# Frustration-driven multi magnon condensates and their excitations

Oleg Starykh, University of Utah, USA

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





### Collaborators



Leon Balents, KITP, UCSB Andrey Chubukov, Univ of Minnesota

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 :))

#### Condensed Matter Physics at the University of Utah

Scanning Probe Microscopy Nano-optics Low Temperature Transport xotic Matter and High Pressure Spin electronics Organic Semiconductors NMR and MRI



University

Strongly Correlated Electron Physics Topological insulators Frustrated magnetism Superconductivity

### Life at (and near) University of Utah



### Outline

- Frustrated magnetism (brief intro)
- emergence of composite orders from competing interactions
- Nematic vs SDW in LiCuVO<sub>4</sub>
  - ✓ 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



PHYSICAL REVIEW B 85, 174404 (2012)

Ising order: spin chirality

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



FIG. 2. The two different minimum energy configurations with magnetic wave vectors  $\vec{Q} = (Q, Q)$  and  $\vec{Q}^* = (Q, -Q)$  with  $Q = 2\pi/3$ , corresponding to  $J_3/J_1 = 0.5$ .

#### Ising nematic in collinear spin system

VOLUME 64, NUMBER 1

PHYSICAL REVIEW LETTERS

1 JANUARY 1990

#### Ising Transition in Frustrated Heisenberg Models

P. Chandra

Corporate Research Science Laboratories, Exxon Research and Engineering Company, Annandale, New Jersey 08801

P. Coleman and A. I. Larkin<sup>(a)</sup> Serin Physics Laboratory, Rutgers University, P.O. Box 849, Piscataway, New Jersey 08854

$$\sigma = \vec{N}_1 \cdot \vec{N}_2 = \pm 1$$

VOLUME 91 NUMBER 17	PHYSICAL	REVIEW	LETTERS	week ending
VULUME 71. INUMBER 17				Z4 UCTUBER Z003

#### Ising Transition Driven by Frustration in a 2D Classical Model with Continuous Symmetry

Cédric Weber,<sup>1,2</sup> Luca Capriotti,<sup>3</sup> Grégoire Misguich,<sup>4</sup> Federico Becca,<sup>5</sup> Maged Elhajal,<sup>1</sup> and Frédéric Mila<sup>1</sup>



FIG. 4. Monte Carlo results for the critical temperature  $T_c$  as a function of the frustrating ratio  $J_2/J_1$ . The line is an extrapolation of the large  $J_2$  data down to  $J_2 = 0$  (see text).

### Outline

- Frustrated magnetism (brief intro)
- emergence of composite orders from competing interactions
- Nematic vs SDW in LiCuVO<sub>4</sub>
  - ✓ 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

### LiCuVO<sub>4</sub> : magnon superconductor?



### High-field analysis: condensate of bound magnon pairs $\langle S^+ \rangle = 0$ $\langle S^+ S^+ \rangle \neq 0$

Ferromagnetic  $J_1 < o$  produces attraction in real space



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<sub>4</sub> 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



# Hidden order

No dipolar order 
$$\langle S_i^+ 
angle = 0$$
  
 $\langle S_i^+ S_j^- 
angle \sim e^{-|i-j|/\xi}$  S<sup>z</sup>=1 gap

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

Magnetic quadrupole moment nematic Symmetry breaking U(1)  $\rightarrow$  Z<sub>2</sub> director can think of a fluctuating fan state

### LiCuVO<sub>4</sub>: NMR lineshape - collinear SDW along **B**



 $-g\mu_{B}S$ 

FIG. 2. Field dependence of the incommensurate wave vector  $k_{ic}$  for applied magnetic fields  $\mathbf{H} \parallel \mathbf{c}$  in LiCuVO<sub>4</sub>. The open symbols

#### Evidence of a Bond-Nematic Phase in LiCuVO<sub>4</sub>

M. Mourigal,<sup>1,2</sup> M. Enderle,<sup>1</sup> B. Fåk,<sup>3</sup> R. K. Kremer,<sup>4</sup> J. M. Law,<sup>4,\*</sup> A. Schneidewind,<sup>5</sup> A. Hiess,<sup>1,†</sup> and A. Prokofiev<sup>6,7</sup>



FIG. 3 (color online). Polarized cross sections measured at T = 70 mK for the magnetic reflections  $\mathbf{Q} = (1, k_{\text{IC}}, 0)$  with  $\mathbf{H} \| \mathbf{c}$  [left panels, (a)–(c)] and  $\mathbf{Q} = (0, -k_{\text{IC}}, 1)$  with  $\mathbf{H} \| \mathbf{a}$  [right panels, (d)–(f)].



