rare Earth Magnetism" http://www.nbi.ku.dk/page40667.htm

Linear response theory

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

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

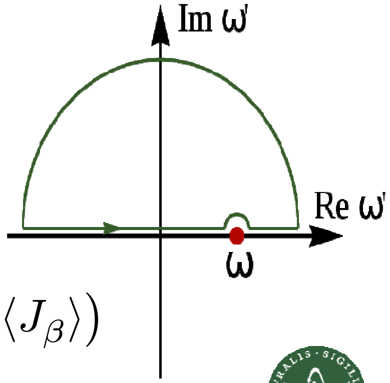
Linear response to a time-~~in~~dependent field: $\delta\langle \vec{J} \rangle = \overline{\overline{\chi}}(\vec{H}_0, T, \omega) \cdot \delta\vec{h}\, e^{-i\omega t}$

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

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

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

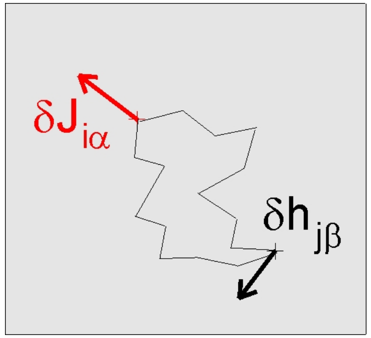
Kramers-Kronig relation: $\chi_{\alpha\beta}(\omega) = \frac{1}{i\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi_{\alpha\beta}(\omega')}{\omega' - \omega} d\omega'$



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



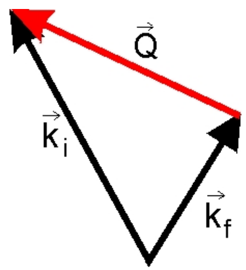
Non-local susceptibility: $\Delta\mathcal{H} = -J_{j\beta}\,\left[\delta h_{j\beta}\, e^{-i\omega t}\right]$

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

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:

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

$$= \delta(\hbar\omega)\sum_{j'}\langle J_{j\alpha}\rangle\,\langle J_{j'\beta}\rangle e^{-i\vec{Q}\cdot(\vec{R}_j-\vec{R}_{j'})} + \frac{1}{\pi}\,\frac{1}{1-e^{-\hbar\omega/k_BT}}\,\chi''_{\alpha\beta}(\vec{Q},\omega)$$

Random-phase approximation (RPA)

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

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

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

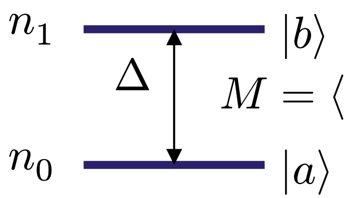
$$\delta\langle\vec{J}_i\rangle = \overline{\overline{\chi}}(ij,\omega)\left[\delta\vec{h}_j e^{-i\omega t}\right] \qquad = \overline{\overline{\chi}}{}^0_i(\omega)\left[\delta\vec{h}_j e^{-i\omega t}\right]_{\rm eff}$$

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

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

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

Magnetic excitations (simple example)



$M = \langle b|J_z|a\rangle$

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

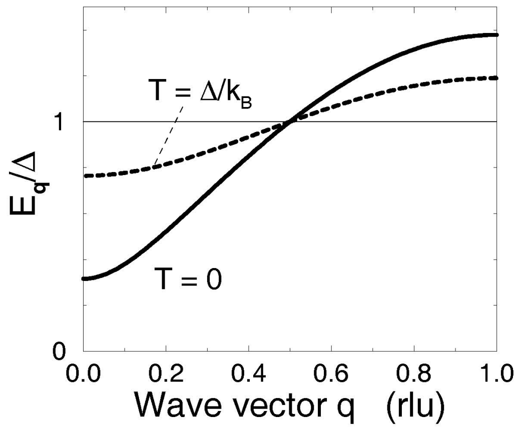
$$\chi_{zz}(\vec{q},\omega) = \chi^0_{zz}(\omega)\{1 + \mathcal{J}(\vec{q})\chi_{zz}(\vec{q},\omega)\}$$

