Spinons in Spatially Anisotropic Frustrated Antiferromagnets

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Outline

Introduction

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Dynamical Structure Factor observed in Cs₂CuCl₄

Elementary Excitations in Antiferromagnets



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Experimental Indications of 2D Spin liquids

- Ground state without long-range order:
- · Triangular lattice: κ-(BETT-TTF)₂Cu₂(CN)₃ Solid ³He in 2D
- · Kagome lattice : $ZnCu_3(OH)_6Cl_2$
- ... No signature of long-range order down to *T*<<*J* [ex. *T*~32 mK, *J*~250 K]



Y.Shimizu et al, PRL 91, 107001 (2003).

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Large Tail of $S(k,\omega)$ in Cs₂CuCl₄





Line shape in Rb₂MnF₄ [square lattice, S=5/2 at $\mathbf{k}=(\pi,0.3\pi)$]

> T.Huberman, *et al.*, Phys. Rev. B. **72** 014413 (2005).

Large Tail of $S(k,\omega)$ in Cs₂CuCl₄

Line shape in Cs₂CuCl₄



Dynamical structure factor $S(k, \omega)$ at $k_x = \pi[1]$

• Spinon :

Power-law divergence with a large tail. Magnon :

δ-functional peak with negligible tail.

[1] R.Coldea, et al., Phys. Rev. B 68, 134424 (2003).

Theoretical proposals for 2D spinons

- J.Alicea, O.I.Motrunich & M.P.Fisher: Phys. Rev. Lett. **95**, 247203 (2005).
- S.V.Isakov, T.Senthil & Y.B.Kim: Phys. Rev. B **72**, 174417 (2005).
- Y.Zhou & X.-G.Wen: cond-mat/0210662.
- F.Wang & A.Vishwanath: Phys. Rev. B **74**, 174423 (2006).
- C.-H.Chung, K.Voelker & Y. B. Kim: Phys. Rev. B **68**, 094412 (2003).

$$S(k,\omega) \propto \frac{1}{\left[\omega^2 - \omega_L(k_x)^2\right]^{1-\eta/2}},$$

 $\eta = 0.74 \pm 0.14$ (experimental fit)

2D Dispersion Relation in Cs₂CuCl₄



Spin-wave theory

- M.Y.Veillette, A.J.A.James & F.H.L.Essler: Phys. Rev. B **72**, 134429 (2005).
- D.Dalidovich, R.Sknepnek, A.J.Berlinsky, J.Zhang & C.Kallin: Phys. Rev. B **73**, 184403 (2006).
- R.Coldea, D.A.Tennant & Z.Tylczynski: Phys. Rev. B 68, 134424 (2003).

$$\omega_{k} = \sqrt{\left(J_{k} - J_{Q}\right)\left[\left(J_{k-Q} + J_{k+Q}\right)/2 - J_{Q}\right]},$$

$$J_{k} = \tilde{J}\cos k'_{x} + 2\tilde{J}'\cos\frac{k'_{x}}{2}\cos\frac{k'_{y}}{2},$$

$$\tilde{J} = 0.61(1) \text{meV}, \quad \tilde{J}' = 0.107(10) \text{meV}.$$

c.f. bare couplings

J=0.374(5)meV, *J*'=0.128(5)meV.

Different Types of Line Shapes in Cs₂CuCl₄



R.Coldea, et al., Phys. Rev. B 68, 134424 (2003).

Unusual Features in Cs₂CuCl₄



Interpretation of experimental results is controversial.

Consistent explanation by a controlled approximation basically without fitting parameter

Model



Highly frustrated antiferromagets : $J_1 = J_2 + J_3$.

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Spatially anisotropic triangular lattice:

 $J_{1}=J_{2}, J_{3}=0$



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• Spatially anisotropic triangular lattice: • Spatially anisotropic J_1 - J_2 model:

 $J'_1 = J'_2, J'_3 = 0$

 $J'_{2}=J'_{3}=J'_{1}/2: \text{critical}$ (x-1,y+1)(x,y+1)(x+1,y+1) $J'_{3},J'_{1},J'_{2},J''_{3}$

Decoupled Chain Limit $(J' \rightarrow 0)$

At J'=0, the system is decomposed into decoupled Heisenberg chains.

The S=1/2 Heisenberg chain is exactly solved by the Bethe ansatz.

