Spin 1/2 Triangular Antiferromagnet Cs_2CuCl_4

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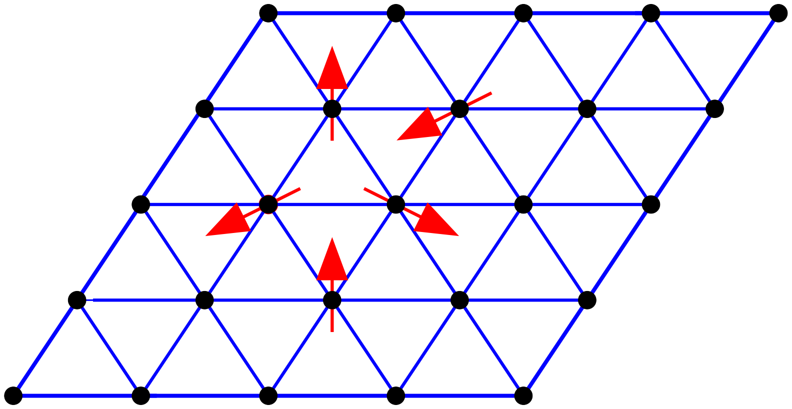
Phys. Rev. B 74, 052402 (2006)

Phys. Rev. B 72, 134429 (2005)

Phys. Rev. B 71, 214426 (2005)



Outline

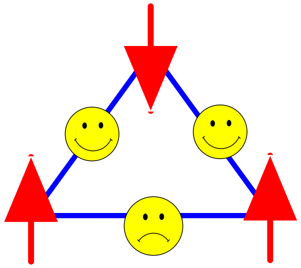
- Geometric Frustration
- Fundamentals of Cs_2CuCl_4
  - ⋆ Structure and Properties
  - ⋆ Experimental Results: Phase Diagram, Excitations
- Mean Field in Zero Field
- Role of Quantum Fluctuations
  - ⋆ Large S Expansion: Spinwaves
  - ⋆ Quantum fluctuation renormalization
  - ⋆ Dynamical Correlation Functions
  - ⋆ Comparison to Experiments
- Conclusion

Frustrated Magnets: Unhappy Magnets

Quantum Fluctuations are generic of Antiferromagnets

$$i\frac{d\mathbf{N}}{dt} = [\mathbf{N}, H] \neq 0$$

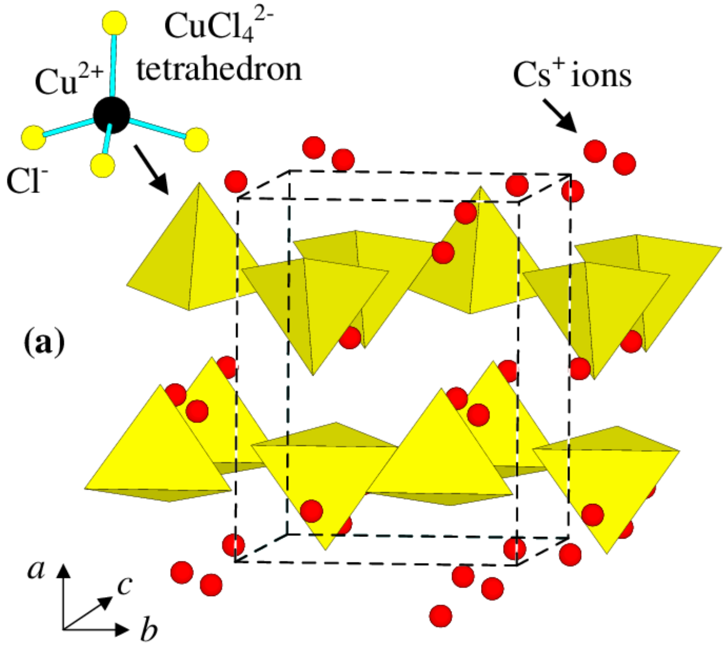
Order parameter is not a constant of motion.



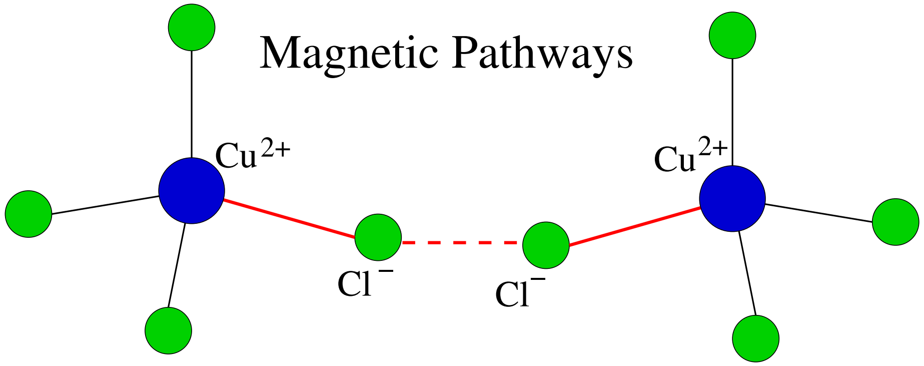
Phase space for fluctuations is enhanced
by geometric frustration

- Competition between mean-field ordering and quantum fluctuations
- Many new collective behavior may emerge
  - ⋆ New Paradigms
  - ⋆ Quantum Spin Liquid: Cs_2CuCl_4 ??

Crystal Structure and Magnetism of Cs_2CuCl_4

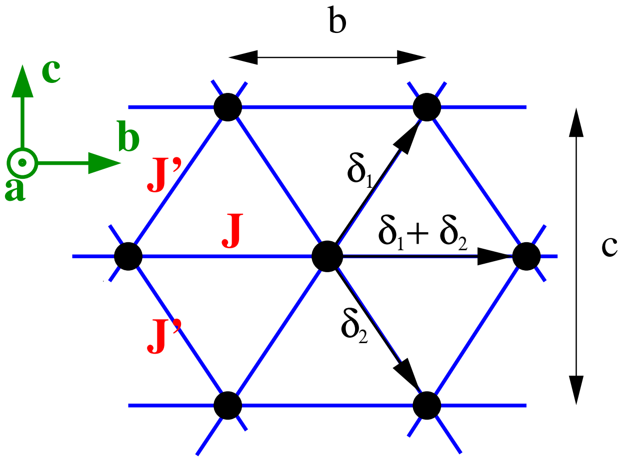


Layers of $S = 1/2$ Cu^{2+} ions coupled in triangular geometry



Low exchange interaction

Cs_2CuCl_4: Quasi 2-D Spin 1/2 Triangular Antiferromagnet



$$\mathcal{H}_0 = \frac{1}{2} \sum_{\mathbf{R},\delta} J_\delta \mathbf{S}_\mathbf{R} \cdot \mathbf{S}_{\mathbf{R}+\delta}$$

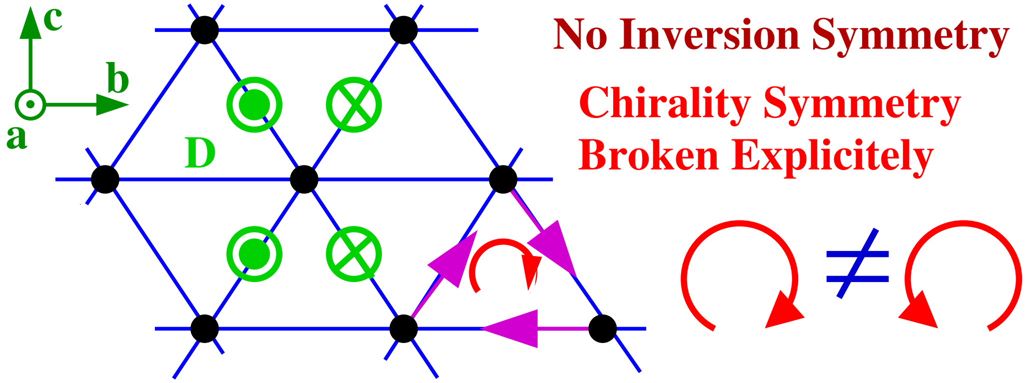
- $J' = 0 \rightarrow$ Non-Interacting Chain
- $J = J' \rightarrow$ Fully Frustrated Triangular Lattice

Experiment: $J'/J = 0.34(3)$, $J = 0.37(4)\text{meV} \approx 4K$

Additional Interactions:

- Coupling $J'' = 0.017\text{meV}$ between Stacked Layers
  Small but ultimately responsible for LRO
- Dzyaloshinskii-Moriya (DM) Interaction :
  Due to absence of inversion symmetry
  Small ($D = 0.020\text{meV}$) but Breaks SU(2) symmetry
  Transverse Field $\neq$ Longitudinal Field

$$\mathcal{H}_{DM} = -\sum_{\mathbf{R}} (-1)^n \mathbf{D} \cdot \left(\mathbf{S}_{\mathbf{R}} \times \left[\mathbf{S}_{\mathbf{R}+\delta_1} - \mathbf{S}_{\mathbf{R}-\delta_1} + \mathbf{S}_{\mathbf{R}+\delta_2} - \mathbf{S}_{\mathbf{R}-\delta_2}\right]\right)$$



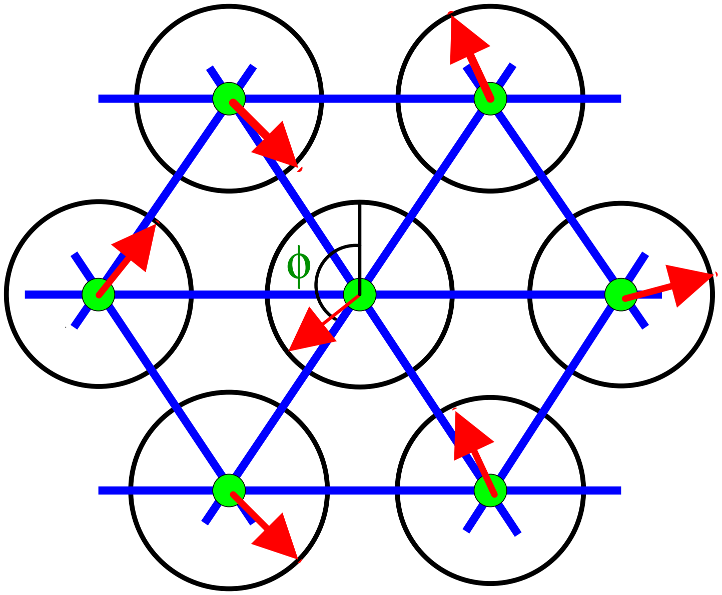
No Inversion Symmetry

Chirality Symmetry Broken Explicitely

Mean Field Result

In Zero field: Cycloid Phase → Incommensurate LRO

$$\begin{aligned} S^b_{\mathbf{R}} &= S\cos(\phi_{\mathbf{R}}) \\ S^c_{\mathbf{R}} &= S\sin(\phi_{\mathbf{R}}) \end{aligned}$$



$$\begin{aligned} \phi_{\mathbf{R}} &= \mathbf{Q}\cdot\mathbf{R}+\alpha \\ \mathbf{Q} &= (0,\pi+2\pi\epsilon,0) \\ \epsilon &\simeq 1/\pi\sin^{-1}\left(\frac{J'}{2J}\right) \end{aligned}$$

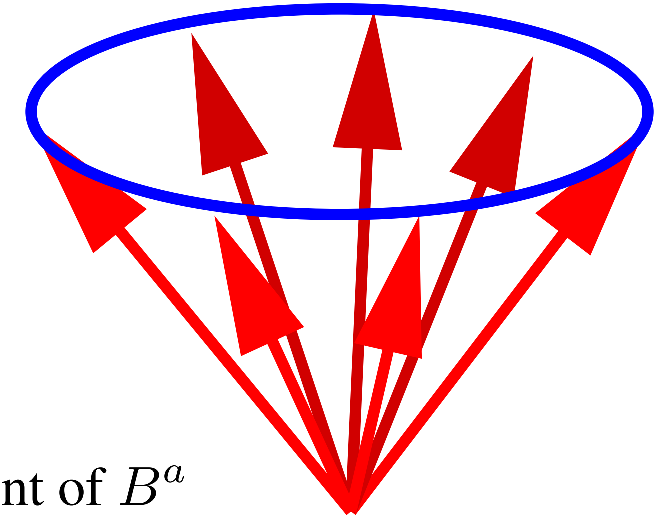
Non-Collinearity promoted by frustration, $\epsilon = 0.053$