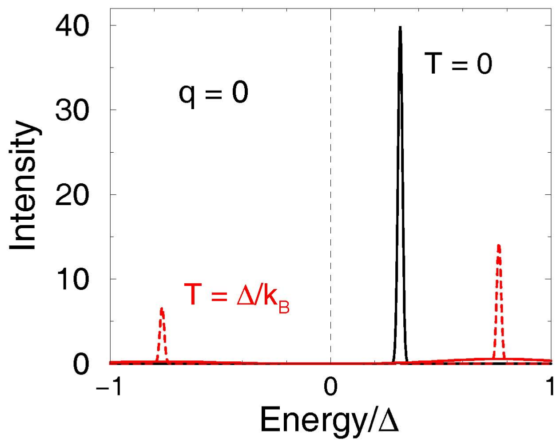
$$\chi_{zz}(\vec{q},\omega) = \frac{\chi^0_{zz}(\omega)}{1 - \chi^0_{zz}(\omega)\mathcal{J}(\vec{q})} = \frac{2n_{01}M^2\Delta}{E^2_{\vec{q}} - (\hbar\omega)^2}, \qquad E^2_{\vec{q}} = \Delta\left[\Delta - 2n_{01}M^2\mathcal{J}(\vec{q})\right]$$

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

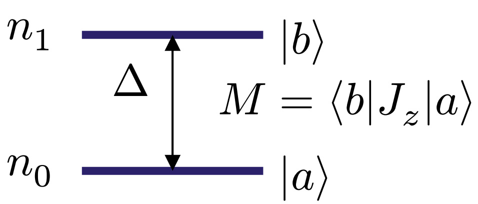
$$\mathcal{S}^{zz}(\vec{q},\omega) = \frac{1}{\pi}\frac{1}{1 - e^{-\hbar\omega/k_BT}}\chi''_{zz}(\vec{q},\omega) = n_{01}M^2\frac{\Delta}{E_{\vec{q}}}\left[(1 + n_{\vec{q}})\,\delta(E_{\vec{q}} - \hbar\omega) + n_{\vec{q}}\,\delta(E_{\vec{q}} + \hbar\omega)\right]$$

$R = 2M^2\mathcal{J}(\vec{0})/\Delta = 0.9$





Magnetic excitations (damping)



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

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

(i) Becker-Fulde-Keller theory [Z. Physik B **28**, 9 (1977), see Manual p.104-112]: Electron-hole pair scattering in metallic systems (RKKY interaction). Dominates in weakly coupled (dilute) systems and in the $T = 0$ limit in interacting (strongly dispersive) metallic rare-earth systems.

(ii) Scattering due to single-site population fluctuations:
$|\text{MF-ground state}\rangle = \prod_i |a\rangle_i$ is not an eigenstate.

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

Diagrammatic high-density $1/z$ 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.

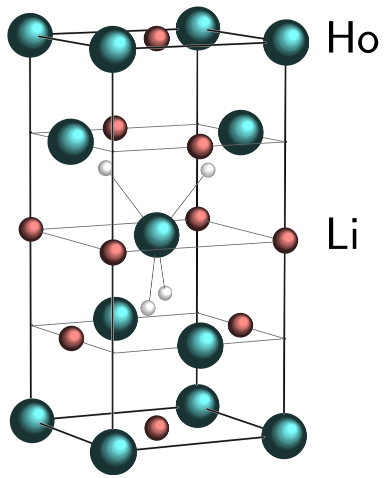
•The MF approximation is a prerequisite for the RPA, and both approximations are most trustworthy for 3D-systems with long range interactions (excluding "highly frustrated systems").

•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 (excluding non or weakly interacting systems).

•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.