Ground state : S=0, $E_0=J(-\ln 2+1/4)L$ (L:length). Triplet states : 2-, 4-, ... spinon states

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2-spinon states can be specified by 2 quasi-momenta (k_{x1}, k_{x2}) .

Alternatively, we can specify them by total momentum k_x and excitation energy ε_{kx} .

 $\begin{cases} k_{x} = k_{x1} + k_{x2} \\ \epsilon_{kx} = \pi J \sin[k_{x}/2] \cos[(k_{x1} - k_{x2})/2] \end{cases}$



2-spinon spectrum of Heisenberg chain

Basis

At J'=0, exact ground state and triplet states are given as follows:



 $|0\rangle_{y}$: Ground state of y-th chain.



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In the small J' regime, restricting the Hilbert space to that spanned by this basis, states with momentum k_x and k_y are expressed as

$$\left|\psi(k_{x},k_{y})\right\rangle = \int d\varepsilon_{k_{x}} D_{k_{x}}(\varepsilon_{k_{x}}) c_{k_{x},k_{y}}(\varepsilon_{k_{x}}) \left|k_{x},\varepsilon_{k_{x}},k_{y}\right\rangle, \quad \left|k_{x},\varepsilon_{k_{x}},k_{y}\right\rangle = \frac{1}{\sqrt{L}} \sum_{y} e^{ik_{y}y} \left|k_{x},\varepsilon_{k_{x}},y\right\rangle.$$

Interchain Interaction

Expressing spin operators in k-space, H_1 term is rewritten as follows:

$$H_{1} = \sum_{k_{x},y} J'(k_{x}) S_{k_{x},y} \cdot S_{-k_{x},y+1}, \quad J'(k_{x}) \equiv J'_{1} + J'_{2} e^{ik_{x}} + J'_{3} e^{-ik_{x}}$$

The expectation value of the interaction H_1 by $|\psi(k_x,k_y)\rangle$ has terms like

 $|0\rangle_{y+2} \rightarrow 1$ y+2: $_{y+2}\langle 0 |$ y+1: $_{y+1}\langle k_x, \varepsilon'_{k_x} | S_{k_x, y+1} \qquad |0\rangle_{y+1} \rightarrow \langle k_x, \varepsilon'_{k_x} | S_{k_x, y+1} | 0 \rangle = A^*(k_x, \varepsilon'_{k_x})$ y : $y\langle 0|$ $S_{-k_x,y}|k_x,\varepsilon_x\rangle_y \rightarrow \langle 0|S_{-k_x,y}|k_x,\varepsilon_{k_x}\rangle = A(k_x,\varepsilon_{k_x})$ y. $y-1: _{y-1}\langle 0| \qquad |0\rangle_{y-1} \rightarrow 1$ $A(k_x,\varepsilon_{k_x}) \equiv \langle 0 | S^{\alpha}_{-k_x,y} | k_x,\varepsilon_{k_x} \rangle_{y}$ $(\alpha = x, y \text{ or } z).$ $\langle \psi(k_x,k_y) | H_1 | \psi(k_x,k_y) \rangle$ $=J'(k_x,k_y)\int d\varepsilon_{k_x}\int d\varepsilon'_{k_x}D_{k_x}(\varepsilon_{k_x})D_{k_x}(\varepsilon'_{k_x})c^*_{k_x,k_y}(\varepsilon_{k_x})c_{k_x,k_y}(\varepsilon'_{k_x})A^*(k_x,\varepsilon_{k_x})A(k_x,\varepsilon'_{k_x}),$ where $J'(k_x, k_y) = 2\{J'_1 \cos k_y + J'_2 \cos(k_x + k_y) + J'_3 \cos(k_x - k_y)\}.$

