Magnon collapse near the Lifshitz point and multipolar phases of frustrated magnets

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New Phases and Emergent Phenomena in Correlated Materials with Strong Spin-Orbit Coupling, KITP, July 24, 2015
Exotic ordered phases

Usual

Quantum spin liquids

ordered states

Usual

Exotic

spin nematic

composite order parameter

$$\mathcal{O}^{\alpha\beta}(r_i, r_j) = \frac{1}{2}(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) - \frac{1}{3}\delta^{\alpha\beta}\langle S_i \cdot S_j \rangle$$
Outline

• Motivation: usual \textit{vs} unusual ordered states
  – emergence of composite orders from competing interactions

• Nematic/SDW states in LiCuVO$_4$
  ✓ spin nematic: “magnon superconductor”
  ✓ collinear SDW: “magnon charge density wave”

• Volborthite kagome antiferromagnet
  – experimental status - magnetization plateau
  – UUD, SDW and \textit{more}

★ Field theory of the Lifshitz point
Magnon binding and pair condensation route to nematic phases

1-magnon

2-magnon bound state

\[ E - E_{\text{FM}} = \varepsilon_1 + h \]

\[ E - E_{\text{FM}} = \varepsilon_2 + 2h \]

For \( d > 1 \) at \( T = 0 \) this is a molecular BEC

= true spin nematic

Formation of molecular fluid
Hidden order

No dipolar order

\[ \langle S_i^+ \rangle = 0 \]
\[ \langle S_i^+ S_j^- \rangle \sim e^{-|i-j|/\xi} \quad S^z=1 \text{ gap} \]

Nematic order

\[ \langle S_i^+ S_{i+a}^+ \rangle \neq 0 \]

Magnetic quadrupole moment

Symmetry breaking \( U(1) \rightarrow Z_2 \)

can think of a fluctuating fan state
High-field analysis: condensate of bound magnon pairs

\[ \langle S^+ \rangle = 0 \quad \langle S^+ S^+ \rangle \neq 0 \]

Ferromagnetic \( J_1 < 0 \) produces attraction in real space

- Chubukov 1991
- Kecke et al 2007
- Kuzian and Drechsler 2007
- Hikihara et al 2008
- Sudan et al 2009
- Zhitomirsky and Tsunetsugu 2010

Shannon, Momoi, Sindzingre PRL 2006
LiCuVO$_4$ : spin nematic?

\[ H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - \hbar \sum_i S_i^z \]

+ weak interchain coupling $J_5$...

estimates:

- $J_1 = -1.6$ meV
- $J_2 = 3.9$ meV
- $J_5 = -0.4$ meV
LiCuVO$_4$ experiment: collinear SDW along \textbf{B}

Hagiwara, Svistov et al, 2011

Buttgen et al 2012, 2014

FIG. 2. Field dependence of the incommensurate wave vector $k_{ic}$ for applied magnetic fields $\mathbf{H} \parallel \mathbf{c}$ in LiCuVO$_4$. The open symbols

$T=1.3$ K $H_{//c}$

$H_{c1}$ $H_{c2}$ $H_{sat}$

$M(\mu_B/Cu^{2+})$ vs $\mu_0H(T)$

$\nu$ NMR spin echo amplitude (arb. units)

$T=0.38$ K $245$ MHz

$\mu_0H$ (T) $g\mu_BS$ $-g\mu_BS$

Field 22 T
Evidence of a Bond-Nematic Phase in LiCuVO₄

No spin-flip scattering above ~ 9 Tesla:

**longitudinal SDW state**

SF = spin flip, ΔS = 1
NSF = no spin flip, ΔS = 0

FIG. 3 (color online). Polarized cross sections measured at T = 70 mK for the magnetic reflections \( Q = (1, k, 0) \) with \( H \parallel c \) [left panels, (a)–(c)] and \( Q = (0, -k, 1) \) with \( H \parallel a \) [right panels, (d)–(f)].
T=0 schematic phase diagram of weakly coupled **nematic** spin chains

Cautionary remark: an impurity effect!