Dipolar and hyperfine interactions in the Ising ferromagnet $LiHoF_4$

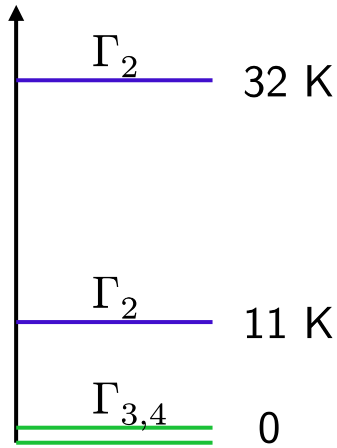
Hund's rules applied on Ho^{3+} ion with ten $4f$ electrons $\Rightarrow$
$S = 2,\ L = 6,\ J = 8$, and $g = 5/4$



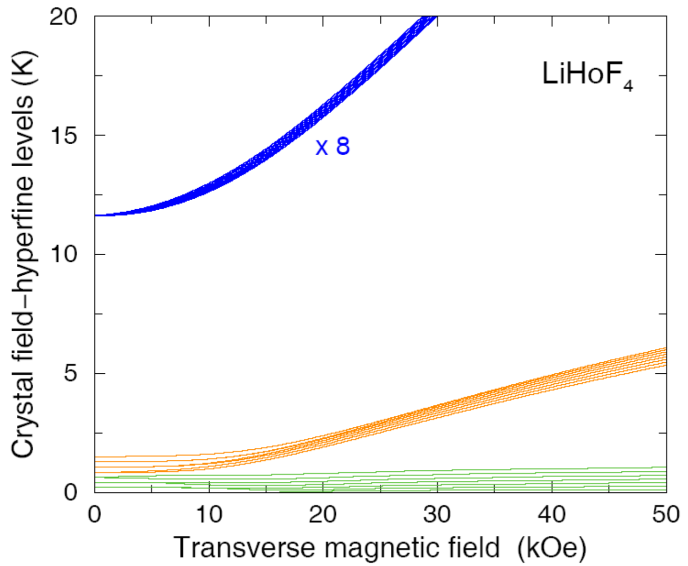
Scheelite lattice (tetragonal space group 88).
The point symmetry of the Ho sites is S_4

$$\mathcal{H}_{CF} = \sum_{\ell=2,4,6} B_\ell^0 O_\ell^0 + \sum_{\ell=4,6} B_\ell^4(c) O_\ell^4(c) + B_6^4(s) O_6^4(s)$$

B_ℓ^m are determined from susceptibility and resonance measurements. The ground state is a doublet where only the matrix element of J_c is non-zero.
At $T \ll 11$ K, the system is effectively an $S = 1/2$ Ising system.



Ising model in a transverse field



Single-ion Hamiltonian:

$$\mathcal{H}(\mathbf{J}_i) = \sum_i \left[\mathcal{H}_{\mathsf{CF}}(\mathbf{J}_i) + A\,\mathbf{J}_i \cdot \mathbf{I}_i - g\mu_B \mathbf{J}_i \cdot \mathbf{H}_\perp \right]$$

$I = 7/2$ for ^{165}Ho and $A = 3.36\ \mu$eV.
$(2J+1) \times (2I+1) = 136$ number of states.

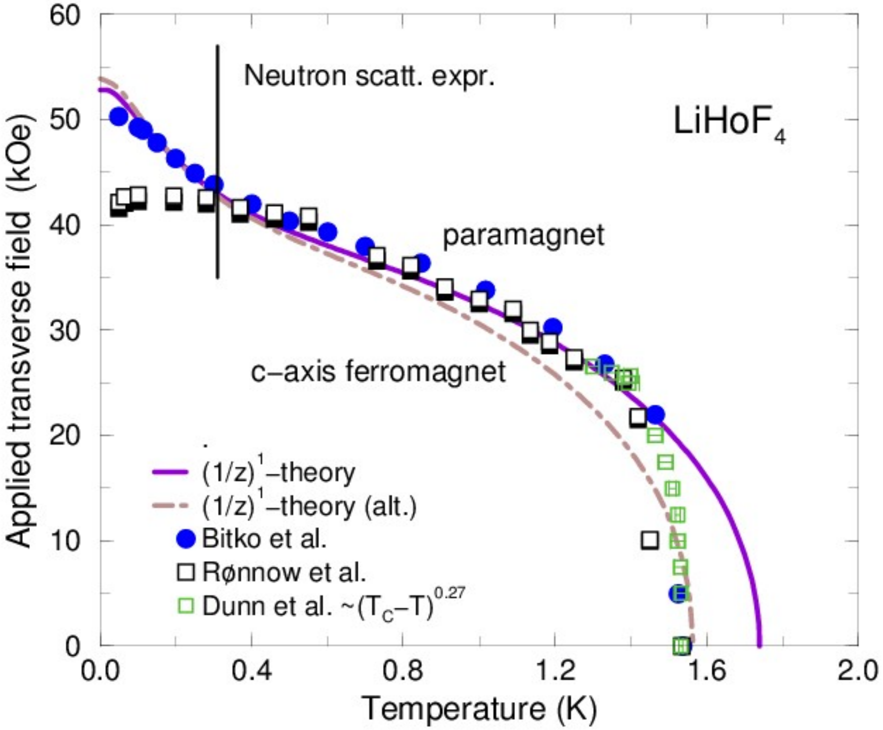
Two-ion dipolar interaction:

