Magnon pairing in quantum spin nematic

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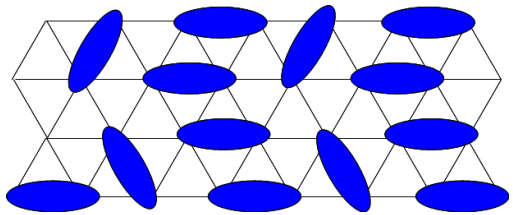
in collaboration with Hiro Tsunetsugu (Tokyo)

Outline

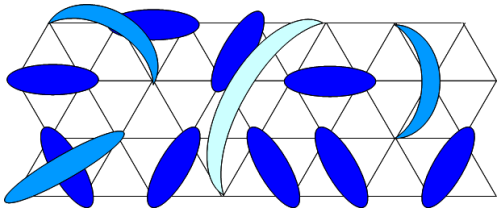
- Introduction
- Bound magnon states: from 1D chain to frustrated square lattice
- Condensate of bound magnon pairs: spin nematic state



Spin Liquids

- ‘Traditional’ quantum magnetism: ordered magnetic states, spin chains, ladders,…
- Mordern theories: possibility of spin liquids in arbitrary dimension short-range RVB, long-range RVB, Z_2, $U(1)$, topological,…
- Recent experiments: triangular organics, (hyper)kagome, ^{3}He monolayers,

anything else exotic?



3 + 1 states of Matter

Matter	Magnetic Matter	Defining property
gas	paramagnet	dilute
liquid	spin-liquid	isotropic
solid	ordered magnets	anisotropic
liquid crystal	**spin-nematic***	partial symmetry breaking

*(a.k.a. multipolar states) described by tensor order parameters, searched for the past 40 years, were rarely observed so far

Spin nematics in literature and in nature

- magnets with $S \geq 1$ and biquadratic exchange $K(\mathbf{S}_i \cdot \mathbf{S}_j)^2$ on site tensor

Blume & Hseich (1969)

$$\left\langle S_i^\alpha S_i^\beta \right\rangle \neq \delta^{\alpha\beta}$$

- 'preformed' CEF multipoles: $\left|J,m\right\rangle \rightarrow \left|\Gamma_n^{(l)}\right\rangle$ such that

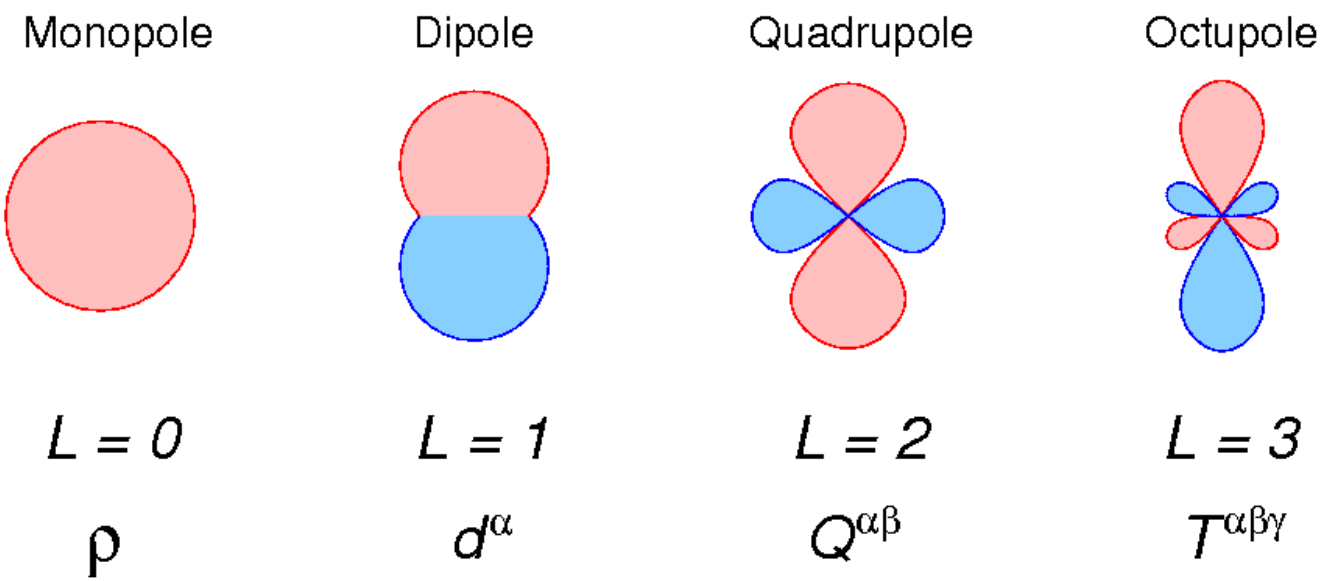
$\left\langle \Gamma_k^{(l)} \middle| J^\alpha \middle| \Gamma_n^{(l)} \right\rangle = 0$, while $\left\langle \Gamma_k^{(l)} \middle| J^\alpha J^\beta \middle| \Gamma_n^{(l)} \right\rangle \neq 0$