Effective Hamiltonian for Small J'

$$\left\langle \psi(k_x,k_y) \middle| H \middle| \psi(k_x,k_y) \right\rangle = \sum_{i,j} \overline{c}_{k_x,k_y}^* \left(\varepsilon_{k_x}^i \right) H_{i,j}^{(k_x,k_y)} \overline{c}_{k_x,k_y} \left(\varepsilon_{k_x}^j \right), \quad \overline{c}_{k_x,k_y} \left(\varepsilon_{k_x}^j \right) = \sqrt{\frac{k_x}{2M}} c_{k_x,k_y} \left(\varepsilon_{k_x}^j \right).$$

$$H_{i,j}^{(k_x,k_y)} = \left(\varepsilon_{k_x}^i + E_0 L \right) \delta_{i,j} + \frac{k_x}{2M} J'(k_x,k_y) A^*(k_x,\varepsilon_{k_x}^i) A(k_x,\varepsilon_{k_x}^j),$$
where
$$\begin{cases} J'(k_x,k_y) = 2 \left\{ J_1' \cos k_y + J_2' \cos(k_x + k_y) + J_3' \cos(k_x - k_y) \right\} \\ A(k_x,\varepsilon_{k_x}) = \left\langle 0 \middle| S_{-k_x,y}^\alpha \middle| k_x,\varepsilon_{k_x} \right\rangle_y \end{cases}$$
(*M* is the number of points in the ω space.



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$$H_{i,j}^{(k_x,k_y)} = (\varepsilon_{k_x}^i + E_0 L) \delta_{i,j} + \frac{k_x}{2M} J'(k_x,k_y) A^*(k_x,\varepsilon_{k_x}^i) A(k_x,\varepsilon_{k_x}^j),$$

$$\text{where} \begin{cases} J'(k_x,k_y) = 2 \{ J_1' \cos k_y + J_2' \cos(k_x + k_y) + J_3' \cos(k_x - k_y) \} \\ A(k_x,\varepsilon_{k_x}) = \langle 0 | S_{-k_x,y}^\alpha | k_x,\varepsilon_{k_x} \rangle_y \end{cases}$$

$$(M \text{ is the number of points in the } \omega \text{ space.}) \end{cases}$$

Dynamical structure factor $S(k,\omega)$ is obtained as

$$S(k,\omega) \approx \sum_{i} \left| \left\langle \mathbf{G.S.} \left| S_{-k}^{\alpha} \right| i(k) \right\rangle \right|^{2} \delta(\omega - e_{i}(k)),$$

where $\left| \left\langle k_{x}, e_{k_{x},k_{y}}^{i}, k_{y} \right| S_{k_{x},k_{y}}^{\alpha} | \mathbf{G.S.} \right\rangle \right|^{2} \approx \left| \sum_{j} \overline{c}_{k_{x},k_{y}}^{i*} \left(\varepsilon_{k_{x}}^{j} \right) A^{*}(k_{x},\varepsilon_{k_{x}}^{j}) \left\{ 1 - J'(k_{x},k_{y}) \sum_{l} \frac{k_{x}}{2M} \frac{\left| A(-k_{x},\varepsilon_{-k_{x}}^{l}) \right|^{2}}{\varepsilon_{k_{x}}^{j} + \varepsilon_{-k_{x}}^{l}} \right\} \right|^{2}$

Transition Rate in the Heisenberg Chain

The transition rate $M(k,\omega)$ between the ground state and 2-spinon states is exactly obtained for the Heisenberg chain by an algebraic analysis based on the (infinite-dimensional symmetry) quantum group[2]:

$$A(k_x,\varepsilon_{k_x})=\sqrt{M(k_x,\varepsilon_{k_x})},$$

$$M(k_x,\varepsilon_{k_x}) = \left| \left\langle 0 \left| S^{\alpha}_{-k_x} \right| k_x, \varepsilon_{k_x} \right\rangle \right|^2 = \frac{1}{4\pi} \exp\left(-\int_0^\infty dx \frac{\cosh(2x)\cos(xt) - 1}{x\sinh(2x)\cosh(x)} e^x \right),$$

where
$$t = \frac{4}{\pi} \ln \left(\frac{\sqrt{\omega_{U}^{k_{x}^{2}} - \omega_{L}^{k_{x}^{2}}} + \sqrt{\omega_{U}^{k_{x}^{2}} - \varepsilon_{k_{x}^{2}}}}{\sqrt{\varepsilon_{k_{x}^{2}}} - \omega_{L}^{k_{x}^{2}}} \right),$$

 $\omega_{U}^{k_{x}} = \pi J \sin \frac{k_{x}}{2}, \quad \omega_{L}^{k_{x}} = \frac{\pi J}{2} \sin k_{x}.$

[2] A.H. Bougourzi, M. Couture and M. Kacir, PRB 54, R12669 (1996).