In Transverse field, i.e. $B^a \neq 0$:

$$\begin{aligned} S^a_{\mathbf{R}} &= S\sin\theta \\ S^b_{\mathbf{R}} &= S\cos\theta\cos\left(\phi_{\mathbf{R}}\right) \\ S^c_{\mathbf{R}} &= S\cos\theta\sin\left(\phi_{\mathbf{R}}\right) \end{aligned}$$

Perpendicular field stabilizes Cone State

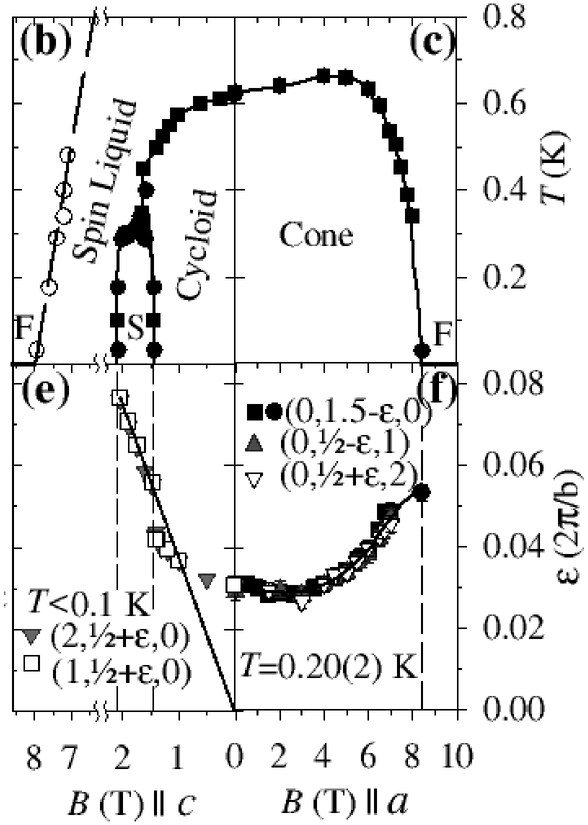
Spins cant along the magnetic field



$$\begin{aligned} \sin\theta &= h^a/h^a_{cr} \\ h^a_{cr} &= 2(J^T_{\mathbf{0}} - J^T_{\mathbf{Q}}) \\ B^a_{cr} &= 8.36\mathrm{T} \\ \text{Exp.: } B^a_{cr} &= 8.44\mathrm{T} \\ \phi_{\mathbf{R}} &= \mathbf{Q}\cdot\mathbf{R} \text{ independent of } B^a \end{aligned}$$

Experimental Phase Diagram R.Coldea et al. PRL, 2001, Ibid, 2002 and Ibid, PRB 2003

- Very Sensitive to Anisotropy
- Incommensurate Ordering in Transverse and Zero field
- Quantum Spin Liquid Phase in Longitudinal Field ?



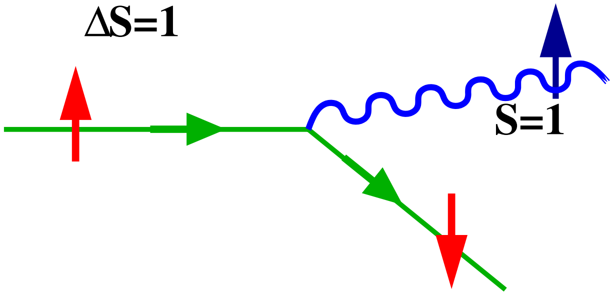
Phase and Excitations

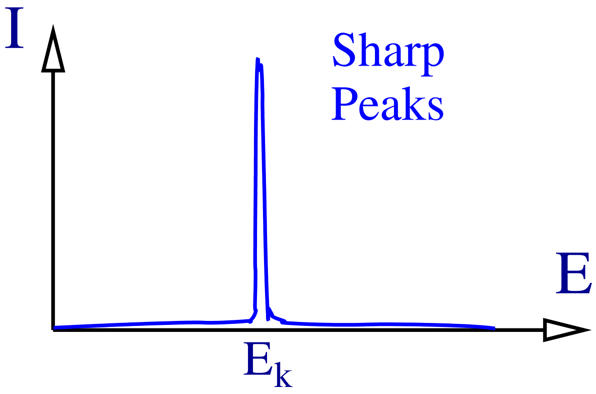
Spin Solid	**Spin Liquid**
Old and Boring	**New Paradigm**
Long Range Order below T_c	Short Range Order
Bragg Peaks	$\langle S_{\mathbf{r}} S_{\mathbf{0}} \rangle \sim e^{-\mathbf{r}/\xi}$
Conventional	Melting of Crystal Order
Symmetry Breaking	Emergent (Gauge) Symmetry
Magnetic Excitations:	
Magnons	Spinons
Goldstone Mode (Gapless)	Gap/Gapless
Spin 1	Spin 1/2
Bosons	Fermions or Bosons

Spin Spectral Function

Spin Solid

Neutron Scattering from Magnons ΔS=1

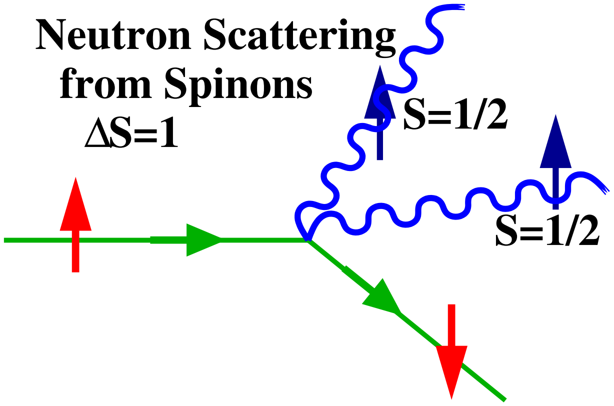


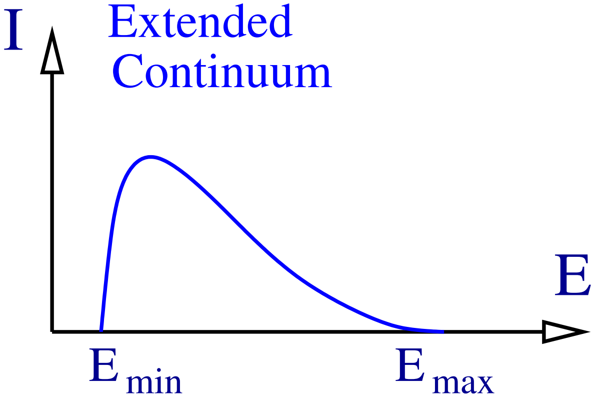


Sharp Peaks

Spin Liquid

Neutron Scattering from Spinons ΔS=1

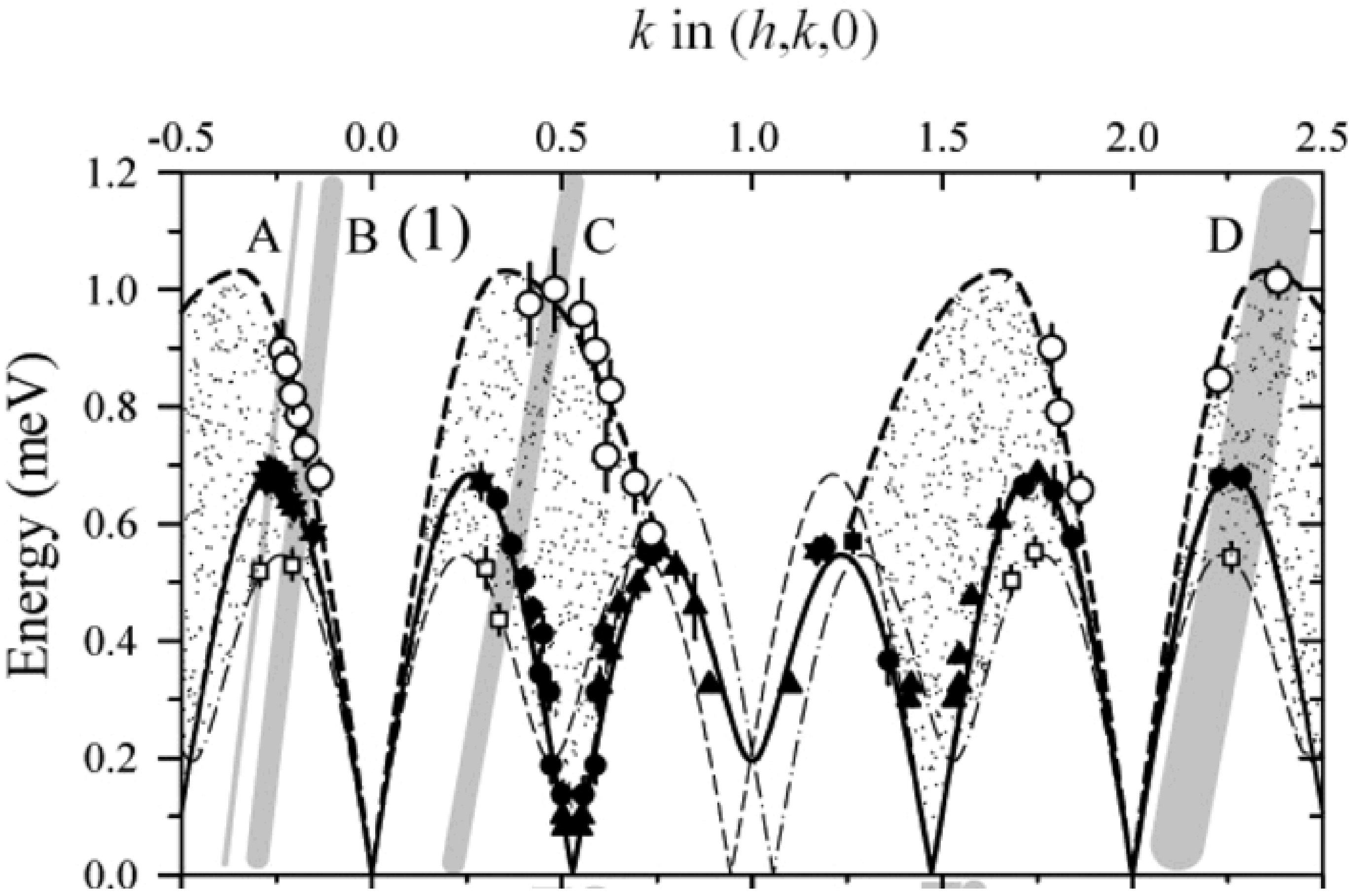




Extended Continuum

Spin Spectral Function

Continuum Scattering in zero-field: Deconfined $S = 1/2$ spinons?



What Makes Cs_2CuCl_4 so Special?

