Frustration-driven multi magnon condensates and their excitations

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Current trends in frustrated magnetism,
ICTP and Jawaharlal Nehru University, New Delhi, India, Feb 9-13, 2015
Collaborators

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*not today but*

closely related findings:
spin-current state at the tip of 1/3 magnetization plateau,
spontaneous generation of orbiting spin currents (ask me for details after the talk :))
Outline

• Frustrated magnetism (brief intro)
  – emergence of composite orders from competing interactions

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

• Volborthite kagome antiferromagnet
  – experimental status - magnetization plateau
  – Nematic, SDW and more
  – Field theory of the Lifshitz point

• Conclusions
Emergent Ising order parameters

Chiral spin liquid in two-dimensional $XY$ helimagnets
A. O. Sorokin and A. V. Syromyatnikov

\[ H = \sum_x (J_1 \cos(\varphi_x - \varphi_{x+a}) + J_2 \cos(\varphi_x - \varphi_{x+2a}) \\
- J_b \cos(\varphi_x - \varphi_{x+b})). \]

**Ising order: spin chirality**

\[ \chi = \sum_{\text{triangle}} \vec{S}_i \times \vec{S}_j \]

Low-Temperature Broken-Symmetry Phases of Spiral Antiferromagnets
Luca Capriotti and Subir Sachdev

**FIG. 2.** The two different minimum energy configurations with magnetic wave vectors $Q = (Q, Q)$ and $Q' = (Q, -Q)$ with $Q = 2\pi/3$, corresponding to $J_3/J_1 = 0.5$. 
Ising nematic in collinear spin system

\[ \sigma = \vec{N}_1 \cdot \vec{N}_2 = \pm 1 \]
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LiCuVO$_4$: magnon superconductor?

estimates:

$J_1 = -1.6$ meV

$J_2 = 3.9$ meV (subject of active debates)

$J_5 = -0.4$ meV
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

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Fig. 1: (Color online) Energy-field diagram for a frustrated quantum magnet close to the saturation field. Dot-dashed lines show lowest one- and two-magnon states. Solid lines represent the ground-state energy for the one-magnon (spin-cone) and the two-magnon (spin-nematic) condensate.

Fig. 2: (Color online) Two-dimensional array of copper ions in LiCuVO$_4$ with principal exchange couplings.

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

1-magnon

2-magnon bound state

Formation of molecular fluid
For $d>1$ at $T=0$ this is a molecular BEC
$= \text{true spin nematic}$

$E - E_{FM} = \varepsilon_1 + h$

$E - E_{FM} = \varepsilon_2 + 2h$

$\varepsilon_2 < 2 \varepsilon_1$: "molecular" bound state
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

nematic director
LiCuVO$_4$: NMR lineshape - collinear SDW along $\mathbf{B}$

Hagiwara, Svistov et al, 2011

Buttgen et al 2012

FIG. 2. Field dependence of the incommensurate wave vector $k_{i_c}$ for applied magnetic fields $\mathbf{H} \parallel \mathbf{c}$ in LiCuVO$_4$. The open symbols
Evidence of a Bond-Nematic Phase in LiCuVO$_4$


No spin-flip scattering above \(~ 9\) Tesla:

**longitudinal SDW state**

$SF = \text{spin flip, } \Delta S = 1$

$NSF = \text{no spin flip, } \Delta S = 0$

FIG. 3 (color online). Polarized cross sections measured at $T = 70$ mK for the magnetic reflections $Q = (1, k_{1C}, 0)$ with $H \parallel c$ [left panels, (a)–(c)] and $Q = (0, -k_{1C}, 1)$ with $H \parallel a$ [right panels, (d)–(f)].
Geometry (motivated by LiCuVO$_4$)

- $J_1 < 0$ (ferro)
- $J_2 > 0$, $J' > 0$ (afm)

in magnetic field

- No true condensation [U(1) breaking] in d=1.

- Inter-chain interaction is crucial for establishing symmetry breaking in d=2.

