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

Oleg Starykh, University of Utah
Leon Balents, KITP, UCSB
Edward Parker, UCSB



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





Exotic ordered phases

spin nematic

composite order parameter

$$\mathcal{O}^{\alpha\beta}(\mathbf{r}_i, \mathbf{r}_j) = \tfrac{1}{2}(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) - \tfrac{1}{3}\delta^{\alpha\beta}\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$$

ordered states

Usual

quantum spin liquids

Exotic

Outline

- Motivation: usual *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

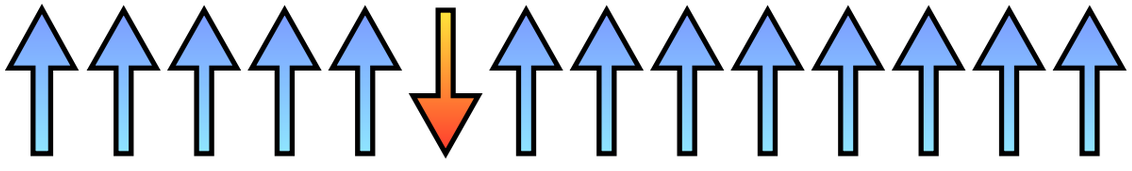
– experimental status - magnetization plateau

– UUD, SDW and ***more***

✴ *Field theory of the Lifshitz point*

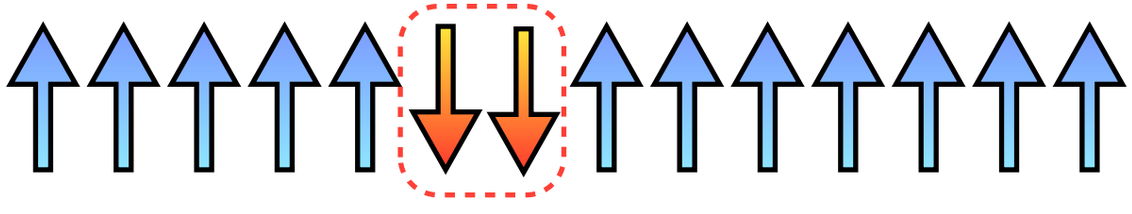
Magnon binding and pair condensation route to nematic phases

1-magnon

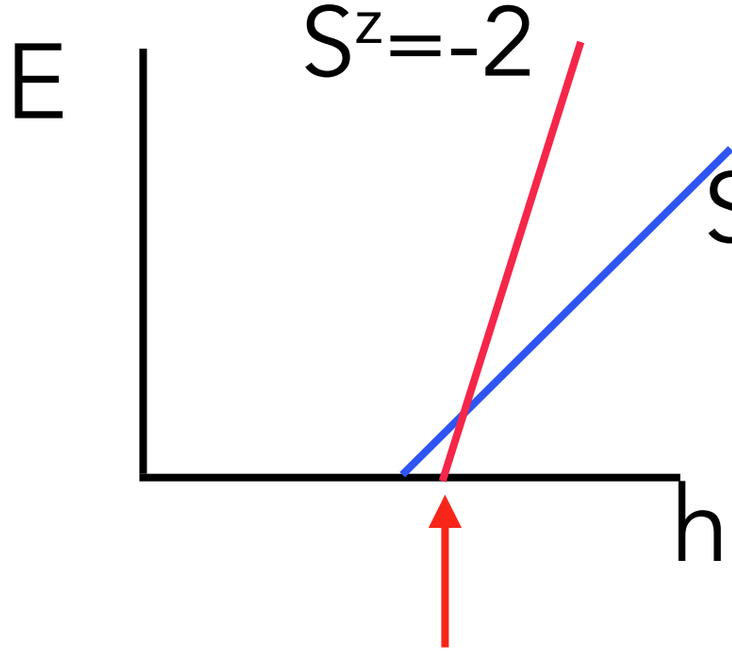


$E\text{-}E_{FM} = \varepsilon_1 + h$

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 $\langle S_i^+ \rangle = 0$

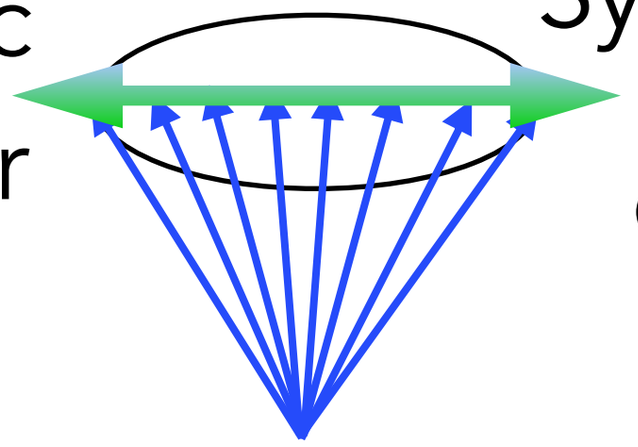
$\langle S_i^+ S_j^- \rangle \sim e^{-|i-j|/\xi}$ S^z=1 gap

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

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

quasi-1d

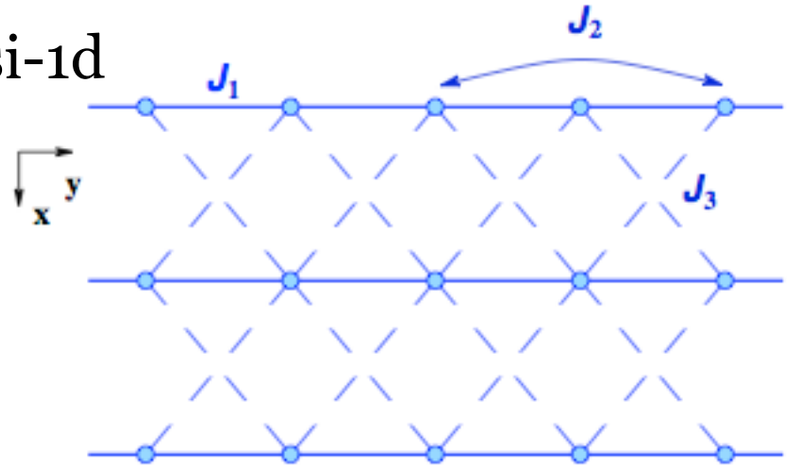


Fig. 2: (Color online) Two-dimensional array of copper ions in $LiCuVO_4$ with principal exchange couplings.

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

2d

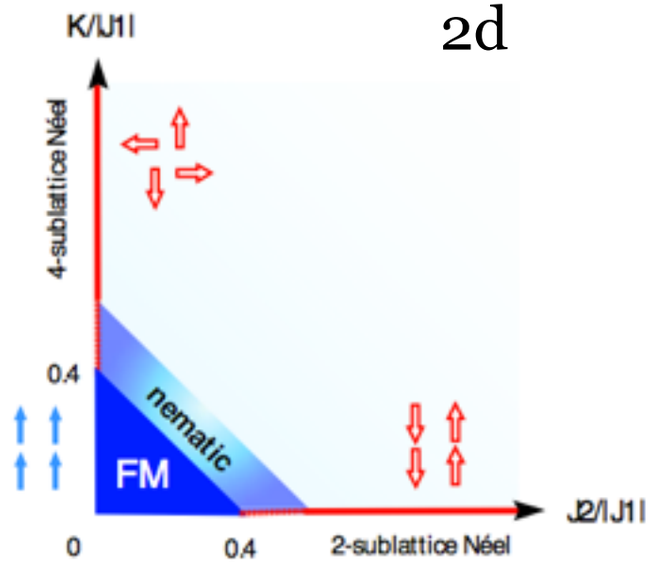
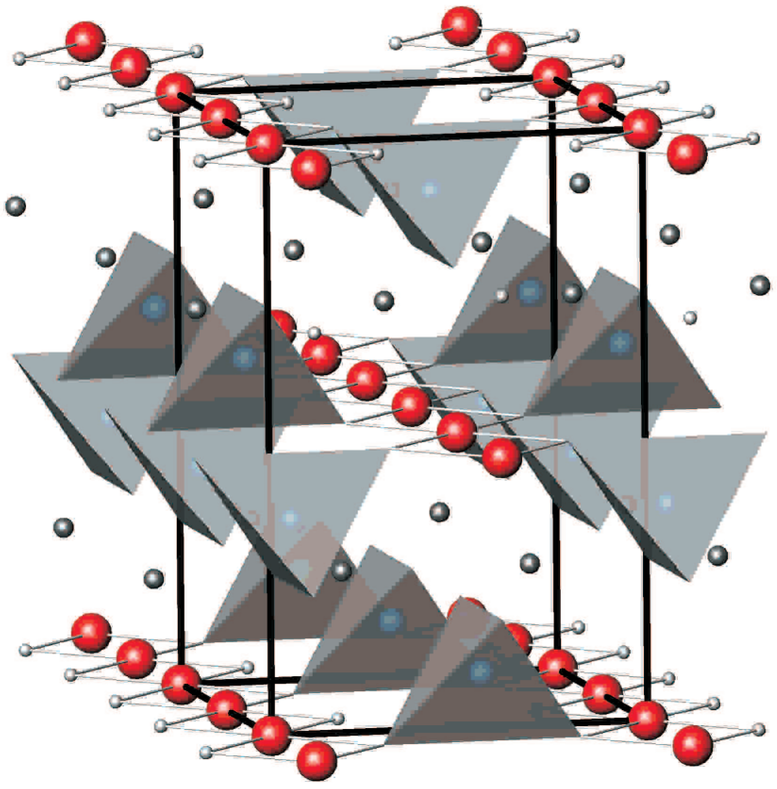
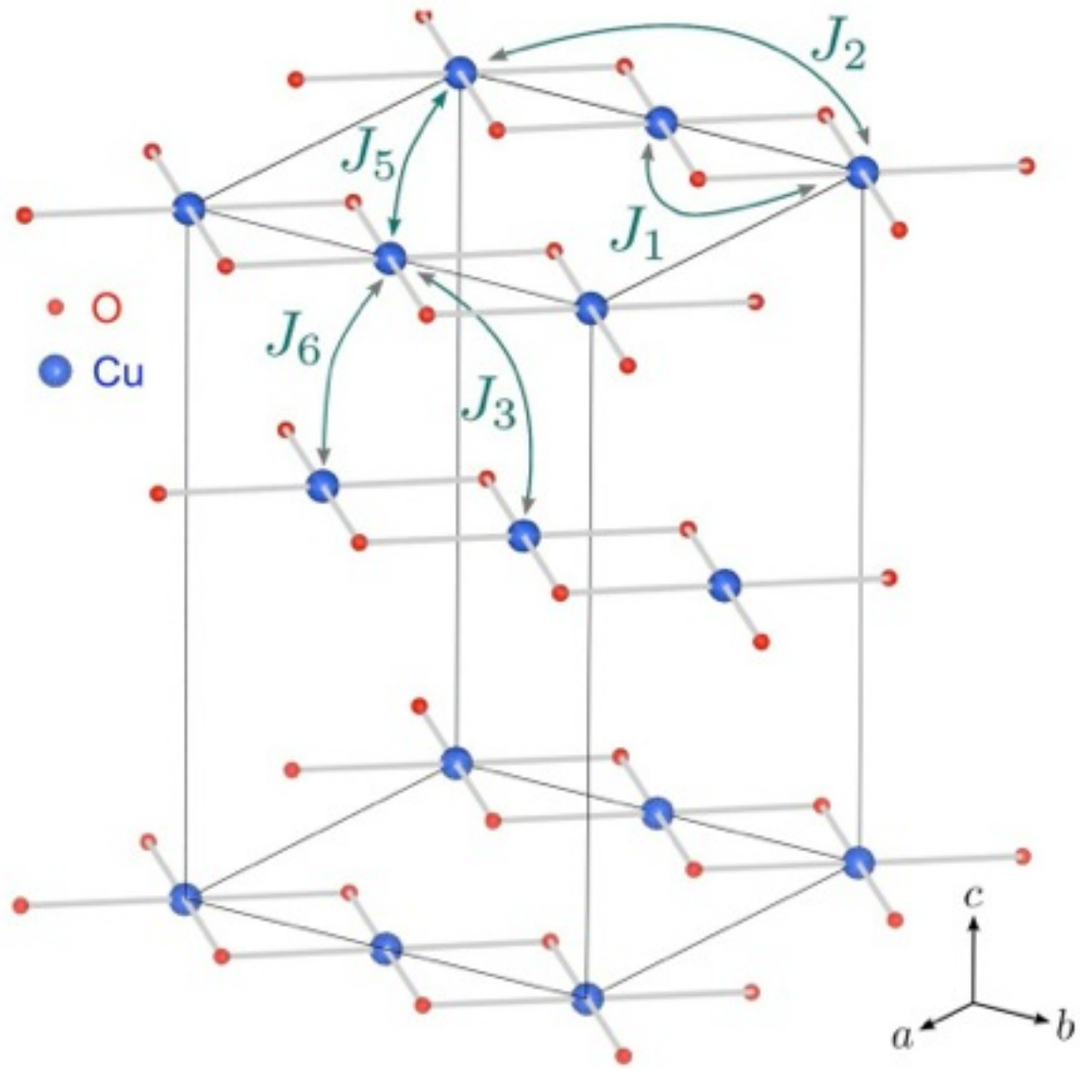


FIG. 1 (color online). Phase diagram for the spin-1/2 J_1-J_2-K Heisenberg model on a square lattice, in the parameter range where $J_1 < 0$ is FM and both $J_2 \geq 0$ and $K \geq 0$ are AF. Solid lines on the $J_2/|J_1|$ and $K/|J_1|$ axes denote known Néel phases. A single nematic phase interpolates between the FM occurring for large FM J_1 and these two, different, AF ordered states.