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

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

Sato et al 2013 Starykh and Balents 2014

Need to study weakly coupled "superconducting" chains

### **Inter-chain interaction** $H_{\text{inter-chain}} = \sum_{y} \int dx \ \vec{S}_{y} \cdot \vec{S}_{y+1} \sim \sum_{y} \int dx \ S_{y}^{+} S_{y+1}^{-} + S_{y}^{z} S_{y+1}^{z}$

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

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

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

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

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**.

$$S_y^z = M - 2n_{\text{pair}} = M - \tilde{A}_1 e^{i\frac{\sqrt{2\pi}}{\beta}\varphi_y^+(x)} e^{ik_{\text{sdw}}x}$$
$$H_{\text{inter-chain}}^z = H_{\text{sdw}} \sim J' \sum_y S_y^z S_{y+1}^z \sim J' \sum_y \int dx \cos\left[\frac{\sqrt{2\pi}}{\beta}(\varphi_y^+ - \varphi_{y+1}^+)\right]$$

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

#### Simple scaling

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

- describes kinetic energy of magnon pairs, linear in magnon pair density  $n_{pair}$ 

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

• describes potential energy of interaction between magnon pairs on neighboring chains, quadratic in magnon pair density  $\eta_{pair}$ 

• 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<sub>pair</sub> < n<sup>\*</sup><sub>pair</sub>
- SDW for  $n_{pair} > n_{pair}^*$

## T=0 schematic phase diagram of weakly coupled **nematic** spin chains



Excitations (via spin-spin correlation functions)

• 2d SDW 
$$\langle S^{z}(\mathbf{r}) \rangle = M + \operatorname{Re}\left(\Phi e^{i\mathbf{k}_{\operatorname{sdw}}\cdot\mathbf{r}}\right)$$

- 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



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)  $\langle S^+(\mathbf{r}) \rangle = 0$
- gapless density fluctuations at k=0



### 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

### Outline

- Frustrated magnetism (brief intro)
- emergence of composite orders from competing interactions
- Nematic vs SDW in LiCuVO<sub>4</sub>
  - ✓ 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

## Volborthite









#### Volborthite's timeline

Formula:  $Cu_3(V_2O_7)(OH)_2 \cdot 2H_2O$ 

System: Monoclinic

Hardness: 3½

Name: Named after Alexander von Volborth (1800-1876), Russian paleontologist, who first noted the mineral.

Colour: Olive-green, ...

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.



time = material quality

### 2014: huge plateau!

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

#### High-field magnetization

more different *MH* curves in a pile of 50 large "thick" arrowhead-shaped crystals <sup>30 days growth</sup>





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 mag. meas. @ Tokunaga & Kindo labs



### Frustrated ferromagnetism

PHYSICAL REVIEW B 82, 104434 (2010)

Coupled frustrated quantum spin- $\frac{1}{2}$  chains with orbital order in volborthite Cu<sub>3</sub>V<sub>2</sub>O<sub>7</sub>(OH)<sub>2</sub>·2H<sub>2</sub>O

O. Janson,<sup>1,\*</sup> J. Richter,<sup>2</sup> P. Sindzingre,<sup>3</sup> and H. Rosner<sup>1,†</sup> <sup>1</sup>Max-Planck-Institut für Chemische Physik fester Stoffe, D-01187 Dresden, Germany <sup>2</sup>Institut für Theoretische Physik, Universität Magdeburg, D-39016 Magdeburg, Germany <sup>3</sup>Laboratoire de Physique Théorique de la Matière Condensée, Univ. P. & M. Curie, Paris, France (Received 9 August 2010; published 30 September 2010)





# Ferrimagnetic state

PHYSICAL REVIEW B 82, 104434 (2010)

Coupled frustrated quantum spin- $\frac{1}{2}$  chains with orbital order in volborthite Cu<sub>3</sub>V<sub>2</sub>O<sub>7</sub>(OH)<sub>2</sub>·2H<sub>2</sub>O

O. Janson,<sup>1,\*</sup> J. Richter,<sup>2</sup> P. Sindzingre,<sup>3</sup> and H. Rosner<sup>1,†</sup> <sup>1</sup>Max-Planck-Institut für Chemische Physik fester Stoffe, D-01187 Dresden, Germany <sup>2</sup>Institut für Theoretische Physik, Universität Magdeburg, D-39016 Magdeburg, Germany <sup>3</sup>Laboratoire de Physique Théorique de la Matière Condensée, Univ. P. & M. Curie, Paris, France (Received 9 August 2010; published 30 September 2010)



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



# Spin chain redux

Frustrated ferromagnetic chain



# Quasi-1d nematic



Hikihara *et al*, 2008 Sudan *et al*, 2009

# Multipolar phases

Frustrated ferromagnetic chain





Hikihara *et al*, 2008

Is it an infinite progression?