- Need to study weakly coupled “superconducting” chains

Sato et al 2013
Starykh and Balents 2014
Inter-chain interaction

\[ H_{\text{inter-chain}} = \sum_y \int dx \ S_y \cdot S_{y+1} \sim \sum_y \int dx \ S_y^+ S_{y+1}^- + S_y^z S_{y+1}^z \]

**Superconducting analogy:** single-particle (magnon) tunneling between magnon superconductors is strongly suppressed at low energy (below the single-particle gap)

\[ H_{\text{inter}}^\perp = \sum_y \int dx \ J' \langle S_y^+ (x) S_{y+1}^- (x + 1) \rangle_{\text{nematic ground state}} \rightarrow 0 \]

**Superconducting analogy:** fluctuations generate two-magnon (Josephson coupling) tunneling between chains. They are generically weak, \( \sim J_1 (J'/J_1)^2 \ll J' \), but responsible for a true **two-dimensional nematic order**

\[ H_{\text{nem}} \sim (J'^2 / J_1) \sum_y \int dx \ [T_y^+ (x) T_{y+1}^- (x) + \text{h.c.}] \]

At the same time, density-density inter-chain interaction does not experience any suppression. It drives the system toward a **two-dimensional collinear SDW order**.

\[ S_y^z = M - 2n_{\text{pair}} = M - \tilde{A}_1 e^{i \frac{\sqrt{2} \pi}{\beta} \varphi_y^+ (x)} e^{i k_{\text{sdw}} x} \]

\[ H_{\text{inter-chain}}^z = H_{\text{sdw}} \sim J' \sum_y S_y^z S_{y+1}^z \sim J' \sum_y \int dx \cos[\frac{\sqrt{2} \pi}{\beta} (\varphi_y^+ - \varphi_{y+1}^+)] \]

Away from the saturation, **SDW** is more relevant [and stronger, via \( J' \gg (J')^2 / J_1 \)] than the **nematic interaction**: coupled 1d nematic chains order in a 2d SDW state.
Simple scaling

\[ H_{\text{nem}} \sim \left( \frac{J'}{J_1} \right)^2 \sum_y \int dx \left[ T_y^+(x)T_{y+1}^-(x) + \text{h.c.} \right] \]

- describes kinetic energy of magnon pairs, linear in magnon pair density \( n_{\text{pair}} \)

\[ H_{\text{inter-chain}} = H_{\text{sdw}} \sim J' \sum_y S_y^z S_{y+1}^z \sim J' \sum_y \int dx \cos\left[ \frac{\sqrt{2}\pi}{\beta} (\varphi_y^+ - \varphi_{y+1}^+) \right] \]

- describes potential energy of interaction between magnon pairs on neighboring chains, quadratic in magnon pair density \( n_{\text{pair}} \)

- Competition \( \frac{(J')^2}{J_1} n_{\text{pair}} \sim J' n_{\text{pair}}^2 \), hence \( n_{\text{pair}} \sim J'/J_1 \)

- Hence:
  - Spin Nematic near saturation, for \( n_{\text{pair}} < n_{\text{pair}}^* \)
  - SDW for \( n_{\text{pair}} > n_{\text{pair}}^* \)
T=0 schematic phase diagram of weakly coupled nematic spin chains

Cautionary remark: maybe impurity effect

$M_{1/2} - O(J'/J)$

Fully Polarized

Spin Nematic

SDW

BEC physics

cf: Sato, Hikihara, Momoi 2013
Excitations (via spin-spin correlation functions)

- 2d SDW \( \langle S^z(r) \rangle = M + \text{Re} \left( \Phi e^{i k_{sdw} \cdot r} \right) \)

- preserves U(1) [with respect to magnetic field] -> hence NO transverse spin waves

- breaks translational symmetry -> longitudinal phason mode at \( k_{sdw} = \pi(1-2M) \) and \( k=0 \)

\[
\hat{S}_z^i(r) = M + \text{Re} \Phi e^{i k_{sdw} \cdot r}
\]

(solitons (kinks) of massive sine-Gordon model which describes 2d ordered state)
Excitations (via spin-spin correlation functions)

- **2d Spin Nematic** \[ \langle S^+(r)S^+(r') \rangle \sim \psi \neq 0 \]

- breaks U(1) but \( \Delta S=1 \) excitations are gapped (magnon superconductor) \[ \langle S^+(r) \rangle = 0 \]

- gapless density fluctuations at \( k=0 \)