NpO_2 , $Ce_xLa_{1-x}B_6$

- spin-nematic order from strong correlations in a system of magnetic dipoles

Multipoles: symmetry classification

- zero magnetic field, irreps of $SO(3)$, classification in L



- finite magnetic fields, irreps of $SO(2)$, classif. in L_z, spin nematic $\Longleftrightarrow$ $L_z = \pm 2$

Magnon BEC in applied field

Bosons

- $U(1)$ gauge invariance
- condensate wavefunction

$$\langle \psi(r) \rangle$$

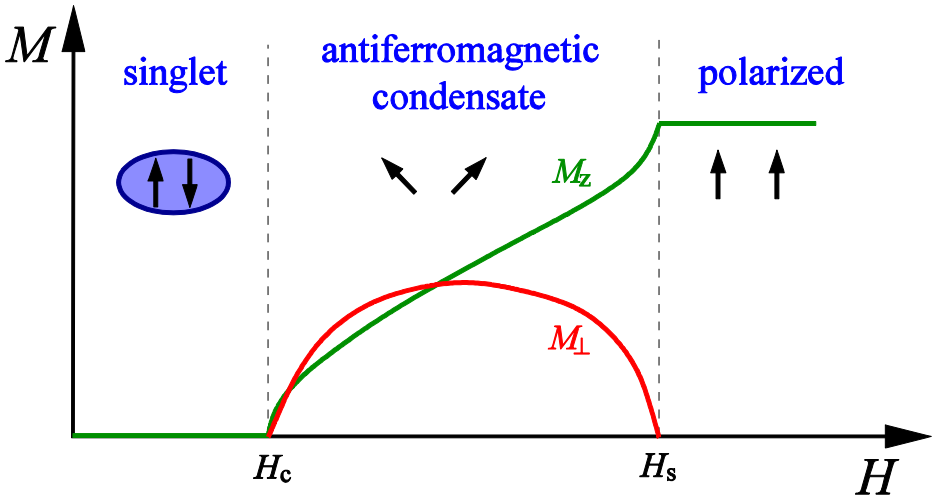
- chemical potential μ

Spins

- $SO(2)$ rotation symmetry
- staggered magnetization

$$M_j^{\perp} = \langle S_j^x + iS_j^y \rangle$$

- magnetic field H

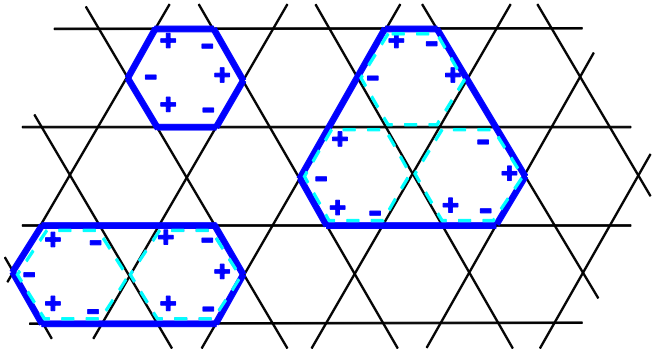


Magnon BEC: quest for new physics

- change bosons: add flavor, valley index, triplet polarization,…

- change kinetic energy: localized magnons in flat bands

Schulenburg et al (2002)