Sato, Hikihara, Momoi 2013; Starykh, Balents 2014
Cold reality

Search for a spin-nematic phase in the quasi-one-dimensional frustrated magnet LiCuVO₄

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“Our results suggest that the theoretically predicted spin-nematic phase, if it exists in LiCuVO₄, can be established only within the narrow field range 40.5<H<41.4 T.”

- so far, extensive experimental evidence for longitudinal SDW
- Spin Nematic phase is constrained to field interval < 1 T right below the saturation field (of the order 40 T)

Have to look elsewhere
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✴ Field theory of the Lifshitz point
Volborthite

Figure 1 | Crystal structure of volborthite Cu·2H·(OH)·2Cl·2H·(OH), the red lines highlight the kagomé lattice made up of Cu atoms.

Figure 2 | Crystal structure of vesignieite, BaCu6(PO4)4(OH)2·H2O.

Absorption spectrum showing broad features at 6700 and 5000 cm⁻¹ in the solid state (γ), which are absent in the liquid state (β).

Absorption spectrum showing broad features at 6700 and 5000 cm⁻¹ in the solid state (γ), which are absent in the liquid state (β).

The magnetic properties of volborthite Cu·2H·(OH)·2Cl·2H·(OH), the red lines highlight the kagomé lattice made up of Cu atoms.

Volborthite from Lisbon Valley, San Juan County, Utah

Volborthite from Lisbon Valley, San Juan County, Utah
Volborthite’s timeline

Formula: \( \text{Cu}_3(\text{V}_2\text{O}_7)(\text{OH})_2 \cdot 2\text{H}_2\text{O} \)
System: Monoclinic
Hardness: 3½
Name: Named after Alexander von Volborth (1800–1876), Russian paleontologist, who first noted the mineral.

A secondary mineral found in the oxidized zones of vanadium–bearing hydrothermal deposits.

At least two different monoclinic space–group variants (C2/m, C2/c) seem to be stable at ambient temperature. Visually similar to vésigniéite.

2001 quantum spin liquid?!
2009 impurity ordering at low \( T \)? magnetization steps?
2012 magnetic order!
2014 magnetization plateau

time = material quality
One-Third Magnetization Plateau with a Preceding Novel Phase in Volborhite


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Huge 1/3 magnetization plateau!
How far in field does the plateau extend?

Meeting abstracts of the Physical Society of Japan 2014

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How far in field does the plateau extend?

Translation:

== Intro ==
- S=1/2 kagome antiferromagnet is a known candidate spin liquid. Under external field, five magnetic plateaus have been theoretically found in S=1/2 antiferromagnet: m=0, 1/9, 1/3, 5/9, and 7/9. Among them, first two are spin liquids, and the later three are long-range orders (Ref. 1).

- Experimentally, a large 1/3 magnetization plateau was recently observed in Volborthite, which extends above 75T.

== Method & Results ==
- We study the magnetic phases of Volborthite in high-field, above 100T, using Faraday rotation.

- The figure shows rotation angle observed at 11K using a laser of wave length 532nm; it is compared to magnetization curve at 1.3K. We used single crystal samples and the field is applied perpendicular to the ab plane.

- The rotation angle show increase around 25T, where rapid increase in magnetization curve is observed. The rotation angle become nearly constant corresponding to the 1/3 magnetic plateau, which extends up to 120T.

this is the best plateau ever: 28 T < plateau < 120 T
(a) V NMR spectra measured on a single-domain piece of a crystal in magnetic fields between 15 and 30 T applied perpendicular to the ab plane at T = 0.4 K.

(b) Magnetization curve of single crystal (top, black line) and its field derivative (bottom, red line) in B_ab at 1.4 K after the subtraction of V-Vleck paramagnetic magnetization (M_{VV}). Magnetization deduced from the center of gravity of the NMR spectra is also plotted (top, blue circles). Expected spin structures in phases II and III are schematically depicted in the inset.

Phase diagram

1/3 plateau

N? SDW

small plateau’s onset field of 27 Tesla, relative to J ~ 100 K, suggest the presence of ferromagnetic exchange interactions.