- Very few evidences of spin liquid behavior for D $>$ 1 on triangular lattice
  - ⋆ NMR on Organic Mott Insulator $\kappa-(ET)_2Cu_2(CN)_3$
  - ⋆ $ZnCu_3(OH)_6Cl_2$ on Kagome lattice

- Why conventional spin wave theory fails?
  - ⋆ Ockham's Razor Principle
    - ∗ Long Range Order at T=0 in most of phase diagram
  - ⋆ Strategy: How far can we push spin-wave theory?
    - ∗ Putting back Quantum Mechanics: $\left[S^i, S^j\right] = i\hbar\epsilon^{ijk}S^k$
    - ∗ Quantitative 1/S expansion
    - ∗ Zero point fluctuations: $E_{QM} = \frac{S}{2}\sum_{\mathbf{k}}\hbar\omega_{\mathbf{k}}$

Quantum Fluctuations To leading order in 1/S

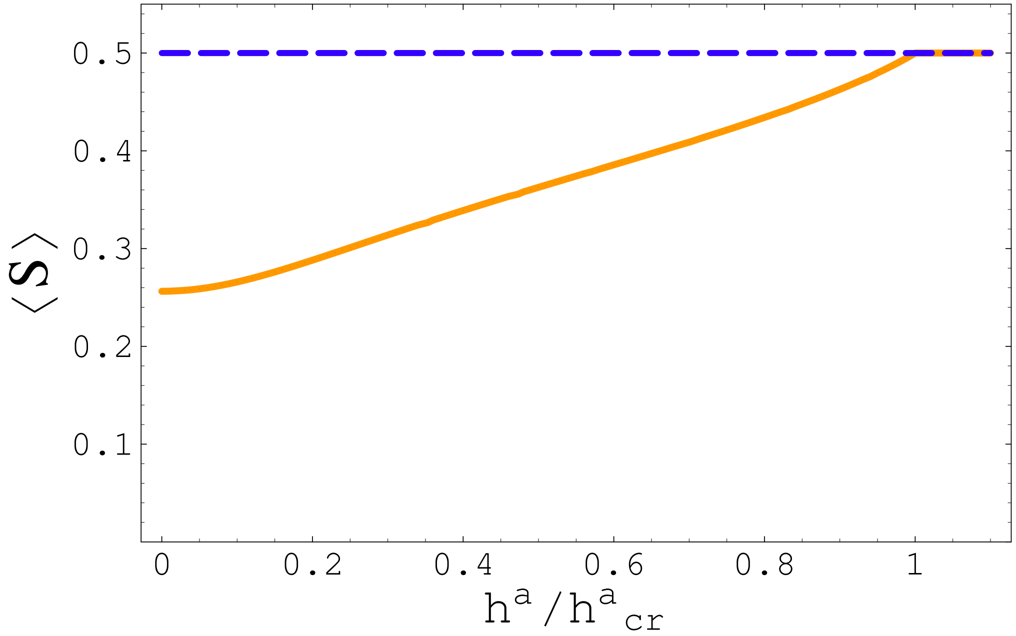
$$E = (S+1)(S)\epsilon_0 + \frac{S}{2}\sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}}.$$

All ground state parameters get renormalized by the quantum fluctuations

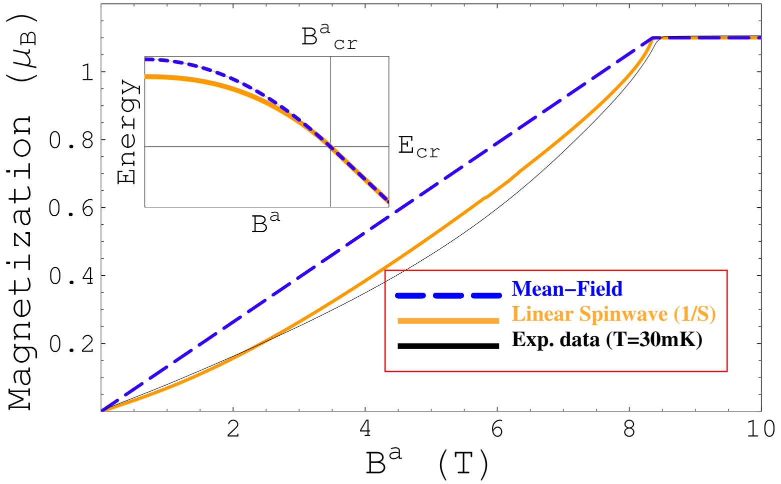
Sublattice
Magnetization

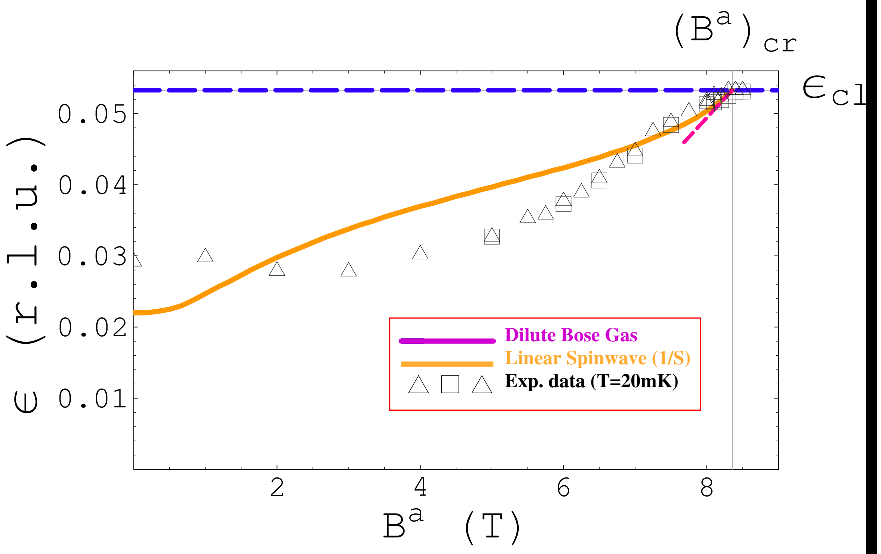
$$\langle S \rangle = S - \frac{1}{N}\sum_{\mathbf{k}} \langle \phi^\dagger_{\mathbf{k}} \phi_{\mathbf{k}} \rangle$$

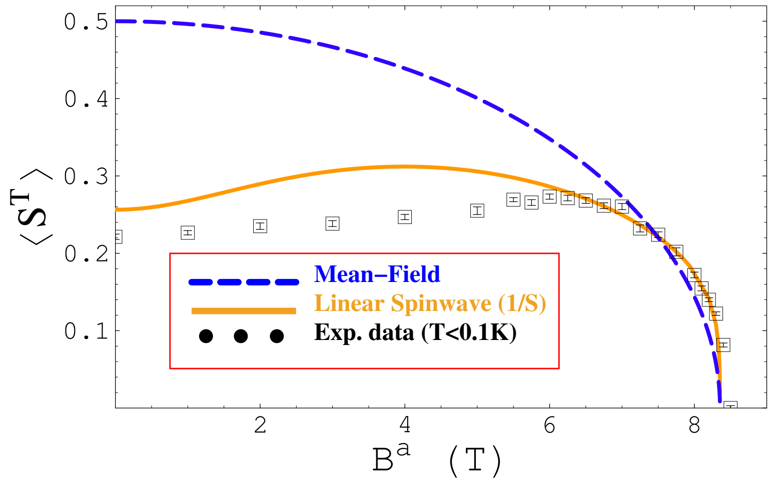
Strong reduction
of ordered moment



Results in Transverse direction





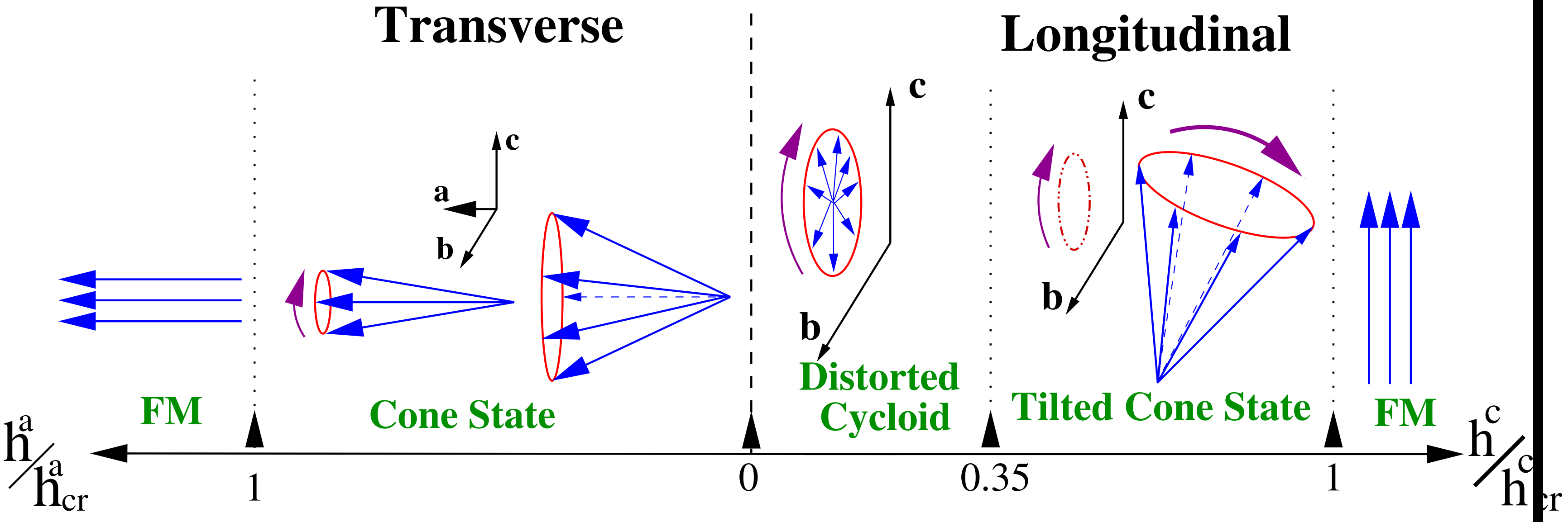


No fitting Parameter!!

Longitudinal Field

More complicated

States not known (analytically) at classical level!!



Inelastic Neutron Scattering

- How to explain large scattering continua within spin wave theory ?

$$S^{\mu\nu}(\mathbf{k},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt \sum_{\mathbf{R}} \langle S^{\mu}_{\mathbf{0}} S^{\nu}_{\mathbf{R}}(t)\rangle e^{+i\omega t - \mathbf{k}\cdot\mathbf{R}}$$

The (unpolarized) inelastic neutron scattering cross section is

$$\frac{d^2\sigma}{d\omega d\Omega} = |f_{\mathbf{k}}|^2 \sum_{\mu\nu} \left(\delta_{\mu\nu} - \hat{\mathbf{k}}_{\mu}\hat{\mathbf{k}}_{\nu}\right) S^{\mu\nu}_{\mathbf{k},\omega},$$

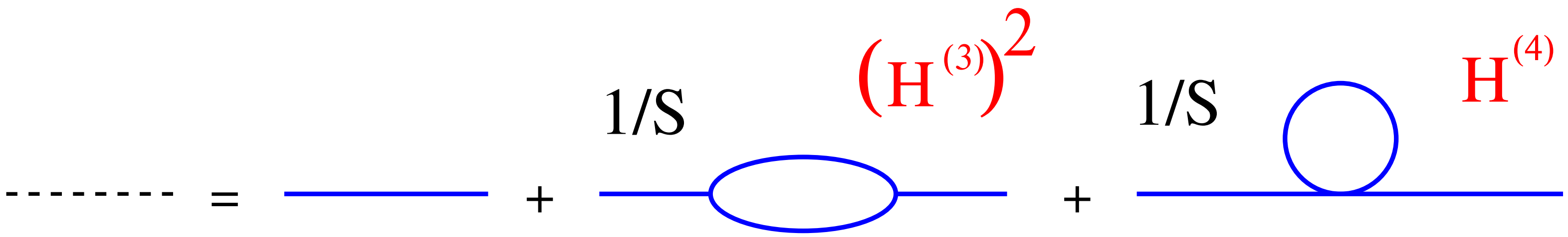
Sum rule for the total scattering per spin= S(S+1)

Three contributions:

- ⋆ $\omega = 0 \rightarrow$ elastic processes: Bragg Peaks $\simeq (S - \Delta S)^2$
- ⋆ $\omega \neq 0$, inelastic processes
  - ∗ one-magnon scattering $\simeq (S - \Delta S)(1 + 2\Delta S)$
  - ∗ two-magnon scattering $\simeq \Delta S(1 + \Delta S)$