# A QCP parent?

Frustrated ferromagnetic chain



# Lifshitz Point

- Unusual QCP: order-to-order transition
- Effective action  $NL\sigma M$

$$S = \int dx d\tau \{ is \mathcal{A}_B[\hat{m}] + \delta |\partial_x \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h \hat{m}_z \}$$
  
Berry tunes two symmetry  
$$\mathcal{A}_B = \frac{\hat{m}_1 \partial_\tau \hat{m}_2 - \hat{m}_2 \partial_\tau \hat{m}_1}{1 + \hat{m}_3} \text{ phase } OCP \text{ allowed interactions}$$
  
term at O(q<sup>4</sup>)

All properties near Lifshitz point obey "one parameter universality" dependent upon u/K ratio

# Lifshitz Point

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

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

$$x \to \sqrt{\frac{K}{|\delta|}} x \qquad \tau \to \frac{K}{\delta^2} \tau$$

 $S = \sqrt{\frac{K}{\delta}} \int dx d\tau \left\{ is \mathcal{A}_B[\hat{m}] + \operatorname{sgn}(\delta) |\partial_x \hat{m}|^2 + |\partial_x^2 \hat{m}|^2 + v |\partial_x \hat{m}|^4 - \bar{h} \hat{m}_z \right\}$ 

 $v = \frac{u}{K}$   $\overline{h} = \frac{hK}{\delta^2}$ 

Large parameter: saddle point!

1

# Saddle point

 $S = \sqrt{\frac{K}{\lambda}} \int dx d\tau \left\{ is \mathcal{A}_B[\hat{m}] + \operatorname{sgn}(\delta) |\partial_x \hat{m}|^2 + |\partial_x^2 \hat{m}|^2 + v |\partial_x \hat{m}|^4 - \bar{h} \hat{m}_z \right\}$ v derives from quantum fluctuations By a spin wave analysis, one finds v ~ -3/(2S) < 0hfirst order  $h_c = \frac{\delta^2}{8K\sqrt{|v|}(1-\sqrt{|v|})}$  $-1 < v < -\frac{1}{4}$ FM local instability of FM state (1-magnon condensation) IC cone

# Phase diagram

Frustrated ferromagnetic chain

First order metamagnetic transition near Lifshitz point

Higher dimensions?

Hikihara et al, 2008



## d > 1

 $S = \int dx d^{d-1}y d\tau \left\{ is \mathcal{A}_B[\hat{m}] + \delta |\partial_x \hat{m}|^2 + c |\partial_y \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h \hat{m}_z \right\}$ 

### • Rescaling:

$$x \to \sqrt{\frac{K}{|\delta|}} x \qquad au \to \frac{K}{\delta^2} au \quad y \to \frac{\sqrt{cK}}{\delta} y$$

 $S = \frac{\sqrt{K^{d}c^{d-1}}}{\delta^{d-1/2}} \int dx d^{d-1}y d\tau \{ is \mathcal{A}_{B}[\hat{m}] + \operatorname{sgn}(\delta) |\partial_{x}\hat{m}|^{2} + |\partial_{x}^{2}\hat{m}|^{2} + |\partial_{y}\hat{m}|^{2} + v|\partial_{x}\hat{m}|^{4} - \bar{h}\hat{m}_{z} \}$ 

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

# Phase diagram



multipolar phases from QCP?