Shannon, Momoi, Sindzingre PRL 2006

$LiCuVO_4$: spin nematic?





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

+ 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 **B**

Hagiwara, Svistov et al, 2011

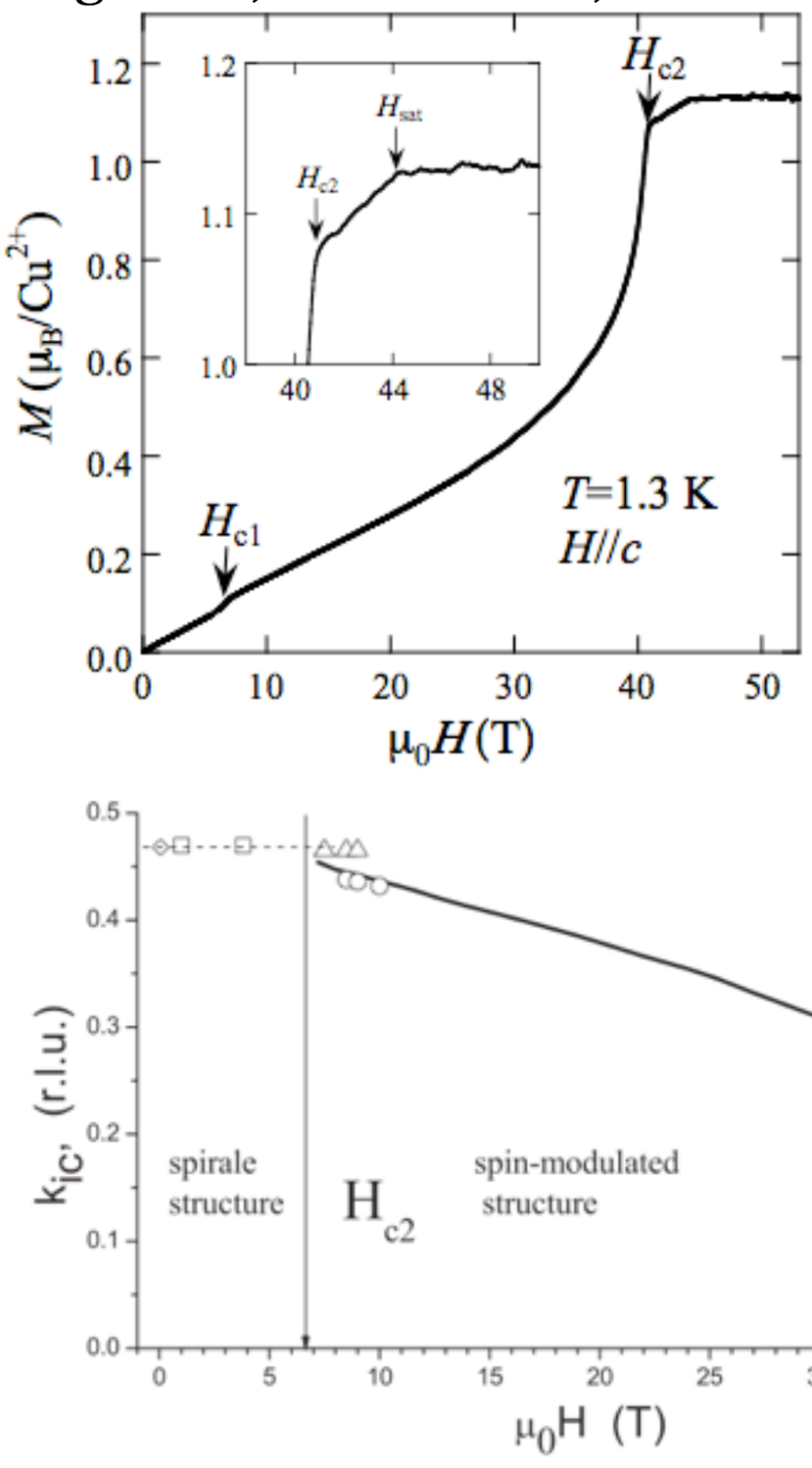
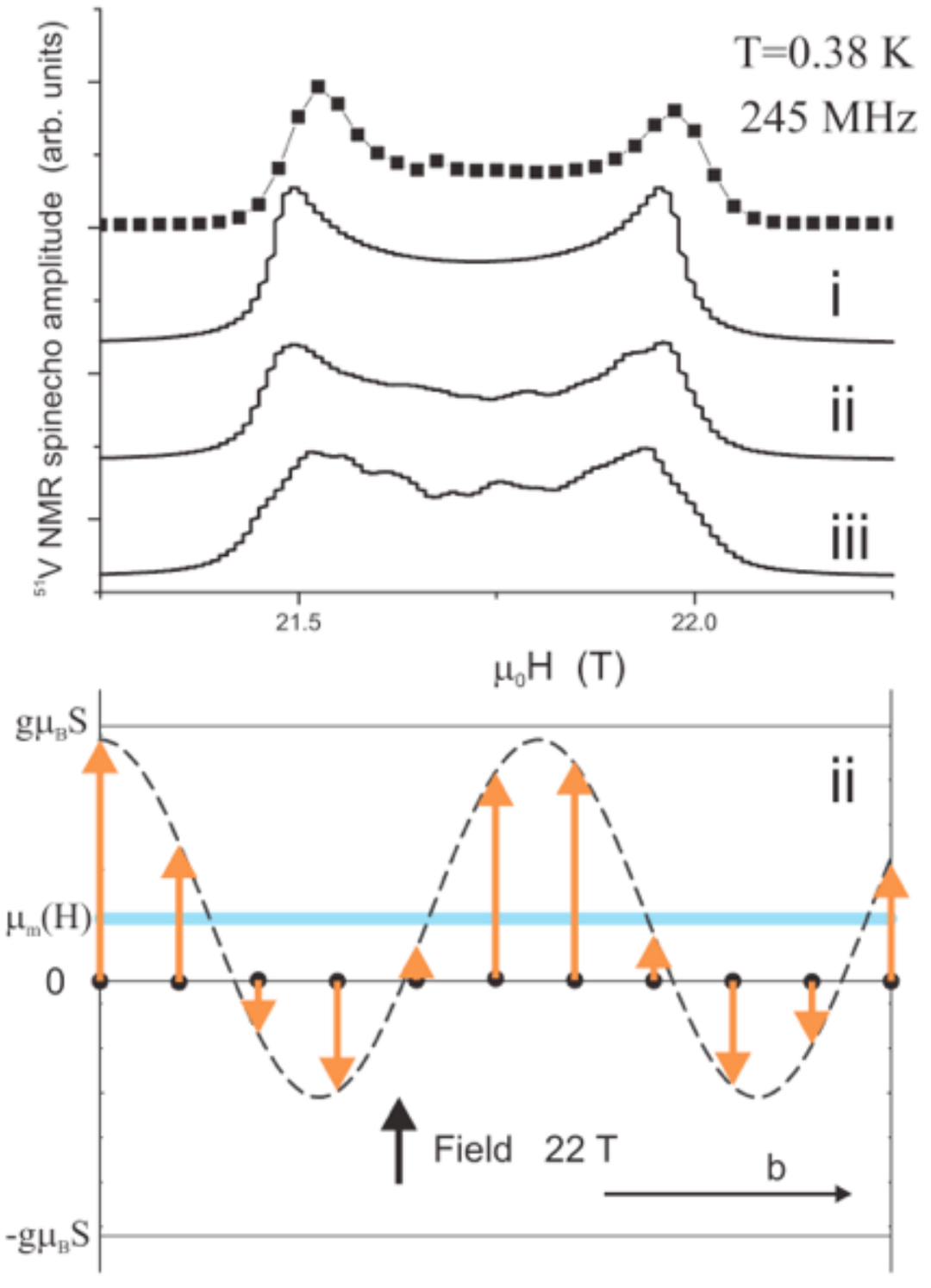


FIG. 2. Field dependence of the incommensurate wave vector k_{ic} for applied magnetic fields **H** ∥ **c** in $LiCuVO_4$. The open symbols

Buttgen et al 2012, 2014

PHYSICAL REVIEW B **85**, 214421 (2012)



longitudinal SDW

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, ΔS = 1
NSF = no spin flip, ΔS = 0

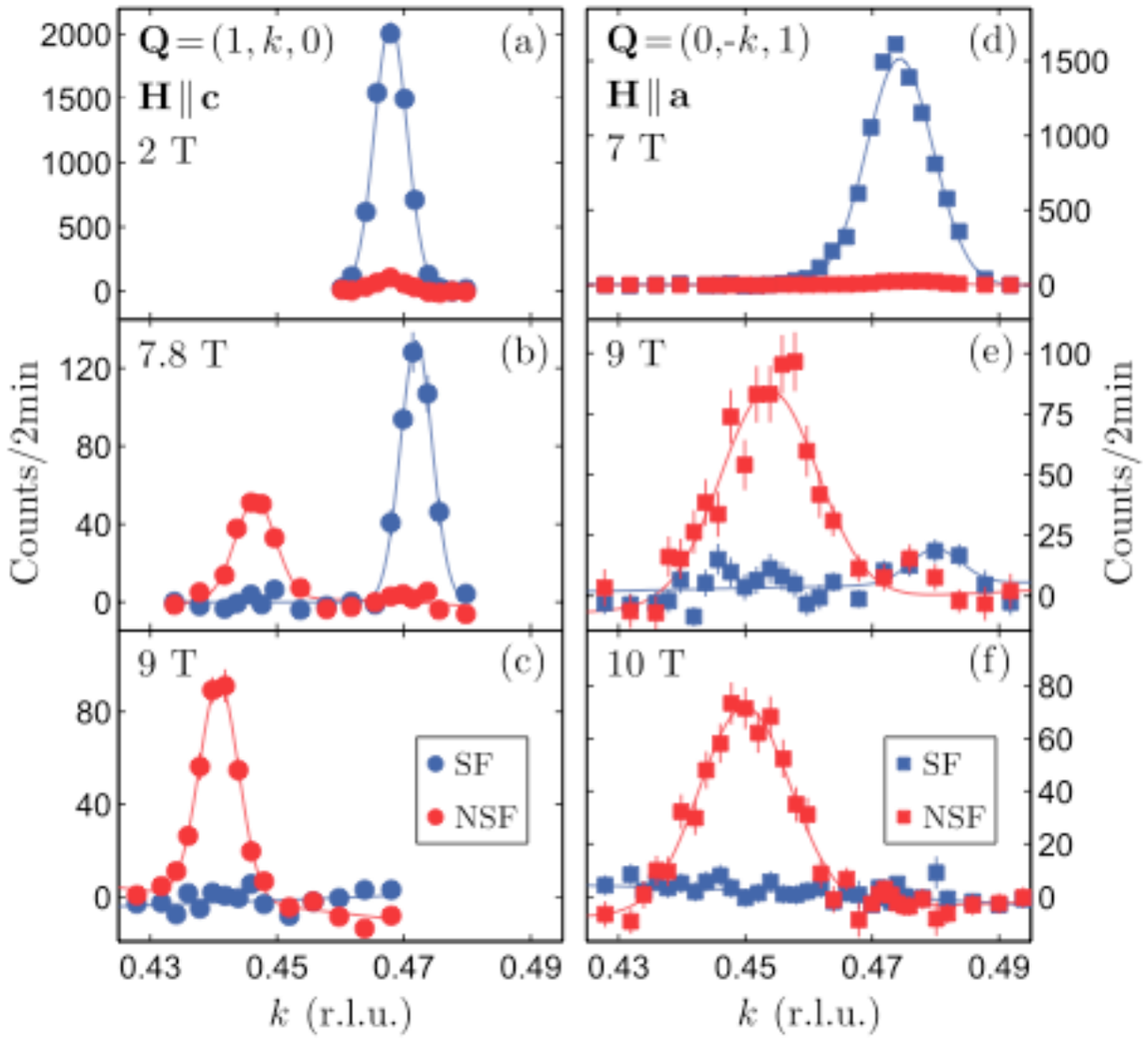
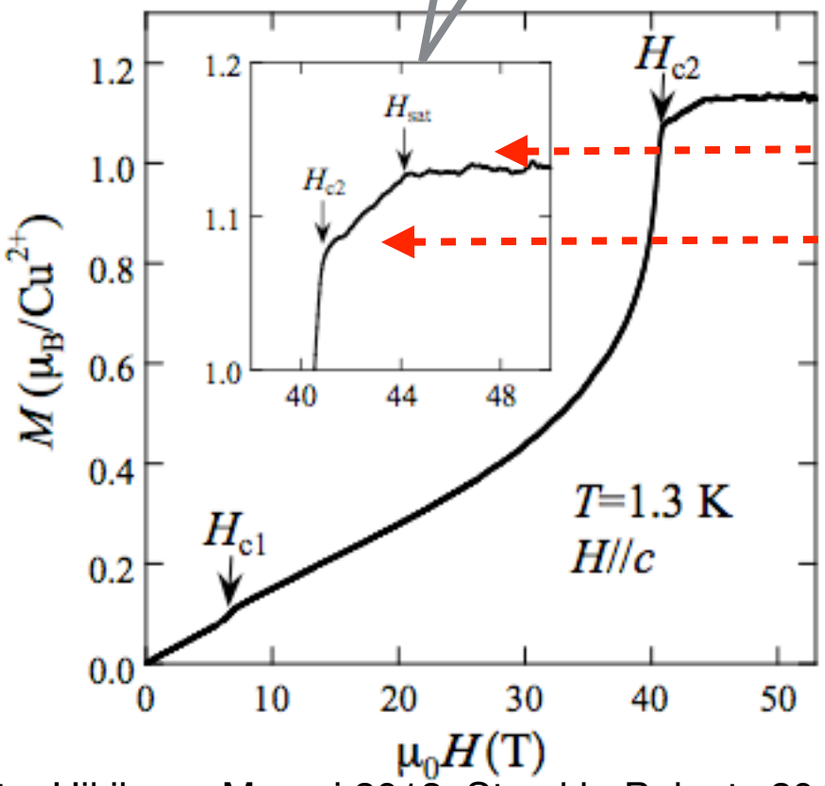


