Spinon spin resonance

Electron spin resonance of spinon gas

Oleg Starykh, University of Utah

In collaboration with:

K. Povarov, A. Smirnov, S. Petrov, Kapitza Institute for Physical Problems, Moscow, Russia,
A. Shapiro, Shubnikov Institute for Crystallography, Moscow, Russia

Rachel Glenn and Mikhail Raikh (U Utah)

Exotic phases of frustrated magnets, KITP, October 8-12, 2012
Outline

- **Main ingredients**
  - spin liquid with spinon Fermi surface
  - DM interaction

- **Case study** $\text{Cs}_2\text{CuCl}_4$: Spinon continuum and ESR

- **Higher dimensional extension**: ESR in the presence of uniform DM interaction
  - from Hubbard to spin liquid with spinon Fermi surface

- Conclusions
Spin liquid with spinon Fermi surface

M. Yamashita et al, Science 2010


electrical insulator,
metal-like thermal conductor
Regarding the nature of excitations:

…the magnetic excitations levels correspond to the deviations from the normal distribution of the magnetic moments which are propagating through the whole crystal and are not localized in a definite place of the lattice. Such magnetic excitations will be called in the following "magnons" (this name was suggested by L. Landau).

Regarding statistics of spin excitations:

“The experimental facts available suggest that the magnons are submitted to the Fermi statistics; namely, when \( T << T_{\text{CW}} \) the susceptibility tends to a constant limit, which is of the order of \( \text{const}/T_{\text{CW}} \) [for \( T > T_{\text{CW}}, \chi=\text{const}/(T + T_{\text{CW}}) \)]. Evidently we have here to deal with the Pauli paramagnetism which can be directly obtained from the Fermi distribution. Therefore, we shall assume the Fermi statistics for the magnons.”

Ref. (5) A. Perrier and Kamerlingh Onnes, Leiden Comm. No.139 (1914)
Dzyaloshinskii-Moriya (DM) interaction

\[ D_{ij} \cdot S_i \times S_j \]

- Reduces symmetry to \( U(1) \) [rotations about D axis]
- Easy plane anisotropy (perp. to D)
- Promotes magnetic order
- Generically stabilizes incommensurate non-collinear (spiral) states
Example: DM in kagome

main DM: orthogonal to kagome plane; staggered between up and down triangles

$$H = \sum_{nn} [J S_i \cdot S_j + D_{ij} \cdot (S_i \times S_j)]$$

Elhajal, Canals, and Lacroix, PRB 2002
Rigol, Singh 2007

Spin liquid ordered

Cepas, Fong, Leung, Lhuillier, PRB 2008
Main idea:

• turn annoying material imperfection (DM) into a probe of exotic spin state and its excitations

• probe small-$q$ excitations by ESR
Cs$_2$CuCl$_4$: consequences of Dzyaloshinskii-Moriya interaction

$\vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j$

- Is known from inelastic neutron scattering data (Coldea et al. 2001-03)
- 3D ordered state - determined by weak residual interactions - interplane and Dzyaloshinskii-Moriya (DM) (OS, Katsura, Balents 2010)

5 different DM terms allowed
Experimental Realization of a 2D Fractional Quantum S

R. Coldea,1,2 D. A. Tennant,2,3 A. M. Tsvelik,4 and Z. Tylczynski

PHYSICAL REVIEW B 68, 134424 (2003)

Extended scattering continua characteristic of spin fractionalization frustrated quantum magnet Cs$_2$CuCl$_4$ observed by neut

R. Coldea,1,2,3 D. A. Tennant,1,3 and Z. Tylczynski4

Very unusual response: broad and strong continuum; spectral intensity varies strongly with 2d momentum ($k_x$, $k_y$)
Highly anisotropic phase diagram of Cs$_2$CuCl$_4$ in magnetic field is explained by DM interactions.

