# 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 :))

# 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

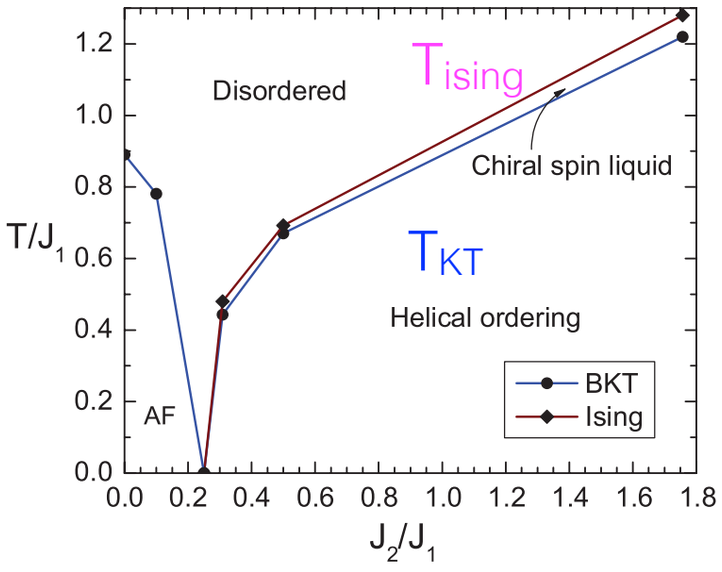
# Emergent Ising order parameters

PHYSICAL REVIEW B **85**, 174404 (2012)

## Chiral spin liquid in two-dimensional $XY$ helimagnets

A. O. Sorokin[1,*] and A. V. Syromyatnikov[1,2,†]

$$H = \sum_{\mathbf{x}} (J_1 \cos(\varphi_{\mathbf{x}} - \varphi_{\mathbf{x}+\mathbf{a}}) + J_2 \cos(\varphi_{\mathbf{x}} - \varphi_{\mathbf{x}+2\mathbf{a}}) - J_b \cos(\varphi_{\mathbf{x}} - \varphi_{\mathbf{x}+\mathbf{b}})),$$




## Ising order: spin chirality

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

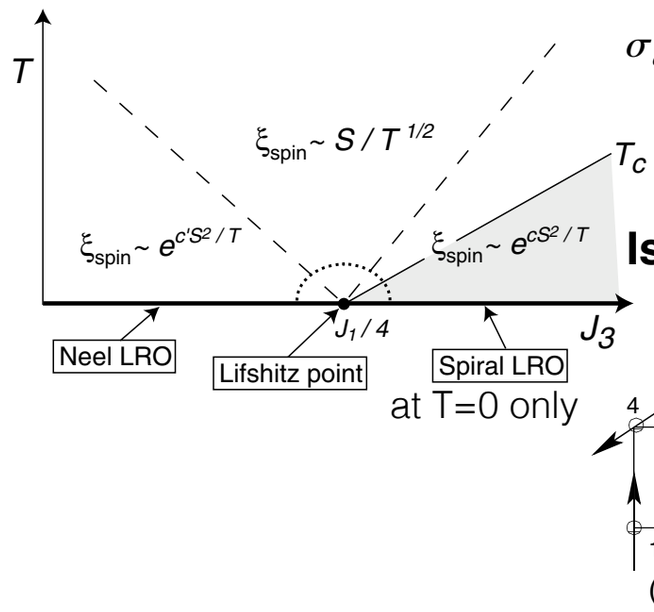
PRL **93,** 257206 (2004) PHYSICAL REVIEW LETTERS week ending 17 DECEMBER 2004

## Low-Temperature Broken-Symmetry Phases of Spiral Antiferromagnets

Luca Capriotti[1,2] and Subir Sachdev[2,3]

$$\hat{H} = J_1 \sum_{\langle i,j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + J_3 \sum_{\langle\langle i,j \rangle\rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j,$$

$$\sigma_a = (\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_3 - \hat{\mathbf{S}}_2 \cdot \hat{\mathbf{S}}_4)_a,$$



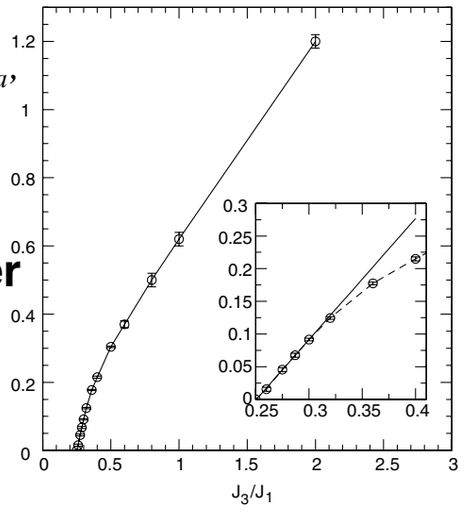




FIG. 2. The two different minimum energy configurations with magnetic wave vectors $\vec{Q} = (Q, Q)$ and $\vec{Q}^{\star} = (Q, -Q)$ with $Q = 2\pi/3$, corresponding to $J_3/J_1 = 0.5$.

# 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 [a]
*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 24 OCTOBER 2003

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

Cédric Weber,[1,2] Luca Capriotti,[3] Grégoire Misguich,[4] Federico Becca,[5] Maged Elhajal,[1] and Frédéric Mila[1]

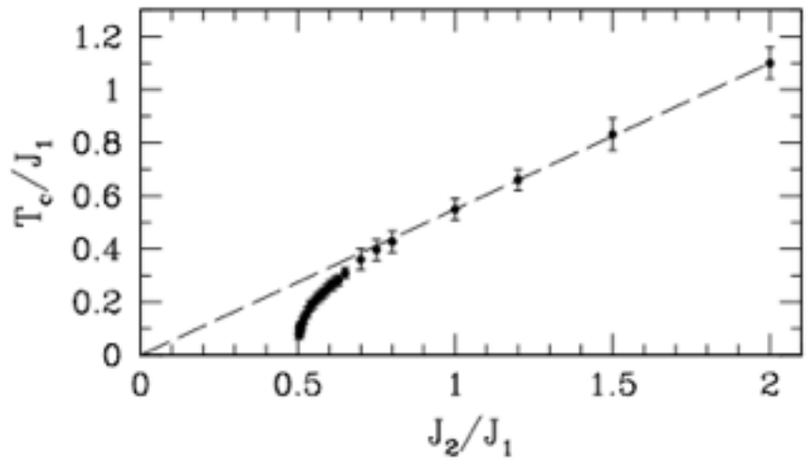
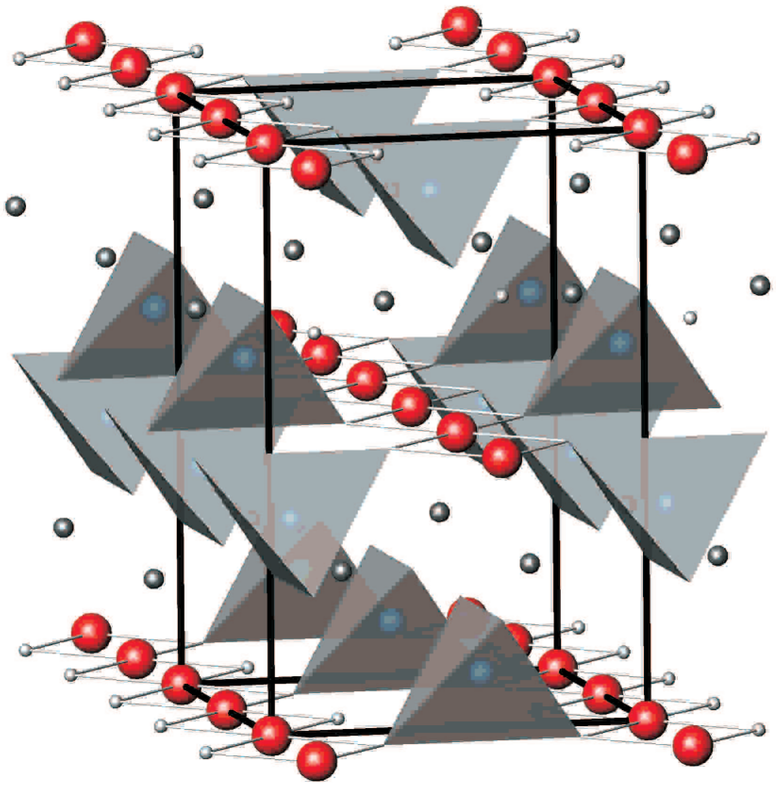


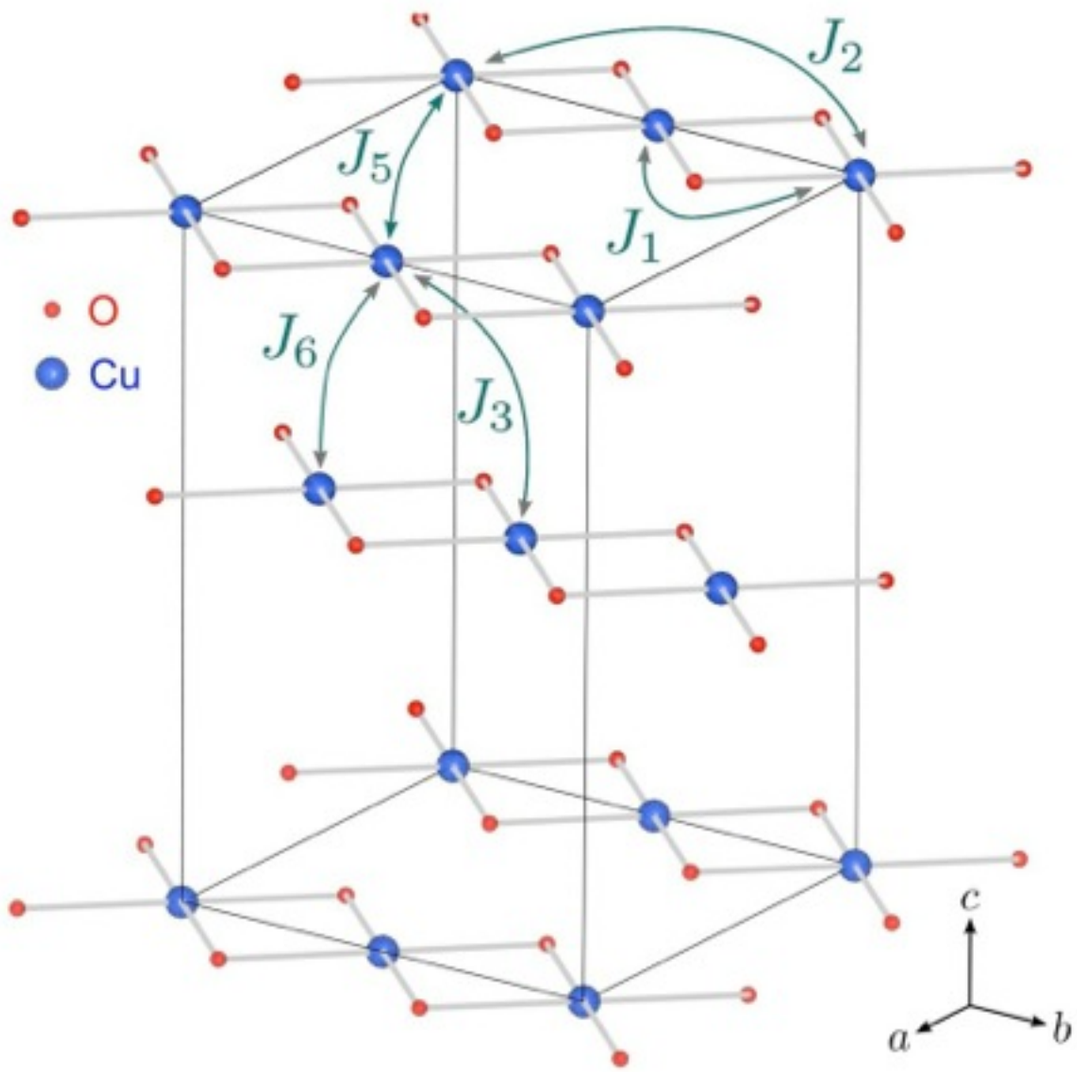

FIG. 4. Monte Carlo results for the critical temperature $T_c$ as a function of the frustrating ratio $J_2/J_1$. The line is an extrapolation of the large $J_2$ data down to $J_2 = 0$ (see text).

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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

$$\langle S^+ \rangle = 0 \qquad \langle S^+ S^+ \rangle \neq 0$$

Ferromagnetic $J_1 < 0$ produces attraction in real space

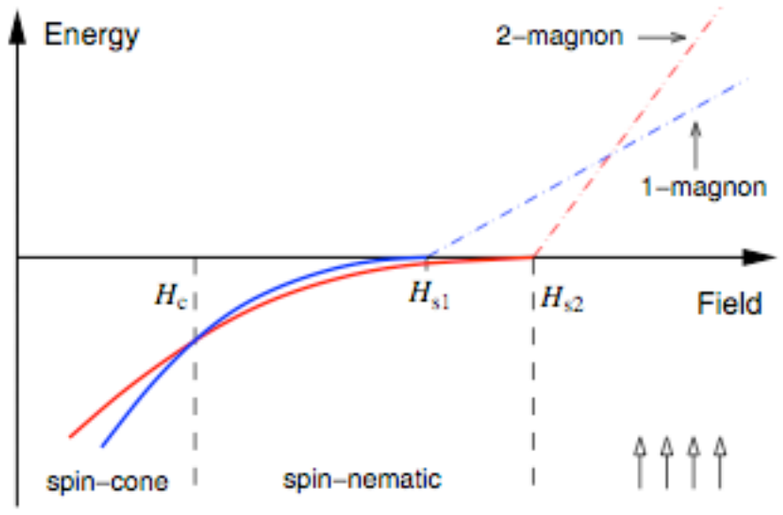



Fig. 1: (Color online) Energy-field diagram for a frustrated quantum magnet close to the saturation field. Dot-dashed lines show lowest one- and two-magnon states. Solid lines represent the ground-state energy for the one-magnon (spin-cone) and the two-magnon (spin-nematic) condensate.