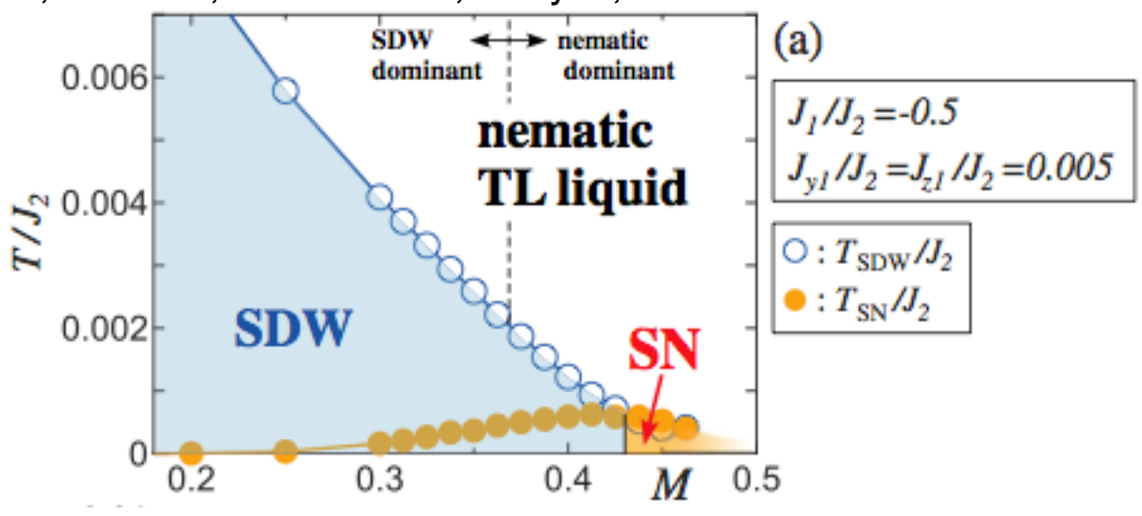
FIG. 3 (color online). Polarized cross sections measured at $T = 70$ mK for the magnetic reflections $\mathbf{Q} = (1, k_{IC}, 0)$ with $\mathbf{H}\|\mathbf{c}$ [left panels, (a)–(c)] and $\mathbf{Q} = (0, -k_{IC}, 1)$ with $\mathbf{H}\|\mathbf{a}$ [right panels, (d)–(f)].

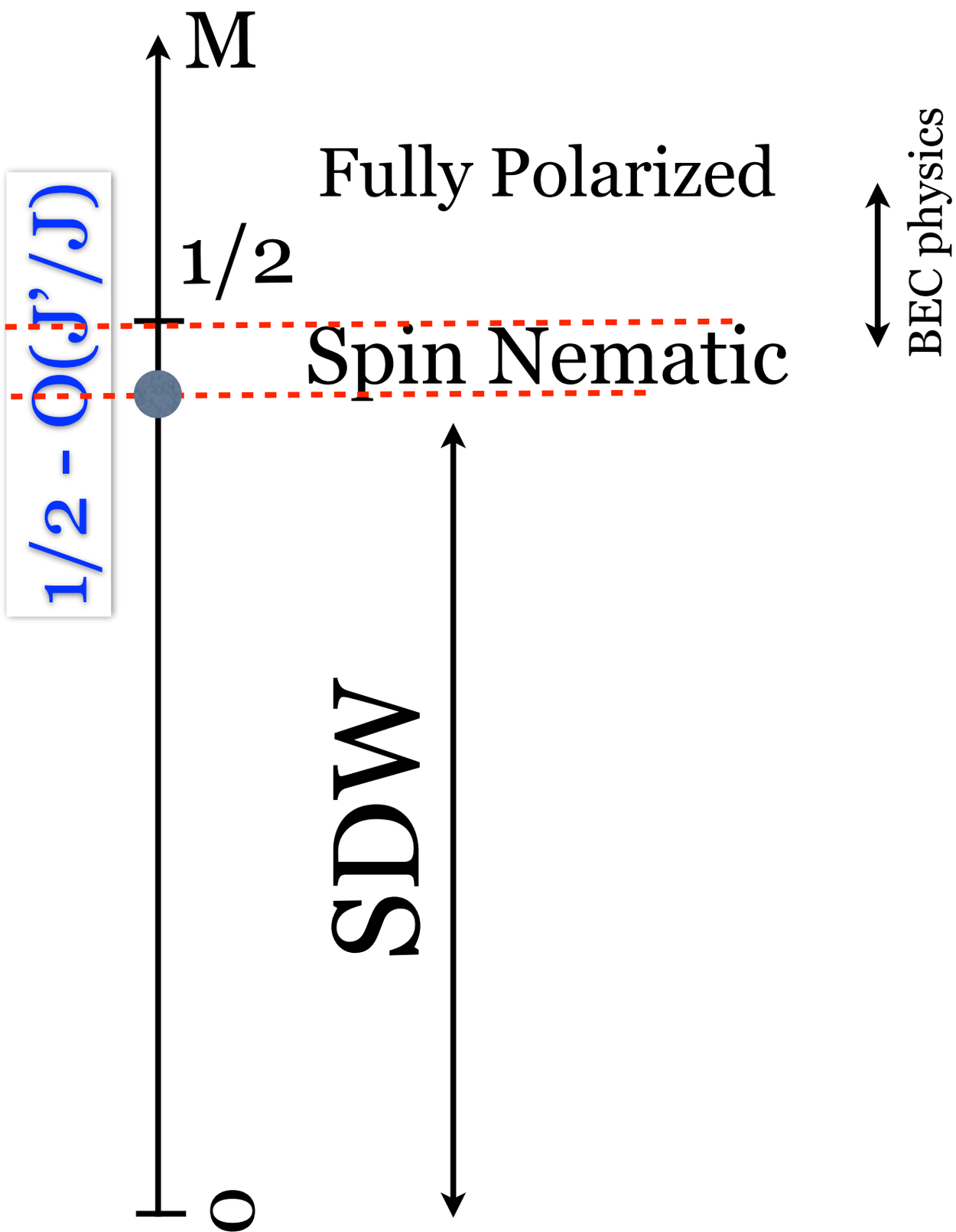
T=0 schematic phase diagram of weakly coupled **nematic** spin chains

Cautionary remark:
an impurity effect !



Sato, Hikihara, Momoi 2013; Starykh, Balents 2014





Cold reality

PHYSICAL REVIEW B **90**, 134401 (2014)

Search for a spin-nematic phase in the quasi-one-dimensional frustrated magnet $LiCuVO_4$

N. Büttgen,[1,*] K. Nawa,[2,3] T. Fujita,[4] M. Hagiwara,[4] P. Kuhns,[5] A. Prokofiev,[6] A. P. Reyes,[5] L. E. Svistov,[7,†] K. Yoshimura,[2] and M. Takigawa[3,‡]

"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

Outline

- Motivation: usual *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”

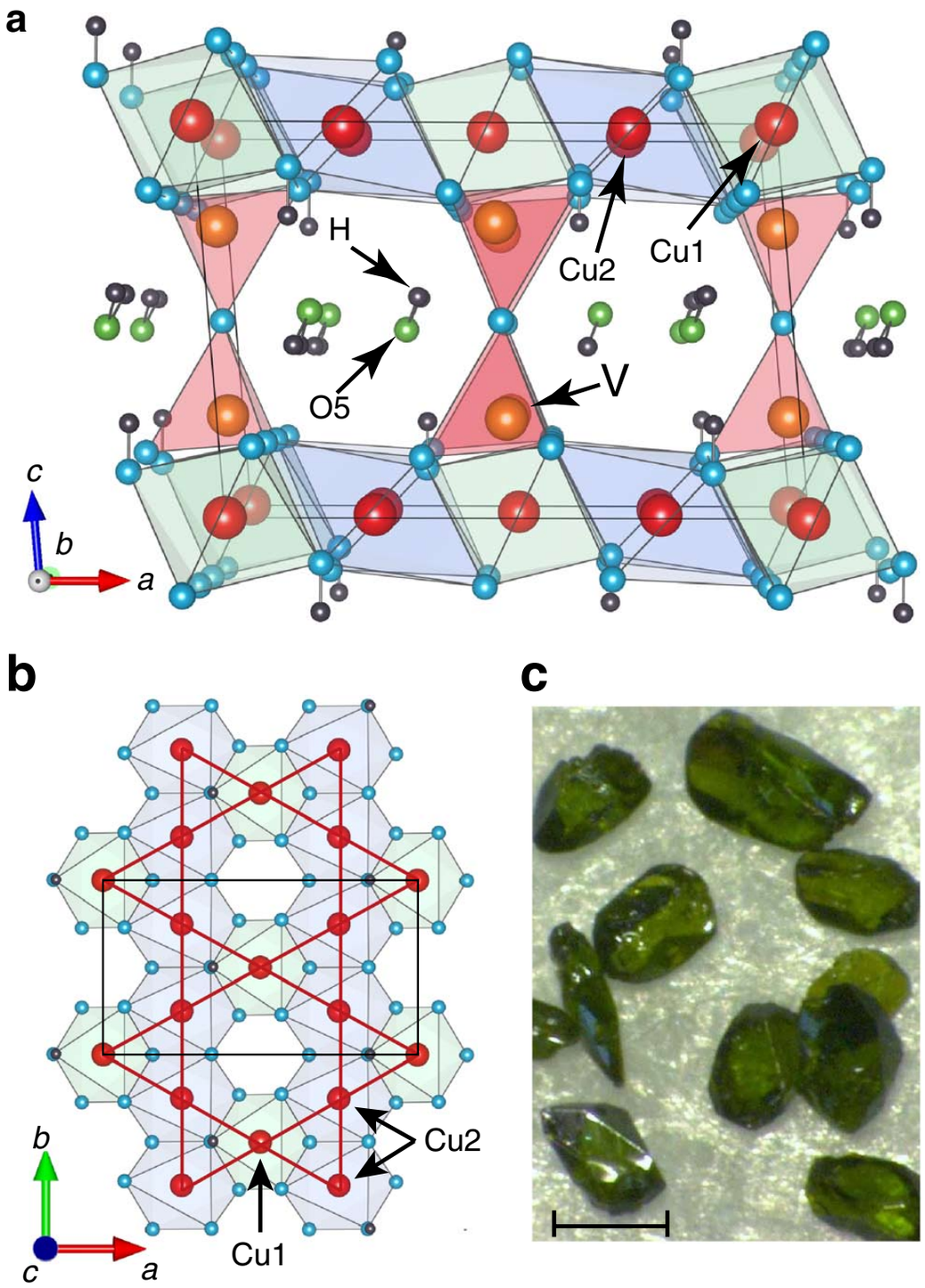
- Volborthite kagome antiferromagnet

– experimental status - magnetization plateau

– UUD, SDW and ***more***

✴ *Field theory of the Lifshitz point*

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.

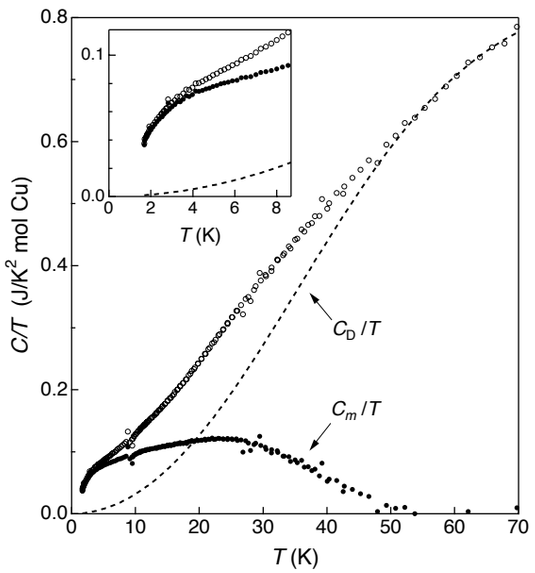
Visually similar to vésigniéite.

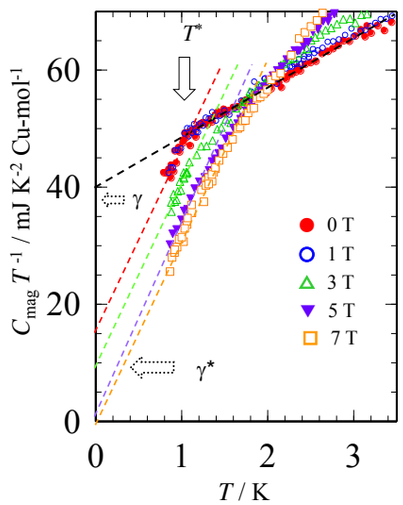
2001 quantum spin liquid?!