\(-\) sector: solitons of massive sine-Gordon model describing 1d **zig-zag chain.**
Energy scale \( J_1 \)

\(+\) sector: solitons of massive sine-Gordon model which describes 2d **ordered** state.
Energy scale \( (J')^2 / J_1 \)

OS, Balents PRB 2014
Intermediate Summary

- Interesting magnetically ordered states: SDW and Spin Nematic
  - Gapped $\Delta S=1$ excitations (no usual spin waves!)
  - Linearly-dispersing *phason* mode with $\Delta S=0$ in 2d SDW
  - SDW naturally sensitive to structural disorder
  - Linearly-dispersing *magnon density* waves in 2d Spin Nematic
  - analogy with superconductor/charge density wave competition
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Volborthite

(a) Crystal structure of volborthite. Points towards the hydroxide ion located above the centre of an 8-vertex region of the kagomé lattice. The elongation along the c-axis is approximately 3%. (b) Perspective view along the [100] direction. The network of Cu–O bonds is depicted. (c) Photograph of single crystals. The red lines highlight the kagomé lattice made up of Cu atoms.

Results

No sign of long-range order has been observed in experiments, mainly due to the omnipresence of disorder or deviations from ideal models in real compounds. It is theoretically predicted that a liquid state is realized at a temperature of around 20 K for polycrystalline samples. An anomaly shows a transition at 290 K for the single crystals only, as shown in the inset of Fig. 2a. Their magnetic susceptibility suddenly increases upon heating and decreases at 290 K, indicating a possible order–liquid phase transition at LT. As a result, a large modification of the magnetic susceptibility is observed. A broad peak at 9 K for Cu1-Cu2 and a cluster of single crystals in arbitrary orientation. An anomaly shows a transition at 290 K for polycrystalline samples.

References


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**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.

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2001 quantum spin liquid?!

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2009 impurity ordering at low T? magnetization steps?

2012 magnetic order!

2014 magnetization plateau

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*time = material quality*
2014: huge plateau!

H. Ishikawa…M. Takigawa…Z. Hiroi, unpublished, 2014

More different $MH$ curves in a pile of 50 large “thick” arrowhead-shaped crystals

30 days growth

Huge 1/3 plateau!

Further optical meas.

@ Takeyama lab

It survives over 120 T!

Kagome plateau or ferrimagnetic state?

coupled to lattice, but already distorted

High-field magnetization

$M$ vs. $B$ for different crystal orientations:

- $B \parallel ab$
- $B \perp ab$

$T = 1.4$ K

$M (\mu_B/Cu)$ vs. $B (T)$

High-field mag. meas.

@ Tokunaga & Kindo labs
FIG. 2(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 the 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

SDW 1/3 plateau

H. Ishikawa et al, unpublished

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

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(Received 9 August 2010; published 30 September 2010)

DFT gets it right!

$J_1 < 0$, $J_2 > 0$, $J' > 0$
Coupled frustrated quantum spin-$\frac{1}{2}$ chains with orbital order in volborthite \( \text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2\cdot2\text{H}_2\text{O} \)

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**Ferrimagnetic state**

\[
J_1 < 0, \quad J_2 > 0, \quad J' > 0
\]

**J** \_\_ FM, **J** \_\_ AF

```
J' AF
polarized chains?!
```
FIG. 2 (color online). (a) $^{51}$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 $V$ Van Vleck paramagnetic magnetization ($M_{VV}$). Magnetization deduced from the center of the 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

1K

SDW

$1/3$ plateau

spin nematic?

may be a spin nematic??

H. Ishikawa et al., unpublished
Spin chain redux

Frustrated ferromagnetic chain

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

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

<table>
<thead>
<tr>
<th>0</th>
<th>1/5</th>
<th>1</th>
</tr>
</thead>
<tbody>
<tr>
<td>FM</td>
<td>quasi-spin-nematic</td>
<td>FM</td>
</tr>
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</table>

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

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Quasi-1d nematic

1d $J_1$-$J_2$ chain is only quasi-spin-nematic

power-law correlations

Ψ $\sim (S^+)^2$ : spin-nematic

ϕ $\sim S^z e^{i q x}$ : SDW

Ψ “dominant”

ϕ “dominant”
Multipolar phases

Frustrated ferromagnetic chain

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

Is it an infinite progression?
A QCP parent?