Anharmonic terms: Spin Wave Interactions

- $\mathcal{H}_I = \mathcal{H}^{(3)} + \mathcal{H}^{(4)} + \cdots$

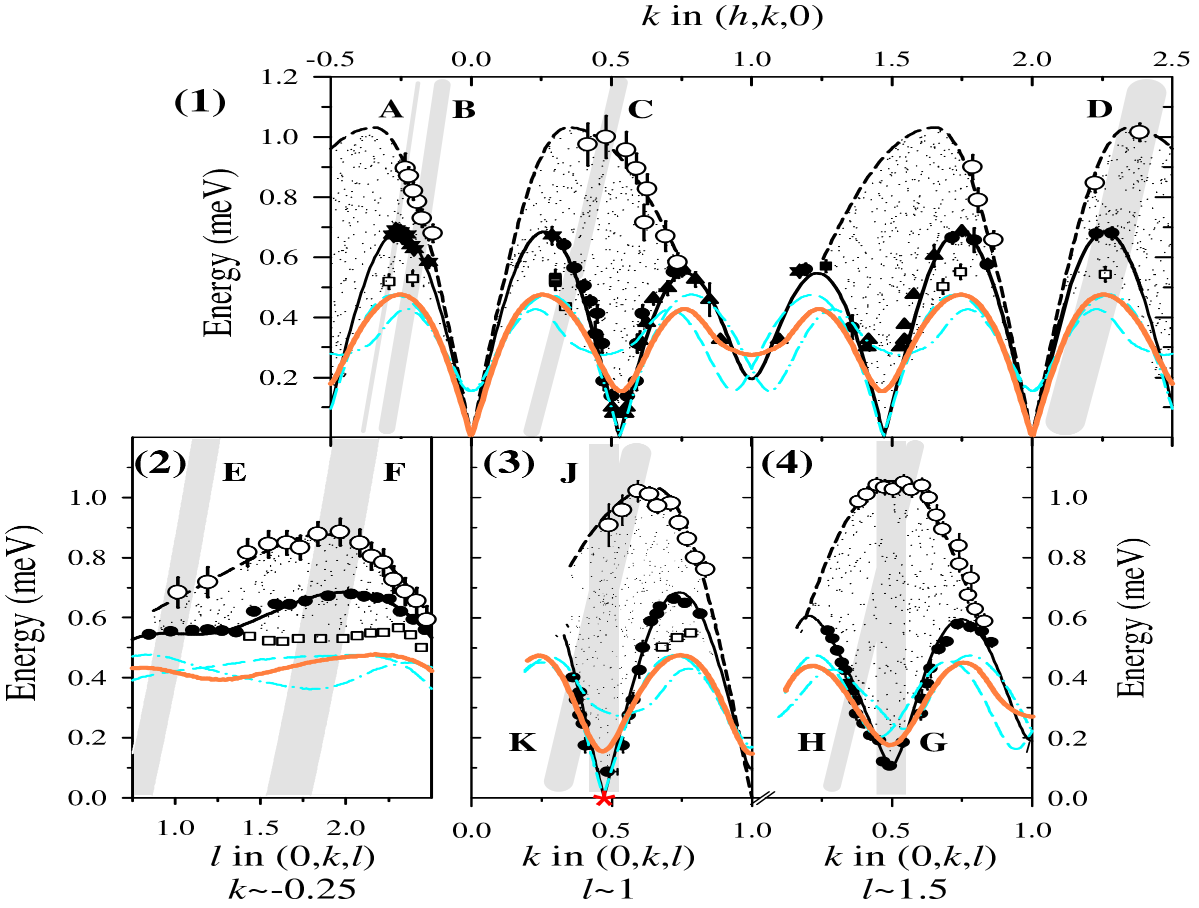


- Strong Interaction
  - ⋆ Low Spin $S = 1/2$
  - ⋆ Frustrated interactions
  - ⋆ Non-collinear order → $\mathcal{H}^{(3)} \neq 0$
    - ∗ Couples longitudinal to transverse spin fluctuations
    - ∗ Frequency dependent diagrams → linear spin waves can decay
    - ∗ Finite linewidth for spin waves

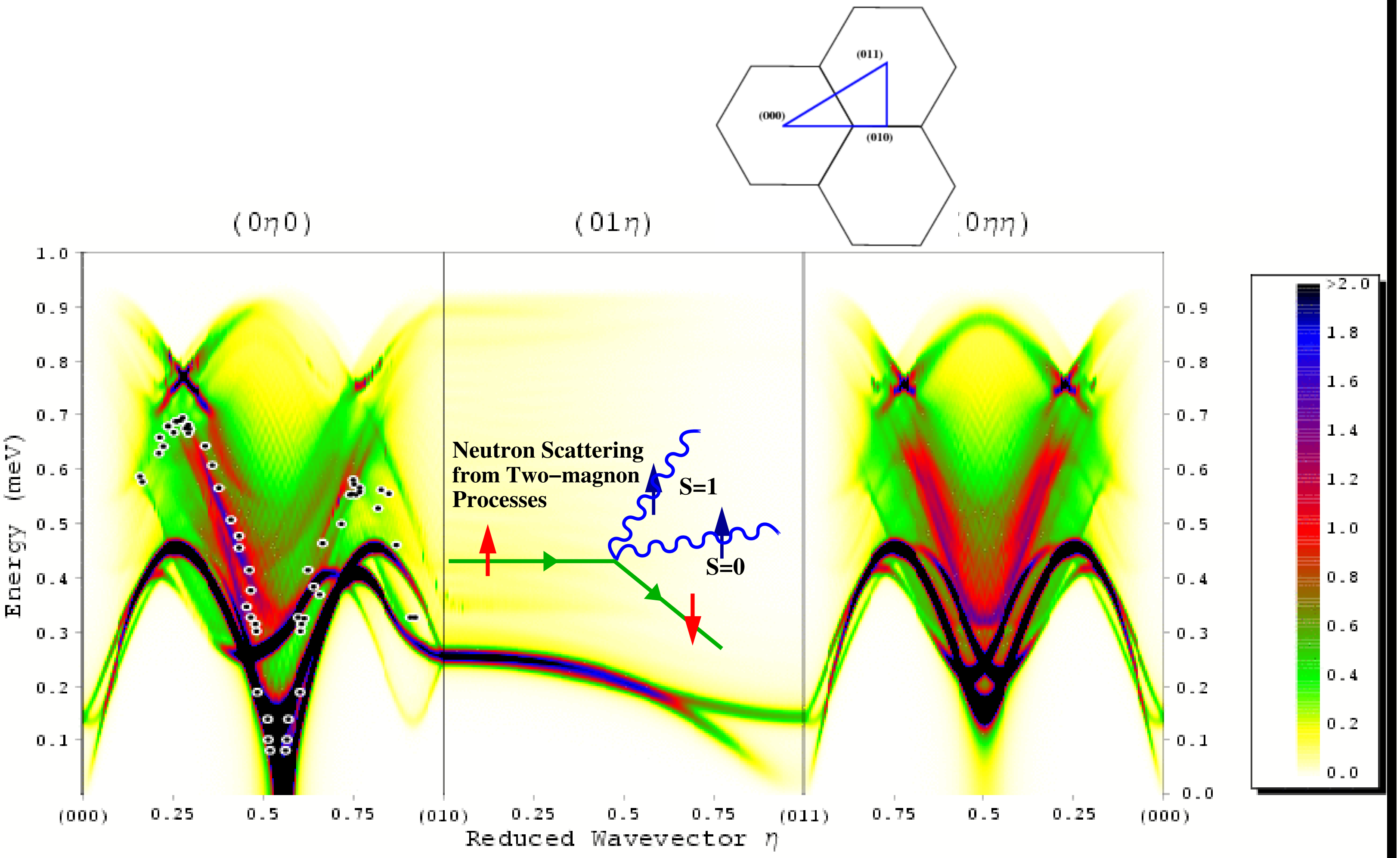
Three Polarizations for Non-Colinear Spin:

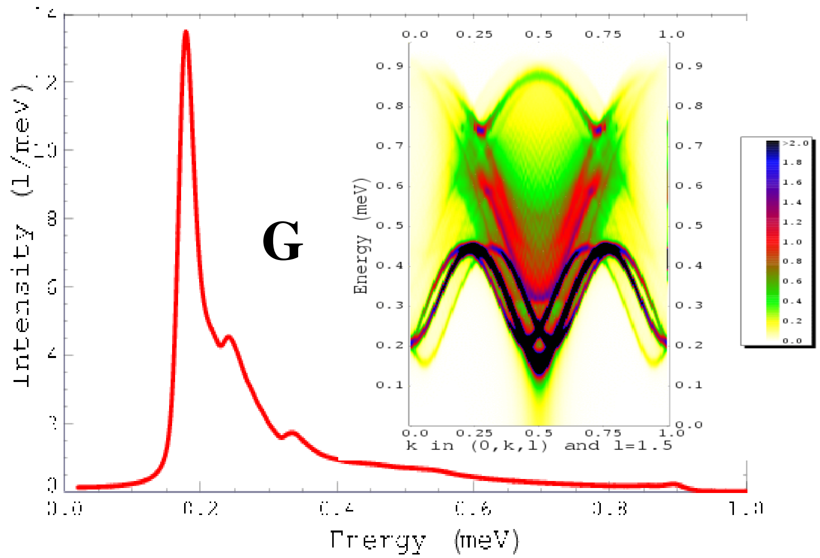
Three Polarizations:Two In-Plane Mode, One Out-of-plane mode.

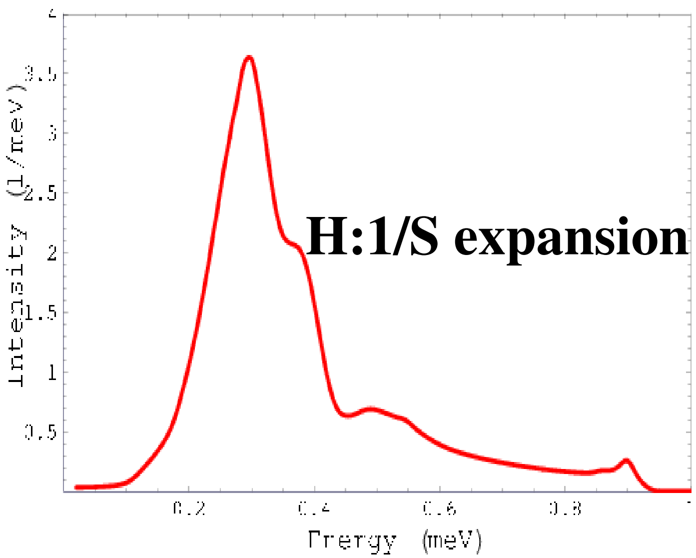
Linear Spin-wave Predictions:

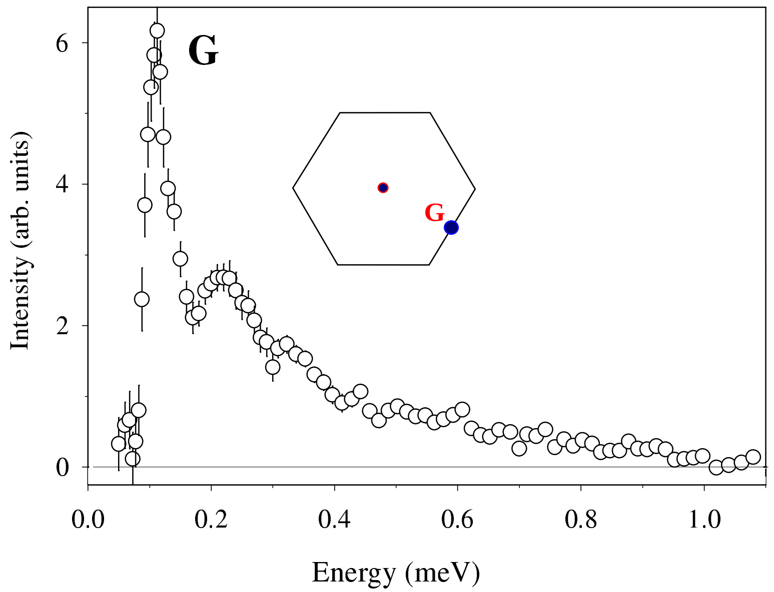


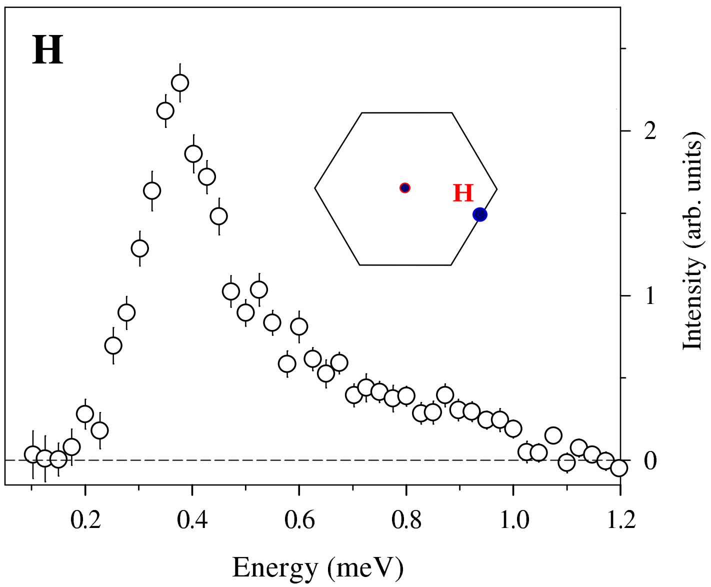
Significant Two-Magnon Scattering for In-plane Fluctuations