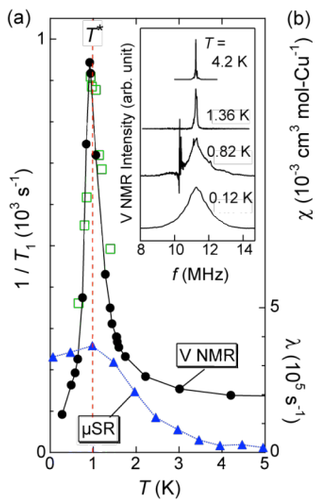
2009 impurity ordering at low T? magnetization steps?

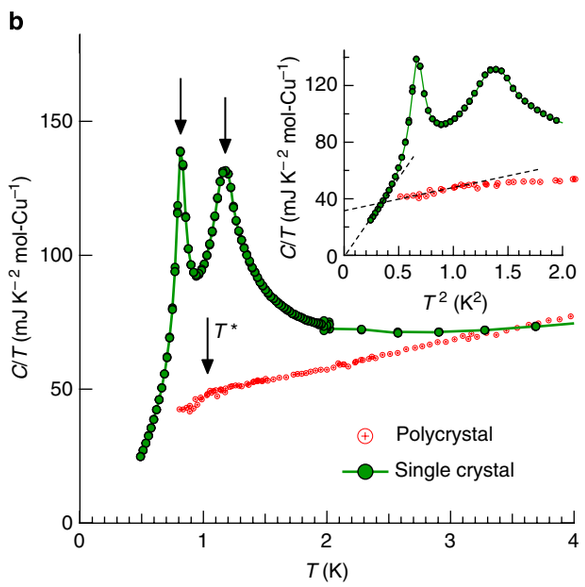
2012 magnetic order !

2014 magnetization plateau









time = material quality

One-Third Magnetization Plateau with a Preceding Novel Phase in Volborthite

H. Ishikawa,[1] M. Yoshida,[1] K. Nawa,[1] M. Jeong,[2] S. Krämer,[2] M. Horvatić,[2] C. Berthier,[2] M. Takigawa,[1] M. Akaki,[1] A. Miyake,[1] M. Tokunaga,[1] K. Kindo,[1] J. Yamaura,[3] Y. Okamoto,[1,4] and Z. Hiroi[1]

[1]*Institute for Solid State Physics, The University of Tokyo, Kashiwa, Chiba 277-8581, Japan*

[2]*Laboratoire National des Champs Magnétiques Intenses, LNCMI-CNRS (UPR3228), UJF, UPS and INSA, B.P. 166, 38042 Grenoble Cedex 9, France*

[3]*Materials Research Center for Element Strategy, Tokyo Institute of Technology, Yokohama 226-8503, Japan*

[4]*Department of Applied Physics, Nagoya University, Nagoya 464-8603, Japan*

(Received 23 November 2014; published 1 June 2015)

Huge 1/3 magnetization plateau !

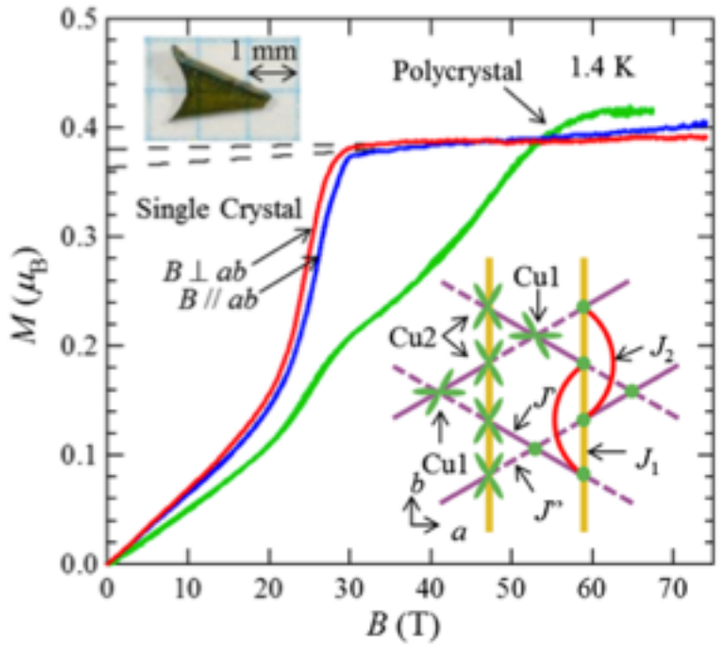


FIG. 1 (color online). Magnetization curves of volborthite measured at 1.4 K on two piles of single crystals in magnetic fields perpendicular (red line) and parallel (blue line) to the *ab* plane, and on a polycrystalline sample (green line) [32]. Shown also are a typical single crystal of volborthite (upper left) and the arrangement of Cu d_{x2-y2} orbitals projected onto the *ab* plane in the low-temperature $P2_1/a$ structure (lower right). J_1 and J_2 represent the NN and NNN interactions in the Cu2 spin chains, respectively. J' and J'' represent the NN interactions between Cu1 and Cu2 spins.

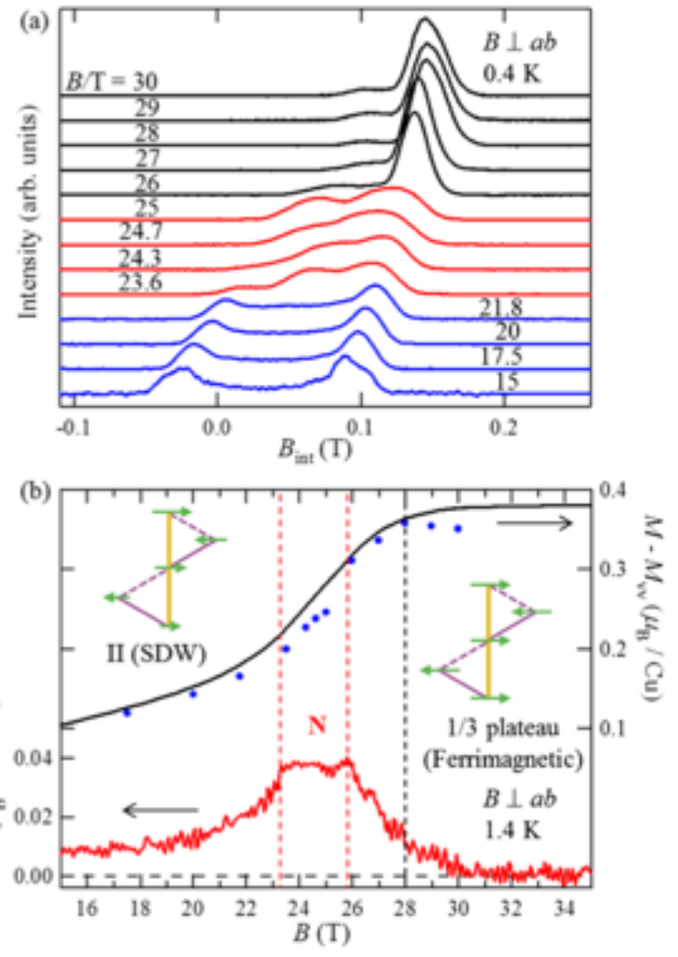


FIG. 2 (color online). (a) ^{51}V NMR spectra measured on a single-domain piece of a crystal in magnetic fields applied perpendicular to the *ab* plane at $T = 0.4$ K. The labeled fields correspond to $B = \nu_0/\gamma(B_{\rm int} = 0)$. (b) Magnetization curve of single crystals (top, black line) and its field derivative (bottom) in $B \perp ab$ at 1.4 K after the subtraction of the Van Vleck paramagnetic magnetization ($M_{\rm VV}$). Magnetization deduced from the center of the gravity of the NMR spectra is also plotted (top, blue circles).

How far in field does the plateau extend?

Meeting abstracts of the Physical Society of Japan 2014

27aAF-9

単結晶 volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ の超強磁場磁化過程

東大物性研，東大院工 A
山下知大 A，宮田敦彦，嶽山正二郎，石川孟，岡本佳比古，広井善二

Magnetization curves of volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ in ultra-high magnetic fields

ISSP, Univ. of Tokyo, Dep. of Appl. Phys., Univ. of Tokyo A
T. Yamashita A, A. Miyata, S. Takeyama, H. Ishikawa, Y. Okamoto, and Z. Hiroi

スピン 1/2 カゴメ格子反強磁性体は、幾何学的フラストレーションによりスピン液体状態が実現する有力な候補と考えられている。最近、その磁化過程の計算が進み、飽和に至るまでに 5 つの磁化プラトーが現れることが示唆された[1]。これら 5 つのプラトーの内、0, 1/9 プラトーはスピン液体状態、1/3, 5/9, 7/9 プラトーは長距離秩序となり、多彩な磁場誘起相の出現が示唆されている。

一方、スピン 1/2 カゴメ反強磁性体を実現する理想的な物質はこれまで見つかっていない。本研究で用いた volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ は、カゴメ格子が歪んでいるが、理想に近い物質の一つとして盛んに研究が行われている。また、近年、volborthite 単結晶試料を用いた磁化測定が 75T まで行われ、幅広い 1/3 プラトーが観測された[2]。

我々は、飽和に至るまでの全磁場誘起相を調べるため、100 T 以上の超強磁場下におけるファラデー回転測定を試みている。図は、波長 532 nm レーザーを用いた 11 K におけるファラデー回転角を 1.3 K における磁化曲線[2]と比較したものである。磁場は単結晶試料の ab 平面に垂直に印加した。ファラデー回転角は磁化の奇数次項で表せ、25 T 近辺における磁化の増加に対応して回転角も増えている。また、1/3 磁化プラトーに対応して回転角も一定になり、この領域は 120 T まで続いている。

本講演では、ファラデー回転測定の結果に加えて、ピックアップコイルを用いた誘導法による磁化測定(<110 T)の結果も合わせて発表する。

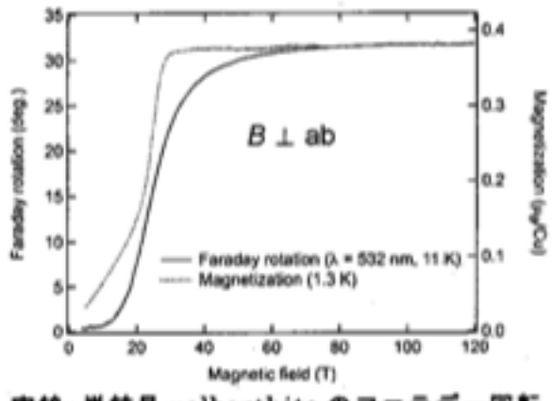


図　実線：単結晶 volborthite のファラデー回転
点線：磁化曲線 [2]

[1] S. Nishimoto, N. Shiba, and C. Hotta, Nature Comm. 4, 2287 (2013).
[2] 石川孟 他，日本物理学会第 69 回年次大会 27aAF-8 (2014).

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**.

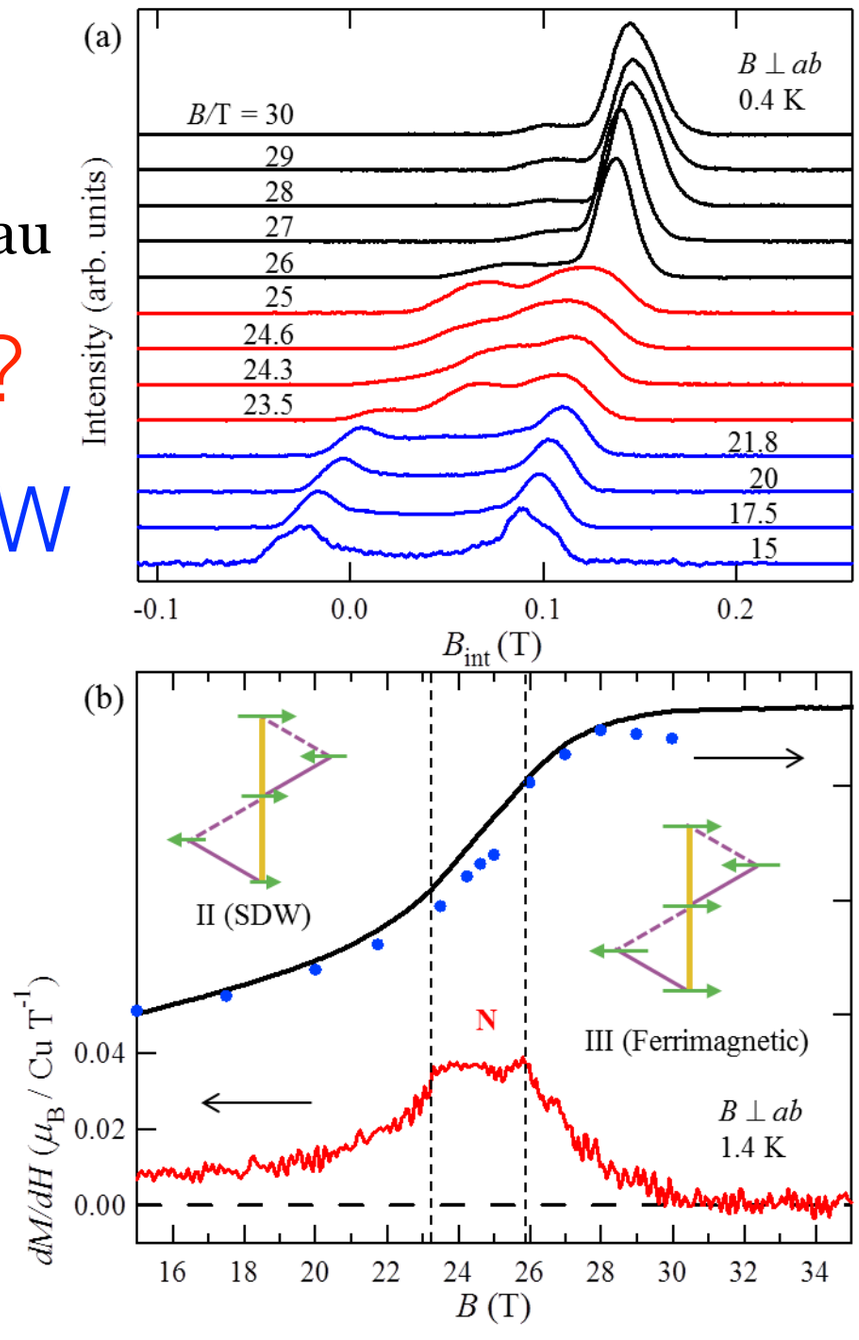
this is the best plateau ever:
28 T < plateau < 120 T

