Spinon magnetic resonance of two-dimensional U(1) spin liquids with Fermi surface

Oleg Starykh, University of Utah
Outline

• **Main ingredients**
  – spin liquid
  – absence of spin-rotational symmetry: spin-orbit, DM, anisotropy...

• **Line shape**: ESR of two-dimensional spinon continuum $YbMgGaO_4$

• **Line width**: ESR of spinons coupled to gauge field

• Conclusions
The big question(s)

**What is quantum spin liquid?**

No broken symmetries. Quantum entangled state: fractionalized excitations = spinons emergent gauge fields

**Which materials realize it?**

Past candidates: Cs2CuCl4, kagome volborthite…
Current candidates: kagome herbertsmithite, α-RuCl3, organic Mott insulators

**How to detect/observe it?**

Neutrons (if good single crystals are available), RIXS, NMR, thermal transport, terahertz optics, ESR
Organic Mott insulators: Spin liquid with spinon Fermi surface?

M. Yamashita et al, Science 2010


electrical insulator, but metal-like thermal conductor
α-RuCl₃: quantized thermal Hall

Majorana quantization and half-integer thermal quantum Hall effect in a Kitaev spin liquid

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Edge Majorana spinons?!
Mott Physics in Organic Conductors with Triangular Lattices

Kazushi Kanoda
Reizo Kato

10.30 am - 11.00 am: Bella Lake
Exploration of the quantum spin liquid state in Ca$_{10}$Cr$_7$O$_{28}$.

2.00 pm - 2.30 pm: Philippe Mendels
Low-$T$ $^{17}$O NMR study of herbertsmithite crystals.

9.30 am -10.00 am: Liang Wu
Antiferromagnetic resonance and terahertz continuum in $\alpha$-RuCl$_3$

10.30 am -11.00 am: Arnab Banerjee (invited)
Magnetic disorder, order and models of $\alpha$-RuCl$_3$

6.00 pm -6.30 pm: Yasuhiro Shimizu
Quantum criticality of Kitaev spin liquid

9.00 am - 9.30 am: Yuji Matsuda (invited)
Thermal Hall effect in a Kitaev spin liquid: A possible signature of Majorana chiral edge current
Spinon
Magnetic Resonance
Electron Spin Resonance (ESR)

ESR measures absorption of electromagnetic radiation by a sample that is (typically) subjected to an external static magnetic field.

Linear response theory:

$$I(q = 0, \omega) = \frac{1}{2} |h|^2 \omega \text{Im} \chi_{\alpha\beta}(q = 0, \omega)$$

$$\chi_{\alpha\beta}(q = 0, \omega) = i \int_0^\infty dt \langle [S^\alpha(t), S^\beta(0)] \rangle e^{i\omega t}$$

For SU(2) invariant systems, completely sharp:

$$I(\omega) = \frac{1}{2} |h|^2 \omega \delta(\omega - B)$$

No matter how exotic the ground state is!

The key point

• Perturbations violating SU(2) symmetry do show up in ESR: line shift and line width!

• turn annoying material “imperfections” (spin-orbit, Dzyaloshinskii-Moriya) into a probe of exotic spin state and its excitations

Condensed matter physics in 21 century: the age of spin-orbit
✓ spintronics
✓ topological insulators, Majorana fermions
✓ Kitaev’s non-abelian honeycomb spin liquid
Probing spinon continuum in one dimension

\begin{align*}
I(\omega) & \propto \delta(\omega - H) \\
\end{align*}

Dender et al, PRL 1997
Uniform Dzyaloshinskii-Moriya interaction

\[ H = \sum_{x,y,z} JS_{x,y,z} \cdot S_{x+1,y,z} - D_{y,z} \cdot S_{x,y,z} \times S_{x+1,y,z} - g\muBH \cdot S_{x,y,z} \]

- Removes DM term from the Hamiltonian (to D² accuracy)
- **Boosts** momentum to \( D/(Ja_0) \)