- change **potential energy**: add attraction, competing *ferro*- and antiferromagnetic bonds

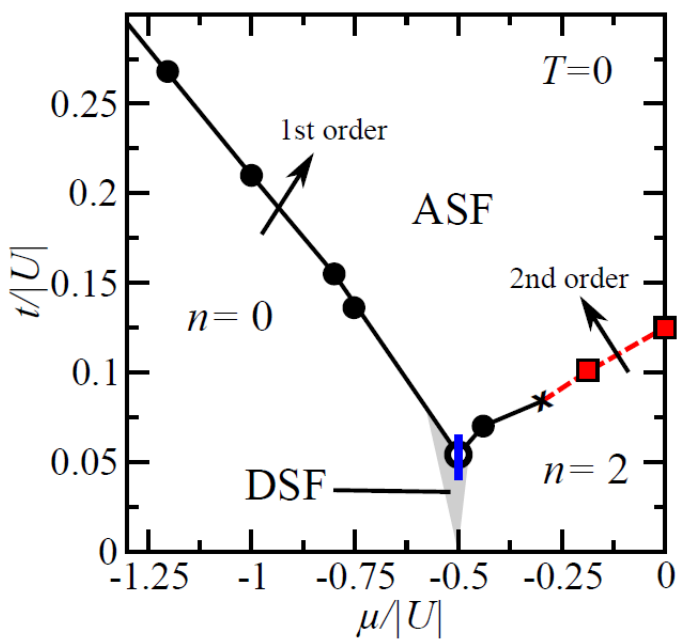
Pair superfluidity in attractive Bose gases

- pair condensation versus density collapse

Valatin (1958), Noziéres (1982)

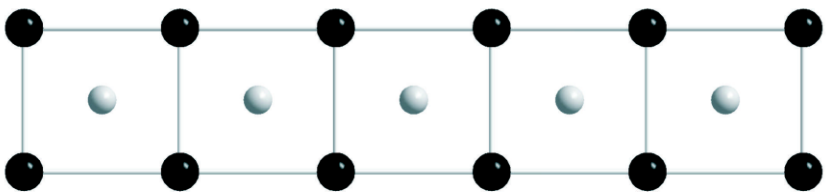
- constrained Bose-Hubbard model, $n_i \leq 2$

Bonnes & Wessel (2011)