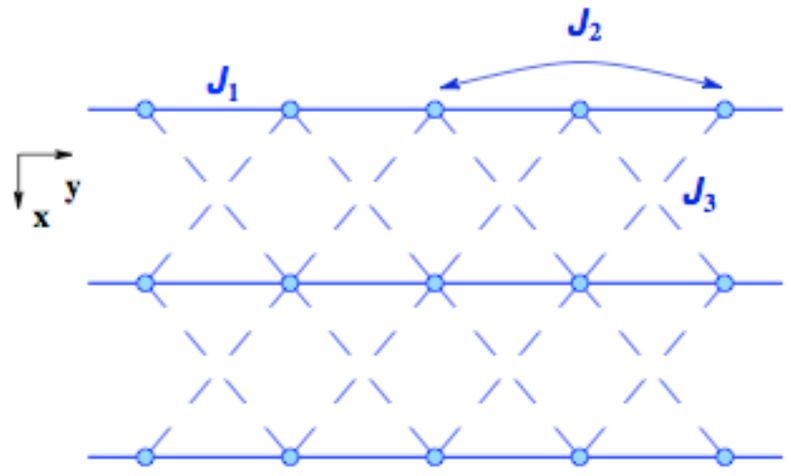
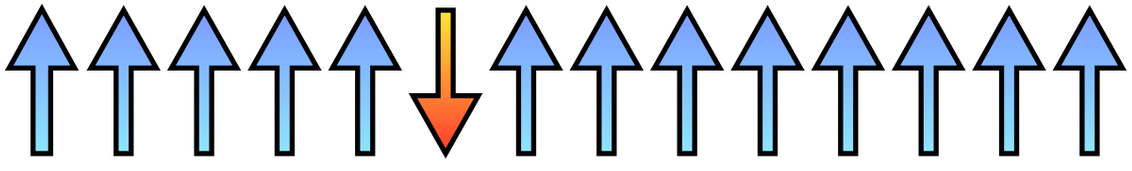



Fig. 2: (Color online) Two-dimensional array of copper ions in $LiCuVO_4$ with principal exchange couplings.

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

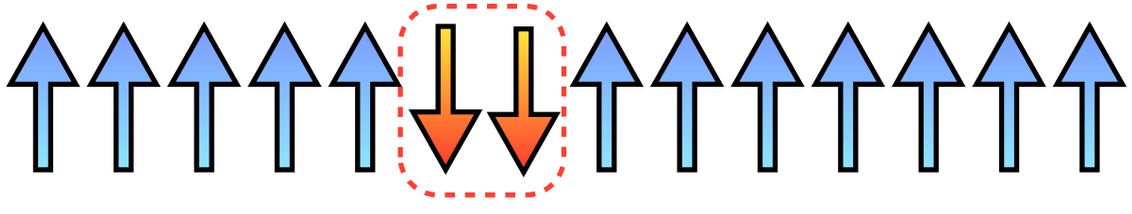
# Magnon binding

1-magnon

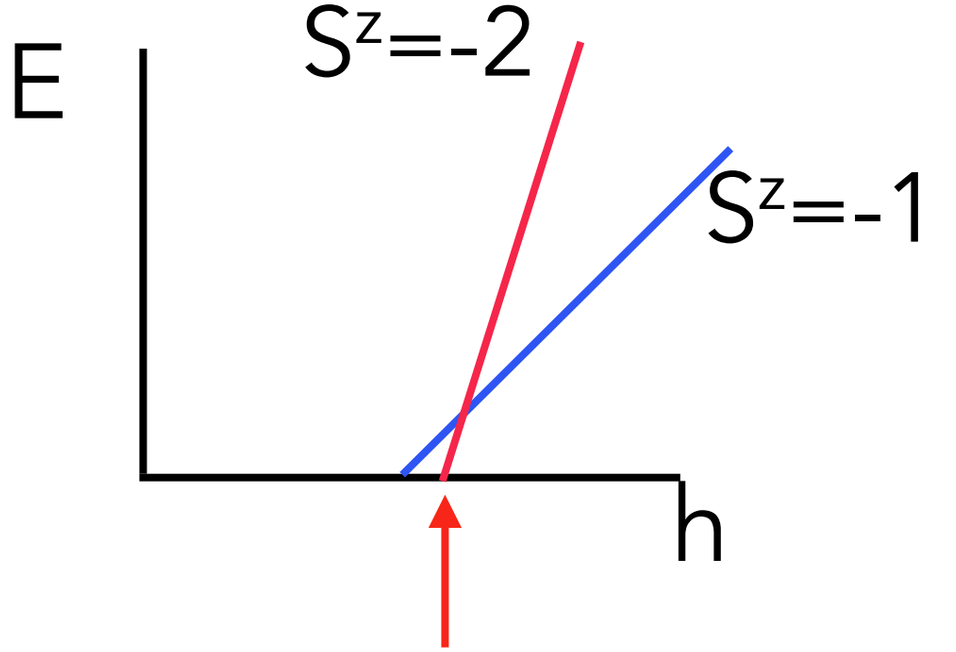


$E\text{-}E_{FM} = \varepsilon_1 + h$

2-magnon bound state



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




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

Formation of molecular fluid

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

= true spin nematic

# Hidden order

No dipolar order $\langle S_i^+ \rangle = 0$

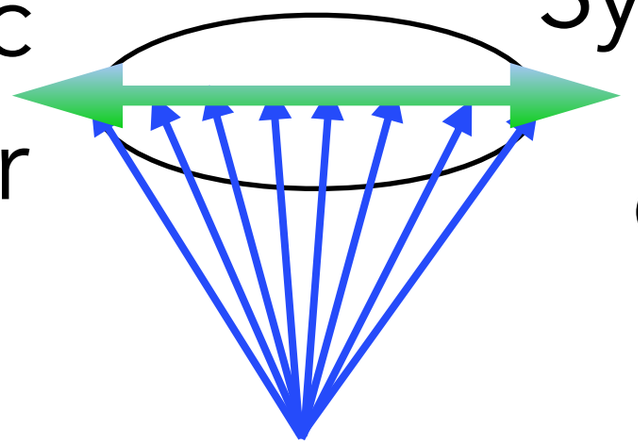
$\langle S_i^+ S_j^- \rangle \sim e^{-|i-j|/\xi}$ $S^z$=1 gap

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

Magnetic quadrupole moment

Symmetry breaking U(1) → $Z_2$

nematic director



can think of a fluctuating fan state

# $LiCuVO_4$: NMR lineshape - collinear SDW along **B**

Hagiwara, Svistov et al, 2011

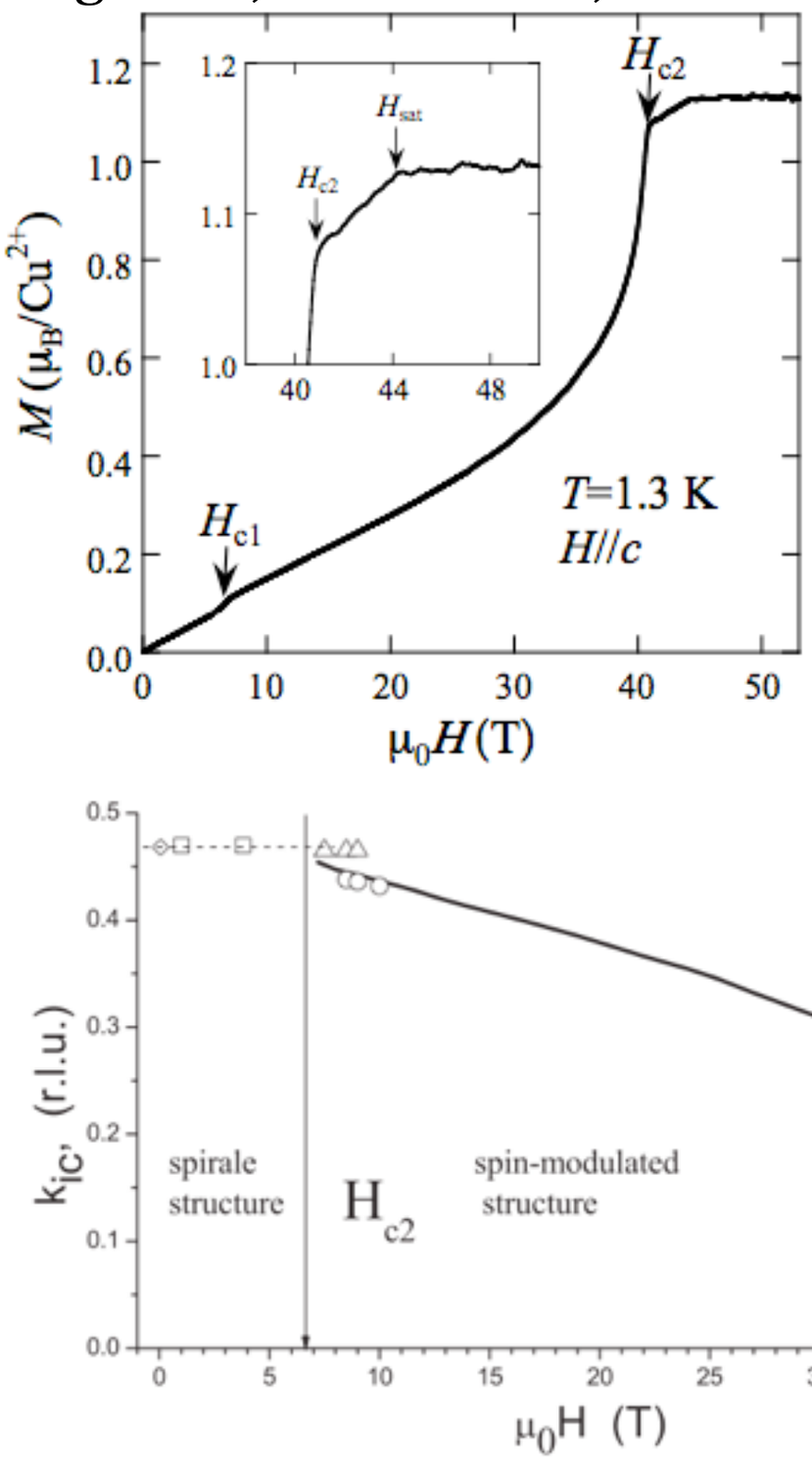
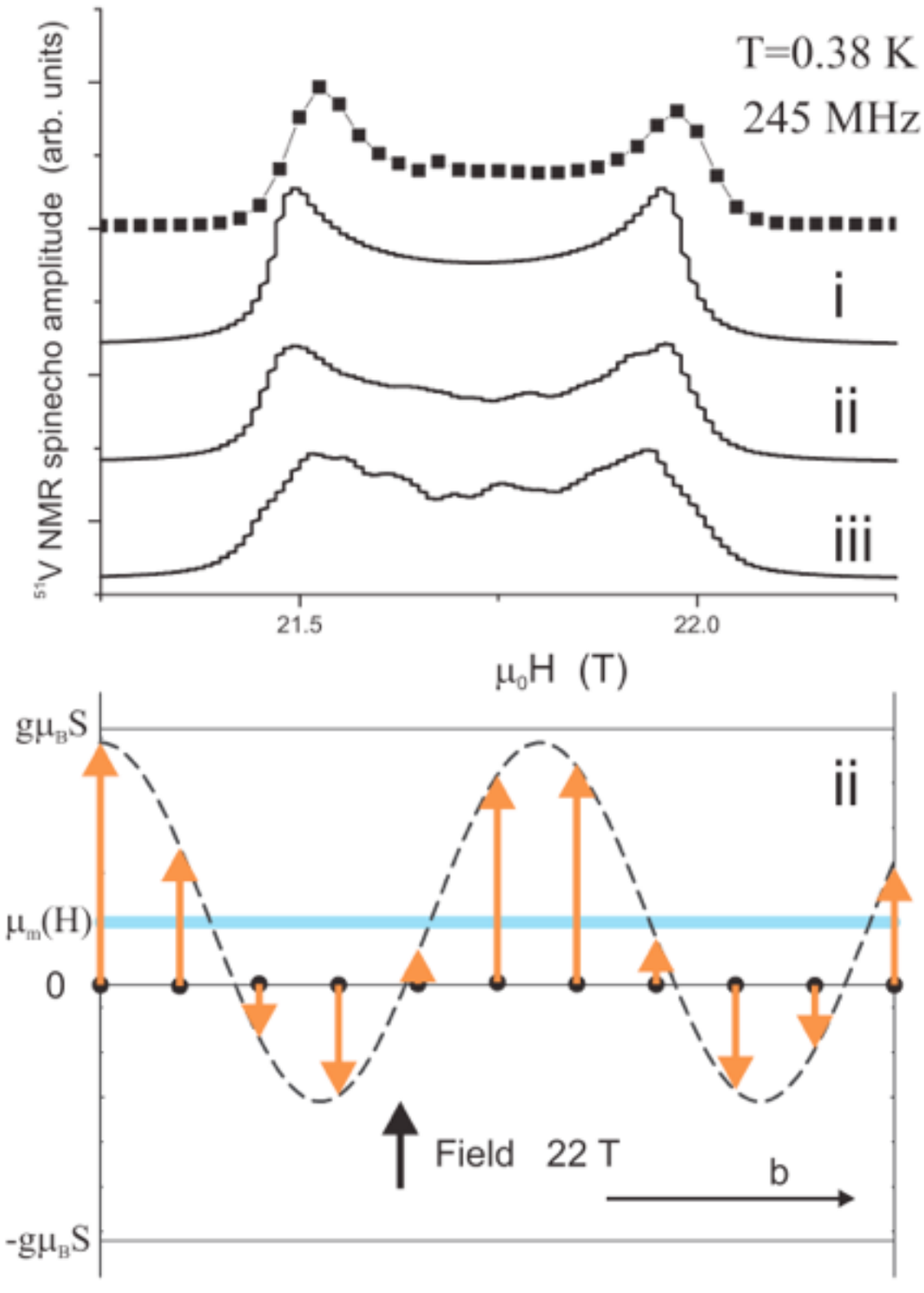



FIG. 2. Field dependence of the incommensurate wave vector $k_{ic}$ for applied magnetic fields **H** ∥ **c** in $LiCuVO_4$. The open symbols

Buttgen et al 2012 PHYSICAL REVIEW B **85**, 214421 (2012)



# Evidence of a Bond-Nematic Phase in $LiCuVO_4$

M. Mourigal,[1,2] M. Enderle,[1] B. Fåk,[3] R. K. Kremer,[4] J. M. Law,[4,*] A. Schneidewind,[5] A. Hiess,[1,†] and A. Prokofiev[6,7]