Unitary rotation about z-axis

\[ S^+(x) \rightarrow S^+(x)e^{i(D/J)x}, S^z(x) \rightarrow S^z(x) \]

\[ q = 0 \rightarrow q = D/(Ja_0) \Rightarrow 2\pi\hbar \nu_{R/L} = g\muBH \pm \pi D/2 \]

\( \nu_{R/L} \):

- \( D=0 \)
- \( D\neq0 \)

\(-2g\muBH/\piJa_0 \quad H \quad 0 \quad +2g\muBH/\piJa_0 \quad H \)

DM interaction allows to probe spinon continuum at finite “boost” momentum
Cs2CuCl4 ESR data

- General orientation of $\mathbf{H}$ and $\mathbf{D}$
- 4 sites/chains in unit cell

\[ (2\pi\hbar \nu_R)^2 = (g_b \mu_B H_b)^2 + [g_a \mu_B H_a + (-1)^z \pi D_a/2]^2 + [g_c \mu_B H_c + (-1)^y \pi D_c/2]^2, \]

\[ (2\pi\hbar \nu_L)^2 = (g_b \mu_B H_b)^2 + [g_a \mu_B H_a - (-1)^z \pi D_a/2]^2 + [g_c \mu_B H_c - (-1)^y \pi D_c/2]^2. \]

- for $\mathbf{H}$ along b-axis only: the “gap” is determined by the DM interaction strength

\[ \Delta = \frac{\pi}{2} \sqrt{D_a^2 + D_c^2} \rightarrow (2\pi\hbar) 13.6 \text{ GHz} \]

Linear in $T$ line width

2D spin liquid: YbMgGaO$_4$

Gapless quantum spin liquid ground state in the two-dimensional spin-1/2 triangular antiferromagnet YbMgGaO$_4$.

Li Y$^1$, Liao H$^2$, Zhang Z$^3$, Li S$^3$, Jin F$^1$, Ling L$^4$, Zhang L$^4$, Zou Y$^4$, Pi L$^4$, Yang Z$^5$, Wang J$^6$, Wu Z$^7$, Zhang Q$^1, 8$.

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Figure from: Nature 540, pp 559–562 (2016).

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Rare-Earth Triangular Lattice Spin Liquid: A Single-Crystal Study of YbMgGaO$_4$

Yuesheng Li, Gang Cher, Wei Tong, Li Pi, Juanjuan Liu, Zhaorong Yang, Xiaoqun Wang, and Qingming Zhang

Phys. Rev. Lett. 115, 167203 – Published 16 October 2015

Muon Spin Relaxation Evidence for the U(1) Quantum Spin-Liquid Ground State in the Triangular Antiferromagnet YbMgGaO$_4$

Yuesheng Li, Dewashibhai Adroja, Pabitra K. Biswas, Peter J. Baker, Qian Zhang, Juanjuan Liu, Alexander A. Tsirlin, Philipp Gegenwart, and Qingming Zhang


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Strong spin-orbit coupling

\[
\mathcal{H} = \sum_{\langle i,j \rangle} [J_{1z}^{zz} S_i^z S_j^z + J_{1}^{zz} (S_i^+ S_j^- + S_i^- S_j^+)] \\
+ J_{1}^{\pm\pm} (\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij} S_i^- S_j^-) \\
- \frac{iJ_{1}^{zz}}{2} (\gamma_{ij} S_i^+ S_j^- - \gamma_{ij} S_i^- S_j^+) + \langle i \leftrightarrow j \rangle] \\
+ \sum_{\langle i,j \rangle} [J_{2z}^{zz} S_i^z S_j^z + J_{2}^{zz} (S_i^+ S_j^- + S_i^- S_j^+)] \\
- \mu_0 B \sum_{i} \left[ g_\perp (H^x S_i^x + H^y S_i^y) + g_\parallel H^z S_i^z \right]
\]
Evidence for a spinon Fermi surface in a triangular-lattice quantum-spin-liquid candidate

Continuous excitations of the triangular-lattice quantum spin liquid YbMgGaO₄

Spinon continuum?

Sign of spinon Fermi surface?

Broad signal in polarized phase?

very disordered
Spinon hypothesis

Spinon mean-field Hamiltonian derived with the help of Projective Symmetry Group (PSG) analysis.