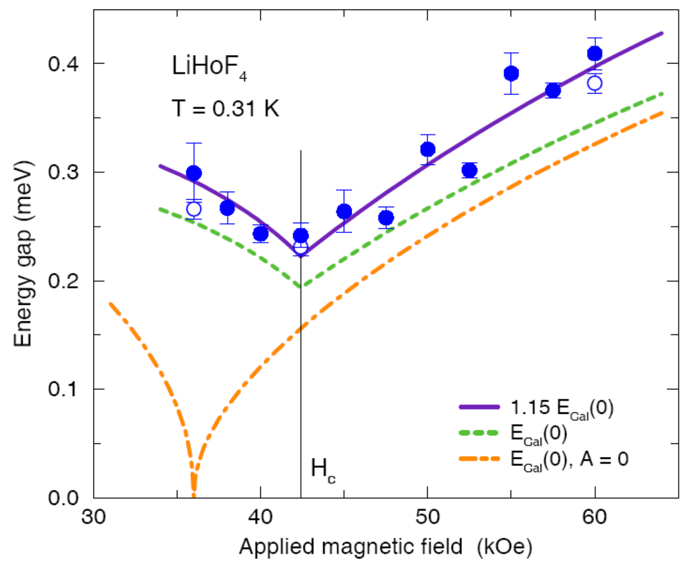
$$\mathcal{H} = \sum_i \mathcal{H}(\mathbf{J}_i) - \tfrac{1}{2} \sum_{ij} \sum_{\alpha\beta} \mathcal{J}_D D_{\alpha\beta}(ij) J_{i\alpha} J_{j\beta} - \tfrac{1}{2} \sum_{ij}^{n.n.} \mathcal{J}_{12}\, \mathbf{J}_i \cdot \mathbf{J}_j$$

$$D_{\alpha\beta}(ij) = \frac{3(r_{i\alpha} - r_{j\alpha})(r_{i\beta} - r_{j\beta}) - |\mathbf{r}_i - \mathbf{r}_j|^2 \delta_{\alpha\beta}}{N |\mathbf{r}_i - \mathbf{r}_j|^5}$$

$\mathcal{J}_D = (g\mu_B)^2 N = 1.165\ \mu$eV and $\mathcal{J}_{12} = -0.1\ \mu$eV (the only fitting parameter).