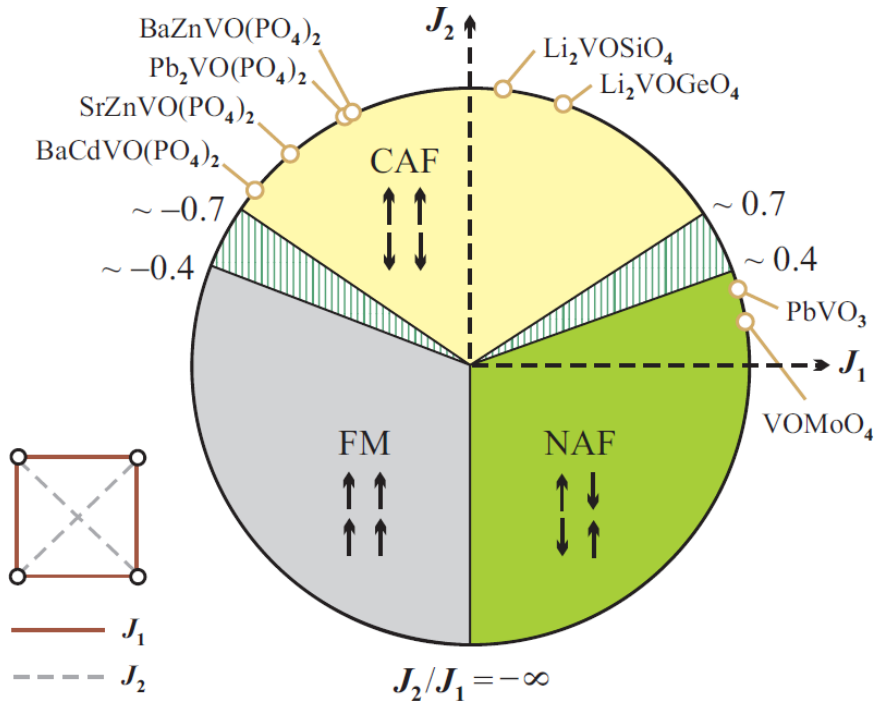
Frustrated ferromagnets

- chain materials $LiCuVO_4$, Li_2CuO_2, Li_2ZrCuO_4…



90° Cu-O-Cu bonds yield FM n.n. exchange $J_1<0$ with AF n.n.n. $J_2>0$

- layered square-lattice afms



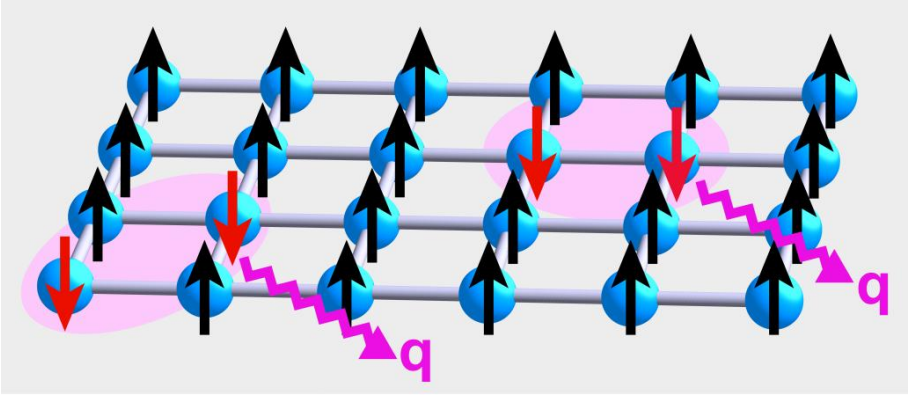
by A. Tsirlin

Magnon pairing in strong fields

- fully polarized state $|0\rangle = |\uparrow\uparrow\uparrow\rangle$ cf. Wortis (1963)

- single spin-flips (magnons) $|\psi_1\rangle = \sum f_i S_i^- |0\rangle$ unstable at H_{s1}

$$\varepsilon_k = H + \tfrac{1}{2}(J_k - J_0)$$



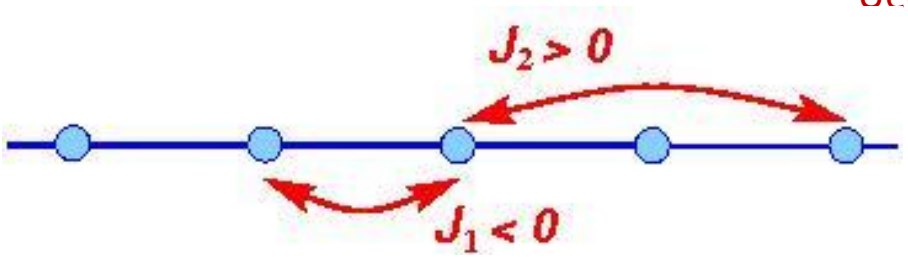
- pair of spin flips $|\psi_2\rangle = \sum f_{ij} S_i^- S_j^- |0\rangle$ unstable at $H_{s2} = H_{s1} + ½E_B$

$$(\varepsilon_2 - \varepsilon_{k/2+q} - \varepsilon_{k/2-q}) f_k(q) = \tfrac{1}{2}\sum_p (J_{p+q} + J_{p-q} - J_{k/2+q} - J_{k/2-q}) f_k(p)$$