No spin-flip scattering above ~ 9 Tesla: **longitudinal SDW state**

SF = spin flip, $\Delta S = 1$
NSF = no spin flip, $\Delta S = 0$

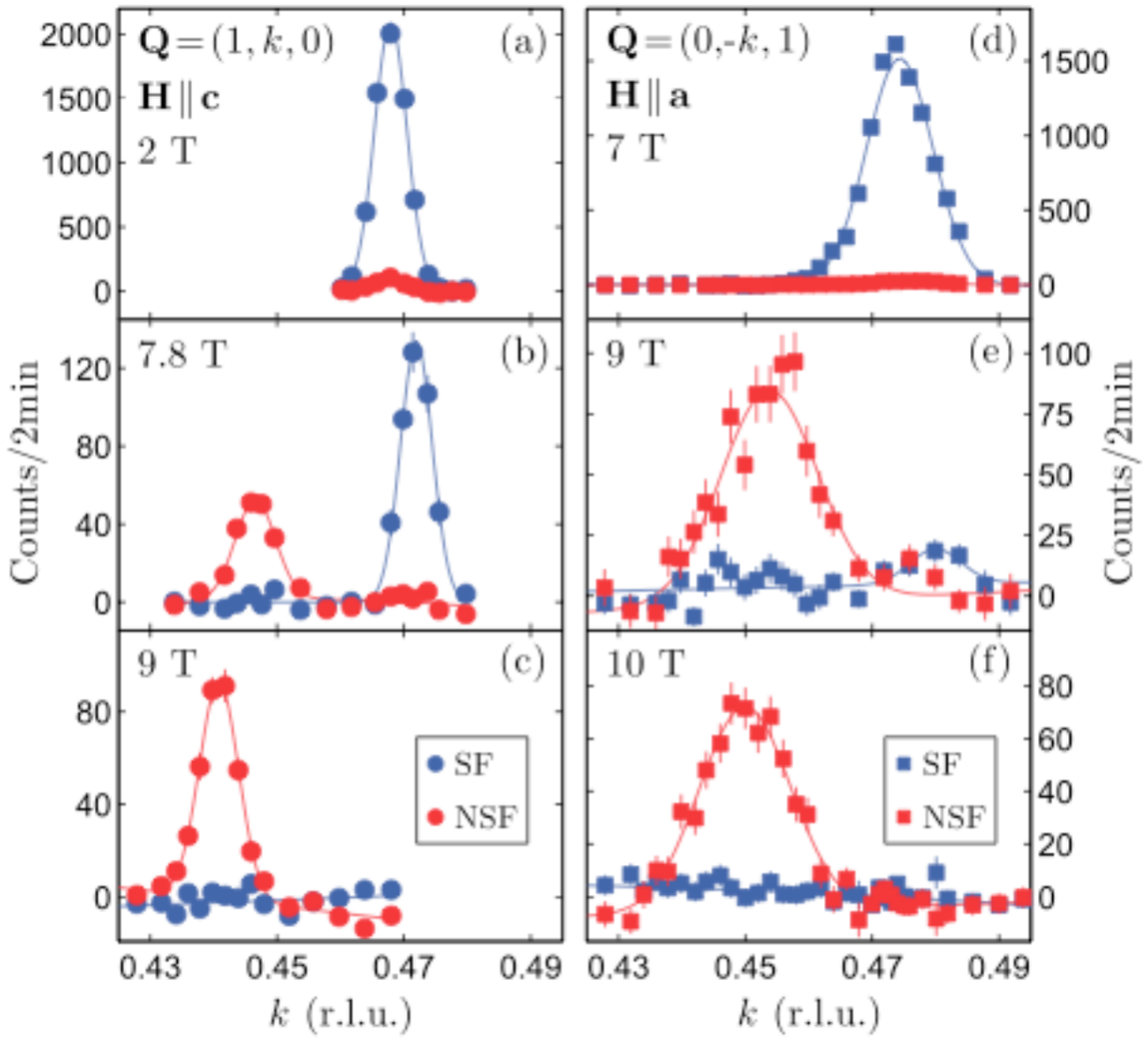
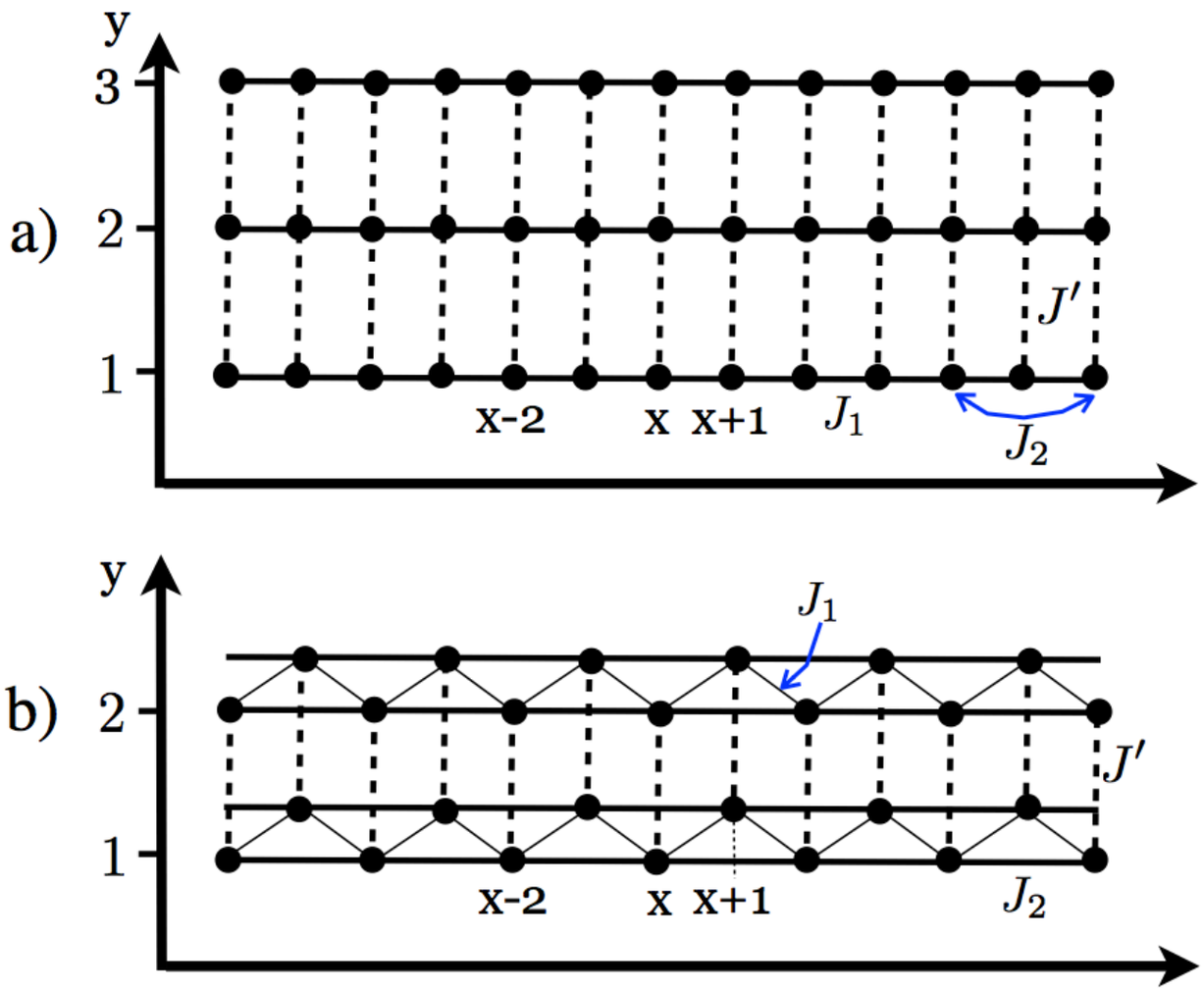
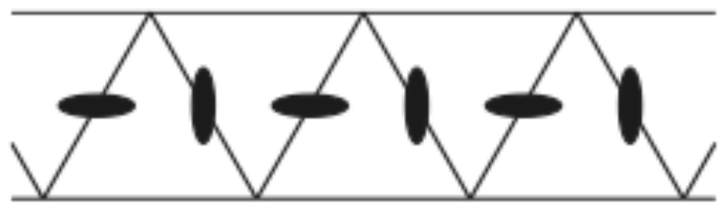



FIG. 3 (color online). Polarized cross sections measured at $T = 70$ mK for the magnetic reflections $\mathbf{Q} = (1, k_{IC}, 0)$ with $\mathbf{H}\|\mathbf{c}$ [left panels, (a)–(c)] and $\mathbf{Q} = (0, -k_{IC}, 1)$ with $\mathbf{H}\|\mathbf{a}$ [right panels, (d)–(f)].

# 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 $H_{\text{inter-chain}} = \sum_y \int dx\ \vec{S}_y \cdot \vec{S}_{y+1} \sim \sum_y \int dx\ S_y^+ S_{y+1}^- + S_y^z S_{y+1}^z$

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, ~ $J_1(J'/J_1)^2 << J'$ , but responsible for a true **two-dimensional nematic order**

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

$$T_y^+(x) \sim S_y^-(x) S_y^-(x+1)$$

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**.

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

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

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 $n_{\rm pair}$

$$H^z_{\rm inter-chain} = H_{\rm sdw} \sim J' \sum_y S^z_y S^z_{y+1} \sim J' \sum_y \int dx \cos[\frac{\sqrt{2}\pi}{\beta}(\varphi^+_y - \varphi^+_{y+1})]$$

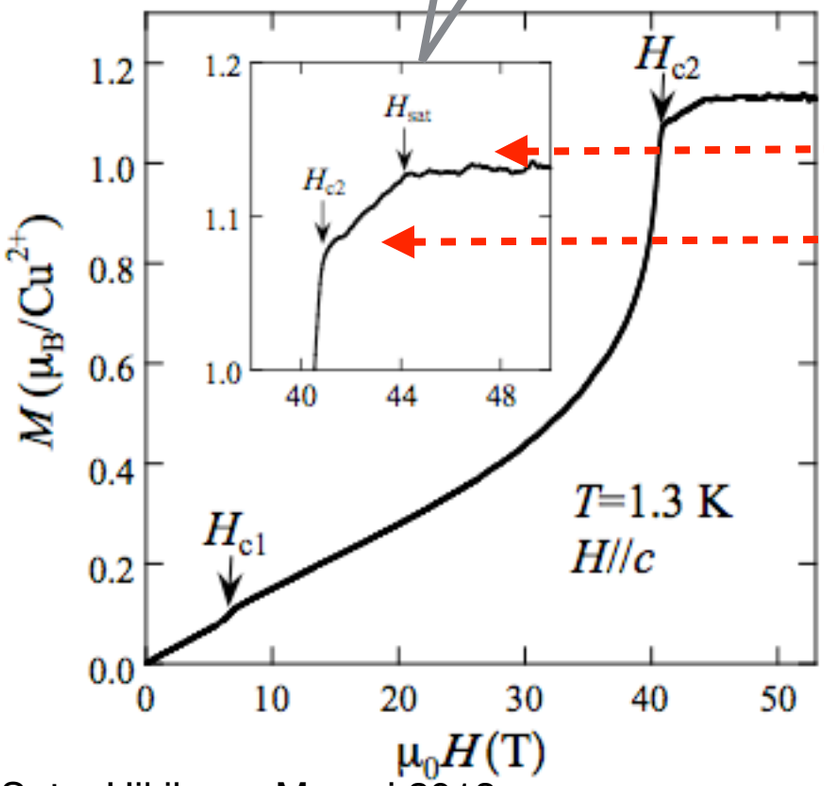
- describes potential energy of interaction between magnon pairs on neighboring chains, quadratic in magnon pair density $n_{\rm pair}$

- Competition $\frac{(J')^2}{J_1} n_{\rm pair} \sim J' n^2_{\rm pair}, \;\; \text{hence} \boxed{n^*_{\rm pair} \sim J'/J_1}$

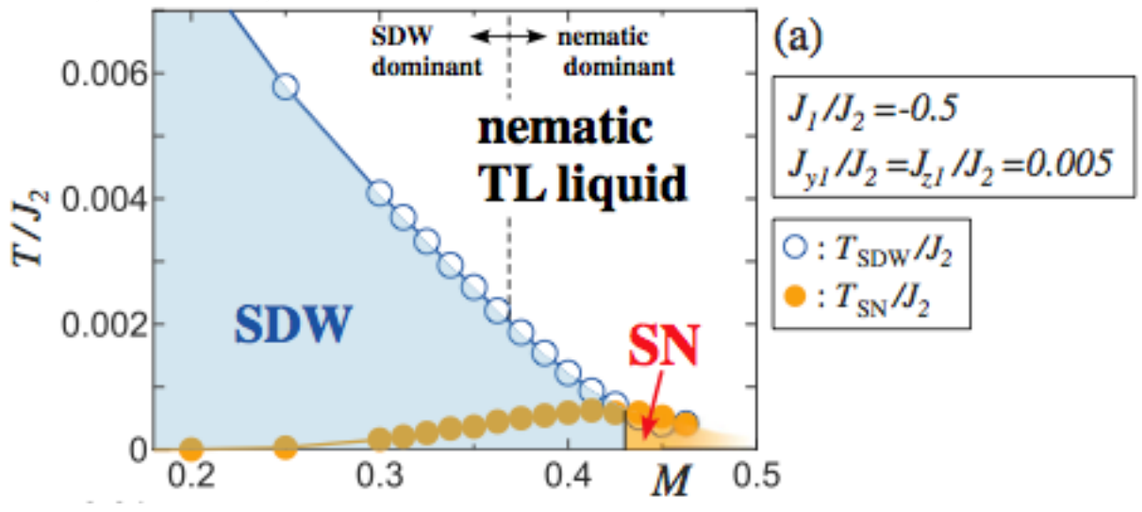
- Hence:
- Spin Nematic *near saturation*, for $n_{pair} < n^*_{pair}$
- SDW for $n_{pair} > n^*_{pair}$

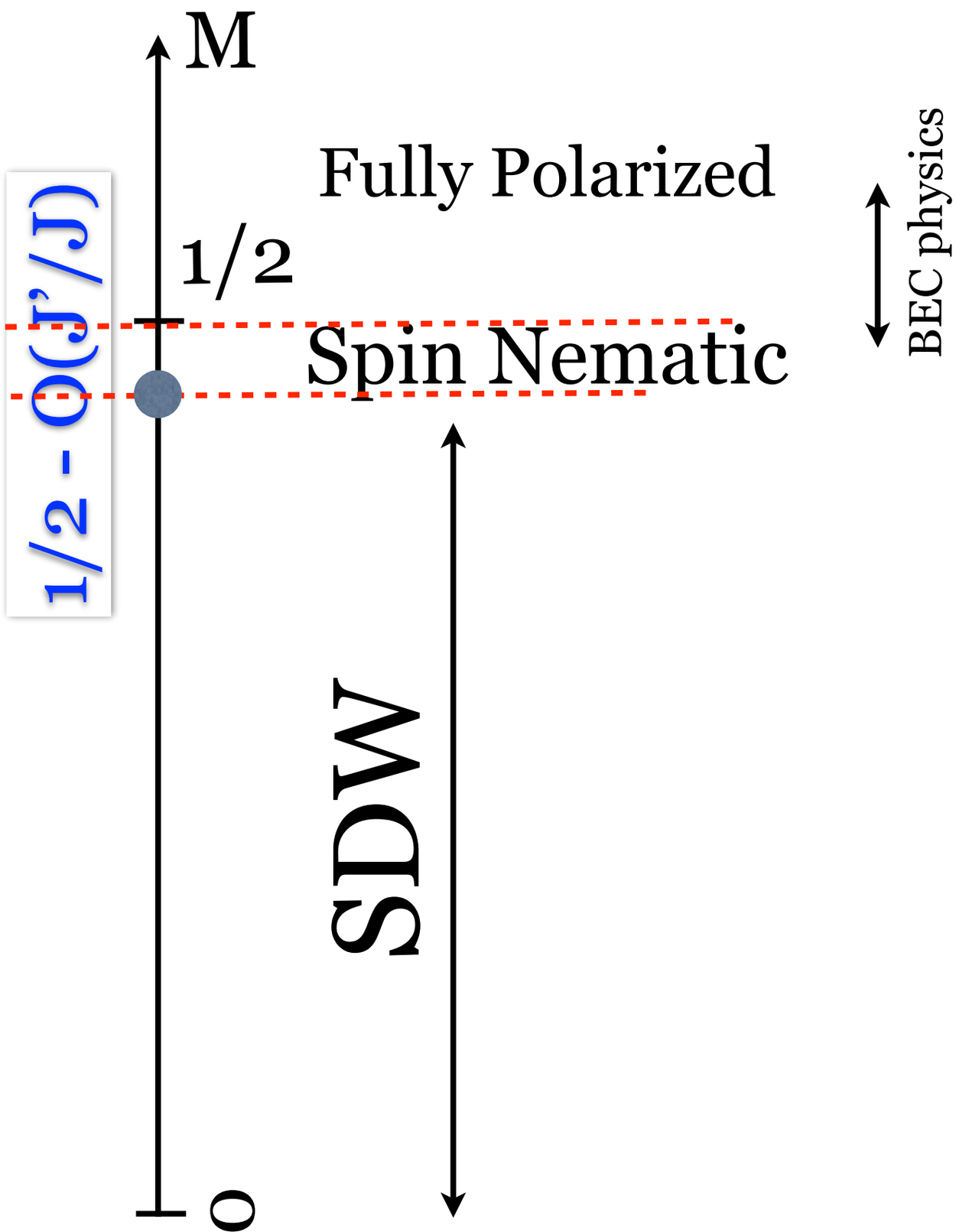
# T=0 schematic phase diagram of weakly coupled **nematic** spin chains