Frustrated ferromagnetic chain

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

\( H/(|J_1|+J_2) \)

“Lifshitz” QCP

FM

quasi-spin-nematic

0 \quad 1/5 \quad 1 \quad J_2/(|J_1|+J_2)
Lifshitz Point

- Unusual QCP: order-to-order transition
- Effective action - NLσM

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

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

Berry phase tuning allows interactions at O(q^4)

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

\[ S = \int \! dxd\tau \left\{ i s \mathcal{A}_B [\hat{m}] + \delta |\partial_x \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h \hat{m}_z \right\} \]

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

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

\[ S = \sqrt{\frac{K}{\delta}} \int \! dxd\tau \left\{ i s \mathcal{A}_B [\hat{m}] + \text{sgn}(\delta) |\partial_x \hat{m}|^2 + |\partial_x^2 \hat{m}|^2 + v |\partial_x \hat{m}|^4 - \hbar \hat{m}_z \right\} \]

Large parameter: saddle point!

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

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

v derives from quantum fluctuations

By a spin wave analysis, one finds \( v \sim -3/(2S) < 0 \)

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

\(-1 < v < -\frac{1}{4}\)

local instability of FM state (1-magnon condensation)
Phase diagram

Frustrated ferromagnetic chain

First order metamagnetic transition near Lifshitz point

Higher dimensions?

Hikihara et al, 2008
\[ d > 1 \]

\[ S = \int dxd^{d-1}yd\tau \{ i\mathcal{A}_B[\hat{m}] + \delta |\partial_x \hat{m}|^2 + c |\partial_y \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h\hat{m}_z \} \]

- Rescaling:

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

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

:. Similar theory applies in \( d > 1 \), and very similar conclusions apply
Crosses show the transition and crossover points obtained from the circulating spin current in the zigzag chain with ferromagnetic guide for the eyes. In this phase, the SDW phase is characterized by long-range order with a wave number $Q$, and dotted lines are the transition functions. Shaded regions in the phase diagram represent the nematic correlation with a wave number $Q$, and the SDW phase is characterized by incommensurate nematic order. At higher magnetic field up to the saturation field, the nematic order becomes dominant, and the multipolar phases, represented in Sec. V, appear at $Q$. Another important feature of the vector chiral phase is that the transverse-spin correlation decays slower than the density-density correlation, it is appropriate to call this phase the triatic phase. The triatic/SDW phases, represented by the vector chiral phase, as shown schematically in Fig. 1, become dominant, and the multipolar phases are a TL liquid of bosons because of the physical identification depicted in Fig. 2. We now real-

\begin{align*}
\prod_{i} (1 + e^{i(k - q_i)l} + e^{-i(k - q_i)l}) &= \prod_{i} e^{i(k - q_i)l} + e^{-i(k - q_i)l}, \\
&= \prod_{i} e^{i(k - q_i)l} + \prod_{i} e^{-i(k - q_i)l} \\
&= e^{i(k - q)l} + e^{-i(k - q)l}.
\end{align*}

The incommensurate nematic order is broken spontaneously. We can also present the nematic correlation with wave number $Q$, triatic correlation with an incommensurate wave number. The transition patterns in Fig. 1. The ground state of the system is the unit of changes in the total spin density-wave phases: SDW, antiferromagnetic SDW, and ferromagnetic SDW. The product

\begin{align*}
\prod_{i} (1 + e^{i(k - q_i)l} + e^{-i(k - q_i)l}) &= \prod_{i} e^{i(k - q_i)l} + e^{-i(k - q_i)l}, \\
&= \prod_{i} e^{i(k - q_i)l} + \prod_{i} e^{-i(k - q_i)l} \\
&= e^{i(k - q)l} + e^{-i(k - q)l}.
\end{align*}

The ground state of the system is the unit of changes in the total spin density-wave phases: SDW, antiferromagnetic SDW, and ferromagnetic SDW.
Origin of multipolar phases

With enough quantum fluctuations, "bubbles" of partially polarized phase may become many-magnon bound states and form multipolar phases.
Origin of multipolar phases

With enough quantum fluctuations, "bubbles" of partially polarized phase may become many-magnon bound states and form multipolar phases.
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