Basic idea: physical spin $S$ is bilinear of spinons $f$, spinons have a bigger symmetry group than spins, which leads to gauge freedom and different classes of possible mean-fields. These classes describe the same spin problem.

Spinon hypothesis

Dirac spectrum!
Spinon mean-field Hamiltonian derived with the help of Projective Symmetry Group (PSG) analysis

**Basic idea:** physical spin $S$ is bilinear of spinons $f$, spinons have bigger symmetry group than spins, this leads to gauge freedom and different classes of possible mean-fields. These classes describe the same spin problem.
Mean-field Hamiltonians

<table>
<thead>
<tr>
<th>Symmetry</th>
<th>Transformation Rules</th>
</tr>
</thead>
</table>
| $T_1$    | $f(x,y)^\uparrow \rightarrow f(x+1,y)^\uparrow$  
           | $f(x,y)^\downarrow \rightarrow f(x+1,y)^\downarrow$ |
| $T_2$    | $f(x,y)^\uparrow \rightarrow f(x,y+1)^\uparrow$  
           | $f(x,y)^\downarrow \rightarrow f(x,y+1)^\downarrow$ |
| $C_2$    | $f(x,y)^\uparrow \rightarrow e^{i\pi/6}f(y,x)^\uparrow$  
           | $f(x,y)^\downarrow \rightarrow e^{-i\pi/6}f(y,x)^\downarrow$ |
| $\bar{C}_6$ | $f(x,y)^\uparrow \rightarrow e^{i\pi/3}f(x-y,x)^\uparrow$  
                      | $f(x,y)^\downarrow \rightarrow -e^{-i\pi/3}f(x-y,x)^\uparrow$ |
| $\mathcal{T}$ | $f(x,y)^\uparrow \rightarrow f(x,y)^\downarrow$  
                         | $f(x,y)^\downarrow \rightarrow -f(x,y)^\uparrow$ |

TABLE I. U1A11 PSG analysis.

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| $T_2$    | $f(x,y)^\uparrow \rightarrow f(x,y+1)^\uparrow$  
           | $f(x,y)^\downarrow \rightarrow f(x,y+1)^\downarrow$ |
| $C_2$    | $f(x,y)^\uparrow \rightarrow -e^{i\pi/6}f(y,x)^\downarrow$  
           | $f(x,y)^\downarrow \rightarrow e^{-i\pi/6}f(y,x)^\uparrow$ |
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                      | $f(x,y)^\downarrow \rightarrow -e^{-i\pi/3}f(x-y,x)^\uparrow$ |
| $\mathcal{T}$ | $f(x,y)^\uparrow \rightarrow f(x,y)^\downarrow$  
                         | $f(x,y)^\downarrow \rightarrow -f(x,y)^\uparrow$ |

TABLE II. U1A01 PSG analysis.

Eight types: U1A00, U1A01, U1A10, U1A11; U1Bxx
SU(2) trivial π-flux

Accidental Symmetry = ideal ESR

$H = \sum_{x,y} \left\{ t_1 \left[i f_{(x+1,y)}^\dagger f_{(x,y)}^\uparrow + i f_{(x,y-1)}^\dagger f_{(x,y)}^\downarrow - i f_{(x,y-1)}^\dagger f_{(x,y)}^\uparrow - i f_{(x+1,y)}^\dagger f_{(x,y)}^\downarrow + i f_{(x+1,y)}^\dagger f_{(x,y)}^\downarrow \right] + t'_1 \left[e^{i\pi/3} f_{(x+1,y)}^\dagger f_{(x,y)}^\uparrow - f_{(x,y-1)}^\dagger f_{(x,y)}^\downarrow + e^{i\pi/3} f_{(x+1,y)}^\dagger f_{(x,y)}^\downarrow + f_{(x,y-1)}^\dagger f_{(x,y)}^\downarrow + e^{i\pi/3} f_{(x+1,y)}^\dagger f_{(x,y)}^\downarrow + f_{(x,y-1)}^\dagger f_{(x,y)}^\downarrow \right] + t'_2 \left[e^{i2\pi/3} f_{(x+1,y-1)}^\dagger f_{(x,y)}^\downarrow + e^{i2\pi/3} f_{(x+1,y-1)}^\dagger f_{(x,y)}^\downarrow + i f_{(x-2,y-1)}^\dagger f_{(x,y)}^\downarrow + e^{i2\pi/3} f_{(x+1,y+2)}^\dagger f_{(x,y)}^\downarrow + e^{i2\pi/3} f_{(x+1,y+2)}^\dagger f_{(x,y)}^\downarrow \right] + h.c. \right\}$