Tokiwa et al, 2006
Veillette, Chalker, Coldea 2005
Starykh, Katsura, Balents 2010

DM controlled cone state
$(-1)^z D'_a \hat{a}$

DM free situation

B $\parallel b$

C - IC transition

B $\parallel c$

DM driven

$(-1)^y D_c \hat{c}$
Cs$_2$CuCl$_4$: **uniform** Dzyaloshinskii-Moriya interaction

$$\vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j$$

Focus on the in-chain DM: for a given chain (y,z) vector D is constant

$$D_{y,z} = (-1)^y D_c \hat{c} + (-1)^z D_a \hat{a}$$
ESR - electron spin resonance

- Simple (in principle) and sensitive probe of magnetic anisotropies (and, also, \( q=0 \) probe: \( S = \Sigma \vec{r} \cdot \vec{S}_r \))

\[
I(h, \omega) = \frac{\omega}{4L} \int dt e^{i\omega t} \langle [S^+(t), S^-] \rangle
\]

- For SU(2) invariant chain in paramagnetic phase

\[
I(H, \omega) \sim \delta(\omega-H) m(H) \quad \text{[Kohn’s Th]} \quad \text{Oshikawa, Affleck PRB 2002}
\]

Zeeman \( H \) along \( z \), microwave radiation \( h \) polarized perpendicular to it.
ESR data (Povarov and Smirnov, Kapitza Institute)

Resonance line is significantly modified with lowering the temperature; modification is strongly anisotropic with respect to field. The lowest temperature $T=1.3 \, \text{K}$ is still twice higher than ordering $T_N$. 

![Shift for H $\parallel b$](image1)

![Shift and splitting for H $\parallel a, c$](image2)
Line splitting

\[ T = 1.3 \, \text{K} \]
\[ \nu = 26.92 \, \text{GHz} \]
\[ H \parallel a \]
Cs$_2$CuCl$_4$  ESR data: $T=1.3$ K  Povarov et al, 2011

- H along b-axis
  
  ✓ gap-like behavior for $\nu > 17$ GHz

  $$2\pi\hbar\nu = \sqrt{(g_b\mu_B H)^2 + \Delta^2}$$

  ✓ loss of intensity for $\nu < 17$ GHz

- H along a-axis: splitting of the ESR line

  $$g_a = 2.20$$
Spin resonance studies of the quasi-one-dimensional Heisenberg antiferromagnet Cs$_2$CuCl$_4$

J.M. Schrama $^{a,*}$, A. Ardavan $^a$, A.V. Semeno $^a$, P.J. Gee $^a$, E. Rzepniewski $^a$, J. Suto $^a$, R. Coldea $^a$, J. Singleton $^a$, P. Goy $^b$

$^a$ Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, UK
$^b$ Abmm, 52 rue Lhomond, 75005 Paris, France

Previous experiments

Physica B 256-258 (1998)
Temperature regimes

Paramagnetic

- individual spins
- universal quasi-classical regime

spin-correlated (spin liquid)

- well-developed correlations along chains;
- but little correlations between chains

ordered phase

- strongly coupled chains;
- 2d (or 3d) description

J S

- correlated spins (high T field theory)

\[ T_0 \sim J e^{-2\pi S} \]

- spinons (low T field theory)

Haldane scale does exist for S=1/2 chains:
- different response for S=1 and S=1/2 chains
- below this temperature

\[ T_N = 0.6 \text{ K} \]

spin waves (at low energy)

C. Buragohain, S. Sachdev
PRB 59 (1999)
Continuum in magnetic field

$H \neq 0$

$S_\perp$ transverse structure factor

$\Delta S = 1$

Dender et al, PRL 1997

**Explanation I: H along DM axis**

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

- **Chain**: \( JS_{x,y,z} \cdot S_{x+1,y,z} \)
- **Uniform DM along the chain**: \( D_{y,z} \cdot S_{x,y,z} \times S_{x+1,y,z} \)
- **Magnetic field**: \( g\mu_B H \cdot S_{x,y,z} \)

Unitary rotation about z-axis:

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

- Removes DM term from the Hamiltonian (to \( D^2 \) accuracy)
- **Boosts** momentum to \( D/(Ja_0) \)

**q = 0 \rightarrow q = D/(Ja_0) \Rightarrow 2\pi h \nu_{R/L} = g\mu_B H \pm \pi D/2**

Dotted lines: \( D=0 \) picture

Oshikawa, Affleck 2002

**Chiral probe:** ESR probes **right- and left-moving modes** (spinons) independently.
Spectrum shift due to the uniform DM

\[ \mathcal{H} = \sum_n J(\mathbf{S}_n \cdot \mathbf{S}_{n+1}) + (\mathbf{D} \cdot [\mathbf{S}_n \times \mathbf{S}_{n+1}]) - \mu_B g(\mathbf{H} \cdot \mathbf{S}_n) \]

Gangadharaiiah, Sun, Starykh, PRB 78 054436 (2008)

Spin transformation \( S^+_n = \tilde{S}^+_n e^{i\alpha n} \quad \alpha = -D/J \) excludes DM interaction, but results in the spectrum shift by momentum \( D/J \). This leads to two ESR peaks.