Cautionary remark:
maybe impurity effect



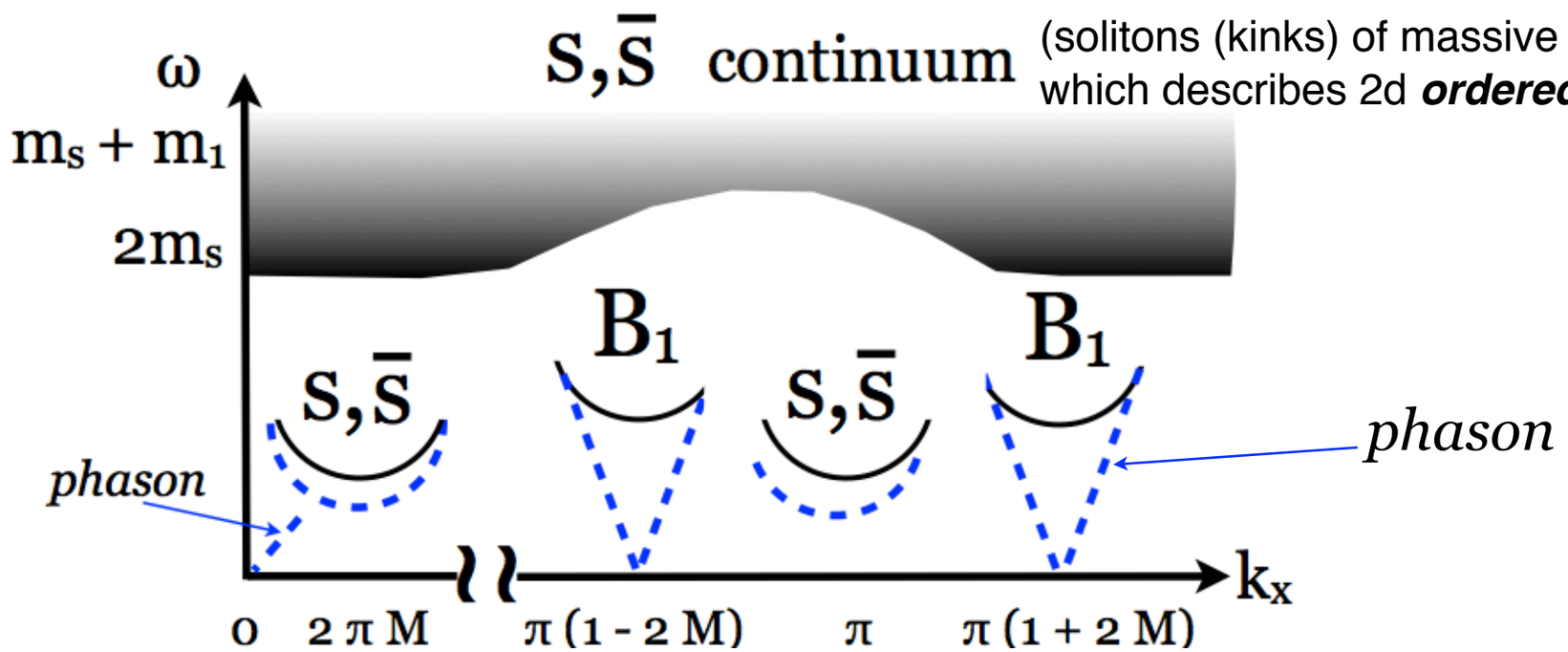

cf: Sato, Hikihara, Momoi 2013



## Excitations (via spin-spin correlation functions)

- 2d SDW $\langle S^z(\mathbf{r})\rangle = M + \mathrm{Re}\left(\Phi e^{i\mathbf{k}_{\mathrm{sdw}}\cdot\mathbf{r}}\right)$
- preserves U(1) [with respect to magnetic field] -> hence NO transverse spin waves
- breaks translational symmetry -> longitudinal phason mode at $k_{sdw} = \pi(1\text{-}2M)$ and k=0




(solitons (kinks) of massive sine-Gordon model which describes 2d ***ordered*** state)

OS, Balents PRB 2014

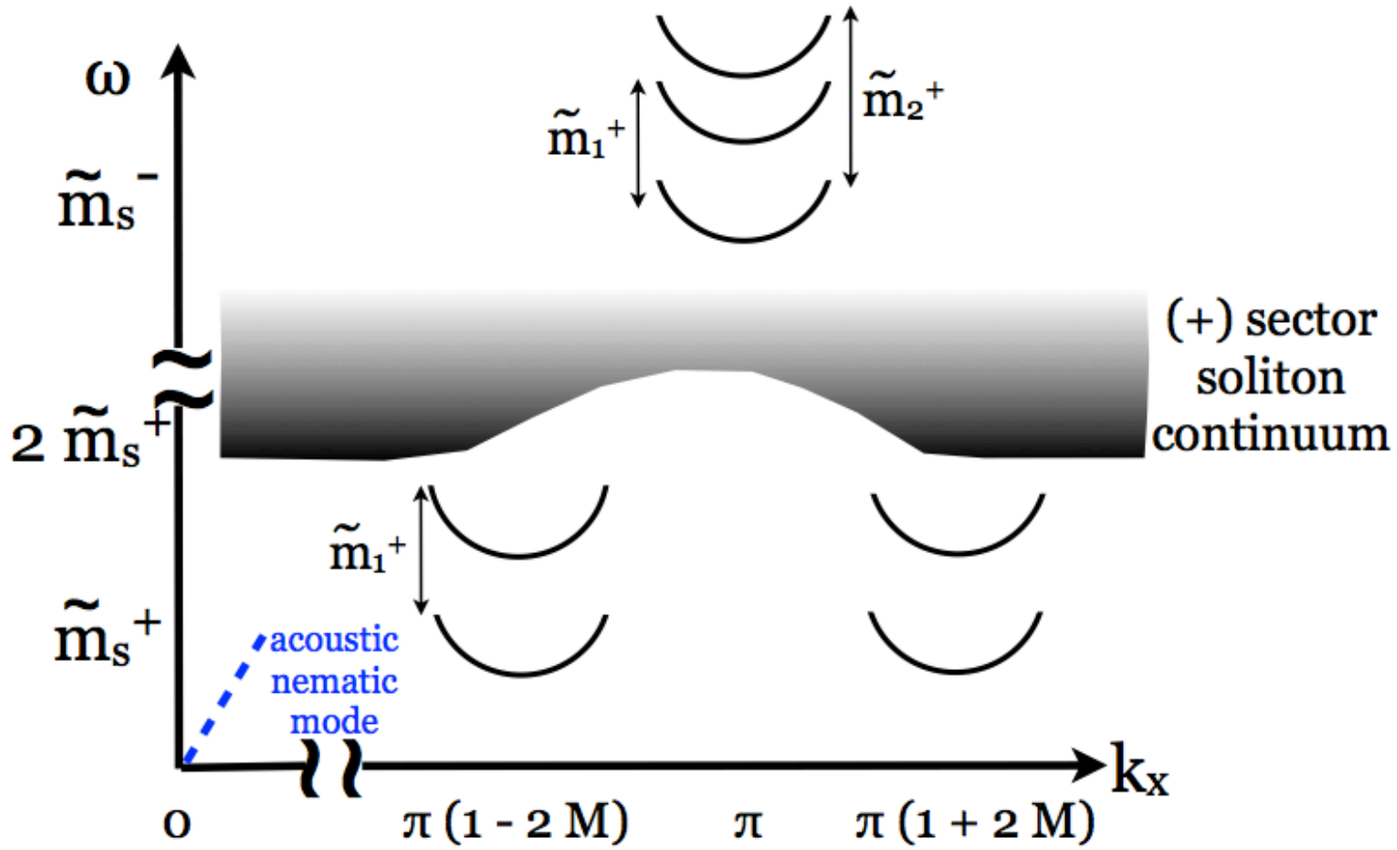
# Excitations (via spin-spin correlation functions)

- 2d Spin Nematic $\langle S^+(r)S^+(r')\rangle \sim \Psi \neq 0$

- breaks U(1) but ΔS=1 excitations are gapped (magnon superconductor) $\langle S^+(\mathbf{r})\rangle = 0$

- 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

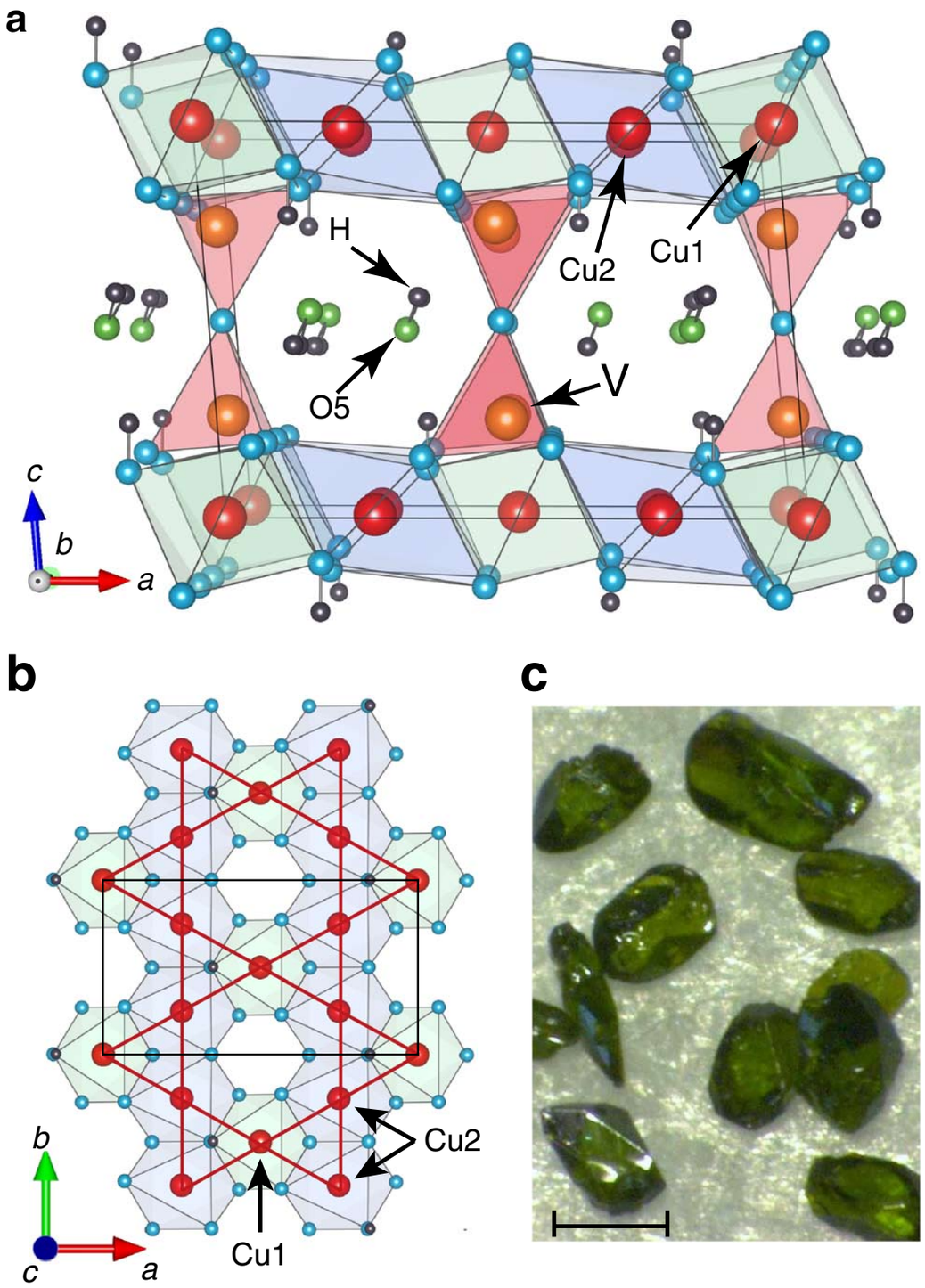
# Intermediate Summary

- 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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  - ✓ 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 **Colour:** Olive-green, ...

**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.

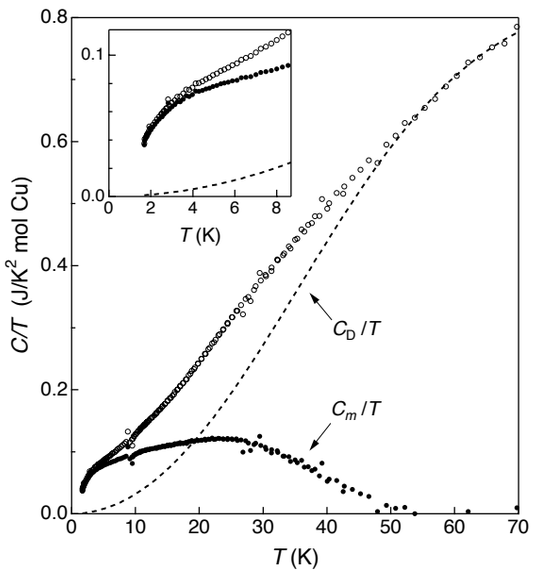
Visually similar to vésigniéite.

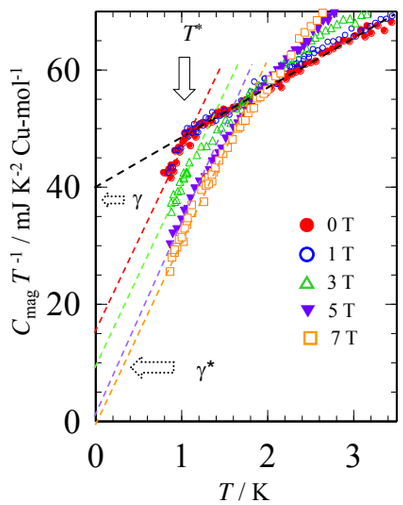
2001 quantum spin liquid?!