Effective Schrödinger Equation for Small J'

Alternatively,

$$\varepsilon_{k_x}\psi_{k_x}(\varepsilon_{k_x})+J'(k)\int d\varepsilon'_{k_x}D(\varepsilon_{k_x})A^*_{k_x}(\varepsilon_{k_x})A_{k_x}(\varepsilon'_{k_x})\psi_{k_x}(\varepsilon'_{k_x})=e_k\psi_{k_x}(\varepsilon_{k_x}).$$



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This equation can be solved analytically, and $S(k,\omega)$ is obtained as

$$S(k,\omega) = \frac{S_{1D}(k_x,\omega)}{\left[1 + J'(k)\chi'_{1D}(k_x,\omega)\right]^2 + \left[\pi J'(k)S_{1D}(k_x,\omega)\right]^2},$$

where
$$\chi'_{1D}(k_x,\omega) = \int_0^\infty d\omega' \frac{S_{1D}(k_x,\omega')}{\omega'-\omega}, \quad S_{1D}(k_x,\omega) = D_{k_x}(\omega) |A_{k_x}(\omega)|^2.$$

This result is similar to RPA: Re $\chi_{1D}(k_x, \omega) = \int_{-\infty}^{\infty} d\omega' \frac{S_{1D}(k_x, |\omega'|)}{\omega' - \omega}$.

Features of $S(k,\omega)$



Interchain interactions effectively vanish. 1D spinons persist:

$$S(k,\omega) \propto \sqrt{\frac{-\ln(\omega - \omega_L(k_x))}{\omega^2 - \omega_L(k_x)^2}}$$

near $\omega \simeq \omega_L(k_x)$.

Spatially anisotropic J_1 - J_2 model with $J'_2=J'_3=J'_1/2=0.24J$

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Bound state of spinons appear below continuum. It is similar to magnon, but does not require ordered ground states. It will be continuously connected to magnon.

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Spectral weight shifts upwards. The peak is broadened in continuum. Anti-bound state appears above the 2-spinon continuum for small k_x .

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Bound State in RPA

The bound state is also obtained by RPA as a pole of dynamical susceptibility:

$$\operatorname{Re}\left[\chi_{2d}^{-1}(k,\omega)\right] = \operatorname{Re}\left[\chi_{1d}^{-1}(k,\omega)\right] + J'(k) = 0$$

From this relation, the energy gap opens as $\Delta E \propto J^2$ with logarithmic



Energy gap ΔE between bound state and lower edge of continuum on a spatially anisotropic triangular lattice at $k=(\pi/4,\pi)$.

: present approach: RPA

Bound State in the Ising Limit In the continuum, spinons are dispersed: $|\uparrow\downarrow\cdots\uparrow\downarrow\uparrow\varsigma\uparrow\downarrow\cdots\uparrow\downarrow\uparrow\varsigma\uparrow\downarrow\cdots\uparrow\downarrow\uparrow\varsigma\uparrow\downarrow\cdots\uparrow\downarrow\uparrow\varsigma\uparrow\downarrow\cdots\uparrow\downarrow\rangle$.



Density-density correlation function $n(\Delta x)$ in the lowest-energy state of the XXZ model in the Ising limit on a spatially anisotropic triangular lattice. (a) Incoherent state at $k_x=0.708\pi$ and (b) bound state at $k_x=0.667\pi$.

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In the continuum, spinons are dispersed: $|\uparrow\downarrow\cdots\uparrow\downarrow\uparrow\diamondsuit\uparrow\downarrow\cdots\uparrow\downarrow\uparrow\diamondsuit\uparrow\downarrow\cdots\uparrow\downarrow\rangle$. In the bound state, spinons are close to each other $(\Delta x \sim 0): |\uparrow\downarrow\cdots\downarrow\uparrow\diamondsuit\uparrow\diamondsuit\uparrow\diamondsuit\uparrow\downarrow\cdots\uparrow\downarrow\rangle$.



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Cs_2CuCl_4

Spatially anisotropic triangular lattice



J=0.374(5)meV, *J*'=0.128(5)meV *J'/J*=0.34(3)

(Interlayer J'' and DM ~ 0.05J)

Cs_2CuCl_4



Unusual Features Observed in Cs₂CuCl₄ by Coldea, et al.