Neutron scattering experiments[1)] near the "QPT" in $LiHoF_4$

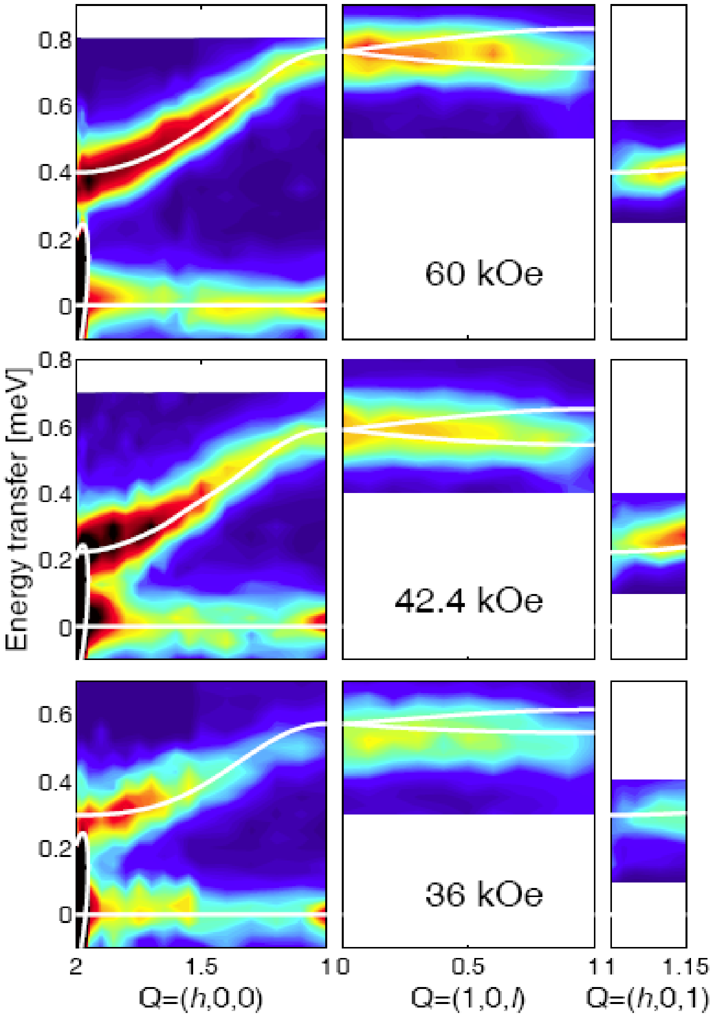


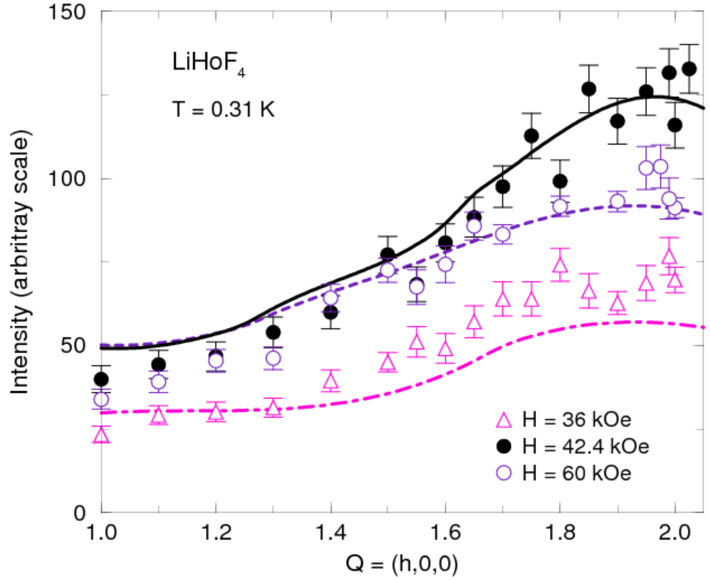


The MF/RPA theory is improved by including the effects of single-site fluctuations (diagrammatic expansion to first order in $1/z$, where z is the coordination number).

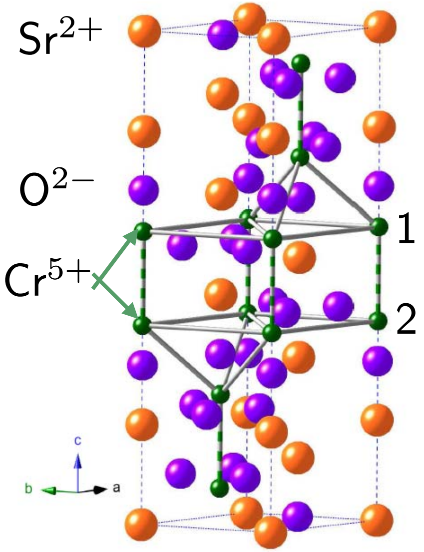
1) Rønnow *et al.*, Science **308**, 389 (2005) and Phys. Rev. B **75**, 054426 (2007)

Neutron scattering experiments near the “QPT” in $LiHoF_4$ (ii)





The singlet-triplet excitations in the dimer system $Sr_3Cr_2O_8$



Hund's rules applied on Cr^{5+} ion with one $3d$ electron $\Rightarrow$ $S = 1/2$ and $L = 2$, but the orbital moment is quenched.