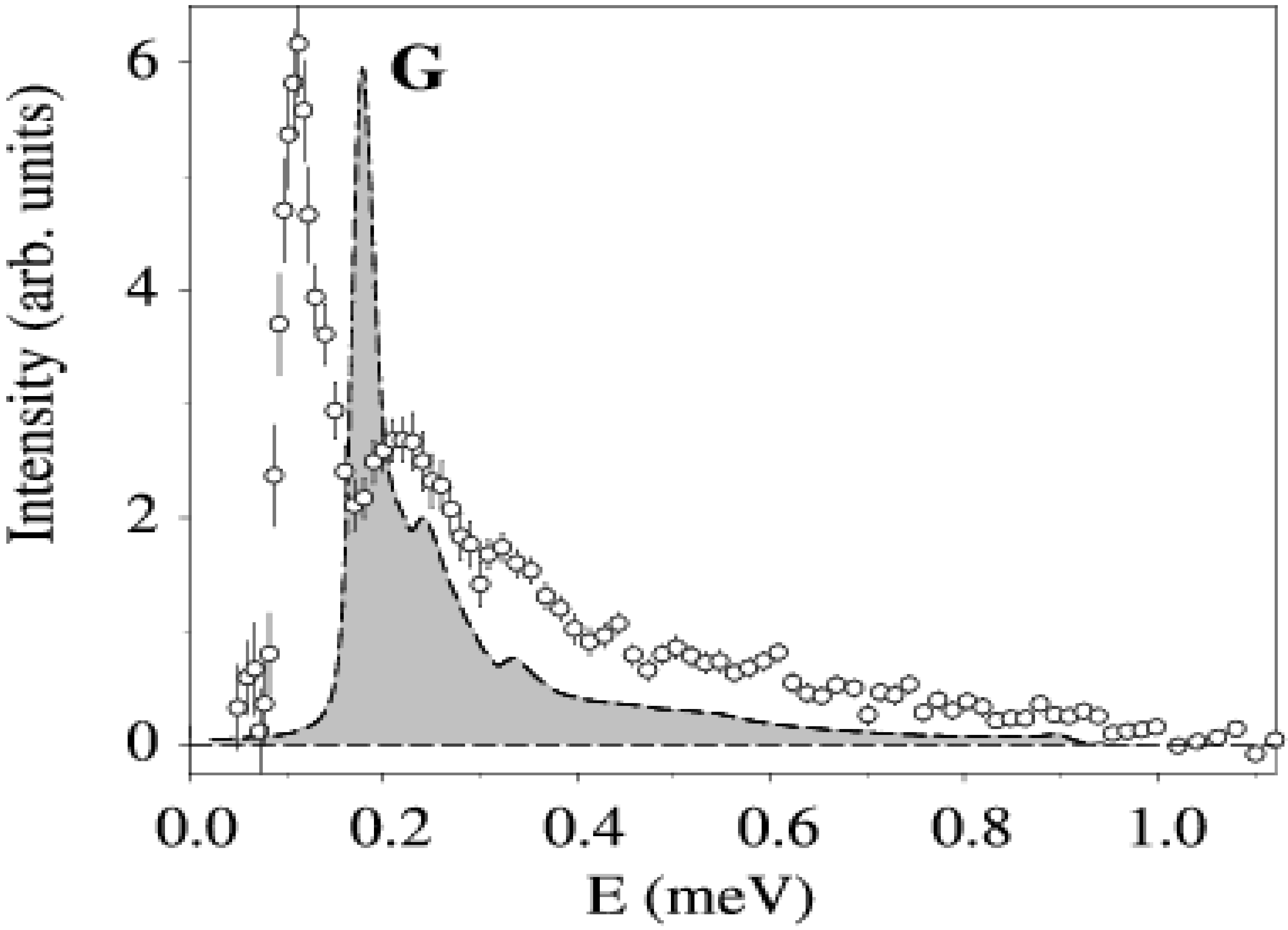



G
Intensity (1/meV)
0
2
4
6
8
10
12
14
0.0
0.2
0.4
0.6
0.8
1.0
Energy (meV)
0.0
0.25
0.5
0.75
1.0
Energy (meV)
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
k in (0,k,1) and l=1.5
>2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0


H:1/S expansion
Intensity (1/meV)
0.5
1
1.5
2
2.5
3
3.5
4
0.2
0.4
0.6
0.8
Energy (meV)


G
G
Intensity (arb. units)
0
2
4
6
0.0
0.2
0.4
0.6
0.8
1.0
Energy (meV)


H
H
Intensity (arb. units)
0
1
2
0.2
0.4
0.6
0.8
1.0
1.2
Energy (meV)


G
Intensity (arb. units)
0
2
4
6
0.0
0.2
0.4
0.6
0.8
1.0
E (meV)

Conclusion

- Phase Diagram: Spin structure dependent on magnetic field orientation
- Dzyaloshinskii-Moriya interaction plays an important role
- $1/S$ expansion in leading order gives good agreement with
    - ⋆ Static Properties: Magnetization, Incommensurate Wavevector, etc..
    - ⋆ Dynamical Spin correlation: Scattering continum, shift of spectral weight
- Cs_2CuCl_4 closer to conventional state of matter than spin liquid ?.

$$\frac{d^2\sigma}{d\omega d\Omega} = |f_{\mathbf{k}}|^2 \left\{ \left(1-\hat{\mathbf{k}}_a^2\right) S^{aa}_{\mathbf{k},\omega} + \left(1+\hat{\mathbf{k}}_a^2\right) S^{bb}_{\mathbf{k},\omega} \right\}. \tag{1}$$

$$S^{aa}_{\mathbf{k},\omega} = -\frac{1}{\pi}\Im\Theta^{0}_{\mathbf{k},\omega}, \tag{2}$$

$$S^{bb}_{\mathbf{k},\omega} = S^{cc}_{\mathbf{k},\omega} = -\frac{1}{\pi}\Im\left[\Theta^{+}_{\mathbf{k}+\mathbf{Q},\omega} + \Theta^{-}_{\mathbf{k}-\mathbf{Q},\omega}\right], \tag{3}$$

$$S^{bc}_{\mathbf{k},\omega} = -S^{cb}_{\mathbf{k},\omega} = -\frac{i}{\pi}\Im\left[\Theta^{+}_{\mathbf{k}+\mathbf{Q},\omega} - \Theta^{-}_{\mathbf{k}-\mathbf{Q},\omega}\right], \tag{4}$$

Magnons in Saturated Field Image the Hamiltonian

- In general ground statea and excitations are non-trivial.
- Beyond B_{cr} → Fully-Polarized state: *Eigenstate* of Hamiltonian, No AF correlations

$$|FM\rangle = |\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\rangle$$

- Excitation in the saturated phase: *Coherent* excitations $\Delta S^z = 1$ of spin-flip states

$$|\phi_{\mathbf{k}}\rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} e^{-i\mathbf{k}\cdot\mathbf{R}} S^-_{\mathbf{R}}$$

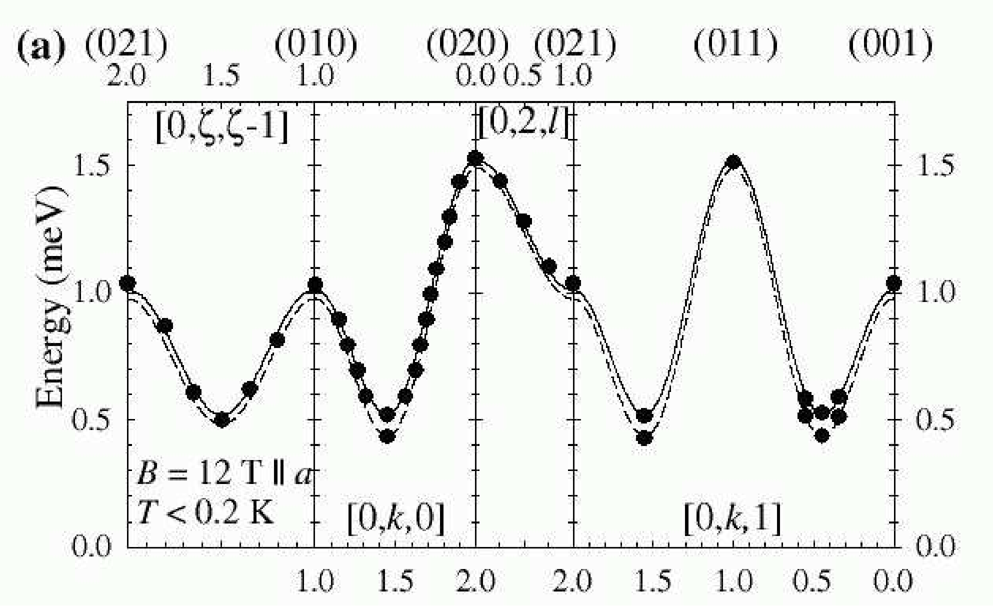
- Magnetization S^z is a good quantum number.
- No Decay, No dressing $\longrightarrow$ No Quantum Fluctuations, $\Sigma(\mathbf{k},\omega) = 0$
- Magnons dispersion $\omega_{\mathbf{k}}$ images the Hamiltonian

The exchange interactions can be read off from the spinwaves

$$\hbar\omega(\mathbf{k}) = J^T_{\mathbf{k}} - J^T_{\mathbf{0}} + g\mu_B B$$

$$J^T_{\mathbf{k}} = \frac{1}{2}\sum_{\delta} J^T_{\delta} \exp(i\mathbf{k}\cdot\delta)$$

$$J^T_{\mathbf{k}} = J_{\mathbf{k}} \pm D_{\mathbf{k}}$$



R. Coldea *et al.*, PRL 88, 137203, (2002)

$$\begin{aligned} J_{\mathbf{k}} &= J\cos(2\pi k) + 2J'\cos(\pi k)\cos(\pi l) + J''\cos(2\pi h) \\ D_{\mathbf{k}} &= 2D\sin(\pi k)\cos(\pi l) \end{aligned}$$

$$J = 0.37(4)\mathrm{meV} \quad , \quad J' = 0.12(8)\mathrm{meV}$$

$$J'' = 0.01(7)\mathrm{meV} \quad , \quad D = 0.02(0)\mathrm{meV}$$

Origin of Dzyaloshinskii-Moriya Interaction

One-electron Hamiltonian

$$\begin{aligned}\mathcal{H}_0 &= \sum_{\mathbf{R}}\sum_{\sigma}\epsilon(\mathbf{R})a^\dagger_\sigma(\mathbf{R})a_\sigma(\mathbf{R}) + \sum_{\mathbf{R}\neq\mathbf{R}'}\sum_{\sigma} b(\mathbf{R}-\mathbf{R}')a^\dagger_\sigma(\mathbf{R})a_\sigma(\mathbf{R}') \\ &+ \sum_{\mathbf{R}\neq\mathbf{R}'}\sum_{\sigma\sigma'} a^\dagger_\sigma(\mathbf{R})\left[\mathbf{C}(\mathbf{R}-\mathbf{R}')\cdot\sigma\right]_{\sigma\sigma'} a_{\sigma'}(\mathbf{R}')\end{aligned}$$

Transfer Integrals

$$\begin{aligned}b(\mathbf{R}'-\mathbf{R}) &= \sum_{\sigma}\int d\mathbf{r}\psi^\star_\sigma(\mathbf{r}-\mathbf{R}')H_1\psi_\sigma(\mathbf{r}-\mathbf{R}) \\ \mathbf{C}(\mathbf{R}'-\mathbf{R}) &= \sum_{\sigma\sigma'}\int d\mathbf{r}\psi^\star_\sigma(\mathbf{r}-\mathbf{R}')\sigma_{\sigma\sigma'}H_1\psi_{\sigma'}(\mathbf{r}-\mathbf{R})\end{aligned}$$

and

$$H_1 = \frac{\mathbf{p}^2}{2m} + V(\mathbf{R}) + \frac{\hbar}{2m^2c^2}\mathbf{S}\cdot\left[\nabla V(\mathbf{R})\times\mathbf{p}\right]$$

Interaction term (Hubbard U)

$$\mathcal{H}_I = U \sum_{\mathbf{R}} \sum_{\sigma\sigma'} a^\dagger_\sigma(\mathbf{R}) a^\dagger_{\sigma'}(\mathbf{R}) a_{\sigma'}(\mathbf{R}) a_\sigma(\mathbf{R})$$