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Strong momentum dependence of line shape

Large Tail at $k'_x = \pi$

At $k'_x = \pi$, $S(k, \omega)$ reduces to that of the Heisenberg chain in the present approach, because J'(k) = 0. $J'(k_x, k_y) = 4J' \cos\left(\frac{k'_x}{2}\right) \cos\left(\frac{k'_y}{2}\right)$.



- : Present result
- : Experimental result of Cs₂CuCl₄ by Coldea, *et al*.[1]

Inset: log-log plot.

There is only one fitting parameter for the overall scale of intensity. [This parameter is inevitable.]

 $S(k,\omega) \propto \sqrt{\frac{-\ln(\omega - \omega_L(k_x))}{\omega^2 - \omega_L(k_x)^2}}$ near $\omega \simeq \omega_L(k_x)$.

Comparison with Power-Law Fit



The experimental data at $k_x = \pi$ have been fitted in a power-law form as

$$S(k,\omega) \propto \frac{1}{\left[\omega^2 - \omega_L(k_x)^2\right]^{1-\eta/2}}$$

where $\omega_L(k_x)$: lower edge of continuum.

 $\eta = 0.74 \pm 0.14$

- : Heisenberg chain (2-spinon)
- : power-law fit with η=0.74

• : experimental data[1]

At $k'_y=0$ and $k'_y=2\pi$, dispersion relation is asymmetric with respect to $k'_x=\pi$. At $k'_y=3\pi$, dispersion relation is symmetric, because $J'(k'_x,k'_y=3\pi)=0$. No fitting parameter



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At $k'_x = -\pi/2$, the sign of J'(k) changes at $k'_y = 3\pi$. No fitting parameter For $k'_y < 3\pi [J'(k) < 0]$, a bound state is formed below the continuum. For $k'_y > 3\pi [J'(k) > 0]$, the peak is broadened in the continuum.



 $S(k,\omega)$ at $k'_x = -\pi/2$ near the lower edge of the continuum by the present method.

$$J'(k_x,k_y) = 4J'\cos\left(\frac{k'_x}{2}\right)\cos\left(\frac{k'_y}{2}\right).$$

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Main peak(♦), upper edge(○) and lower edge(□) estimated experimentally by Coldea, *et al*.[1].

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Bound State and Broad Peak

For J'(k) < 0, $S(k,\omega)$ has a sharp peak of the bound state. No fitting parameter For J'(k) > 0, $S(k,\omega)$ has a broad peak in the continuum.



- -: Present result (2-spinon)
- : Experimental result of Cs₂CuCl₄ by Coldea, *et al*.[1]

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Summary ~ Features in $S(k,\omega)$ ~

Three distinctive features of dynamical structure factor $S(\mathbf{k},\omega)$ are obtained depending on the momentum, which is classified by the sign of $\mathcal{J}'(k)$.

J'(k) = 0: The interchain interactions effectively vanish. Spinons of the Heisenberg chain persist at these momenta.

Bound states of spinons appear below the continuum,

 $J'(\mathbf{k}) < 0$: where a spinon pair moves coherently like a magnon. This bound state does not require ordered ground state This bound state does not require ordered ground states.

Spectral weight shifts upwards.

 $J'(\mathbf{k}) > 0$: The peak is broadened in the continuum. Anti-bound state is observed above 2-spinon continuum.

In the present approach, power-law behaviors appear only at $J'(k_x,k_y)=0$. The exponent $\eta=1$ (1D Heisenberg chain).

Summary ~ Comparison with Experiments ~

Unusual experimental features in Cs₂CuCl₄ have been consistently explained by the present approach basically without fitting parameter.

- Asymmetry of the dispersion relation :
 - Bound state below the continuum for J'(k) < 0.
 - Broad peak in the continuum for J'(k) > 0.
- Momentum dependence of line shape :

Bound states are actually observed experimentally for J'(k) < 0 as a sharp peak.

• Large tail at $k_x = \pi$:

 $S(\mathbf{k},\omega)$ at $J'(\mathbf{k})=0 \xrightarrow{\text{present approximation}} S_{1D}(k_x,\omega)$ (1D Heisenberg chain). Interpretation : descendants of 1D spinons strongly persist at $J'(\mathbf{k})=0$.

Intensity