Phase diagram

1/3 plateau

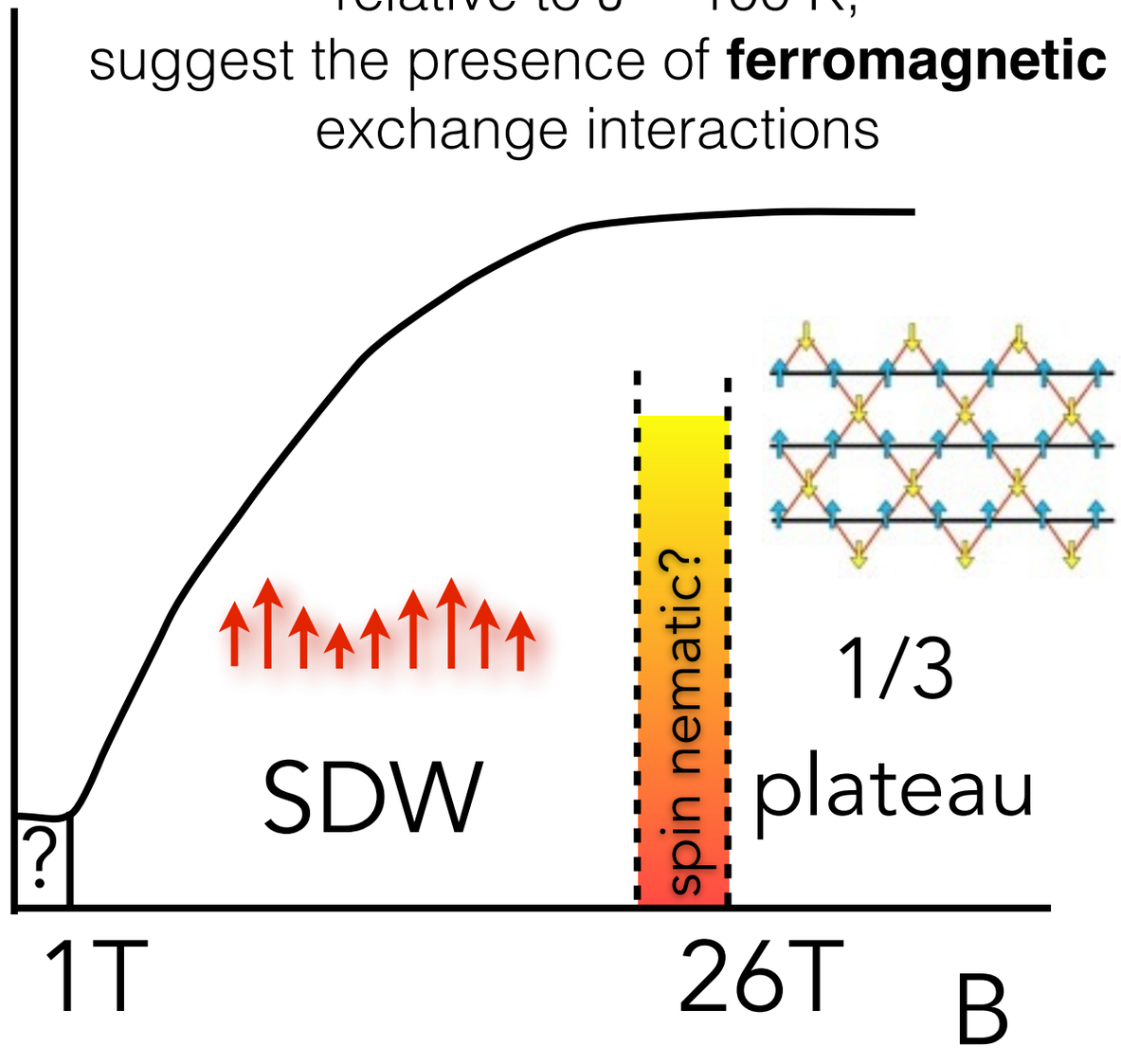
N?

SDW



H. Ishikawa *et al*, PRL 2015

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



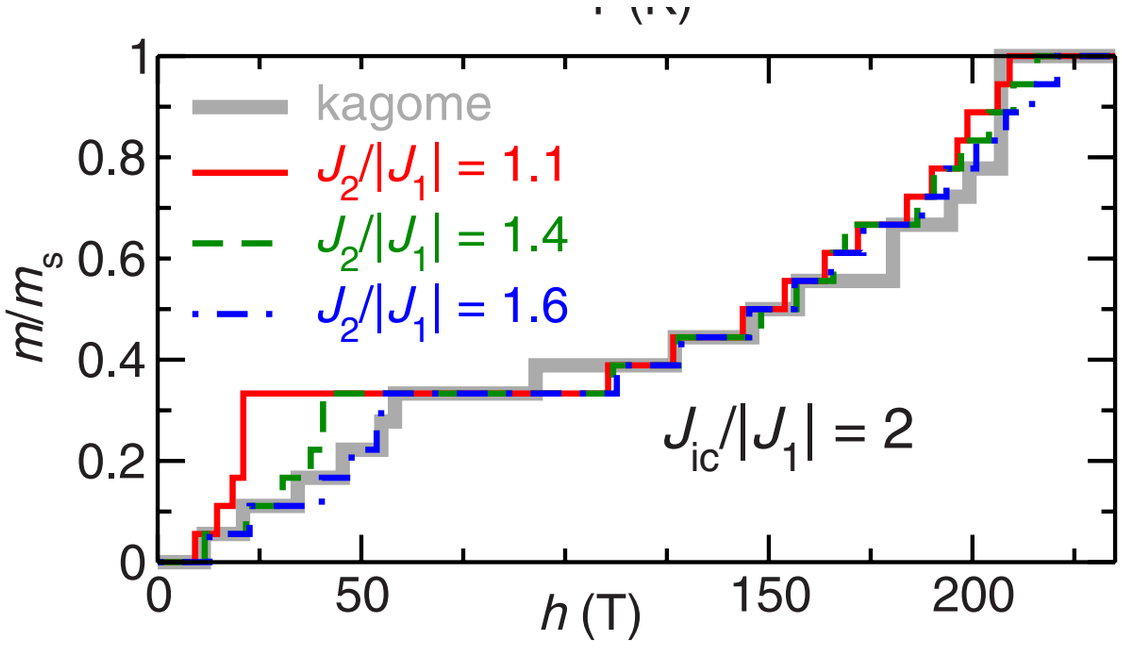
may be a spin nematic??

Frustrated ferromagnetism

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

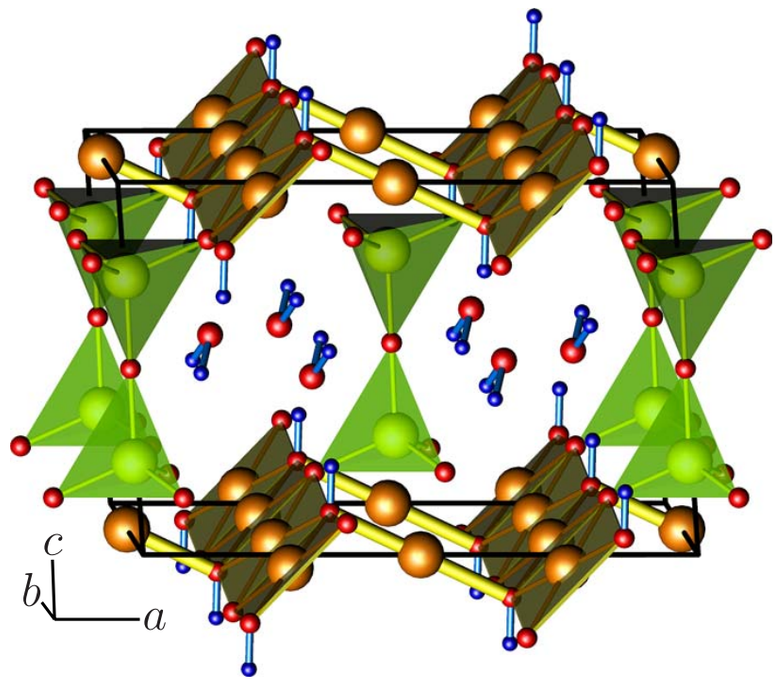
Coupled frustrated quantum spin-$\frac{1}{2}$ chains with orbital order in volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$

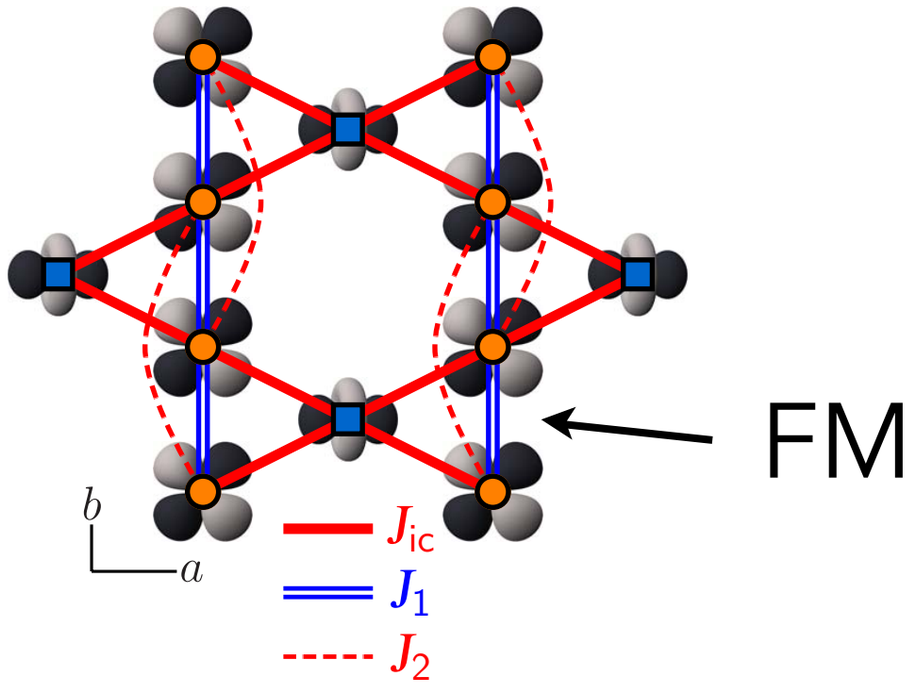
O. Janson,[1,*] J. Richter,[2] P. Sindzingre,[3] and H. Rosner[1,†]
[1]*Max-Planck-Institut für Chemische Physik fester Stoffe, D-01187 Dresden, Germany*
[2]*Institut für Theoretische Physik, Universität Magdeburg, D-39016 Magdeburg, Germany*
[3]*Laboratoire de Physique Théorique de la Matière Condensée,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



DFT gets it right!