H. Ishikawa et al, PRL 2015

may be a spin nematic?
Frustrated ferromagnetism

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Coupled frustrated quantum spin-$\frac{1}{2}$ chains with orbital order in volborthite Cu$_3$V$_2$O$_7$(OH)$_2$·2H$_2$O

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DFT gets it right!

\[ J_1 < 0, \; J_2 > 0, \; J'_2 > 0 \]
Coupled frustrated quantum spin-$\frac{1}{2}$ chains with orbital order in volborthite Cu$_3$V$_2$O$_7$(OH)$_2$$\cdot$2H$_2$O

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$J_1 < 0, J_2 > 0, J' > 0$

J$_1$ FM, J$_2$ AF

J' AF

coupled frustrated quantum spin-$\frac{1}{2}$ chains with orbital order in volborthite Cu$_3$V$_2$O$_7$(OH)$_2$$\cdot$2H$_2$O

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Spin chain redux

Frustrated ferromagnetic chain

\[ H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - h \sum_i S_i^z \]

\[ \frac{H}{|J_1|+J_2} \]

FM

quasi-spin-nematic

\[ J_2 \text{ AF} \]

\[ J_1 \text{ FM} \]
Numerics: nematicity and 1st order seem connected?

1d chain

2d square lattice

Sudan et al, 2009

Shannon, Momoi, Sindzingre PRL 2006
Quantum Lifshitz point

Frustrated ferromagnetic chain

\[ H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - h \sum_i S_i^z \]

\[ \frac{H}{|J_1| + J_2} \]

“Lifshitz”

QCP

FM

quasi-spin-nematic

0 1/5

1 J_2/(|J_1| + J_2)
It is time for a blackboard discussion
Lifshitz Point

• Unusual QCP: order-to-order transition
• Effective action - NLσM for unit vector \( \mathbf{m} \)

\[
S = \int dx d\tau \left\{ i s A_B [\hat{m}] + \delta |\partial_x \hat{m}|^2 + K |\partial_x \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h \hat{m}_z \right\}
\]

\[
A_B = \frac{\hat{m}_1 \partial_\tau \hat{m}_2 - \hat{m}_2 \partial_\tau \hat{m}_1}{1 + \hat{m}_3}
\]

Berry phase QCP allowed interactions at \( O(q^4) \)

tunes

two symmetry

term \( \delta \propto |J_1| - 4J_2 \)

All properties near Lifshitz point obey “one parameter universality” dependent upon \( u/K \) ratio
Lifshitz Point

\[ S = \int dx d\tau \left\{ isA_B[\hat{m}] + \delta |\partial_x \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - \hbar \hat{m}_z \right\} \]

- Intuition: behavior near the Lifshitz point should be semi-classical, since “close” to FM state which is classical

\[ x \to \sqrt{\frac{K}{|\delta|}} x \quad \tau \to \frac{K}{\delta^2} \tau \]

\[ S = \sqrt{\frac{K}{\delta}} \int dx d\tau \left\{ isA_B[\hat{m}] + \text{sgn}(\delta) |\partial_x \hat{m}|^2 + |\partial_x^2 \hat{m}|^2 + v |\partial_x \hat{m}|^4 - \bar{\hbar} \hat{m}_z \right\} \]

Large parameter: saddle point!

\[ v = \frac{u}{K} \quad \bar{\hbar} = \frac{hK}{\delta^2} \]
Saddle point

\[ S = \sqrt{\frac{K}{\delta}} \int dx d\tau \left\{ i s A_B[\hat{m}] + \text{sgn}(\delta)|\partial_x \hat{m}|^2 + |\partial^2_x \hat{m}|^2 + v|\partial_x \hat{m}|^4 - h\hat{m}_z \right\} \]

-1 < v < -1/4 derives from quantum fluctuations

Large S >> 1: v \sim -3/(2S) < 0

S=1/2: v = -5/8

Order parameter discontinuity

\[ \left(\sqrt{4|v|} - 1\right)/|v| \ll 1 \text{ for } v \approx -1/4 \]