Magnon pairs in 1D

- frustrated spin chain

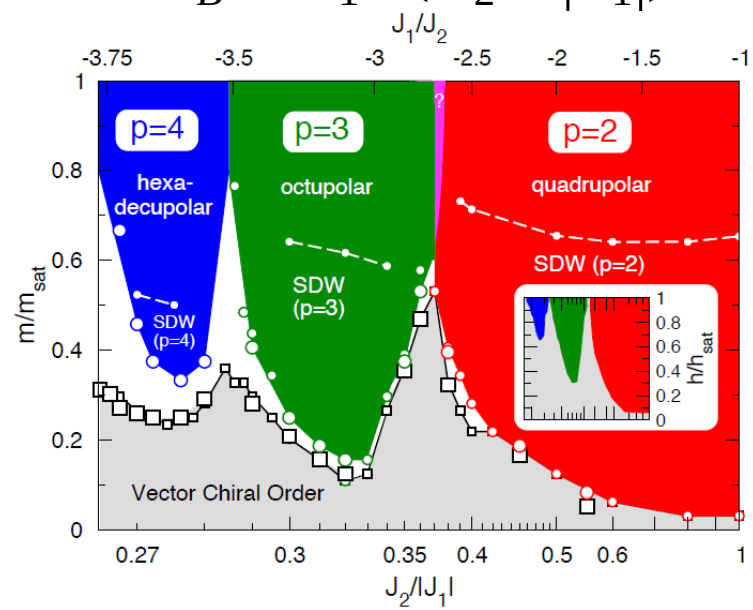
Chubukov (1991), Kuzian et al (2007), Ueda et al (2009)



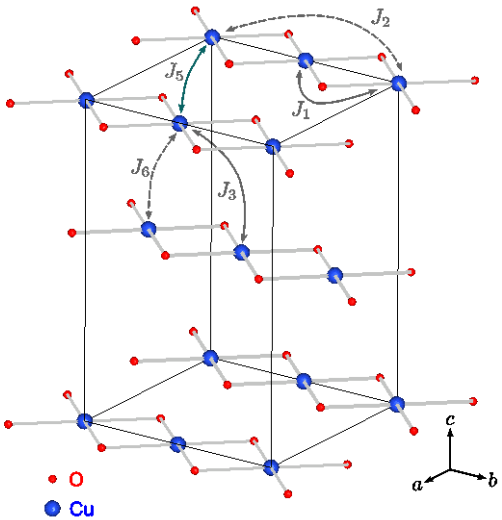
- bound magnon pairs with $k = \pi$ and $E_B = J_1^2 / (J_2 + |J_1|)$

- numerical studies

Honecker et al (2006), Hikihara et al (2008), Sudan et al (2009) Nishimoto et al (2011)