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





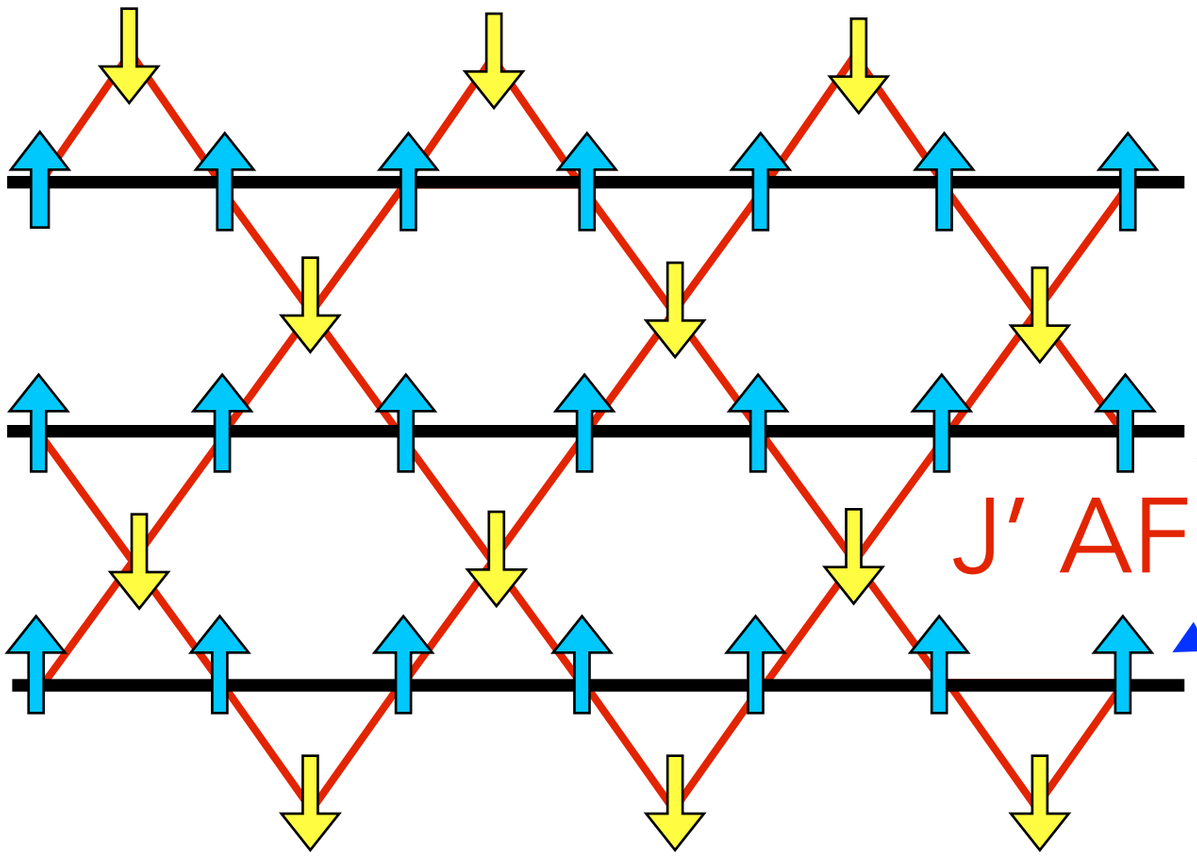
Ferrimagnetic state

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

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

O. Janson,[1,*] J. Richter,[2] P. Sindzingre,[3] and H. Rosner[1,†]
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(Received 9 August 2010; published 30 September 2010)

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



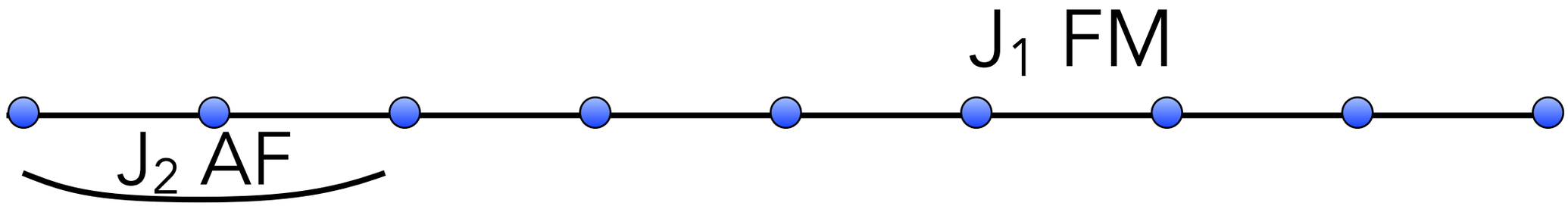
J_1 FM, J_2 AF

J’ AF

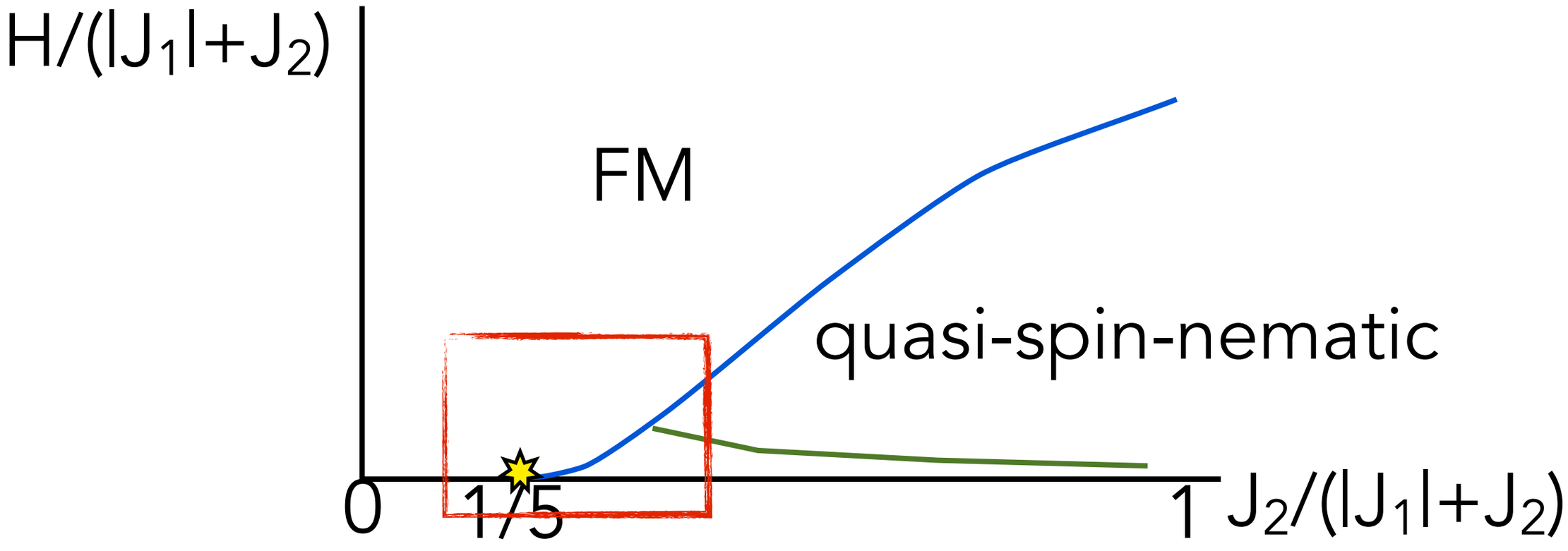
polarized chains?!

Spin chain redux

Frustrated ferromagnetic chain

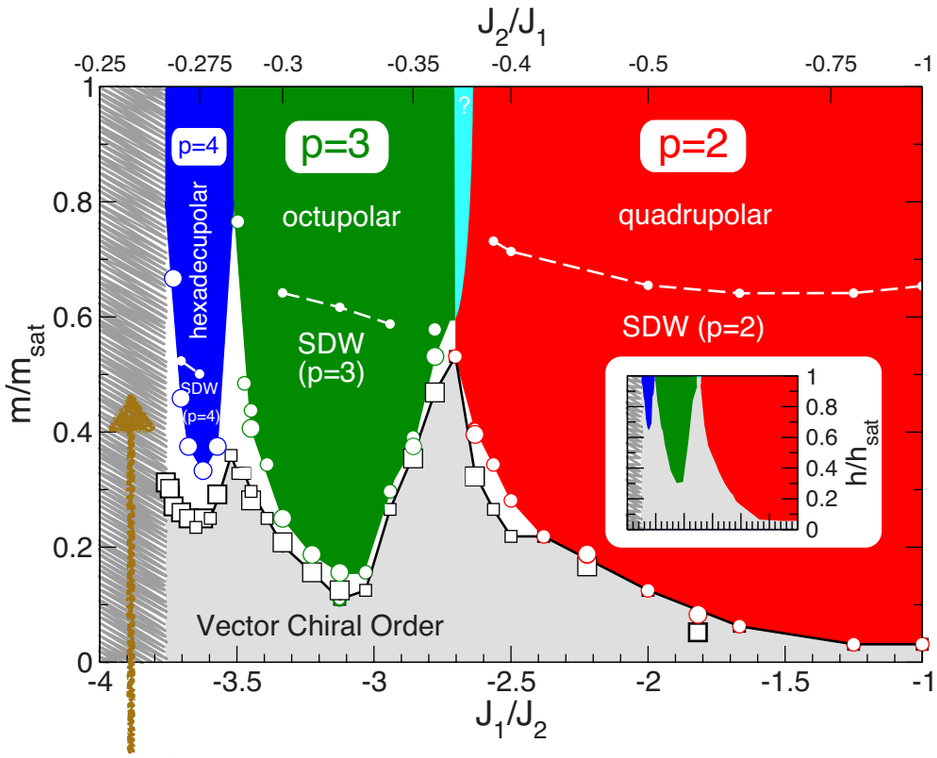


$$H = J_1 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2} - h \sum_i S_i^z$$



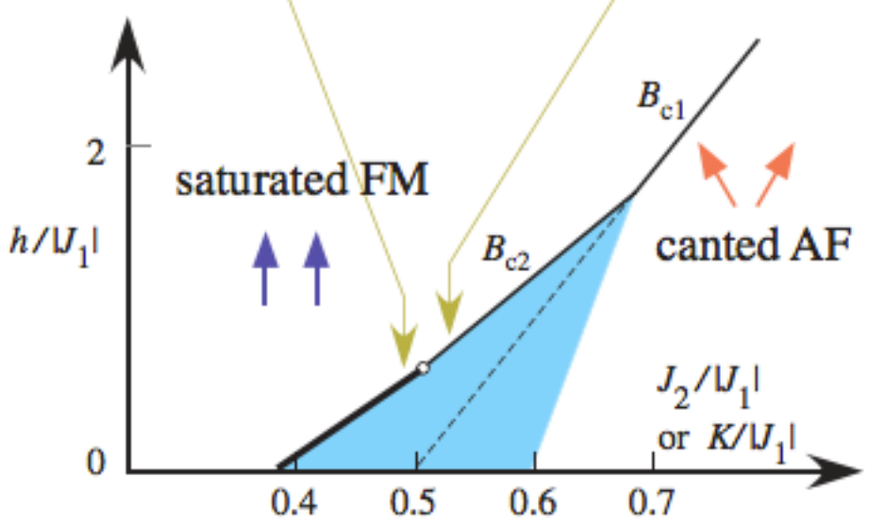
Numerics: nematicity and 1st order seem connected?