FIG. S-5. Spinon dispersions $E_{1,2}(k)$ along the line $\Gamma$-$K$-$M$-$K$-$\Gamma$ for U1A11 and U1A01 states.

Calculation of parameters - Iaconis et al, 2018
Spinon magnetic resonance (low T)

AC magnetic field couples to the total spin

$$S_r^a = \frac{1}{2} f_{\alpha}^r \sigma_\alpha^a f_{r \beta}$$

$$V(t) = he^{-i\omega t} \mathbf{n} \cdot \frac{1}{2} \sum_r (f_{\uparrow r}, f_{\downarrow r}) \sigma \left( \frac{f_{\uparrow r}}{f_{\downarrow r}} \right)$$

Rate of energy absorption

$$I(\omega) = -\omega \chi''_{nn}(\omega) |\hbar|^2 / 2$$

Dynamic susceptibility at $q=0$

$$\chi_{nn}(\omega) = \frac{1}{4N} \sum_k \frac{n_{k\alpha} - n_{k\beta}}{\omega + E_\alpha(k) - E_\beta(k) + i0}$$

$$\times (U_k^+ \sigma^a U_k)_{\alpha\beta} (U_k^+ \sigma^b U_k)_{\beta\alpha} \hat{n}^a \hat{n}^b$$

Absorption without external static field!

van Hove singularities

$\theta = \pi/2$

$\theta = \pi/4$

$\theta = 0$
Additional extremum in the spinon spectrum due to symmetry-enforced Dirac touching at K point.

Absorption without external static field!

With magnetic field along Z

\[ \theta = \pi/4 \]
\[ \theta = \pi/2 \]
\[ \theta = 0 \]

Spinon Fermi surface state, accidental SU(2) threshold frequency is determined by \( B_z \)

\[ \sin^2 \theta \delta(\omega - B_z) \]

U1A00

U1A11

additional singularity
Existing ESR in YbMgGaO₄


Minimum temperature: 1.8 K

Lower the temperature to see the spinon effect!

T ~ 0.1 K
Organic Mott insulators: Spin liquid with spinon Fermi surface?

**Spin liquid?**

M. Yamashita et al, Science 2010

Spin-orbit interaction is present in closely related materials

**EtMe₃Sb[Pd(dmit)₂]₂ (dmit-131)**

**Et₂Me₂Sb[Pd(dmit)₂]₂ (dmit-221)**

Non-magnetic charge-ordered
Organic Mott insulators: Spin liquid with spinon Fermi surface?

M. Yamashita et al, Science 2010

Spin liquid?

Spin-orbit interaction is present in closely related materials

Non-magnetic charge-ordered

EtMe₃Sb[Pd(dmit)₂]₂ (dmit-131)

Et₂Me₂Sb[Pd(dmit)₂]₂ (dmit-221)

Importance of spin-orbit coupling in layered organic salts

We investigate the spin-orbit coupling (SOC) effects in α- and κ-phase BEDT-TTF and BEDT-TSF organic salts. Contrary to the assumption that SOC in organics is negligible due to light C, S, and H atoms, we show the relevance of such an interaction in a few representative cases. In the weakly correlated regime, SOC manifests primarily in the opening of energy gaps at degenerate bare touching points. This effect becomes especially important for Dirac semimetals such as α-(ET)₂Ir. Furthermore, in the magnetic insulating phase, SOC results in additional anisotropic exchange interactions, which provide a compelling explanation for the puzzling field-induced behavior of the quantum spin-liquid candidate κ-(ET)₂Cu₂(CN)₃. We conclude by discussing the importance of SOC for the description of low-energy properties in organics.
Linewidth at (relatively) high T

- Spinon band structure determines line shape of absorption (discussed previously).