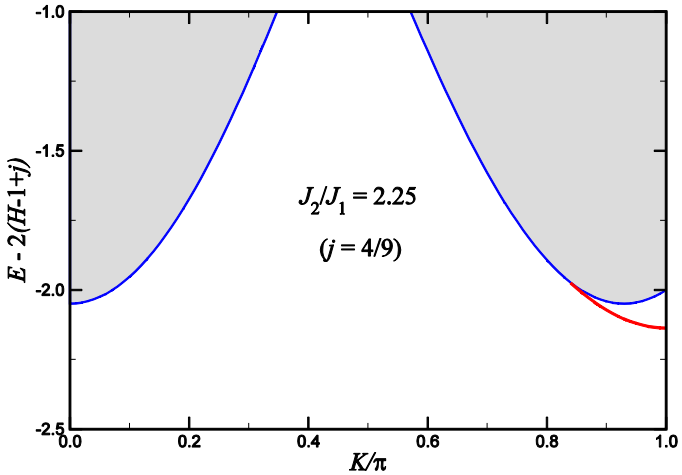


Magnon pairs in $LiCuVO_4$

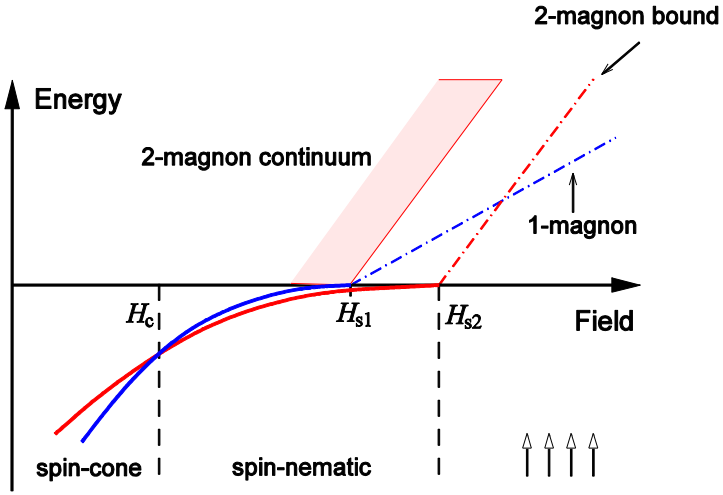


- weakly coupled chains

J_1 = -1.6meV, J_2 = 3.6meV, J_5 ~ -0.4meV Enderle et al (2004)



- pair condensation

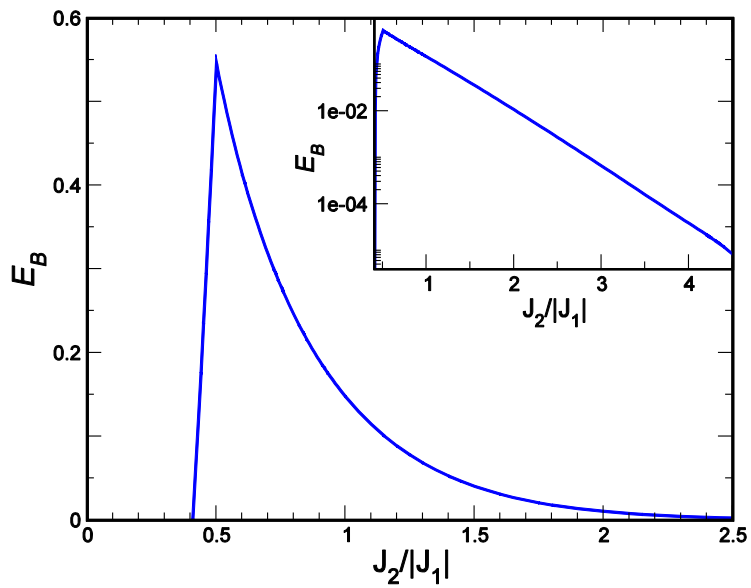


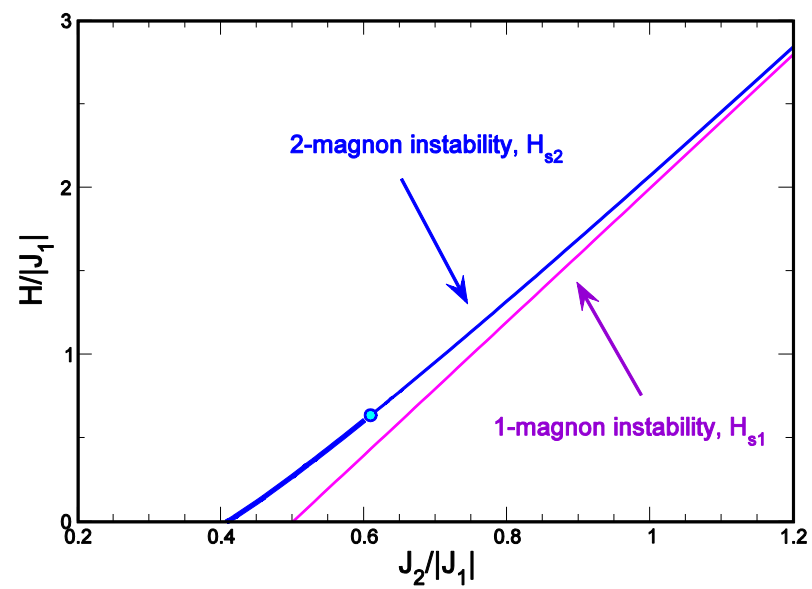
H_{s2} = 47.1T and H_{s1} = 46.5T

Tsunetsugu & MZ (2010)