1d chain



Sudan et al, 2009

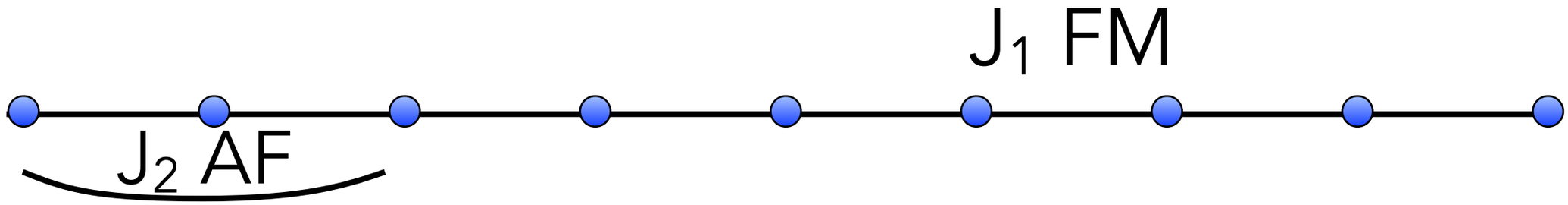
2d square lattice



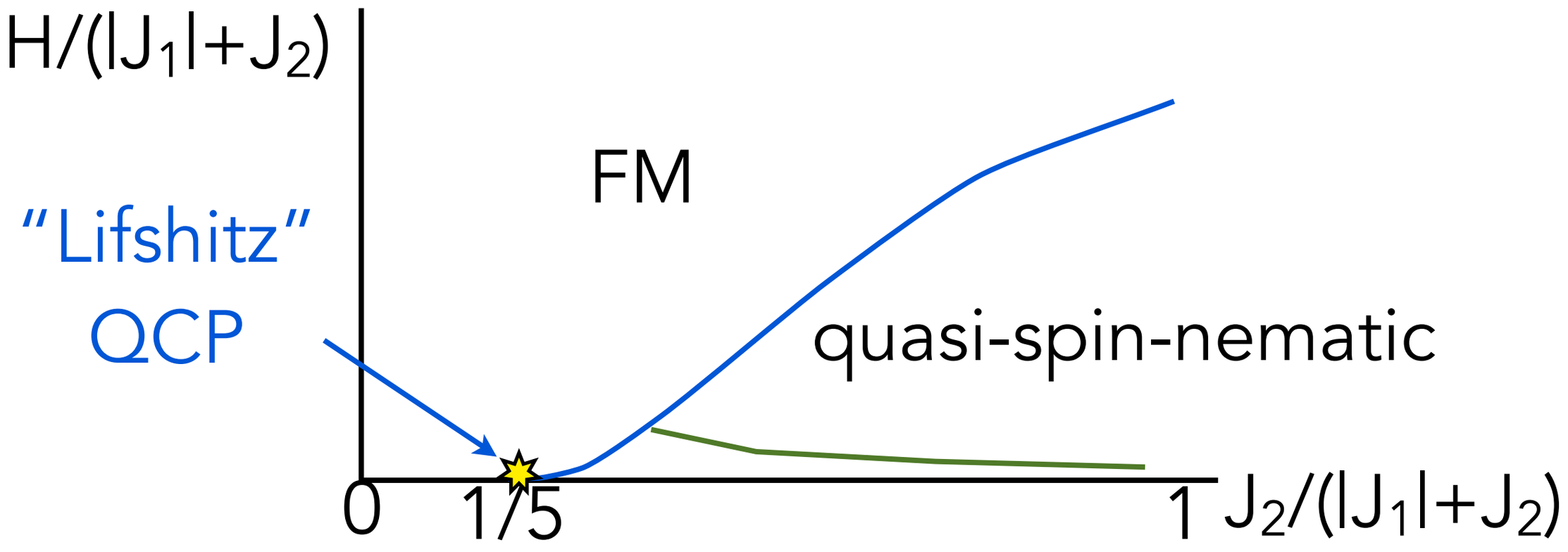
Shannon, Momoi, Sindzingre PRL 2006

Quantum Lifshitz point

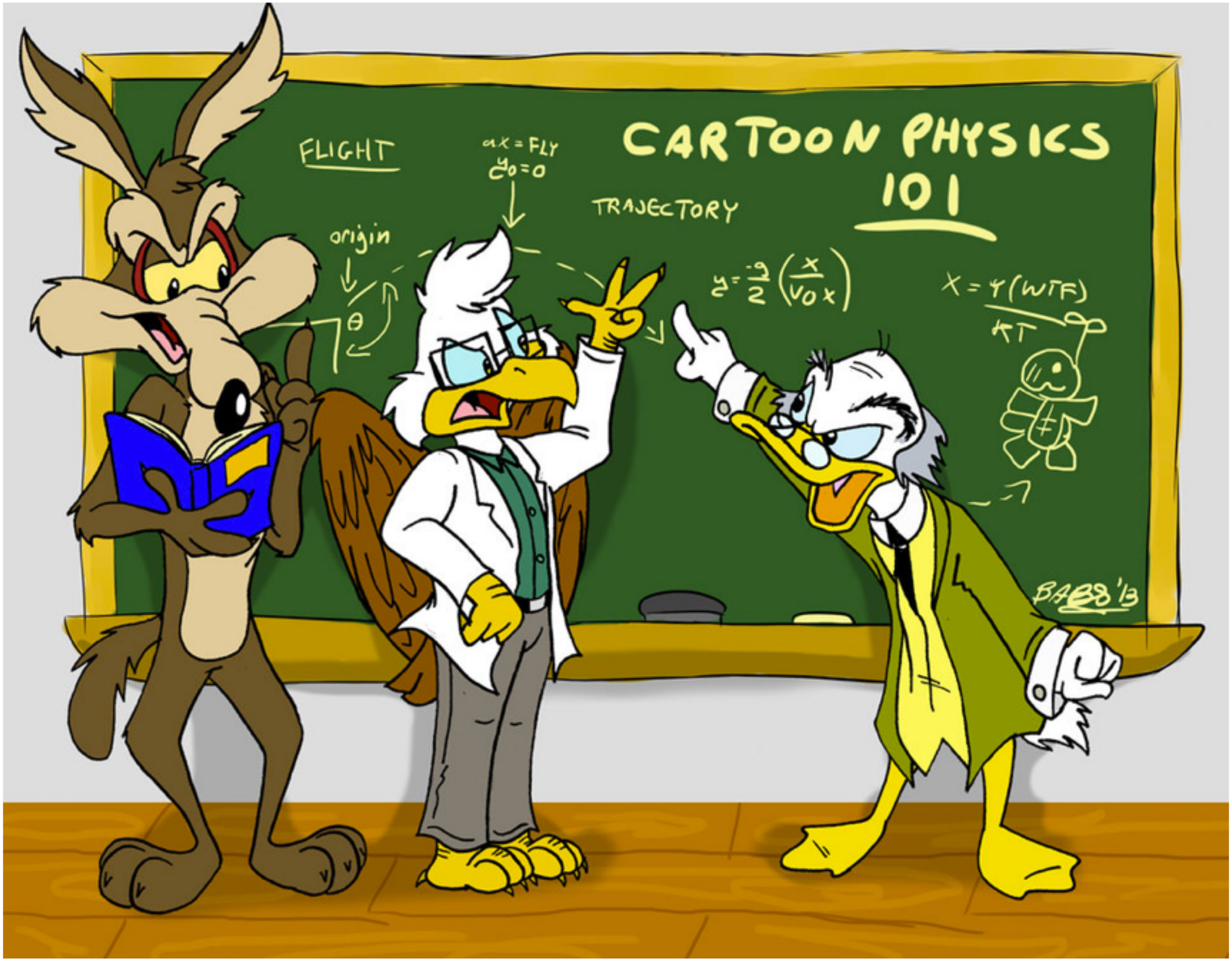
Frustrated ferromagnetic chain



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



It is time for a blackboard discussion



Lifshitz Point

- Unusual QCP: order-to-order transition
- Effective action - NL**σ**M for unit vector **m**

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

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

tunes QCP $\delta \propto |J_1| - 4J_2$

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!

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

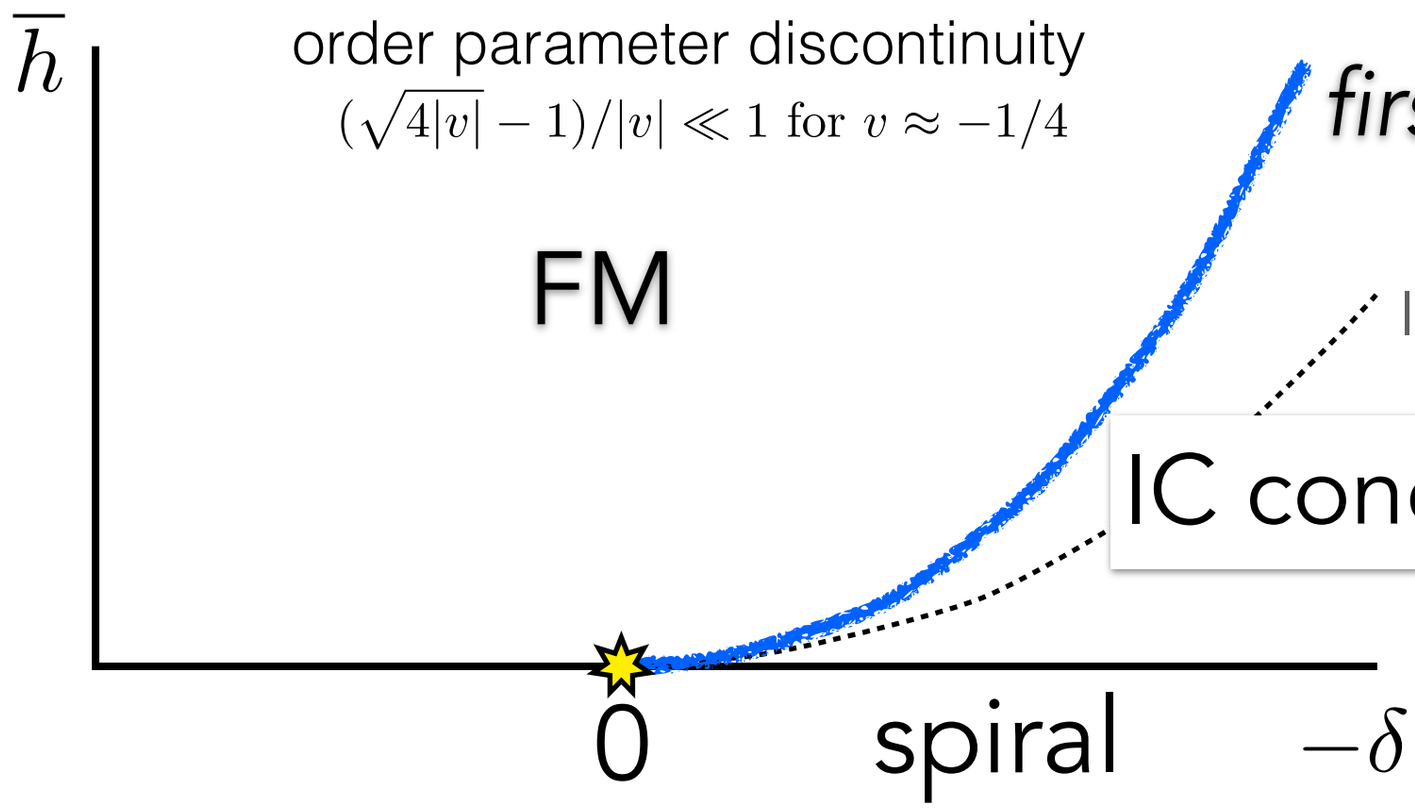
Saddle point

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

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

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

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



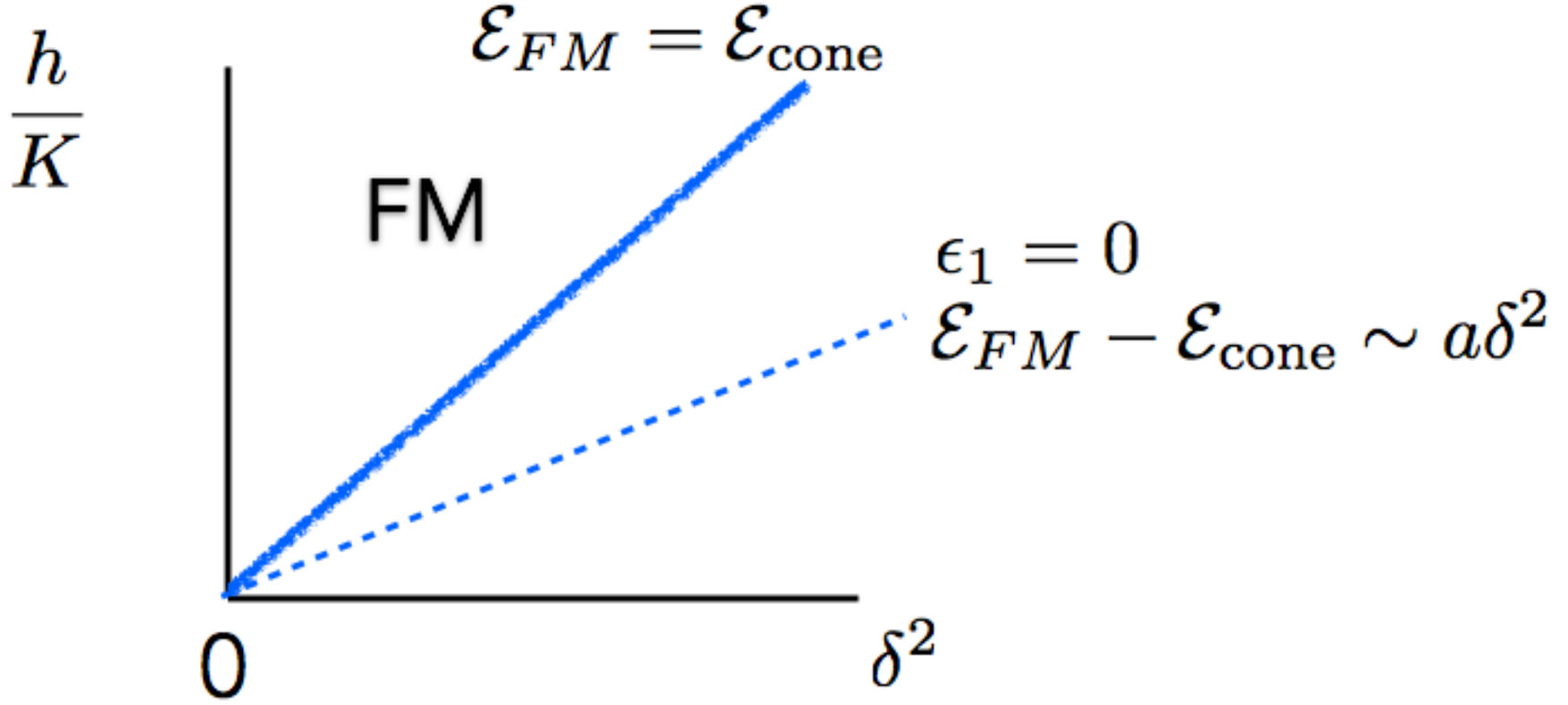
first order $h_c=\dfrac{\delta^2}{8K\sqrt{|v|}(1-\sqrt{|v|})}$

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

local instability of FM state
(1-magnon condensation)

$$\hat{m}=\begin{pmatrix}|\Psi|\cos(qx+\phi)\\ \pm|\Psi|\sin(qx+\phi)\\ \sqrt{1-|\Psi|^2}\end{pmatrix}$$

Metamagnetic endpoint?



Quantum corrections
penalize E_{cone} but not E_{FM}

$$\Delta\mathcal{E}_{\text{cone}} = +f(v)\delta^{5/2}$$

Metamagnetic endpoint?