Interaction between spins in second-order perturbation theory

$$E^{(2)}_{\mathbf{R},\mathbf{R}'} = J_{\mathbf{R},\mathbf{R}'} \mathbf{S}(\mathbf{R})\cdot\mathbf{S}(\mathbf{R}') + \mathbf{D}^M_{\mathbf{R},\mathbf{R}'}\cdot\mathbf{S}(\mathbf{R})\times\mathbf{S}(\mathbf{R}') + \mathbf{S}(\mathbf{R}) \overset{\leftrightarrow}{\Gamma}_{\mathbf{R},\mathbf{R}'} \mathbf{S}(\mathbf{R}')$$

$$\begin{aligned}
J_{\mathbf{R},\mathbf{R}'} &= 4/U |b(\mathbf{R}-\mathbf{R}')|^2 \\
\mathbf{D}^M_{\mathbf{R},\mathbf{R}'} &= 4i/U \left[b(\mathbf{R}-\mathbf{R}')\mathbf{C}(\mathbf{R}'-\mathbf{R}) - b(\mathbf{R}'-\mathbf{R})\mathbf{C}(\mathbf{R}-\mathbf{R}') \right] \\
\overset{\leftrightarrow}{\Gamma}_{\mathbf{R},\mathbf{R}'} &= 4/U \left[\mathbf{C}(\mathbf{R}-\mathbf{R}')\mathbf{C}(\mathbf{R}'-\mathbf{R}) + \mathbf{C}(\mathbf{R}'-\mathbf{R})\mathbf{C}(\mathbf{R}-\mathbf{R}') \right. \\
&- \left. \overset{\leftrightarrow}{I}\, \mathbf{C}(\mathbf{R}-\mathbf{R}') \cdot \mathbf{C}(\mathbf{R}'-\mathbf{R}) \right]
\end{aligned}$$

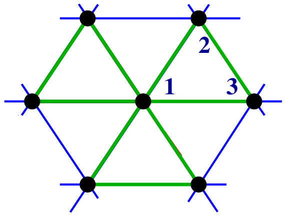
In general

$$\begin{aligned}
b(\mathbf{R}-\mathbf{R}') &= \left[b(\mathbf{R}'-\mathbf{R}) \right]^\star \\
\mathbf{C}(\mathbf{R}-\mathbf{R}') &= \left[\mathbf{C}(\mathbf{R}'-\mathbf{R}) \right]^\star
\end{aligned}$$

Symmetry of Hamiltonian

Spin symmetries

- In Zero-field: $U(1) \times Z_2$
  - ⋆ Spin rotation around $\mathbf{D} \to U(1)$
  - ⋆ Z_2 results from invariance under $\mathbf{R} \to -\mathbf{R}$ and $S^a \to -S^a, S^b \to S^b, S^c \to -S^c$
  - ⋆ $U(1)$ symmetry → Conservation of quantum number S^a



  - ⋆ Chiral scalar associated with Z_2 symmetry $K = \sum_{\triangle} \mathbf{S}_1 \cdot (\mathbf{S}_2 \times \mathbf{S}_3)$
- In transverse field ($B^a \neq 0$): Z_2 broken explicitly, only $U(1)$ left
- In longitudinal field: U(1) broken, only Z_2 left.

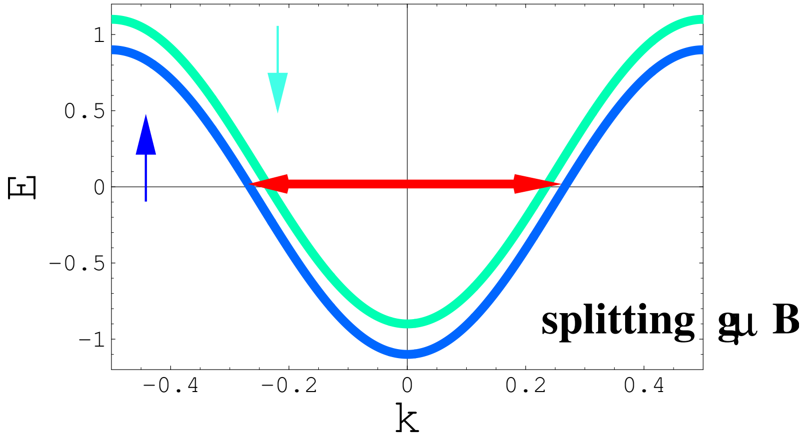
Field-induced incommensuration due to Spinons

$$k_F = 1/2 + \epsilon$$

Incommensuration induced by the applied field

$$\epsilon = \frac{g\mu_B B}{\nu}$$

ϵ can exceed ϵ_{cl}



Band Filling of spinons

Bosonic Hamiltonian

$$
\begin{aligned}
\mathcal{H}_0 &= NS^2\left(J^T_{\mathbf{Q}} - \frac{(h^a)^2}{4\left[J^T_{\mathbf{0}} - J^T_{\mathbf{Q}}\right]}\right), \\
\mathcal{H}_2 &= NSJ^T_{\mathbf{Q}} + \frac{S}{2}\sum_{\mathbf{k}}(A_{\mathbf{k}} + C_{\mathbf{k}})\left(\phi^\dagger_{\mathbf{k}}\phi_{\mathbf{k}} + \phi_{\mathbf{k}}\phi^\dagger_{\mathbf{k}}\right) \\
&\quad -B_{\mathbf{k}}\left(\phi^\dagger_{-\mathbf{k}}\phi^\dagger_{\mathbf{k}} + \phi_{-\mathbf{k}}\phi_{\mathbf{k}}\right),
\end{aligned}
$$

$$
\begin{aligned}
A_{\mathbf{k}} &= \frac{1}{2}\left(2J_{\mathbf{k}} + J^T_{\mathbf{Q}+\mathbf{k}} + J^T_{\mathbf{Q}-\mathbf{k}} - 4J^T_{\mathbf{Q}} + \left[J^T_{\mathbf{Q}+\mathbf{k}} + J^T_{\mathbf{Q}-\mathbf{k}} - 2J_{\mathbf{k}}\right]\frac{(h^a)^2}{(h^a_{cr})^2}\right) \\
B_{\mathbf{k}} &= \frac{1}{2}\left[2J_{\mathbf{k}} - J^T_{\mathbf{Q}+\mathbf{k}} - J^T_{\mathbf{Q}-\mathbf{k}}\right]\left(1 - \left(\frac{h^a}{h^a_{cr}}\right)^2\right), \\
C_{\mathbf{k}} &= \left[J^T_{\mathbf{Q}+\mathbf{k}} - J^T_{\mathbf{Q}-\mathbf{k}}\right]\frac{h^a}{h^a_{cr}}.
\end{aligned}
$$

$$
\mathcal{H}_2 = NSJ^T_{\mathbf{Q}} + S\sum_{\mathbf{k}}\omega_{\mathbf{k}}\left(\gamma^\dagger_{\mathbf{k}}\gamma_{\mathbf{k}} + \frac{1}{2}\right),
$$

where $\omega_{\mathbf{k}} = \sqrt{A^2_{\mathbf{k}} - B^2_{\mathbf{k}}} + C_{\mathbf{k}}$.

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_4 + \cdots \,, \tag{5}$$

where $\mathcal{H}_n$ is proportional to $S^{2-n/2}$ and consists of normal ordered products of n boson operators. The $\mathcal{H}_1$ term is absent, because the ordering wave vector is determined by minimizing the mean-field energy. Linear spin wave theory takes into account only the terms $\mathcal{H}_0$ and $\mathcal{H}_2$ of the expansion. The higher order terms represent interactions between magnons. The leading terms in the expansion are

$$
\begin{aligned}
\mathcal{H}_0 &= NS^2 J^T_{\mathbf{Q}}, \\
\mathcal{H}_2 &= NSJ^T_{\mathbf{Q}} + S\sum_{\mathbf{k}} A_{\mathbf{k}}\left(\phi^\dagger_{\mathbf{k}}\phi_{\mathbf{k}} + \phi_{-\mathbf{k}}\phi^\dagger_{-\mathbf{k}}\right) - B_{\mathbf{k}}\left(\phi^\dagger_{-\mathbf{k}}\phi^\dagger_{\mathbf{k}} + \phi_{-\mathbf{k}}\phi_{\mathbf{k}}\right), \\
\mathcal{H}_3 &= \frac{i}{2}\sqrt{\frac{S}{2N}}\sum_{\mathbf{1},\mathbf{2},\mathbf{3}} \delta_{\mathbf{1+2+3}}\left(C_{\mathbf{1}} + C_{\mathbf{2}}\right)\left(\phi^\dagger_{-\mathbf{3}}\phi_{\mathbf{2}}\phi_{\mathbf{1}} - \phi^\dagger_{\mathbf{1}}\phi^\dagger_{\mathbf{2}}\phi_{-\mathbf{3}}\right), \\
\mathcal{H}_4 &= \frac{1}{4N}\sum_{\mathbf{1},\mathbf{2},\mathbf{3},\mathbf{4}} \Big\{\left[\left(A_{\mathbf{1+3}} + A_{\mathbf{1+4}} + A_{\mathbf{2+3}} + A_{\mathbf{2+4}}\right) - \left(B_{\mathbf{1+3}} + B_{\mathbf{1+4}} + B_{\mathbf{2+3}}\right.\right. \\
&+ \left.\frac{2}{3}\left(B_{\mathbf{2}} + B_{\mathbf{3}} + B_{\mathbf{4}}\right)\left(\phi^\dagger_{\mathbf{1}}\phi_{-\mathbf{2}}\phi_{-\mathbf{3}}\phi_{-\mathbf{4}} + \phi^\dagger_{\mathbf{1}}\phi^\dagger_{\mathbf{2}}\phi^\dagger_{\mathbf{3}}\phi_{-\mathbf{4}}\right)\right\}\delta_{\mathbf{1+2+3+4}}.
\end{aligned}
$$

Theoretical Approaches to Cs_2CuCl_4

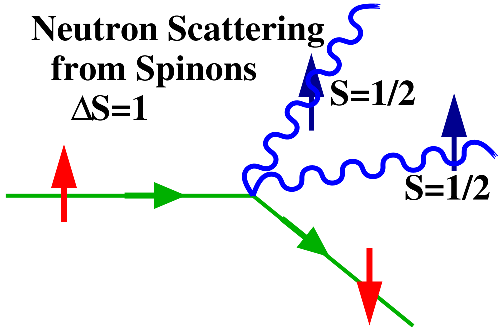
- Bosonic Sp(N) Large-N Mean Field Theory

  C.H. Chung, J.B. Marston and R.H. McKenzie, J. Phys. Cond. Matt. **13**, 5159 (2001)

  - ⋆ Based on SU(2) $\approx$ Sp(1), Expansion in 1/N
  - ⋆ Support Deconfined Spin 1/2 Bosonic Spinons
  - ⋆ Spinons are gapped
- SU(2) Slave-Boson Mean Field Theory Y. Zhou and X.-G Wen, cond-mat/0210662
  - ⋆ Support Deconfined Spin 1/2 Fermionic Spinons
  - ⋆ Spinons are gapless

Spin Spectral function in terms of spinons: *Experimental signature*

C.H. Chung, K. Voelkler and Y.B. Kim, PRB **68**, 094412 (2003)



Transverse vs Longitudinal Field:

Why such a large difference ?

Study the high field region by mapping to a Low Density Bose Gas:

controlled approximation

Transverse Field:

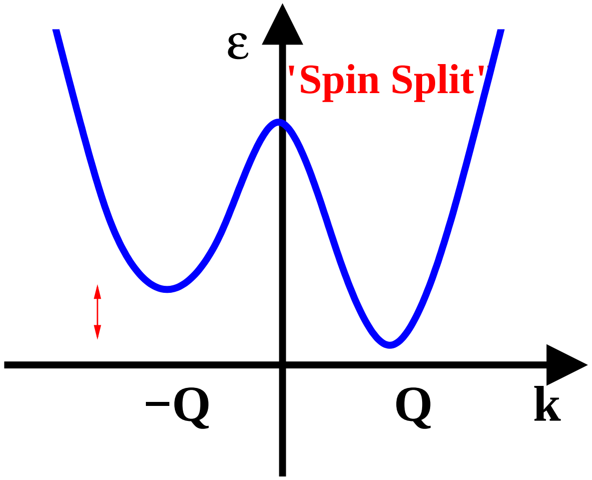
$$S_R^a = 1/2 - \phi_R^\dagger \phi_R, \; S_R^+ = \phi_R, \; S^- = \phi_R^\dagger$$

Hardcore Constraint: $n_R = \phi_R^\dagger \phi_R = \{0, 1\}$

$\epsilon_k - \mu = J_k - J_0 + D_k + B^a$ and $V_k = J_k + U$

$J_k = 1/2 \sum_\delta J_\delta \cos(k \cdot \delta)$

$$\mathcal{H} = (\epsilon_k - \mu)\phi_k^\dagger \phi_k + V_q \phi_{k+q}^\dagger \phi_{k'-q}^\dagger \phi_k \phi_{k'}$$



Degeneracy Lifted by the DM term

RG Language: DM term is relevant.