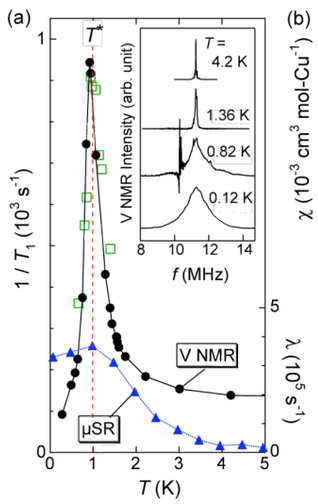
2009 impurity ordering at low T? magnetization steps?

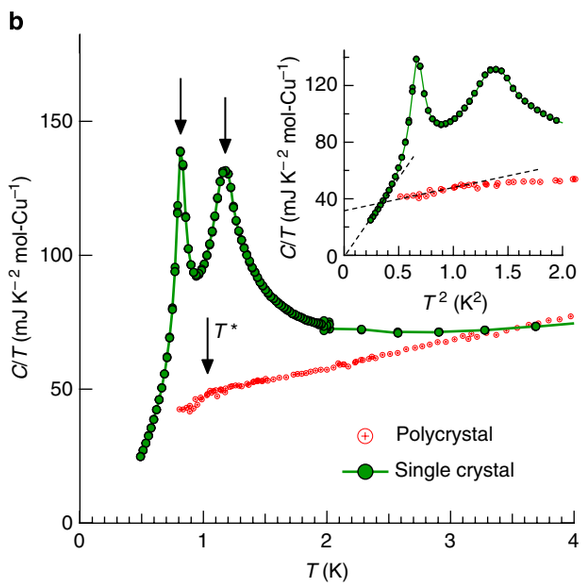
2012 magnetic order !

2014 magnetization plateau




*time = material quality*

# 2014: huge plateau!

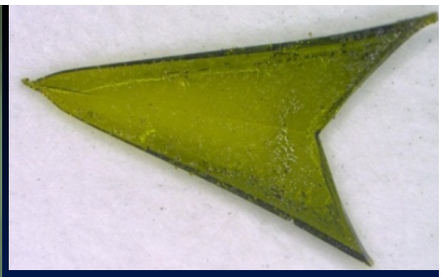
H. Ishikawa…M.Takigawa…Z.Hiroi, unpublished, 2014

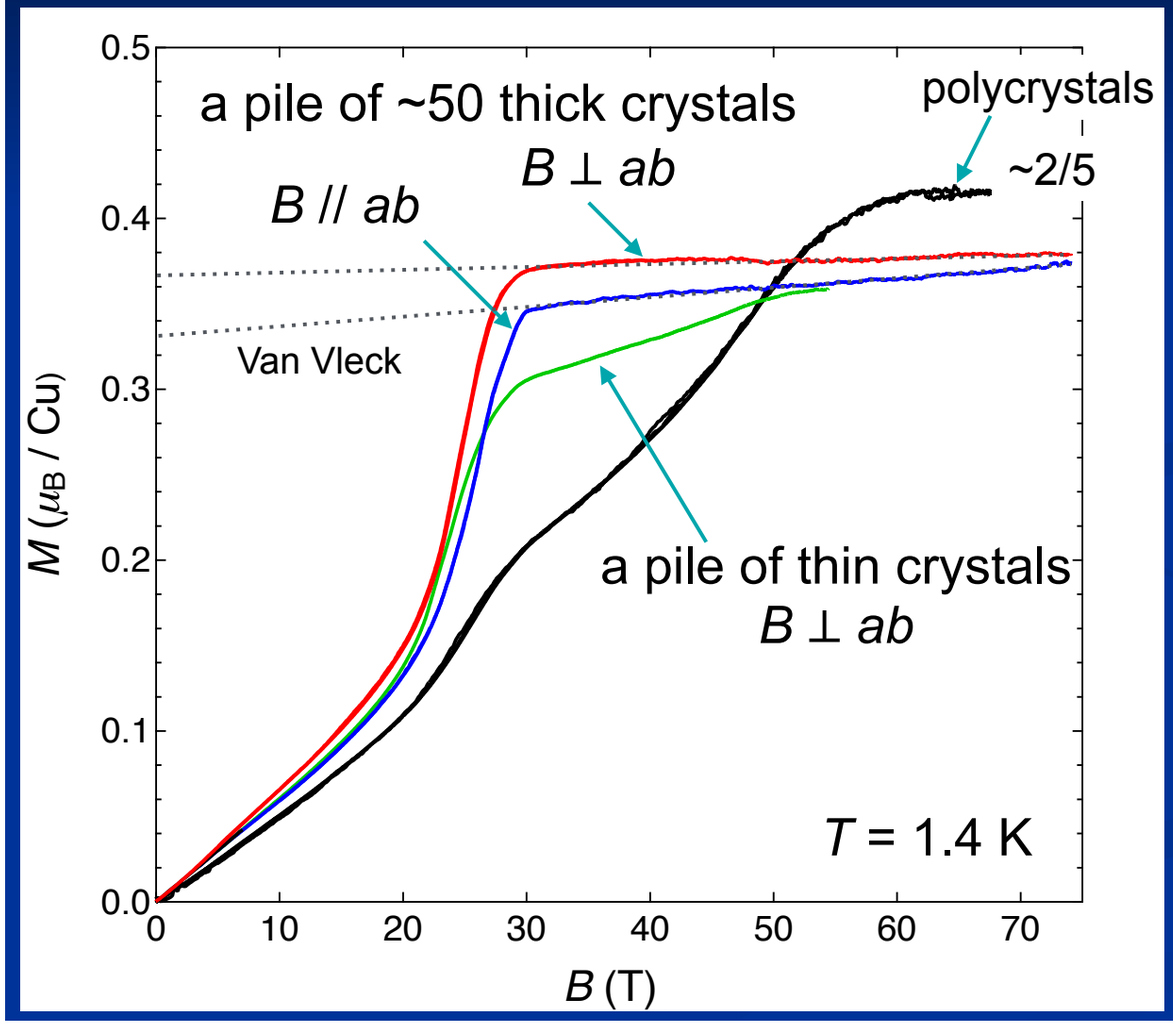
## High-field magnetization

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

30 days growth








Huge 1/3 plateau!

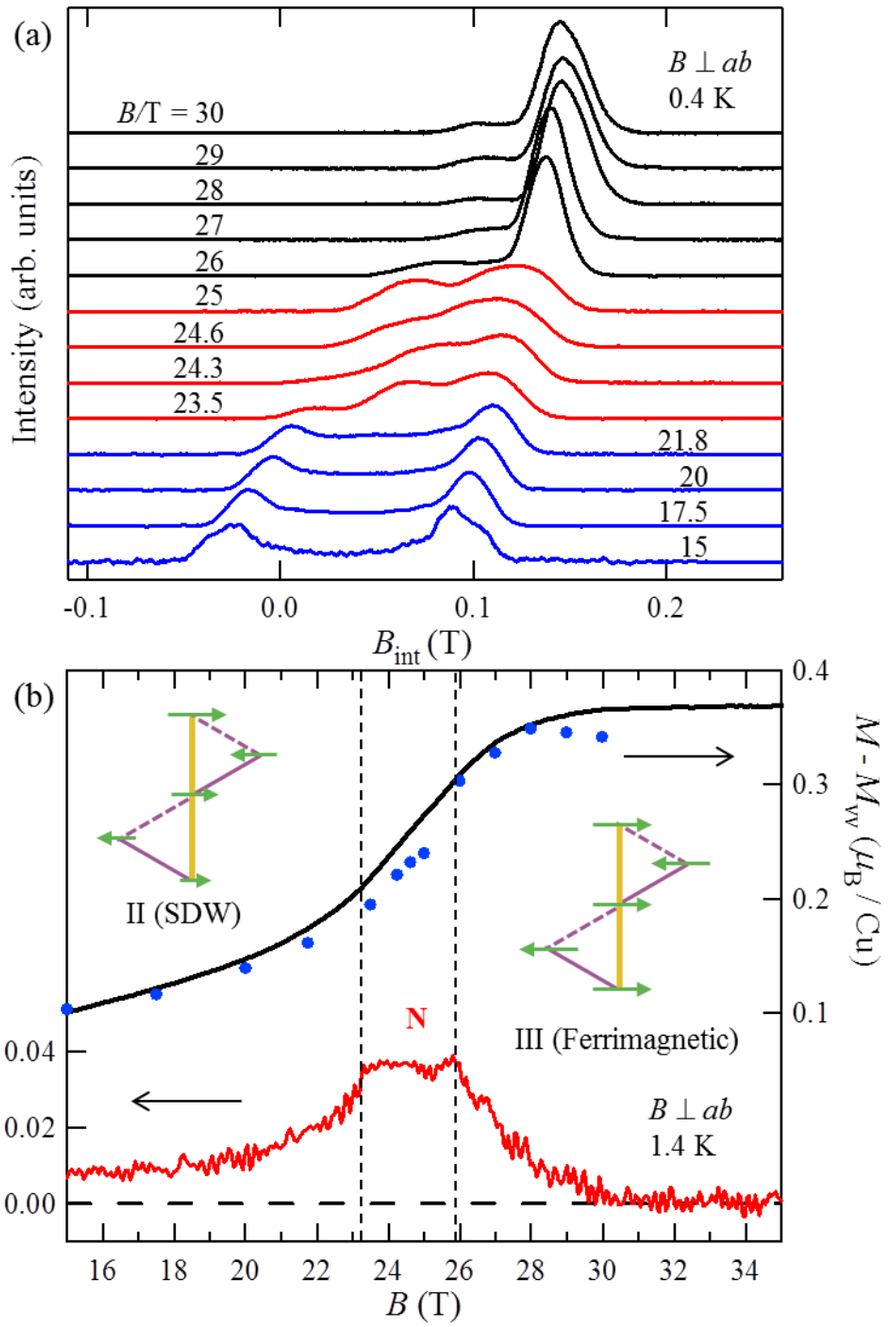
further optical meas.
@ Takeyama lab
It survives over 120 T!

*Kagome plateau or ferrimagnetic state?*

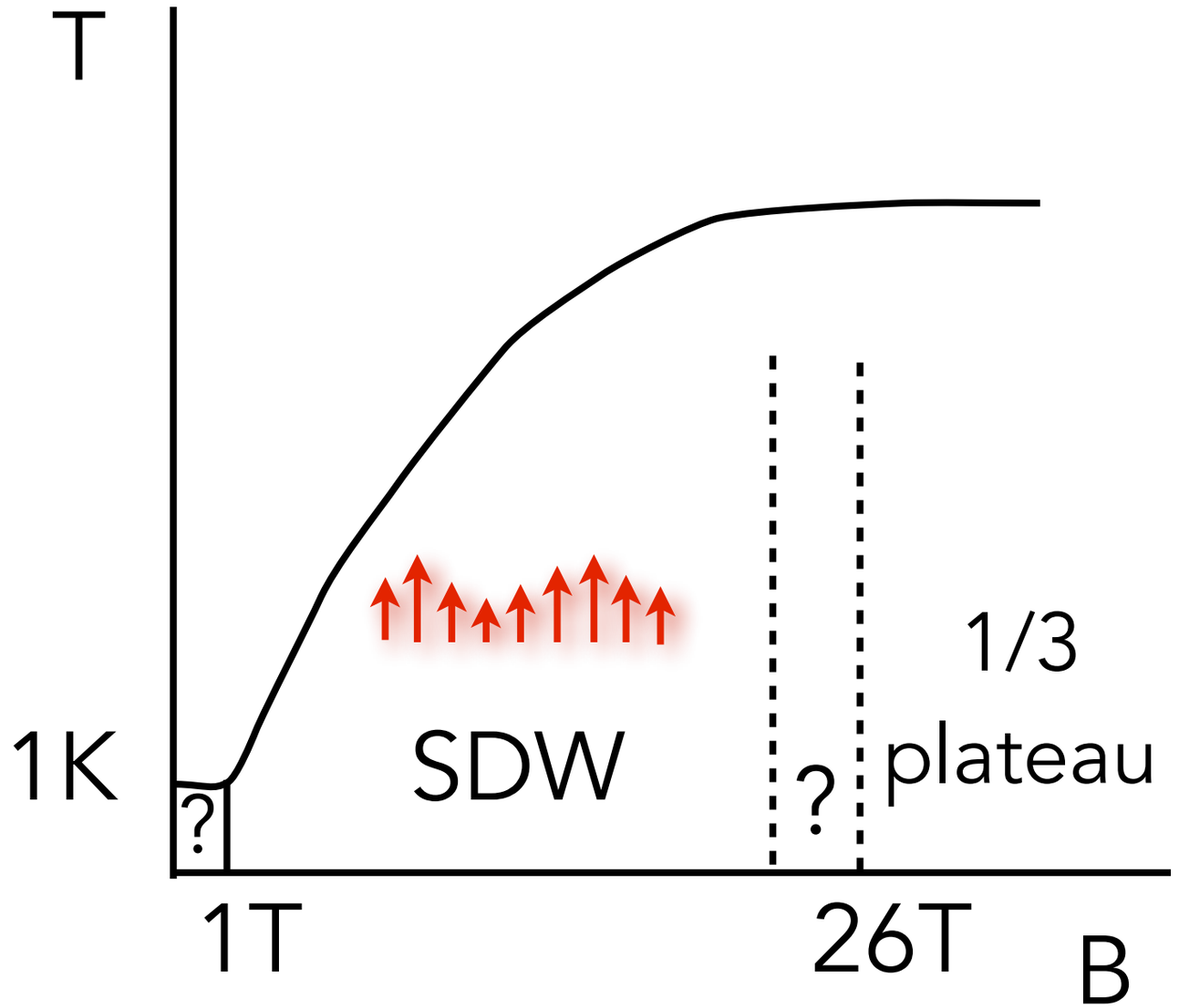
*coupled to lattice, but already distorted*

high-field mag. meas.
@ Tokunaga & Kindo labs

# Phase diagram




H. Ishikawa *et al*, unpublished



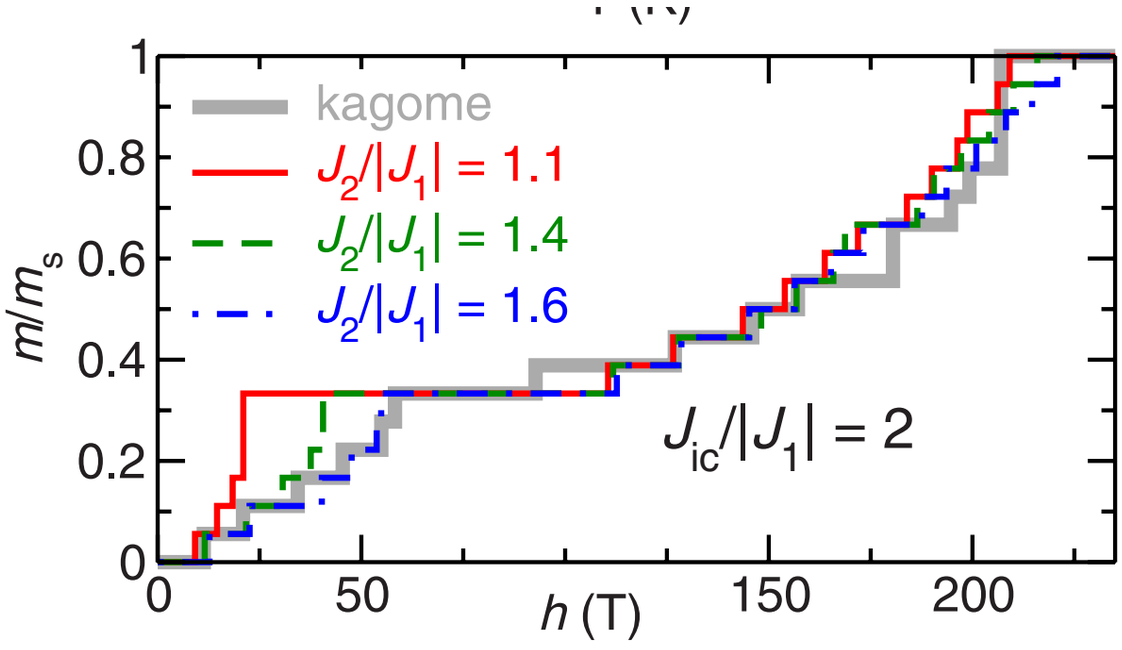

our interpretation

# Frustrated ferromagnetism

PHYSICAL REVIEW B **82**, 104434 (2010)