Karimi, Affleck, PRB 2011
Explanation II: arbitrary orientation

Relevant spin degrees of freedom

- Spin-1/2 AFM chain = half-filled (1 electron per site, $k_F=\pi/2a$) fermion chain

\[
H_{\text{dirac}} = iv \int dx \sum_{s=\uparrow,\downarrow} (\Psi_L^{\dagger}s \partial_x \Psi_L^s - \Psi_R^{\dagger}s \partial_x \Psi_R^s)
\]

- $q=0$ fluctuations: right (R) and left (L) spin currents

\[
\vec{M}_{R/L} = \Psi_{R/L,s}^\dagger \frac{\vec{\sigma}_{ss'}}{2} \Psi_{R/L,s'}
\]

- $2k_F (= \pi/a)$ fluctuations: charge density wave $\varepsilon$, spin density wave $N$

Staggered Magnetization $N$

\[
\begin{align*}
N^+ &\sim \Psi_{R\uparrow}^\dagger \Psi_{L\downarrow} + \text{h.c.} & \text{Spin flip } \Delta S=1 \\
N^- &\sim \Psi_{R\uparrow}^\dagger \Psi_{L\uparrow} - \Psi_{R\downarrow}^\dagger \Psi_{L\downarrow} + \text{h.c.}
\end{align*}
\]

Staggered Dimerization

\[
\varepsilon \sim i \left( \Psi_{R\uparrow}^\dagger \Psi_{L\uparrow} + \Psi_{R\downarrow}^\dagger \Psi_{L\downarrow} - \text{h.c.} \right) & \Delta S=0
\]

Susceptibility

- $1/q$

- $\chi_{1d}(q)$

- $1/q$

- $1/q$

- $1/q$

*Must be careful: often spin-charge separation must be enforced by hand*
**Explanation II: arbitrary orientation**

\[
\mathbf{H} = \frac{2\pi v}{3} \left[ (\bar{M}_R)^2 + (\bar{M}_L)^2 \right] - \frac{vD}{J} [M_R^d - M_L^d] - g\mu_B H [M_R^z + M_L^z]
\]

- **unperturbed chain**
- **uniform DM along the chain**
- **magnetic field**

Uniform DM produces internal momentum-dependent magnetic field along \(d\)-axis

- Total field acting on right/left movers \( g\mu_B \bar{H} \pm \hbar \nu \bar{D}/J \)

- Hence ESR signals at \( 2\pi \hbar \nu_{R/L} = \left| g\mu_B \bar{H} \pm \hbar \nu \bar{D}/J \right| \)

- Polarization: for \(H=0\) maximal absorption when microwave field \(h_{mw}\) is perpendicular to the internal (DM) one. Hence \(h_{mw} \parallel b\) is most effective.

Povarov et al, PRL 2011
Gangadharaiah, Sun, OS, PRB 2008
Explanation III: arbitrary orientation

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

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

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

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

$$\left(2\pi\hbar\nu_L\right)^2 = (g_b\mu_B H_b)^2 + [g_a\mu_B H_a - (-1)^z \pi D_a/2]^2$$
$$+ [g_c\mu_B H_c - (-1)^y \pi D_c/2]^2.$$
This explanation suggests:

1) ESR absorption in the absence of H
   \[ \nu \sim \sqrt{D_a^2 + D_c^2} \]

2) strong polarization dependence in zero field

The largest absorption occurs when microwave field h(t) is lined along crystal b-axis, h || b [so that it is perpendicular to the D vector in a-c plane]
Extension to two-dimensional spin liquids with spinon Fermi surface

“This could be the discovery of the century. Depending, of course, on how far down it goes”
Higher dimensional extension (weak Mott insulators)

- origin of DM: spin-orbit tunneling in Hubbard model
  \[ \hat{H} = \sum_{i,j} \{ c_{i,\alpha}^+ (-t \delta_{\alpha\beta} + i \hat{\lambda}_{i,j} \cdot \vec{s}_{\alpha\beta}) c_{j,\beta} + \text{H.c.} \} + U \sum_i n_{i\uparrow} n_{i\downarrow}. \]

