Neutron scattering from spinons in the anisotropic triangular quantum magnet Cs$_2$CuCl$_4$

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Collaborators:

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Outline
Explore 2D frustrated quantum magnet Cs₂CuCl₄ with magnetization & neutrons
1. Phase diagram
2. Measurement of Hamiltonian
3. Spin excitations
4. Summary

What are the effects of frustration and quantum fluctuations (S=1/2) in this 2D antiferromagnet?

Crystal structure and magnetism of Cs₂CuCl₄

Layers of S=1/2 Cu²⁺ ions coupled in a triangular geometry

J, J' antiferromagnetic

Low antiferromagnetic superexchange J ~ 4 K => saturation fields ~ 9 T

Large high-purity single crystals grow from solution
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**Magnetic susceptibility**

Local moment Curie-Weiss behaviour at high $T$

Low-field magnetization vs $T$

Magnetic order

Broad peak characteristic of short-range antiferromagnetic correlations

**Phase diagram in applied field**

Incommensurate magnetic Bragg peaks indicate spiral order

Non-collinear spiral promoted by frustration

Perpendicular field stabilizes cone

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Cross-over phase diagram

Magnetization vs T at low field

LRO stabilized by interlayer couplings

Fully-polarized ground state (fluctuations suppressed)

Thermally induced spin flips in the saturated phase lead to exponential decay in M(T)

Magnon dispersion in the saturated phase gives Hamiltonian

- neutrons flip over one spin

\[ H = \sum_{\langle ij \rangle} J_{ij} S_i^+ S_j^- - h \sum_i S_i^z \]

coherent propagation of spin-flip states (if Hamiltonian conservs \( S^z \))

\[ |\varphi_q\rangle = \frac{1}{|N|} \sum_i e^{i \varphi} |i\rangle \]

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Magnon dispersion at saturation

Neutron scattering observe

S=1 magnons

Fourier transform of couplings $J(q)$

$J = 0.374 \text{ meV}$

$J' \approx \frac{1}{3}$

2D Hamiltonian

$J''/J = 4.5(5)$ % interlayer exchange

$D_\alpha/J = 5.3(5)$ % DM anisotropy

Transition to cone order: Bose condensation of magnons

Bragg peaks appear where the gap closes

2nd order gap-closing transition

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**Observe non-mean-field scaling laws**

Order parameter $<S^z>$:

$$<S^z> \sim (B_c - B)^\beta$$

$\beta = 0.5$

Exponent consistent with 2D BEC (3D XY universality)

**Renormalization of ordering wavevector $Q$**

Strong quantum renormalization $\varepsilon_0/\varepsilon_c = 0.56$

At high field could attribute this to magnon interactions in the condensed phase

**Renormalization of incommensuration by fluctuations**

Approximate solutions to the $S=1/2$ quantum problem give:

$$\frac{\varepsilon_0}{\varepsilon_c} = 0.43(1)$$

Series expansions, WeiHong et al (1999)

$= 0.70(3)$ Large-N, Chung et al (2001)

$= 0.56$ observed
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**Phase diagram in applied field**

- **Spin liquid**
- **3D LRO**
- **Critical**
- **Ferromagnetic**

**Incommensurate low-energy dispersion in zero field**

Low-energy dispersion shows incommensurate 2D modulations

- Magnetic Bragg peaks
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Incommensurate high-energy dispersion in zero field

Strong asymmetry and transverse dispersion due to frustration $J'$

Dispersion relations in zero field

Magnetic Bragg peaks

2D incommensurate modulations observed both at low and high energies

Semi-classical velocity for bare $(J,J')$ couplings

⇒ quantum renormalization $R=1.65$

(quantum solution for $1D S=1/2$ chain gives $R=1.57$)

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Excitation lineshapes and breakdown of the spin-wave description

Spin-wave theory for a spiral predicts 3 magnon modes $\Delta S^z = 0, +1, -1$

Even higher-order processes in LSWT cannot explain the dominant continuum scattering

Analysis of the dominant continuum scattering

Dominant continuum scattering at medium-high energies not described by 2-magnons

At very low energies, see a sharp $S=1$ magnon
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### Analysis of the dominant continuum scattering

Continuum scattering at high energies described by a power-law form

\[ S(k, \omega) = |k| \frac{\delta(\omega - \omega_0)}{\omega^2 - \omega_0^2} \]

At very low energies see a sharp $S=1$ magnon

### Excitations in the spin-liquid phase above $T_N$

The sharp $S=1$ magnon disappears above $T_N$

Dispersion maintains incommensurate 2D modulations

Transverse dispersion

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**Continuum of scattering and S=1/2 spinons**

Make analogy with 1D where spin-wave description breaks down

=> power-law correlations and spin-1/2 spinons

KCuF3 prototypical 1D system

- Interchain couplings small $J'/J \approx 5\%$
- unfrustrated, commensurate
- high energies dispersion essentially 1D

- Interchain couplings large $J'/J \approx 34\%$ (2D)
- frustrated
- incommensurate dispersion, shows strong 2D modulations at all energies

**Physical picture of excitations**

$S=1/2$ spinons at all energy scales

$S=1/2$ spinons at medium energies
$S=1$ magnon at low energies

$S=1/2$ Spinons + weak attractive interaction (mean field)

$S=1$ magnon only

Continuum of de-confined $S=1/2$ spinons

Bound state ($S=1$ magnon)
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**Magnetization curve in Cs$_2$CuCl$_4$**

- **Nonlinear shape due to fluctuations**
- **Saturation field 42% higher than 1D case ($J'$=0)**
- **Include 1st order corrections to 1/S spin-wave theory for 2D ($J, J'$)**

**Summary & conclusions**

1. Cs$_2$CuCl$_4$ is spin-1/2 HAF on anisotropic triangular lattice (2D) : $J'/J \sim 1/3$

2. See excitations continua in neutron scattering, as characteristic of deconfined $S=1/2$ spinons

3. Dispersion relations show 2D incommensurate modulations

4. Intensities described by a 2-spinon power-law lineshape

5. Excitations velocity renormalized from classical ($R=1.65$)

6. Observe large incommensuration effects in field on the scale of band filling of $S=1/2$ spinon orbitals