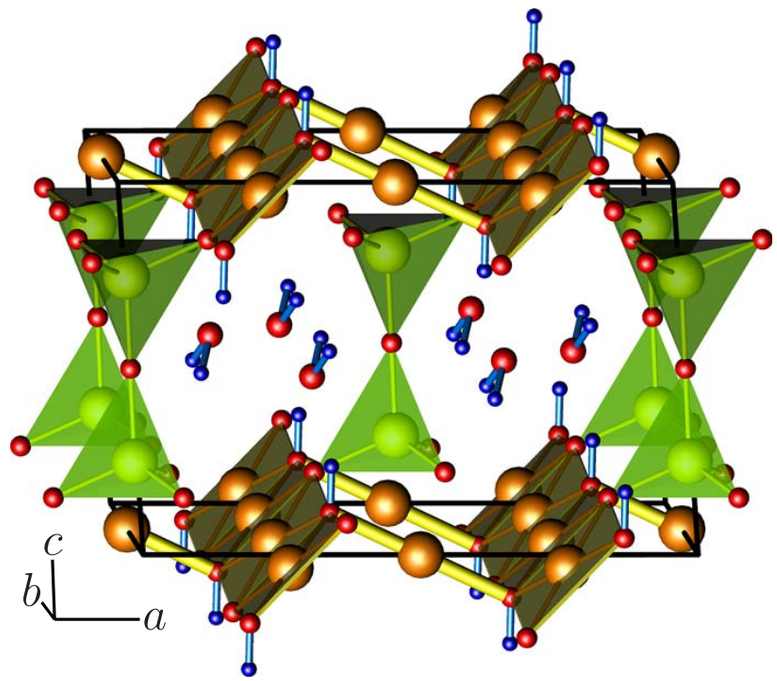
**Coupled frustrated quantum spin-$\frac{1}{2}$ chains with orbital order in volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$**

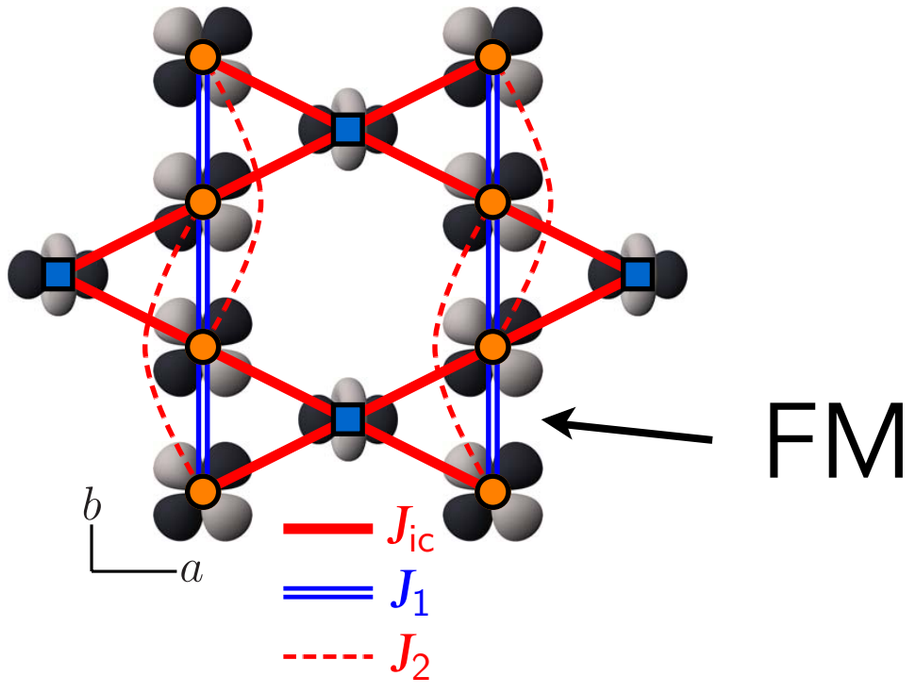
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(Received 9 August 2010; published 30 September 2010)




DFT gets it right!

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



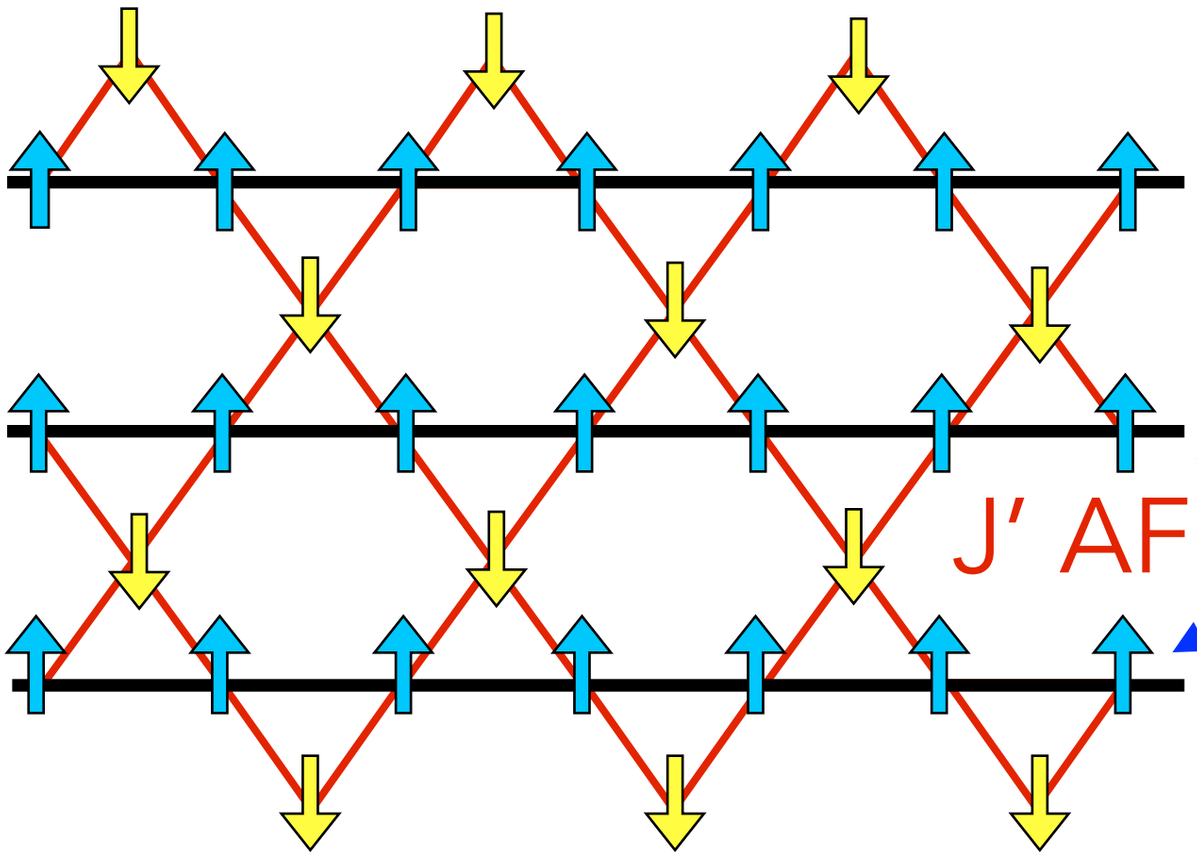
# Ferrimagnetic state

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**Coupled frustrated quantum spin-$\frac{1}{2}$ chains with orbital order in volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$**

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

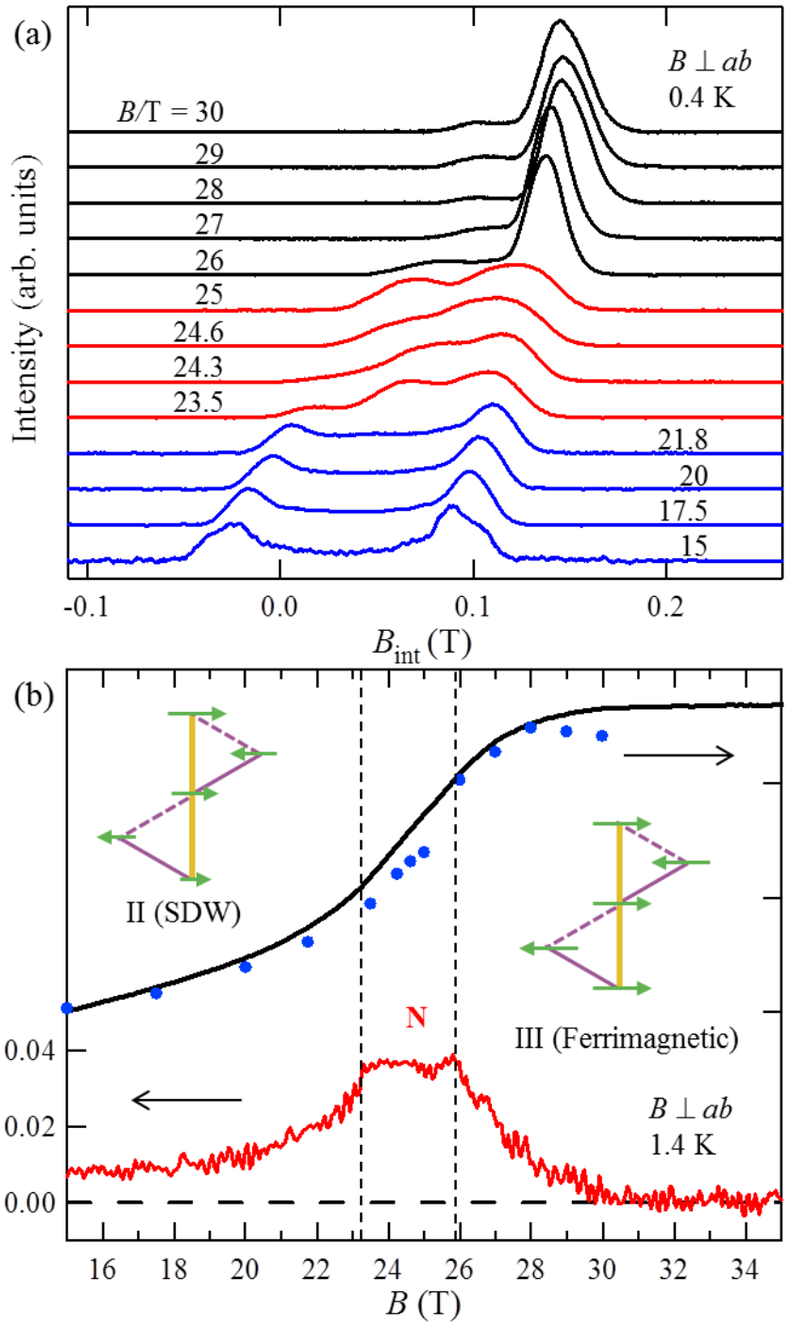


$J_1$ FM, $J_2$ AF

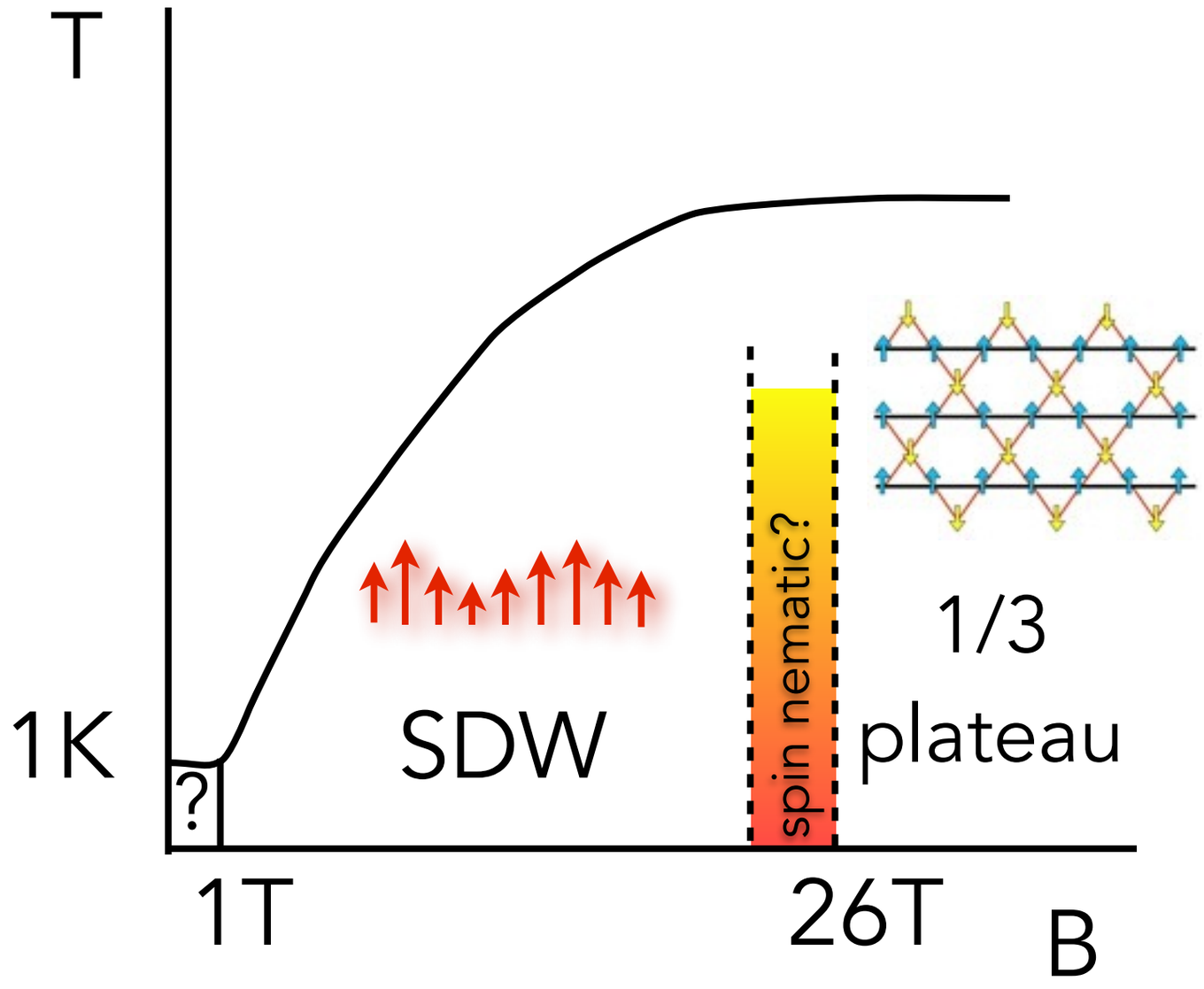
J’ AF

polarized chains?!