- 2D square lattice with **uniform spin-orbit** interaction \((YBa_2Cu_3O_{6+x})\)
  \[ \hat{\lambda}_{i,j} = \lambda \hat{z} \times (\vec{r}_i - \vec{r}_j) \]

- (Lattice) spin-orbit interaction of Rashba type
  \[ \hat{H}_{\text{SO}}(k) = -2\lambda \sum_k c_{k,\alpha}^+ \{ \hat{s}_x \sin[k_y] - \hat{s}_y \sin[k_x] \} c_{k,\beta} \]

- Transition to **spinons** via **slave-rotor** formulation
  \[ c_{r,\sigma} = \int_{r,\sigma} e^{i \theta_r} \]

- (mean-field) Rashba Hamiltonian for free spinons \( (f_{r,s}) \)
  \[ \hat{H}_f = \sum_{i,j} f_{i,\alpha}^+ \left( -t \delta_{\alpha\beta} + (i \hat{\lambda}_{i,j}^{\text{eff}} \cdot \vec{s}_{\alpha\beta}) \right) f_{j,\beta} - \vec{H} \cdot f_{i,\alpha}^+ \vec{s}_{\alpha\beta} f_{j,\beta} \]

Coffey, Rice, Zhang 1991
Shekhtman, Entin-Wolhman, Aharony 1992
Bonesteel 1993
Florens and Georges 2004
S.-S. Lee and P. A. Lee 2005
Glenn, OS, Raikh, PRB 2012
Estimates for 2d spinon gas using Rashba model as an example

\[ \alpha_R (p_x \sigma_y - p_y \sigma_x) \]

no DM

\[ \text{in-plane } \mathbf{H} \]

DM

\[ \text{in-plane } \mathbf{H} \]

splitting of Fermi surfaces

\[ \sqrt{\Delta_{SO}^2 + \Delta_Z^2 + 2 \Delta_{SO} \Delta_Z \sin \phi} \]

Raikh, Chen 1999

Energy absorption due to microwave \( h(t) \)

ESR signal

\[ \chi'' \]

the width of the line \( \sim \min(\mathbf{H}, \alpha_R k_F) \)

line shape is strongly polarization-dependent:

\[ [\omega - \omega_{\min/\max}]^{-1/2} \quad \text{for } h(t) \text{ perpendicular to } \mathbf{H} \]

\[ [\omega - \omega_{\min/\max}]^{1/2} \quad \text{for } h(t) \text{ parallel to } \mathbf{H} \]
Non-trivial lineshape due to vertical transitions between asymmetric subbands

The width of the line $\sim \min(H, \alpha_R k_F)$ line shape is strongly polarization-dependent:

\[ [\omega - \omega_{\text{min/max}}]^{-1/2} \quad \text{for } h(t) \text{ perpendicular to } H \]

\[ [\omega - \omega_{\text{min/max}}]^{1/2} \quad \text{for } h(t) \text{ parallel to } H \]

\[ \chi'' \sim \omega \sigma(\omega) \]

Glenn, OS, Raikh PRB 2012
Conclusions

- **DM** can be used to probe exotic spin liquids
- 1D: **ESR** is a chiral probe of critical spinons (neutral fermions)
  - measurements at small momentum $\sim D/J$
  - allows to extract parameters of DM (spin-orbit) interaction
- Higher-dimensional extension: DM breaks $SU(2)$ and provides access to spinon Fermi surface
Another geometry: **staggered DM**

Oshikawa, Affleck, Essler, Tsvelik

\[ \sum_{x} (-1)^{x} D \cdot S_{x} \times S_{x+1} \]

but: single ESR line!
Uniform vs staggered DM

\[ \vec{D} \cdot \vec{S}_n \times \vec{S}_{n+1} \]

h=0: free spinons
finite h: free spinons
but subject to momentum-dependent magnetic field

ESR: generically two lines
✓ splitting
✓ shift
✓ width (?)

\[ (-1)^n \vec{D} \cdot \vec{S}_n \times \vec{S}_{n+1} \]

free spinons
confined spinons
generate strongly relevant transverse magnetic field \((-1)^n \frac{\vec{D} \times \vec{h}}{2J} \cdot \vec{S}_n\)
that binds spinons together

✓ shift
✓ width

single line