- Interactions determine h,T-dependent line width!

\[ L_{u(1)} = \psi_\alpha^\dagger \left( \partial_t - i A_0 + \epsilon (\nabla - i \vec{A}) \right) \psi_\alpha \]

Rashba-like perturbation due to spin orbit interaction

\[ \delta L_R = \alpha_R \psi_\alpha^\dagger \left( (p_x + A_x) \sigma^y - (p_y + A_y) \sigma^x \right) \psi_\alpha \]

Gauge-invariant response functions of fermions coupled to a gauge field

Yong Baek Kim, Akira Furusaki, Xue-Gang Wen, and Patrick A. Lee
Mori-Kawasaki formalism

Retarded spin GF

\[ G^R_{S^+ S^-}(\omega) \propto \frac{1}{(\omega - h - \Sigma(\omega))} \]

Line width

\[ \eta(\omega = h) = \text{Im} \Sigma(\omega = h) = - \frac{\text{Im}\{G^R_{\mathcal{AA}^\dagger}(\omega)\}}{2\langle S^z \rangle} \]

Perturbation is encoded in the composite operator (depends on polarization of microwave radiation!)

\[ \mathcal{A} = [\delta H_R, S^+] = -2i\alpha_R \sum_{p,q} \psi^\dagger_{p+q} \sigma^z \psi_p (A_x,q - iA_y,q) \]

\[ \eta(h) \sim \alpha_R^2 \int d\epsilon [1 + n_B(\epsilon) + n_B(h - \epsilon)] \text{Im} G^R_{S^z_q S^z_{-q}}(\epsilon) \text{Im} G^R_{A^-_q A^+_q}(h - \epsilon) \]

Gauge field propagator

\[ \text{Im} G^R_{A^-_q A^+_q}(\nu) = \frac{\gamma q \nu}{\gamma^2 \nu^2 + \chi^2 q^6} \]

\[ \nu \sim q^3 \]

Landau damping,

‘Particle-hole’ spinon continuum

\[ \text{Im} G^R_{S^z_q S^z_{-q}}(\epsilon) = \frac{m}{2\pi} \frac{\epsilon}{\sqrt{\nu^2 q^2 - \epsilon^2}} \Theta(\nu q - |\epsilon|) \]
Preliminary results for perturbed U(1) spin liquid

\( T = 0, \ h \gg T \)

\[ \eta \sim \alpha_{R}^{2} \omega^{5/3} / h \sim h^{2/3}, \ h > 0 \]

\[ \eta \sim \omega^{2/3}, \ h = 0 \]

\( T > 0, \ h \ll T \)

\[ \eta = \frac{1}{2 \chi_{u} h} \left( \frac{mT}{8 \pi \chi} + \tilde{c}_{0} T^{5/3} f\left(\frac{h}{T}\right) \right) \sim \frac{T}{h} + T^{2/3} \]

Scaling function \( f(x) \)

\[ f(x) \rightarrow -4.4x \text{ for } x \ll 1; \ f(x) \rightarrow 0.75x^{5/3} \text{ for } x \gg 1 \]
Conclusion:

Spinon magnetic resonance is generic feature of spin liquids with significant spin-orbit interaction and fractionalized excitations

Main features:

- broad continuum response
- zero-field absorption (polarized terahertz spectroscopy)
- strong polarization dependence
- van Hove singularities of spinon spectrum
- interesting and varying $h,T$ dependence of the resonance line width

Already checked in one dimension!
Spinon magnetic resonance has been observed and studied experimentally in quasi-1d materials $\text{Cs}_2\text{CuCl}_4$ and $\text{K}_2\text{CuSO}_4\text{Br}_2$ with uniform DM interaction: K. Povarov, A. Smirnov, OS et al, Phys. Rev. Lett. 107, 037204 (2011).