First order

\[ h_c = \frac{\delta^2}{8K\sqrt{|v|(1 - \sqrt{|v|})}} \]

-1 < v < -1/4

Local instability of FM state

(1-magnon condensation)

\[ \hat{m} = \left( \begin{array}{c} |\Psi| \cos(qx + \phi) \\ \mp |\Psi| \sin(qx + \phi) \\ \sqrt{1 - |\Psi|^2} \end{array} \right) \]
Metamagnetic endpoint?

\[ \frac{h}{K} \]

\[ \mathcal{E}_{FM} = \mathcal{E}_{cone} \]

\[ \epsilon_1 = 0 \]

\[ \mathcal{E}_{FM} - \mathcal{E}_{cone} \sim a\delta^2 \]

FM

Quantum corrections penalize \( E_{cone} \) but not \( E_{FM} \)

\[ \Delta \mathcal{E}_{cone} = +f(v)\delta^{5/2} \]
Metamagnetic endpoint?

\[ S = \int dx d\tau \left\{ i s A_B[\hat{m}] + \delta |\partial_x \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - \hbar \hat{m}_z \right\} \]

\[ \hat{m} = \sqrt{2 - n_1^2 - n_2^2 (n_1 \hat{e}_1(x) + n_2 \hat{e}_2(x)) + (1 - n_1^2 - n_2^2) \hat{e}_3(x)} \]

\[ \hat{e}_1 \times \hat{e}_2 = \hat{e}_3 = \hat{m}^{sp}(x) \]

\[ \eta = n_1 + i n_2 \quad \bar{\eta} = n_1 - i n_2 \]

\[ S = S_{sp} + \int dx \ d\tau \left\{ \bar{\eta} \partial_\tau \eta + H(\bar{\eta}, \eta) \right\} + O(\eta^3) \]

Bogoliubov transformation gives correction to GS energy
Metamagnetic endpoint?

\[ \frac{h}{K} \]

\[ \mathcal{E}_{FM} = \mathcal{E}_{cone} \]

\[ \epsilon_1 = 0 \]

\[ \mathcal{E}_{FM} - \mathcal{E}_{cone} \sim a \delta^2 \]

\[ -f(v) \delta^{5/2} \]

Corrected first order curve bends slightly downward to intersect second order line.
Instabilities

- Choose $E_{FM} = 0$

But the bound states cannot get arbitrarily deep - low density approximation is violated
A natural speculation

Instabilities

• Choose $E_{FM}=0$

Expect that $n$-boson bound states bend with increasing $n$ to approach continuum line.
Summary

Lifshitz point is a "parent" of many phases

\[ S = \int dx d\tau \left\{ i s A_B [\hat{m}] + \delta |\partial_x \hat{m}|^2 + K |\partial^2_x \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h \hat{m}_x \right\} \]
Summary

• Spin chains keep showing up in unexpected places

✓ Nematic physics of frustrated ferromagnets

✓ Explored Lifshitz point as a “parent” for multipolar states and metamagnetism
\[ S = \int dxd^{d-1}y \tau \left\{ isA_B[\hat{m}] + \delta |\partial_x \hat{m}|^2 + c |\partial_y \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h \hat{m}_z \right\} \]

- Rescaling:

\[ x \to \sqrt{\frac{K}{|\delta|}} x \quad \tau \to \frac{K}{\delta^2} \tau \quad y \to \frac{\sqrt{cK}}{\delta} y \]

\[ S = \frac{\sqrt{K^{d}C^{d-1}}}{\delta^{d-1/2}} \int dxd^{d-1}y \tau \left\{ isA_B[\hat{m}] + \text{sgn}(\delta) |\partial_x \hat{m}|^2 + |\partial_x^2 \hat{m}|^2 + |\partial_y \hat{m}|^2 + v |\partial_x \hat{m}|^4 - \bar{h} \hat{m}_z \right\} \]

:. Similar theory applies in \( d>1 \), and very similar conclusions apply