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

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

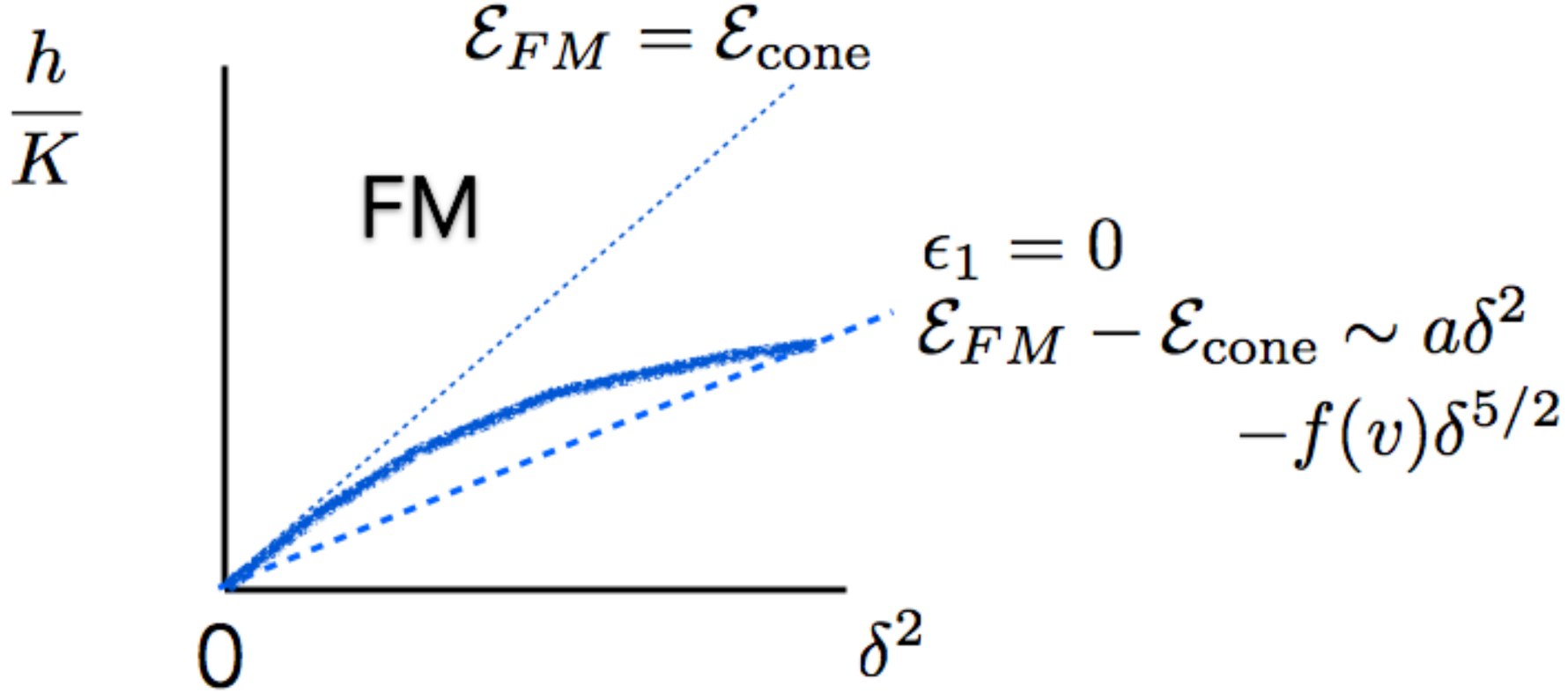
$$\hat{e}_1 \times \hat{e}_2 = \hat{e}_3 = \hat{m}^{sp}(x)$$

$$\eta = n_1 + in_2 \qquad \overline{\eta} = n_1 - in_2$$

$$S = S_{\rm sp} + \int dx\, d\tau\, \{\overline{\eta}\partial_\tau \eta + H(\overline{\eta}, \eta)\} \quad + O(\eta^3)$$

Bogoliubov transformation gives
correction to GS energy

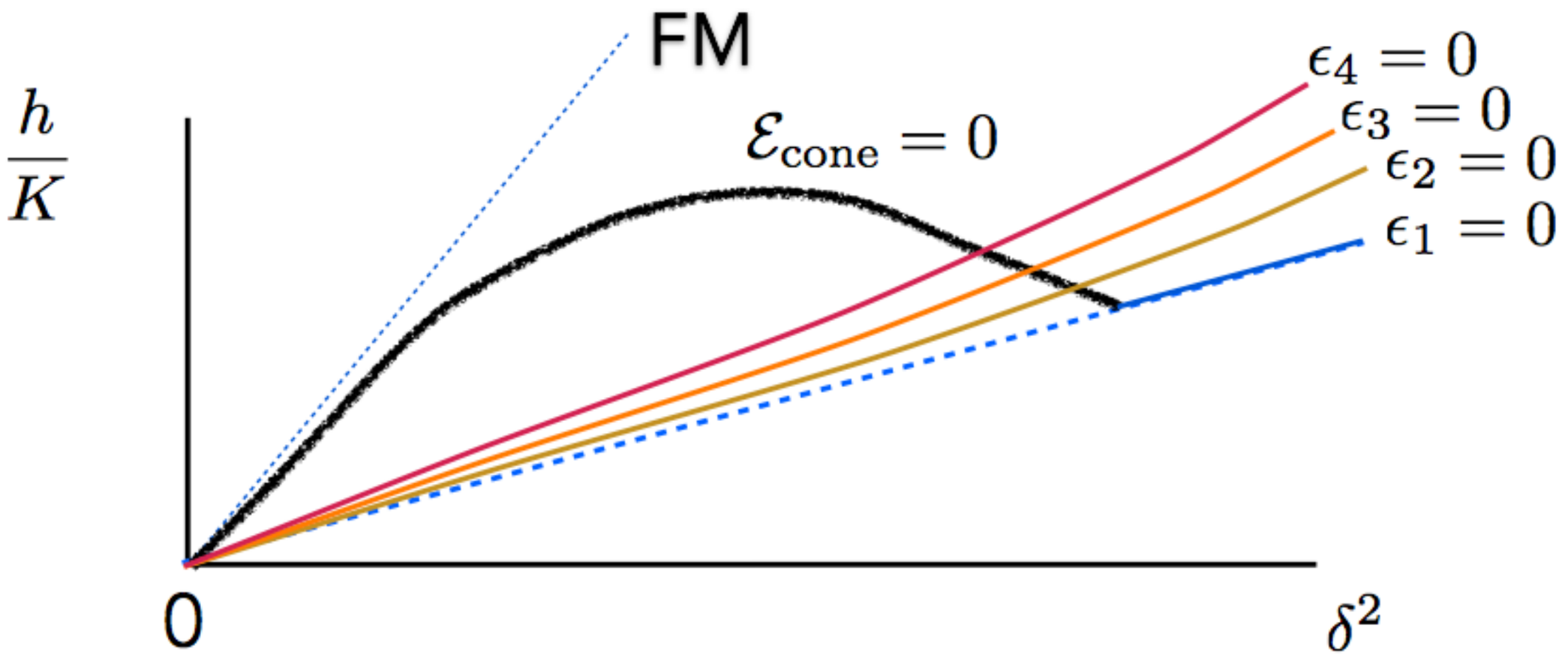
Metamagnetic endpoint?



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

Instabilities

- Choose $E_{FM}=0$

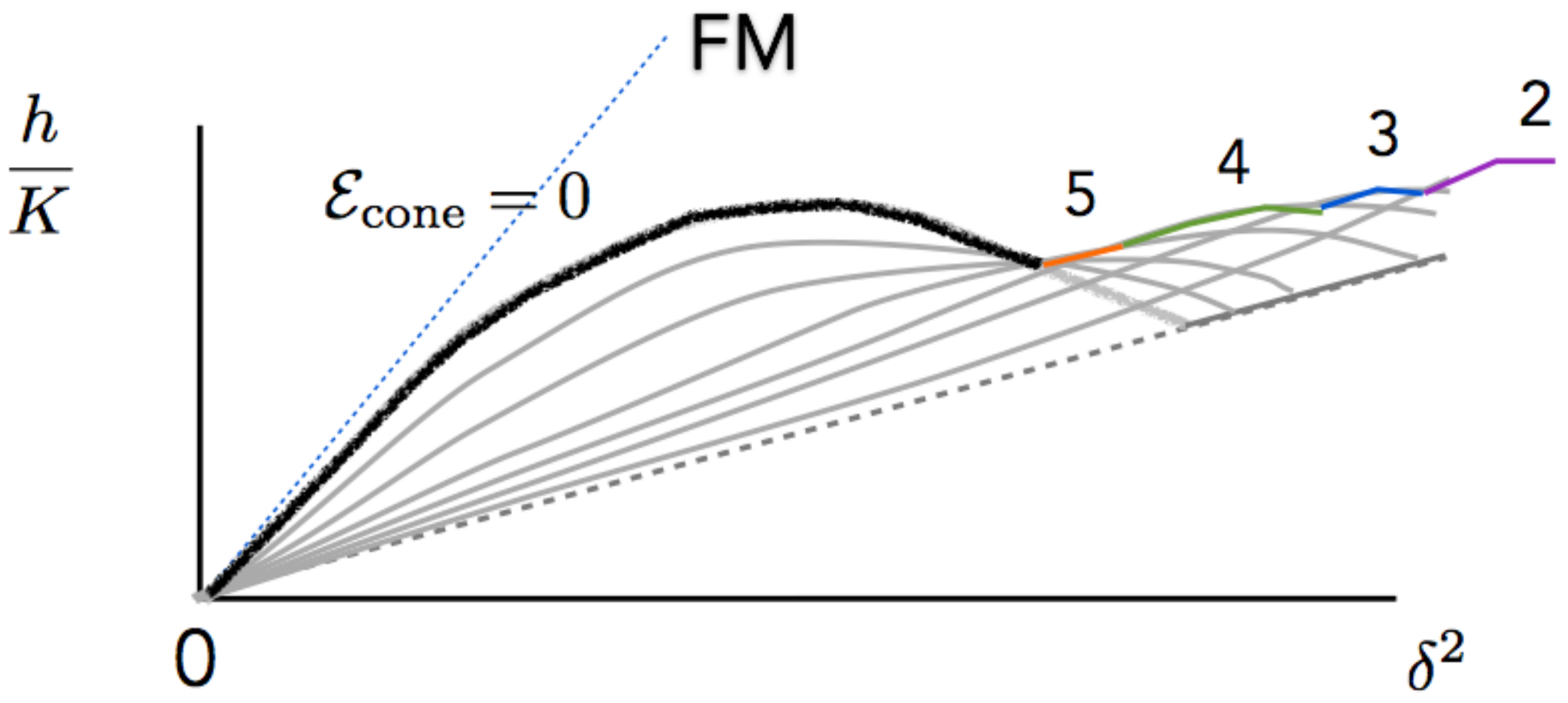


But the bound states cannot get arbitrarily deep - low density approximation is violated

A natural speculation

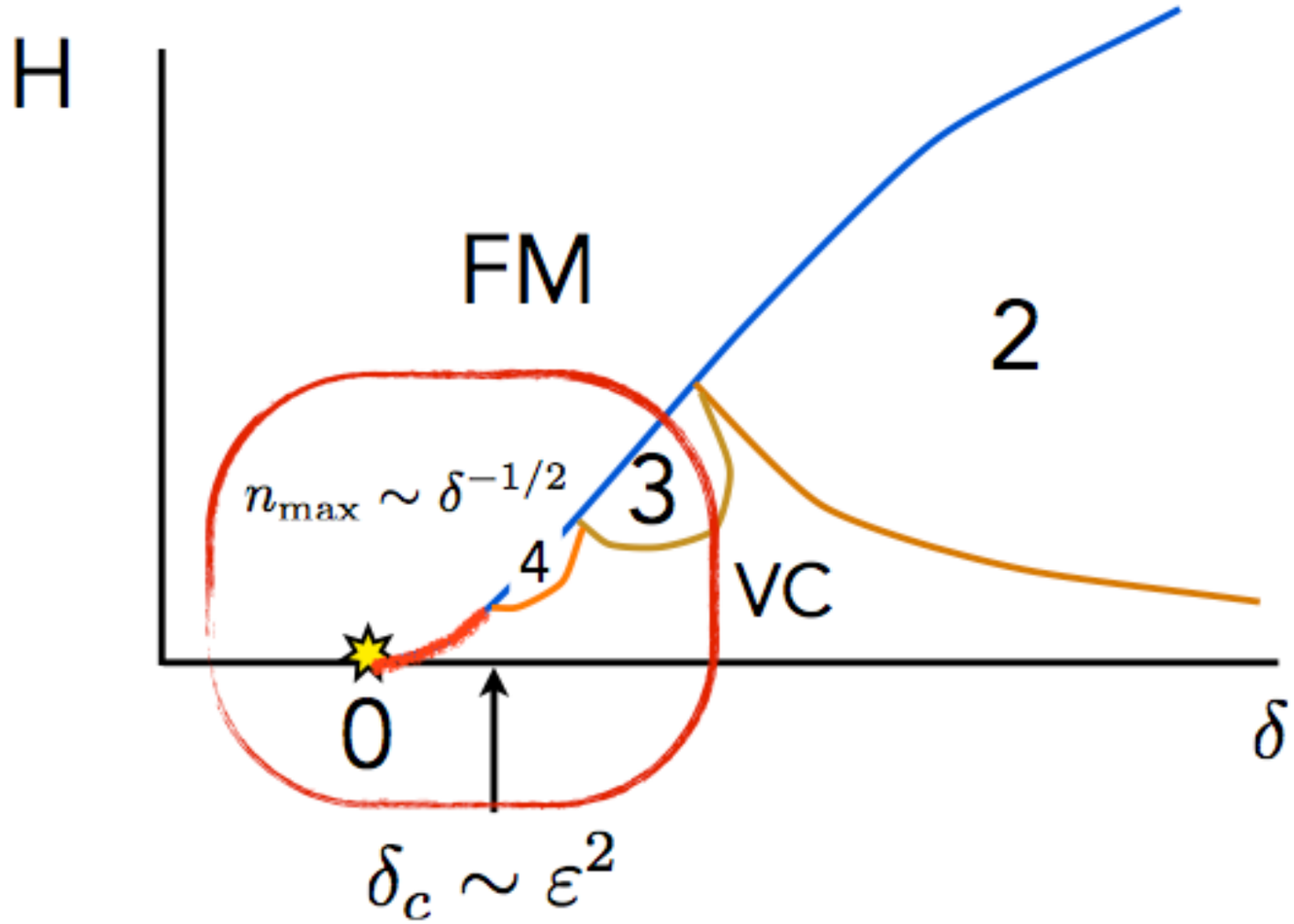
Instabilities

- 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

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

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

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