# Phase diagram



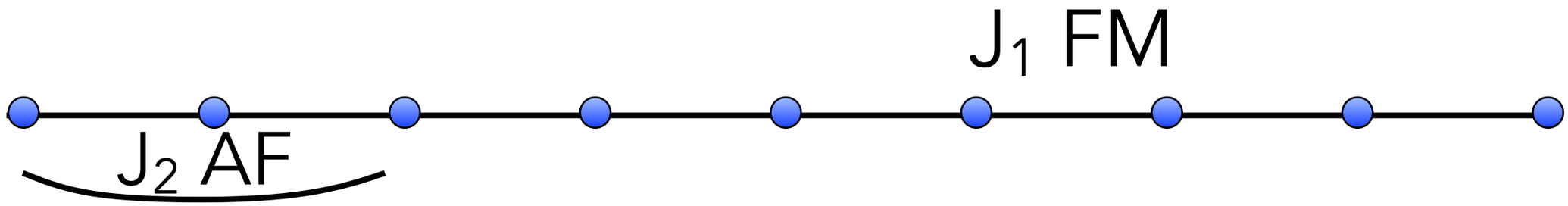

H. Ishikawa *et al*, unpublished



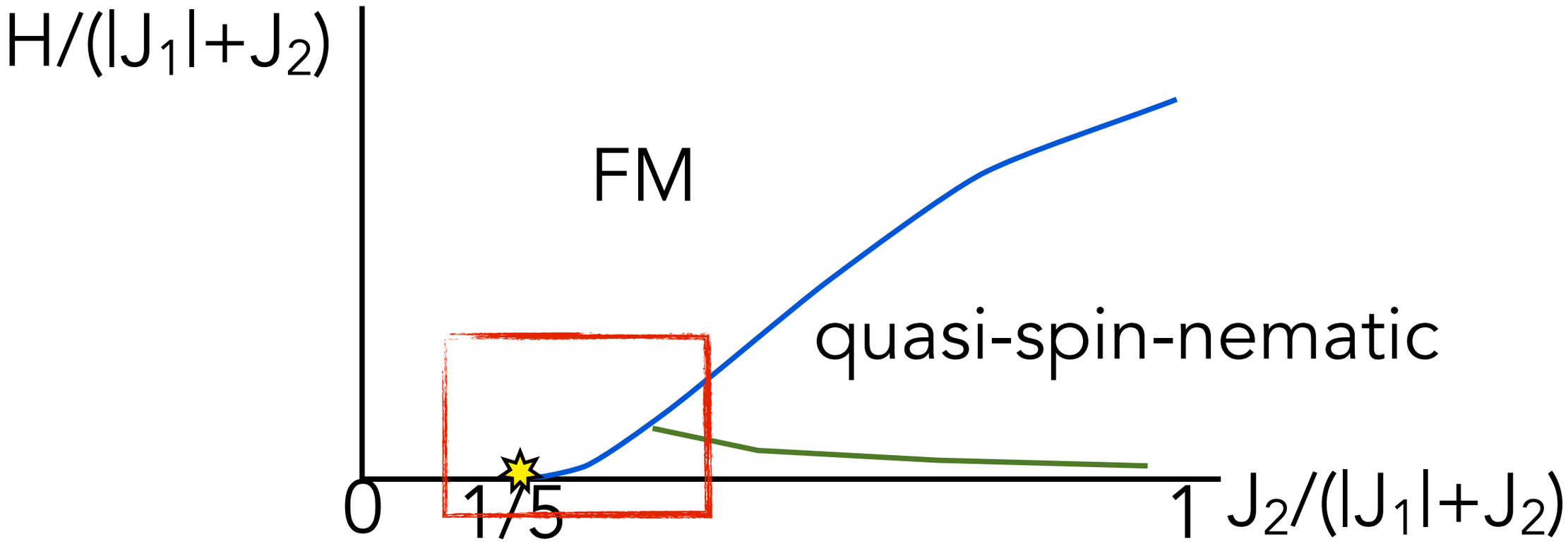

may be a spin nematic??

# Spin chain redux

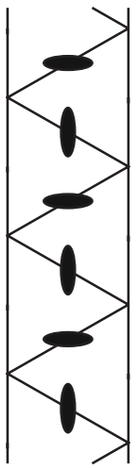
Frustrated ferromagnetic chain




$$H = J_1 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2} - h \sum_i S_i^z$$



# Quasi-1d nematic



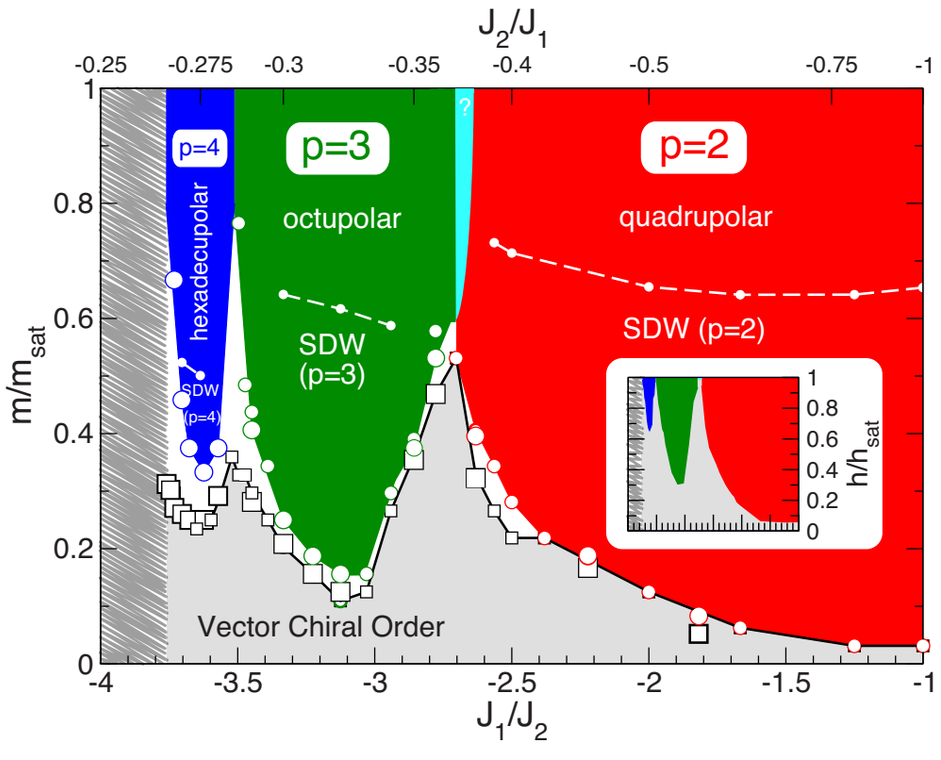
1d $J_1$-$J_2$ chain is only *quasi*-spin-nematic

power-law correlations

$\Psi \sim (S^+)^2$ : spin-nematic

$\phi \sim S^z e^{i q x}$ : SDW

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

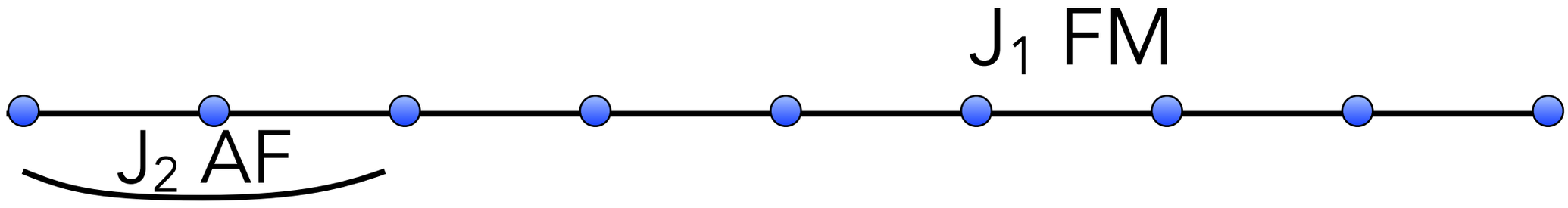



$\Psi$ "dominant"

$\phi$ "dominant"

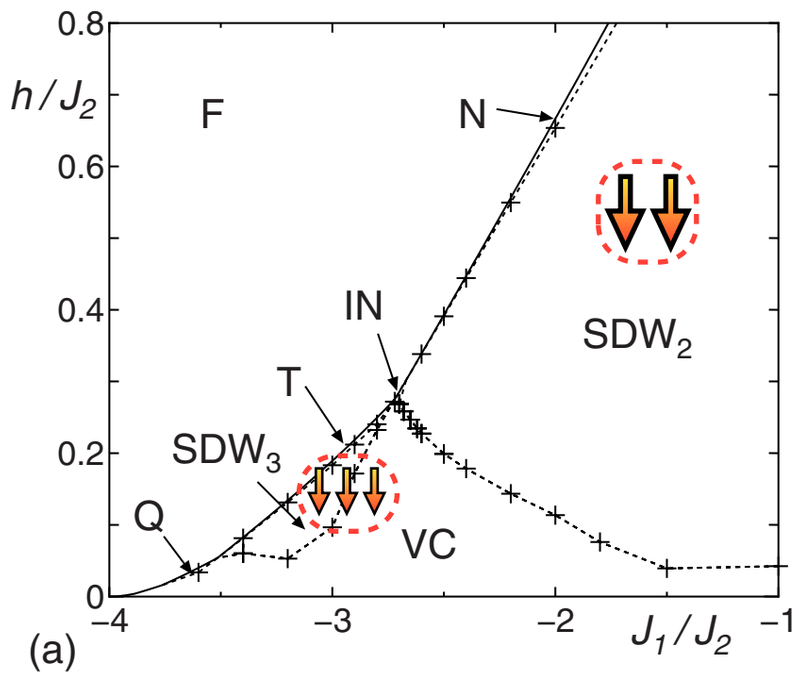
# Multipolar phases

Frustrated ferromagnetic chain




$$H = J_1 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2} - h \sum_i S_i^z$$

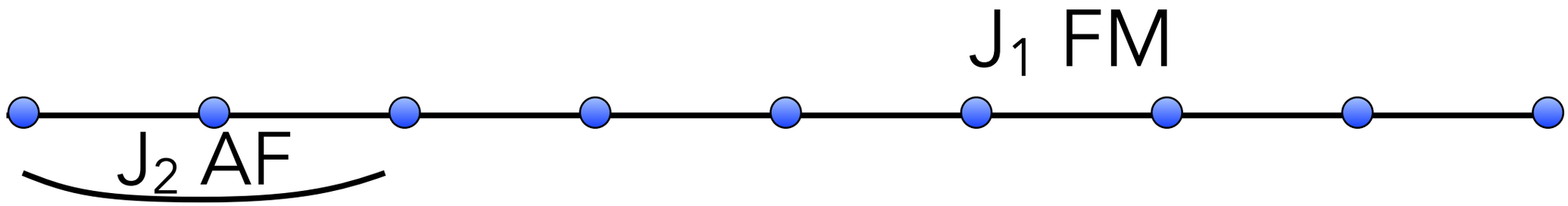
Hikihara *et al*, 2008



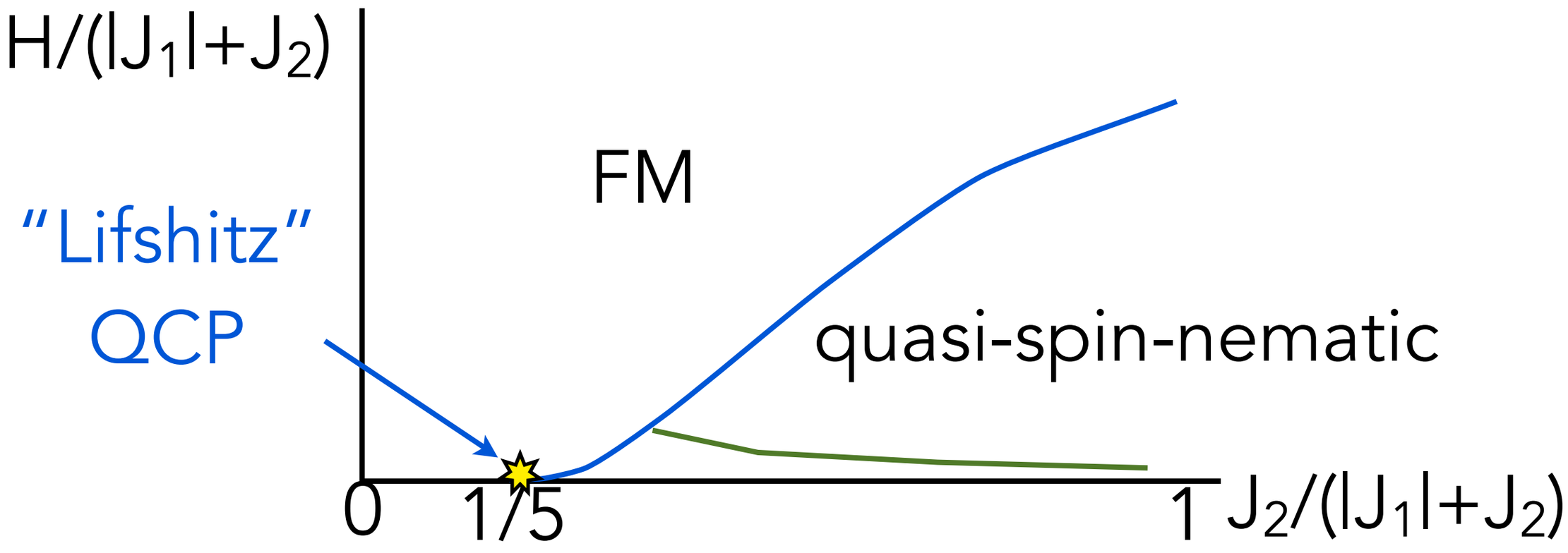

Is it an infinite progression?