Low Effective Action ⇒ Scalar Bose Gas

$$\mathcal{H} = \left(\frac{k^2}{2m} - \mu\right) a^\dagger_{\uparrow k} a_{\uparrow k} + \tilde{V}_q a^\dagger_{\uparrow k+q} a^\dagger_{\uparrow k'-q} a_{\uparrow k} a_{\uparrow k'}$$

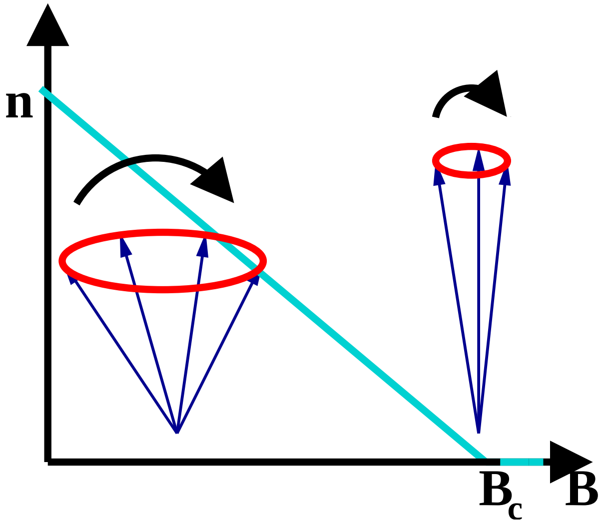
U(1) symmetry → O(2) Spin Rotation

Only Two Types of Order at T=0 M.P.A. Fisher et al. PRB, 1989 **Condensation of Bosons ↔ BEC of magnons**

BEC $\langle a^\dagger_{\uparrow k=0}\rangle = \sqrt{n_{\uparrow 0}}$ for $B^a_{cr} - B^a > 0$

Mean Field: $n_{\uparrow 0} = \frac{(B^a_{cr} - B^a)}{2V_{eff\, k=0}}$

$$\begin{aligned} S^a_R &= 1/2 - n_{\uparrow 0} \\ S^b_R &= \sqrt{n_{\uparrow 0}} \cos(Q \cdot R + \alpha) \\ S^c_R &= \sqrt{n_{\uparrow 0}} \sin(Q \cdot R + \alpha) \end{aligned}$$



Longitudinal Direction: Accessing from High Fields:

$$\begin{aligned}\mathcal{H} &= (\epsilon_{\mathbf{k}} - \mu)\,\phi^\dagger_{\mathbf{k}}\phi_{\mathbf{k}} + \frac{1}{2\sqrt{N}}\left(D_{\mathbf{k},\mathbf{k}'}\phi^\dagger_{\mathbf{k}+\mathbf{k}'}\phi_{\mathbf{k}}\phi_{\mathbf{k}'} + h.c\right) \\ &+ \frac{V_{\mathbf{q}}}{2N}\phi^\dagger_{\mathbf{k}+\mathbf{q}}\phi^\dagger_{\mathbf{k}'-\mathbf{q}}\phi_{\mathbf{k}}\phi_{\mathbf{k}'}\end{aligned}$$

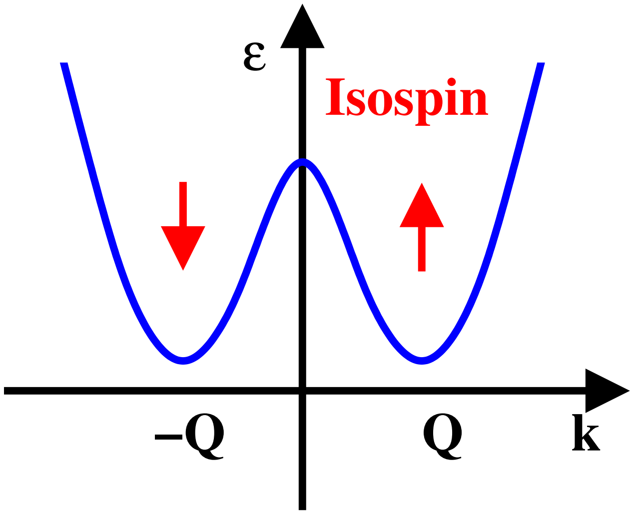
where $\epsilon_{\mathbf{k}} - \mu = J_{\mathbf{k}} - J_0 + h$

Low Energy Description: Spin-1/2 Bose Gas

Degenerate Minima at $\mathbf{k} = \{-\mathbf{Q}, \mathbf{Q}\}$

Z_2 symmetry is present to all order.

$$\begin{aligned}\phi_{\mathbf{k}} &\to -\phi_{-\mathbf{k}} \\ \phi^\dagger_{\mathbf{k}} &\to -\phi^\dagger_{-\mathbf{k}}\end{aligned}$$



Effective Hamiltonian

Slowly fluctuating isospin variables $(a_\uparrow(\mathbf{x}), a_\downarrow(\mathbf{x}))$

$$\phi_{\mathbf{R}} = \left(a_{\uparrow\mathbf{k}} e^{i(\mathbf{k}+\mathbf{Q})\cdot\mathbf{R}} + a_{\downarrow\mathbf{k}} e^{i(\mathbf{k}-\mathbf{Q})\cdot\mathbf{R}}\right) \Theta(\Lambda - |\mathbf{k}|)$$

$$\begin{aligned} \mathcal{H} &= \left(\frac{\mathbf{k}^2}{2m} - \mu\right)\left[a^\dagger_{\downarrow\mathbf{k}} a_{\downarrow\mathbf{k}} + a^\dagger_{\uparrow\mathbf{k}} a_{\uparrow\mathbf{k}}\right] \\ &+ V^0(n_\downarrow + n_\uparrow)^2 + V^z(n_\downarrow n_\uparrow) + g(n_\downarrow + n_\uparrow)\left(a^\dagger_\uparrow a^\dagger_\downarrow + h.c\right) \end{aligned}$$

$V^z > 0$ unlike bosons less repulsive than like Bosons.

Spin 1/2: Internal degree of Freedom

Internal degrees of Freedom yield Richer Structures:

Single particle condensate $|\Psi_{SPC}\rangle = \exp\left(-\phi_\alpha a^\dagger_{\alpha 0}\right)|0\rangle$

$$\begin{aligned}
\langle a_\uparrow\rangle \neq 0 \text{ and } \langle a_\downarrow\rangle = 0 \quad &\longrightarrow \quad \text{Cone state with positive cyclicity} \\
\langle a_\uparrow\rangle = 0 \text{ and } \langle a_\downarrow\rangle \neq 0 \quad &\longrightarrow \quad \text{Cone state with negative cyclicity} \\
\langle a_\uparrow\rangle = -\langle a_\downarrow\rangle \neq 0 \quad &\longrightarrow \quad \text{Spin fan phase}
\end{aligned}$$

All these states have long range order

Proposal: Pair Condensate P. Nozieres et al., J.Phys. 43, 1133 (1982)

$$|\Psi_{PC}\rangle = \exp\left(\sum_{\mathbf{k}} \lambda_{\alpha\beta}(\mathbf{k}) a^\dagger_{\alpha\ \mathbf{k}} a^\dagger_{\beta\ -\mathbf{k}}\right)|0\rangle$$

Only the composite particle has long range order. No long range order in terms of spin-spin correlation function.

Pair Condensate

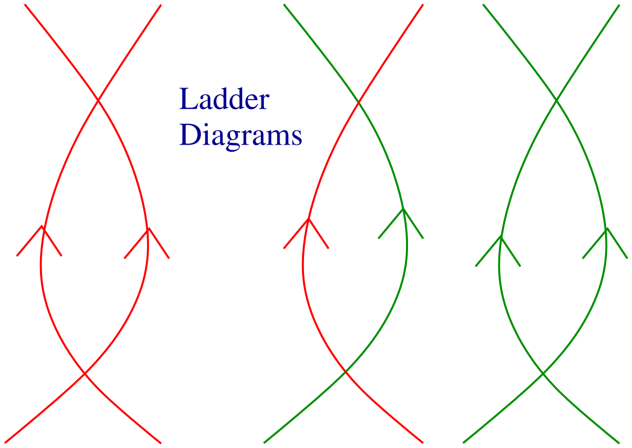
- Favoured when bosons with unlike spins *attract*
  i.e. $V^0 - 2V^z < 0$
  - ⋆ Bare Values: Unlike bosons are repulsive
  - ⋆ Favours cone state (state with LRO)

Large Fluctuations in 2-D, We need Renormalized Values.

Renormalization Group Flow to first loop

At the critical point, namely, T=0, at $\mu = 0$

$$\begin{aligned} \frac{dV^0}{dl} &= -\Gamma\left[(V^0)^2 - (V^z)^2 + 2g^2\right] \\ \frac{dV^z}{dl} &= -\Gamma\left[(V^0)^2 + (V^z)^2 - 2g^2\right] \\ \frac{dg}{dl} &= -\Gamma g\left(V^0 + V^z\right) \end{aligned}$$



In the large l limit:

$$
\begin{aligned}
V^0(0) &\sim \frac{1}{2l} \\
\frac{V^z(l)}{V^0(l)} &= \frac{\sqrt{1-\left(\frac{4g(0)}{V^0(0)}\right)^2}-1}{\sqrt{1-\left(\frac{4g(0)}{V^0(0)}\right)^2}+2} < 0 \\
\frac{g(l)}{V^0(l)+V^z(l)} &= \frac{g(0)}{V^0(0)+V^z(0)}
\end{aligned}
$$

- The renormalized interactions are weaker but do not generate an attraction between $a_\downarrow$ and $a_\uparrow$.
- Pair Condensate is not favoured.
- Conventional Ordering is favoured in high field (cone state)

The boson Green's function at zero temperature is expressed as

$$G_{\mathbf{k},\omega} = -i\int_{-\infty}^{\infty} dt e^{i\omega t}\left\langle T\begin{bmatrix} \phi_{\mathbf{k}}(t) \\ \phi^{\dagger}_{-\mathbf{k}}(t) \end{bmatrix}\left[\phi^{\dagger}_{\mathbf{k}}(0)\phi_{-\mathbf{k}}(0)\right]\right\rangle, \qquad (10)$$

where T stands for the time ordering operator and $\langle\ldots\rangle$ denotes a ground state average. The inverse of the unperturbed Green's function is given by a 2×2 matrix that can be represented in terms of the identity matrix σ^0 and the Pauli matrices σ

$$G^{(0)-1}_{\mathbf{k},\omega} = (-2SA_{\mathbf{k}} + i\eta)\sigma^0 + 2SB_{\mathbf{k}}\sigma^x + \omega\sigma^z, \qquad (11)$$

where $\eta = 0^+$. The self-energy is defined by the Dyson equation,

$$G^{-1}_{\mathbf{k},\omega} = G^{(0)-1}_{\mathbf{k},\omega} - \Sigma_{\mathbf{k},\omega}, \qquad (12)$$

and can be parameterized as

$$\Sigma_{\mathbf{k},\omega} = O_{\mathbf{k},\omega}\sigma^0 + X_{\mathbf{k},\omega}\sigma^x + Z_{\mathbf{k},\omega}\sigma^z. \qquad (13)$$

The leading order (in $1/S$) contributions to the self-energy can be divided into two parts

$$\Sigma_{\mathbf{k},\omega} = \Sigma^{(4)}_{\mathbf{k}} + \Sigma^{(3)}_{\mathbf{k},\omega}. \qquad (14)$$

Here $\Sigma^{(4)}_{\mathbf{k}}$ denotes the vacuum polarization contribution that arises in first order perturbation theory in