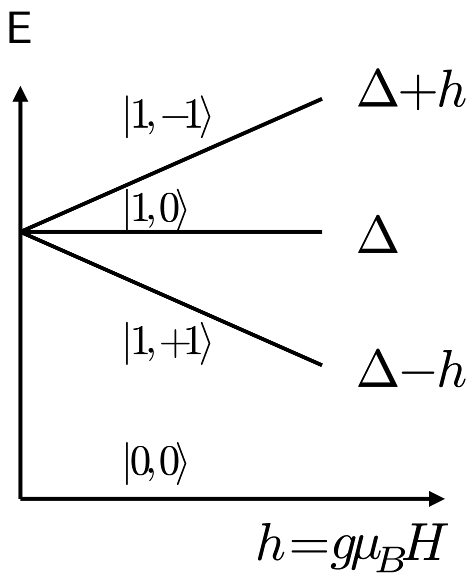
$$\mathcal{H} = -\frac{1}{2}\sum_{ij}\mathcal{J}_H(ij)\vec{s}_i \cdot \vec{s}_j - \sum_i g\mu_B \vec{s}_i \cdot \vec{H}$$

The green pairs (12) of Cr^{5+} ions are strongly coupled, i.e. $|\mathcal{J}_H(12)| = \Delta$ is much larger than any other exchange parameter.

ith dimer: $\vec{S}_i^{\pm} = \vec{s}_{i1} \pm \vec{s}_{i2}$ $\Rightarrow$ paramagnetic MF eigenstates:

$S^+ = 0$ (singlet) and

$S^+ = 1, \; S_z^+ = -1, 0, 1$



Excitations operators:

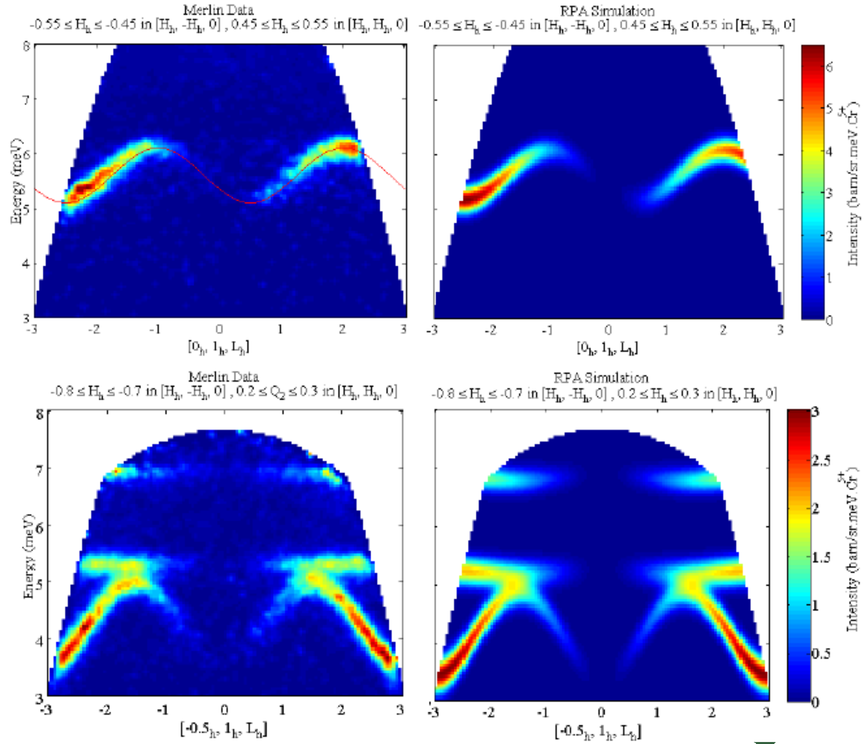
$S_x^-, \; S_y^-, \; S_z^-$

Dispersion relation:

$[E(\vec{q})]^2 = \Delta[\Delta - 2\mathcal{J}(\vec{q})]$

at $T = 0$ and $H = 0$.