# Origin of multipolar phases







First order transition: partially polarized state *coexists* with plateau one

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

# Origin of multipolar phases







First order transition: partially polarized state *coexists* with plateau one

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

### Quasi-1d nematic

1d J<sub>1</sub>-J<sub>2</sub> chain

H FM "dominant" SN "dominant" SDW SDW<sub>2</sub> J'/J c.f. J'/J ~ 0.1 in LiCuVO<sub>4</sub>

True nematic occurs in narrow range near FM state

O. Starykh + LB, 2013

# Quasi-1d nematic

SN

J′/J

1d J<sub>1</sub>-J<sub>2</sub> chain

"dominant" SN

"dominant" SDW



SDW<sub>2</sub>



q = 1/4 - M/2

# 2001: a QSL?



smooth thermodynamics

no spontaneous fields

# 2009: Impurity ordering at 1K? Fermionic QSL?



spinons?



c.f. herbertsmithite:

 $x_{imp} \sim 5\%$ 

# 2009: Impurity ordering at 1K? Fermionic QSL?



## 2012: Ordering transitions! Not a QSL





Amazing how much sample quality matters!

### Frustrated ferromagnetism

PHYSICAL REVIEW B 82, 104434 (2010)

Coupled frustrated quantum spin- $\frac{1}{2}$  chains with orbital order in volborthite Cu<sub>3</sub>V<sub>2</sub>O<sub>7</sub>(OH)<sub>2</sub>·2H<sub>2</sub>O

O. Janson,<sup>1,\*</sup> J. Richter,<sup>2</sup> P. Sindzingre,<sup>3</sup> and H. Rosner<sup>1,†</sup> <sup>1</sup>Max-Planck-Institut für Chemische Physik fester Stoffe, D-01187 Dresden, Germany <sup>2</sup>Institut für Theoretische Physik, Universität Magdeburg, D-39016 Magdeburg, Germany <sup>3</sup>Laboratoire de Physique Théorique de la Matière Condensée, Univ. P. & M. Curie, Paris, France (Received 9 August 2010; published 30 September 2010)



Prospects of observing novel quantum liquids





### Quasi-1d nematic

1d J<sub>1</sub>-J<sub>2</sub> chain



Interchain coupling

~ J'  $\phi_y \phi_{y+1} + (J')^2 / J \Psi_y \Psi_{y+1}$ extra suppression of spinnematic order in quasi-1d limit

#### Nematic chain

S<sup>z</sup>-S<sup>z</sup> (SDW) channel: in-chain J<sub>1</sub>< o gaps out relative mode  $\varphi_y^- = (\varphi_{y,\text{odd}} - \varphi_{y,\text{even}})/\sqrt{2}$  $H_{\text{intra-chain}} = \sum \int dx J_1 \sin[\pi M] \cos\left[\frac{\sqrt{8\pi}}{\beta}\varphi_y^{-}\right] \text{ local pair formation}$ y, even  $J_1$ -J<sub>2</sub> chain  $\langle S^+ \rangle = 0$ quantum-disordered,  $S_y^+(x) \sim (-1)^x A_3 e^{i\frac{\beta}{\sqrt{2}}\theta_y^+(x)} e^{i(-1)^x\frac{\beta}{\sqrt{2}}\theta_y^-(x)}$ decays *exponentially*:  $S^{z} = 1$  excitations are gapped Standard (in 1d) power-law decay: critical nematic spin correlations,  $T_y^+ = S_y^+(x)S_y^+(x+1) \sim e^{i\sqrt{2}\beta\theta_y^+(x)}$ but U(1) *is preserved*  $\langle T^+ \rangle = \langle S^+ S^+ \rangle \to 0$ 

Physical picture: 1d magnon "superconductor"

Kolezhuk, Vekua (2005); Hikihara et al. (2008); Sato, Hikihara, Momoi (2013)

### 2d commensurate SDW (such as 1/3 magnetization plateau)



0.45

Cs<sub>2</sub>CuBr<sub>4</sub>

T = 0.4 K





vanishing spectral weight as  $k_x \rightarrow 0$  (anisotropic)

$$\chi_{2d}^{zz}(k_x,k_y,\omega) = \frac{vk_x^2/\beta^2}{v^2k_x^2 + v_{\perp}^2k_y^2 - \omega^2},$$

phason (longitudinal)  $\chi_{2d}^{zz}(q,\pi+q_y,\omega) \sim \frac{Z_{zz;2d}}{(v^2q^2+v_{\perp}^2q_y^2)-\omega^2}$ 

### Single crystals of volborthite



### H. Yoshida's crystal

- 1. natural leaf crystals, long time ago
- 2. low-quality polycrystalline samples by precipitation, 2001
- 3. high-quality polycrystalline samples by hydrothermal annealing, 2009
- 4. small single crystals, 2012
- 5. large arrowhead-shaped crystals, 2013

mn



# Spin nematic redux

Frustrated ferromagnetic chain