$\mathcal{H}_4$. It is frequency independent and purely real. On the other hand, $\Sigma^{(3)}_{\mathbf{k},\omega}$ denotes the contribution in second order perturbation theory in the three-magnon interaction $\mathcal{H}_3$. It incorporates the effects of magnon decay. Using Eq. (11), the $\Sigma^{(4)}$ contribution to the self-energy is found to be of the form

$$\begin{aligned}
O^{(4)}_{\mathbf{k}} &= A_{\mathbf{k}} + \frac{2S}{N}\sum_{\mathbf{k}'}\frac{1}{\omega_{\mathbf{k}'}}\left[\left(\frac{1}{2}B_{\mathbf{k}} + B_{\mathbf{k}'}\right)B_{\mathbf{k}'}\right. \\
&+ \left.\left(A_{\mathbf{k}-\mathbf{k}'} - B_{\mathbf{k}-\mathbf{k}'} - A_{\mathbf{k}'} - A_{\mathbf{k}}\right)A_{\mathbf{k}'}\right], \\
X^{(4)}_{\mathbf{k}} &= -B_{\mathbf{k}} + \frac{2S}{N}\sum_{\mathbf{k}'}\frac{1}{\omega_{\mathbf{k}'}}\left[\left(B_{\mathbf{k}} + B_{\mathbf{k}'}\right)A_{\mathbf{k}'}\right. \\
&+ \left.\left(A_{\mathbf{k}-\mathbf{k}'} - B_{\mathbf{k}-\mathbf{k}'} - A_{\mathbf{k}'} - \frac{1}{2}A_{\mathbf{k}}\right)B_{\mathbf{k}'}\right], \\
Z^{(4)}_{\mathbf{k}} &= 0.
\end{aligned} \qquad (15)$$

The contribution $\Sigma^{(3)}$ is most easily evaluated in the Boguliobov basis (γ) and is equal to

$$
\begin{aligned}
O^{(3)}_{\mathbf{k},\omega} &= \frac{-S}{16N}\sum_{\mathbf{k}'}\left\{\left[\Phi^{(1)}(\mathbf{k}',\mathbf{k}-\mathbf{k}')\right]^2+\left[\Phi^{(2)}(\mathbf{k}',\mathbf{k}-\mathbf{k}')\right]^2\right\}\left[\frac{1}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-\mathbf{k}'}-\omega-i\eta}+\frac{}{\omega_{\mathbf{k}'}}\right.\\
X^{(3)}_{\mathbf{k},\omega} &= \frac{-S}{16N}\sum_{\mathbf{k}'}\left\{\left[\Phi^{(1)}(\mathbf{k}',\mathbf{k}-\mathbf{k}')\right]^2-\left[\Phi^{(2)}(\mathbf{k}',\mathbf{k}-\mathbf{k}')\right]^2\right\}\left[\frac{1}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-\mathbf{k}'}-\omega-i\eta}+\frac{}{\omega_{\mathbf{k}'}}\right.\\
Z^{(3)}_{\mathbf{k},\omega} &= \frac{-S}{16N}\sum_{\mathbf{k}'}\left\{2\Phi^{(1)}(\mathbf{k}',\mathbf{k}-\mathbf{k}')\Phi^{(2)}(\mathbf{k}',\mathbf{k}-\mathbf{k}')\right\}\left[\frac{1}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-\mathbf{k}'}-\omega-i\eta}-\frac{1}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-\mathbf{k}'}}\right.
\end{aligned}
$$

where

$$
\begin{aligned}
\Phi^{(1)}(\mathbf{k}',\mathbf{k}-\mathbf{k}') &= \left(C_{\mathbf{k}'}+C_{\mathbf{k}-\mathbf{k}'}\right)\left(u_{\mathbf{k}'}+v_{\mathbf{k}'}\right)\left(u_{\mathbf{k}-\mathbf{k}'}+v_{\mathbf{k}-\mathbf{k}'}\right)-2C_{\mathbf{k}}\left(u_{\mathbf{k}'}v_{\mathbf{k}-\mathbf{k}'}+v_{\mathbf{k}'}u_{\mathbf{k}-\mathbf{k}'}\right)\\
\Phi^{(2)}(\mathbf{k}',\mathbf{k}-\mathbf{k}') &= C_{\mathbf{k}'}\left(u_{\mathbf{k}'}+v_{\mathbf{k}'}\right)\left(u_{\mathbf{k}-\mathbf{k}'}-v_{\mathbf{k}-\mathbf{k}'}\right)+C_{\mathbf{k}-\mathbf{k}'}\left(u_{\mathbf{k}-\mathbf{k}'}+v_{\mathbf{k}-\mathbf{k}'}\right)\left(u_{\mathbf{k}'}-v_{\mathbf{k}'}\right).
\end{aligned}
$$

1 Dynamical Correlation Function

Inelastic neutron scattering experiments probe the dynamical structure factor $S^{\mu\nu}_{\mathbf{k},\omega}$. The latter is defined as the Fourier transform of the dynamical spin-spin correlation function

$$S^{\mu\nu}_{\mathbf{k},\omega} = \int_{-\infty}^{\infty} \frac{dt}{2\pi\hbar} e^{-i\omega t} \langle S^{\mu}_{-\mathbf{k}}(0) S^{\nu}_{\mathbf{k}}(t) \rangle . \tag{18}$$

Here $\mu, \nu = (a, b, c)$ and the Fourier-transformed spin operators are defined by $S^{\mu}_{\mathbf{k}} = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} S^{\mu}_{\mathbf{R}} e^{-i\mathbf{k}\cdot\mathbf{R}}$.

It is convenient to introduce the time-ordered correlation function in the rotated coordinate system

$$F^{\alpha\beta}_{\mathbf{k},\omega} = -i \int_{-\infty}^{\infty} dt e^{-i\omega t} \langle T S^{\alpha}_{-\mathbf{k}}(0) S^{\beta}_{\mathbf{k}}(t) \rangle , \tag{19}$$

where $\alpha, \beta = (x, y, z)$. The dynamical structure factor is related to the imaginary part of the time ordered correlation function in the following way

$$S^{aa}_{\mathbf{k},\omega} = -\frac{1}{\pi} \Im F^{xx}_{\mathbf{k},\omega} , \tag{20}$$

$$S^{bb}_{\mathbf{k},\omega} = S^{cc}_{\mathbf{k},\omega} = -\frac{1}{\pi} \Im \left[\Theta^{+}_{\mathbf{k}+\mathbf{Q},\omega} + \Theta^{-}_{\mathbf{k}-\mathbf{Q},\omega} \right] , \tag{21}$$

$$S^{bc}_{\mathbf{k},\omega} = -S^{cb}_{\mathbf{k},\omega} = -\frac{i}{\pi} \Im \left[\Theta^{+}_{\mathbf{k}+\mathbf{Q},\omega} - \Theta^{-}_{\mathbf{k}-\mathbf{Q},\omega} \right] , \tag{22}$$

where

$$\Theta^{\pm}_{\mathbf{k},\omega} = \frac{1}{4}\left\{F^{zz}_{\mathbf{k},\omega} + F^{yy}_{\mathbf{k},\omega} \pm i\left(F^{zy}_{\mathbf{k},\omega} - F^{yz}_{\mathbf{k},\omega}\right)\right\}. \tag{23}$$

To proceed further, we expand the dynamical correlation functions up to the first subleading order in $1/S$. The two diagonal parts of the transverse fluctuations are

$$\begin{aligned} F^{xx}_{\mathbf{k},\omega} &= \frac{S}{2}c_x^2 Tr\left[\left(\sigma^0 - \sigma^x\right)G_{\mathbf{k},\omega}\right], \\ F^{yy}_{\mathbf{k},\omega} &= \frac{S}{2}c_y^2 Tr\left[\left(\sigma^0 + \sigma^x\right)G_{\mathbf{k},\omega}\right], \end{aligned} \tag{24}$$

where the Green's function is given by Eq. (12) and where

$$\begin{aligned} c_x &= 1 - \frac{1}{4SN}\sum_{\mathbf{k}}\left(2v_{\mathbf{k}}^2 - u_{\mathbf{k}}v_{\mathbf{k}}\right), \\ c_y &= 1 - \frac{1}{4SN}\sum_{\mathbf{k}}\left(2v_{\mathbf{k}}^2 + u_{\mathbf{k}}v_{\mathbf{k}}\right). \end{aligned} \tag{25}$$

We note that these results are valid only up to the first subleading order in $1/S$. The mixing of transverse and longitudinal fluctuations is expressed as

$$\begin{aligned} i\left(F^{yz}_{\mathbf{k},\omega} - F^{zy}_{\mathbf{k},\omega}\right) &= c_y\left\{P^{(1)}_{\mathbf{k},\omega} Tr\left[\left(1 + \sigma^x\right)G_{\mathbf{k},\omega}\right]\right. \\ &+ \left. P^{(2)}_{\mathbf{k},\omega} Tr\left[\sigma^z G_{\mathbf{k},\omega}\right]\right\}, \end{aligned} \tag{26}$$

where

$$P^{(1)}_{\mathbf{k},\omega} = \frac{S}{4N} \sum_{\mathbf{k}'} \Phi^{(1)}\left(\mathbf{k}',\mathbf{k}-\mathbf{k}'\right)\left(u_{\mathbf{k}'}v_{\mathbf{k}-\mathbf{k}'}+v_{\mathbf{k}'}u_{\mathbf{k}-\mathbf{k}'}\right)\left[\frac{1}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-\mathbf{k}'}-\omega-i\eta} + \frac{}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-}}\right.$$

$$P^{(2)}_{\mathbf{k},\omega} = \frac{S}{4N} \sum_{\mathbf{k}'} \Phi^{(2)}\left(\mathbf{k}',\mathbf{k}-\mathbf{k}'\right)\left(u_{\mathbf{k}'}v_{\mathbf{k}-\mathbf{k}'}+v_{\mathbf{k}'}u_{\mathbf{k}-\mathbf{k}'}\right)\left[\frac{1}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-\mathbf{k}'}-\omega-i\eta} - \frac{}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-}}\right.$$

Finally the longitudinal fluctuations are

$$F^{zz}_{\mathbf{k},\omega} = F^{(0)zz}_{\mathbf{k},\omega} + F^{(1)zz}_{\mathbf{k},\omega}. \tag{29}$$

Here $F^{(0)zz}$ and $F^{(1)zz}$ denote the leading and subleading contributions respectively and are given by

$$F^{(0)zz}_{\mathbf{k},\omega} = -\frac{1}{2N}\sum_{\mathbf{k}'}\left(u_{\mathbf{k}'}v_{\mathbf{k}-\mathbf{k}'}+v_{\mathbf{k}'}u_{\mathbf{k}-\mathbf{k}'}\right)^2\left[\frac{1}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-\mathbf{k}'}-\omega-i\eta}+\frac{1}{\omega_{\mathbf{k}'}+\omega_{\mathbf{k}-\mathbf{k}'}+\omega-i}\right.$$

$$F^{(1)zz}_{\mathbf{k},\omega} = \frac{1}{2S}\left\{\left(P^{(1)}_{\mathbf{k},\omega}\right)^2 Tr\left[\left(\sigma^0+\sigma^x\right)G_{\mathbf{k},\omega}\right]+\left(P^{(2)}_{\mathbf{k},\omega}\right)^2 Tr\left[\left(\sigma^0-\sigma^x\right)G_{\mathbf{k},\omega}\right]+2P^{(1)}_{\mathbf{k},\omega}P^{(2)}_{\mathbf{k},\omega}\right.$$

The (unpolarized) inelastic neutron scattering cross section is

$$\begin{aligned}\frac{d^2\sigma}{d\omega d\Omega} &= |f_{\mathbf{k}}|^2 \sum_{\mu\nu} \left(\delta_{\mu\nu} - \hat{\mathbf{k}}_\mu \hat{\mathbf{k}}_\nu\right) S^{\mu\nu}_{\mathbf{k},\omega}, \\ &= |f_{\mathbf{k}}|^2 \left\{ \left(1 - \hat{\mathbf{k}}_a^2\right) S^{aa}_{\mathbf{k},\omega} + \left(1 + \hat{\mathbf{k}}_a^2\right) S^{bb}_{\mathbf{k},\omega} \right\}. \qquad (32)\end{aligned}$$