Diana Quintero-Castro: Time-of-flight experiment at ISIS (Merlin) $\Rightarrow$.



The lineshape of the singlet-triplet excitations in the dimer system $Sr_3Cr_2O_8$

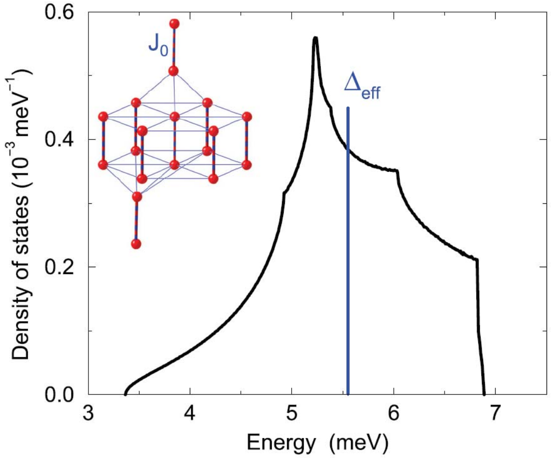


FIG. 1. (Color online) The density of states of singlet-triplet excitations in $Sr_3Cr_2O_8$ in the zero temperature limit. It is determined from the RPA model in Ref. [3] as an average over the three twinned domains. The inset shows the spin-dimer structure in $Sr_3Cr_2O_8$. The

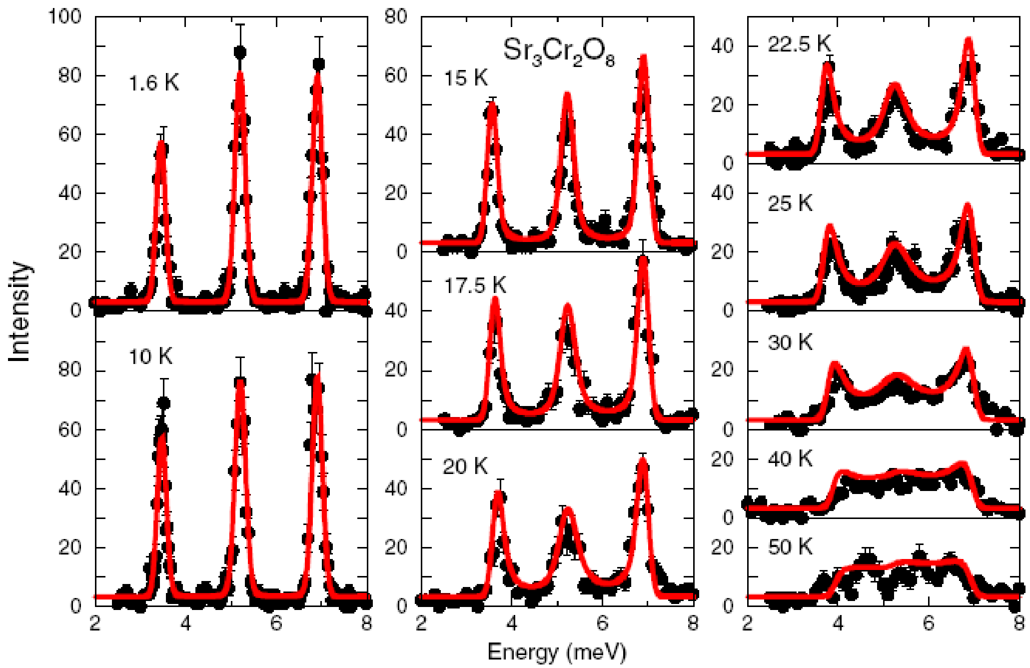


FIG. 2. (Color online) Constant Q scans at (0.5,0.5,3) measured at different temperatures on V2-FLEX. The solid lines are the calculated response functions assuming the fitted experimental resolution function to be independent of temperature. The three peaks derive from the three equivalent crystallographic twins.

Including the fluctuations to leading order in $1/z$, the diagrammatic high-density expansion, is found to offer an accurate description of the singlet-triplet excitations ($R = 0.63$). After J. Jensen *et al.*, Phys. Rev. B **89**, 134407 (2014).