# A QCP parent?

Frustrated ferromagnetic chain




$$H = J_1 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2} - h \sum_i S_i^z$$



# Lifshitz Point

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

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

$\mathcal{A}_B = \frac{\hat{m}_1 \partial_\tau \hat{m}_2 - \hat{m}_2 \partial_\tau \hat{m}_1}{1 + \hat{m}_3}$ Berry phase term

tunes QCP

two symmetry allowed interactions at $O(q^4)$

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

# Lifshitz Point

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

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

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

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

Large parameter:
saddle point!

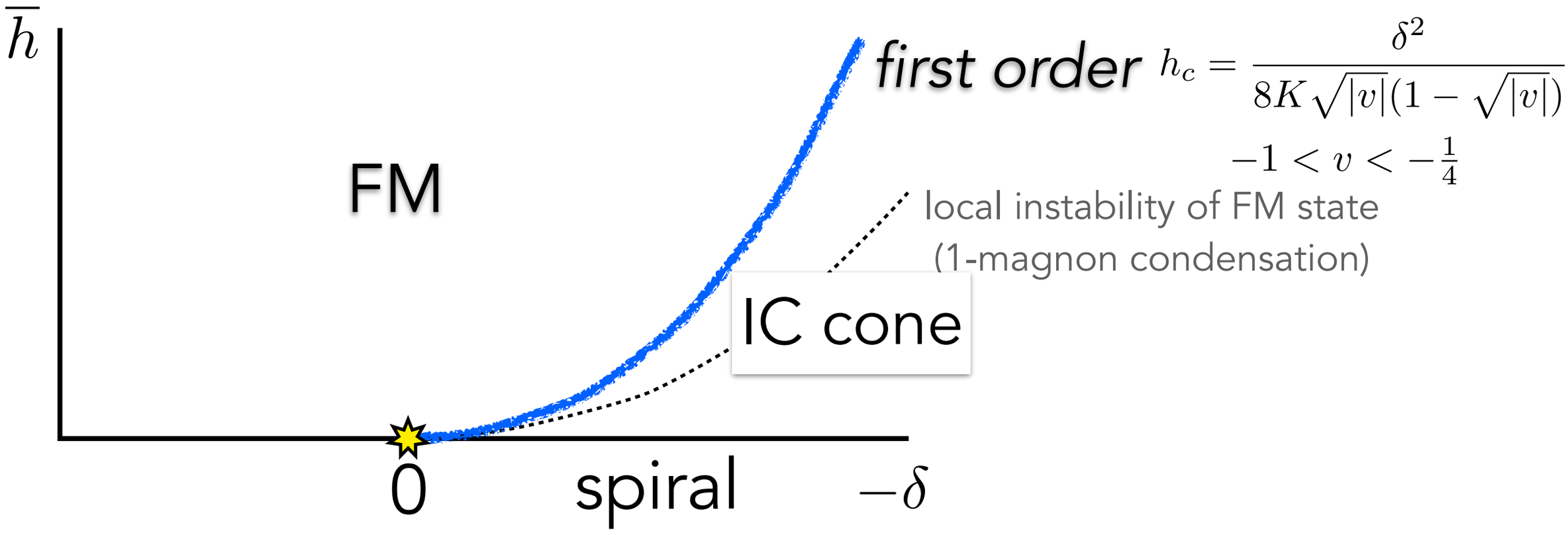
$$v = \frac{u}{K} \qquad \overline{h} = \frac{hK}{\delta^2}$$

# Saddle point

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

v derives from quantum fluctuations

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




*first order* $h_c = \dfrac{\delta^2}{8K\sqrt{|v|}(1-\sqrt{|v|})}$

$-1 < v < -\frac{1}{4}$

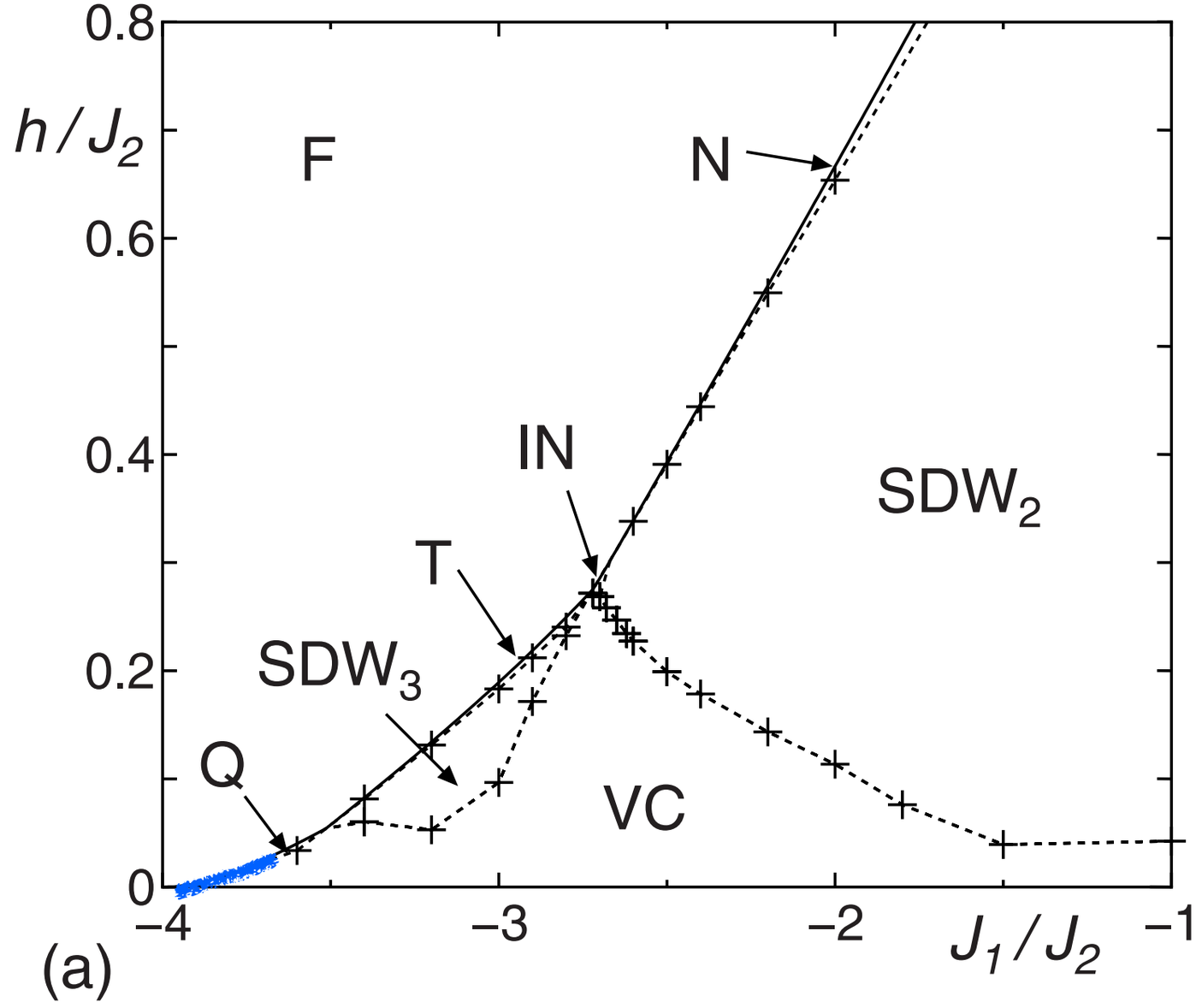
# Phase diagram

Frustrated ferromagnetic chain

First order metamagnetic transition near Lifshitz point

Higher dimensions?

Hikihara et al, 2008



# d>1

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

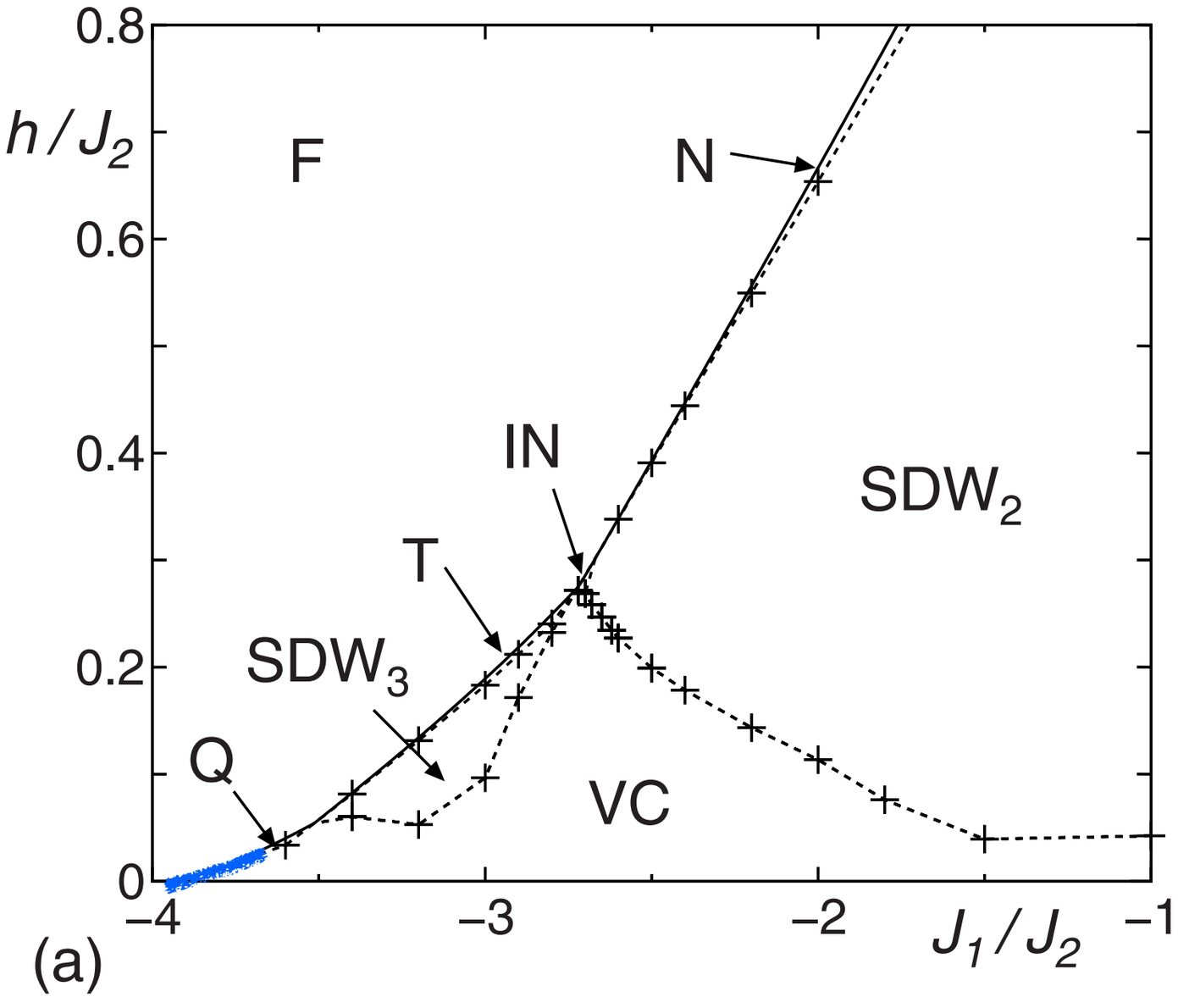
- Rescaling:

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

$$S = \frac{\sqrt{K^d C^{d-1}}}{\delta^{d-1/2}} \int dx d^{d-1} y d\tau \{ is\mathcal{A}_B[\hat{m}] + \mathrm{sgn}(\delta)|\partial_x \hat{m}|^2 + |\partial_x^2 \hat{m}|^2 + |\partial_y \hat{m}|^2 + v|\partial_x \hat{m}|^4 - \overline{h}\hat{m}_z \}$$

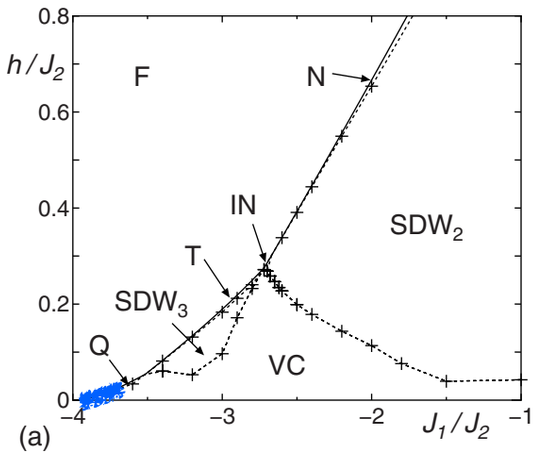
∴ Similar theory applies in d>1, and very similar conclusions apply

# Phase diagram




multipolar phases
from QCP?

# Origin of multipolar phases



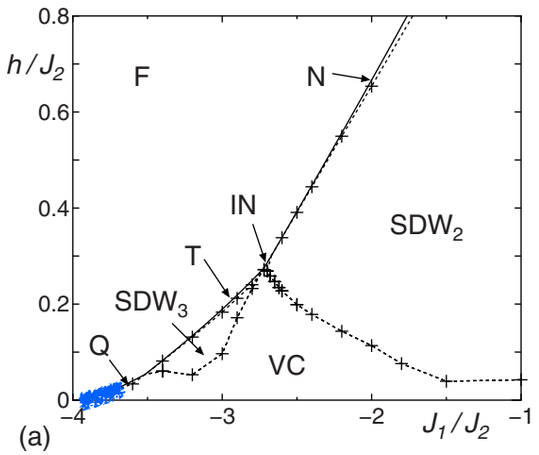


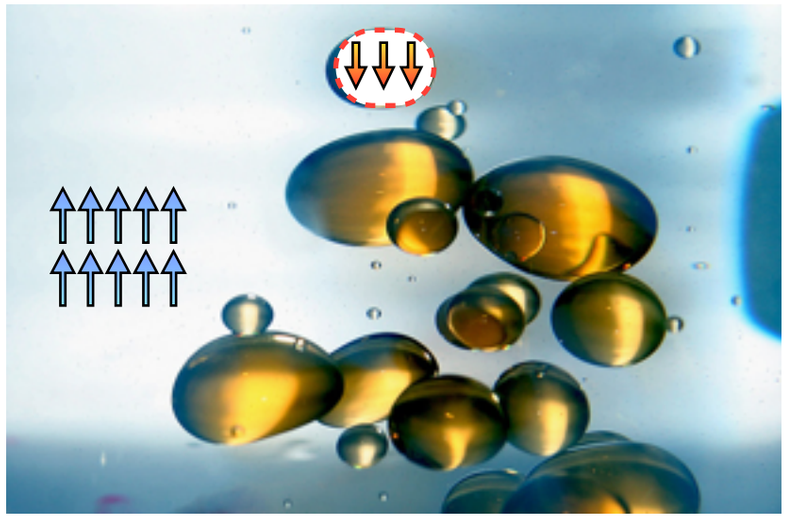



First order transition: partially polarized state *coexists* with plateau one

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