Magnon pairs in 2D: J_1-J_2 SAFM

- bound pairs with ***k***=(0,0)





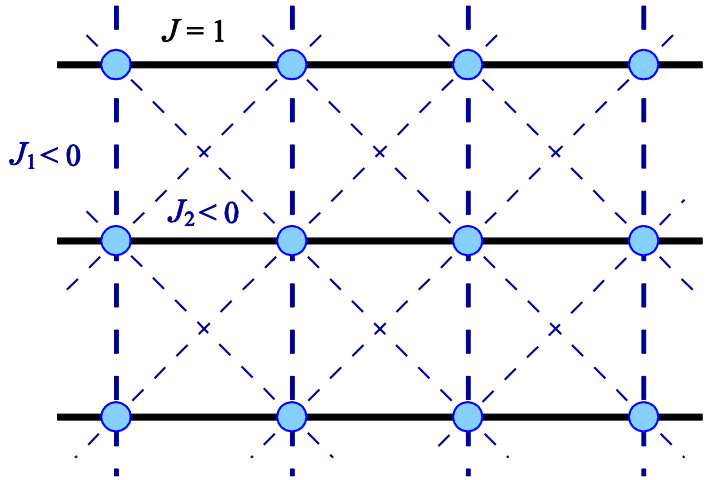
$$f_k(q) \propto (\cos q_x - \cos q_y)$$

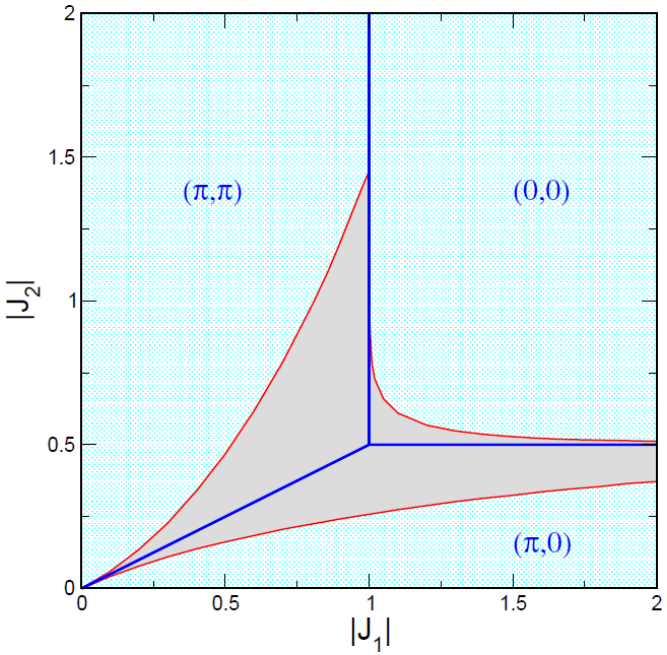
$$E_B = 2je^{-\pi j}$$

cf . Shanon et (2006)

Magnon pairs in 2D

- antiferromanetic chains coupled by frustrating ferromagnetic bond





- frustration is essential as it suppresses the kinetic energy of single spin flips

Condensate of magnon pairs

Tsunetsugu & MZ (2010)

- Bogolyubov theory via the coherent boson states

$$\langle \psi | a_0 | \psi \rangle \neq 0 \;\Rightarrow\; |\psi\rangle = e^{\psi a_0^+}|0\rangle \qquad (a_0|\psi\rangle = \psi|\psi\rangle)$$

- single-magnon condensate = canted AF state

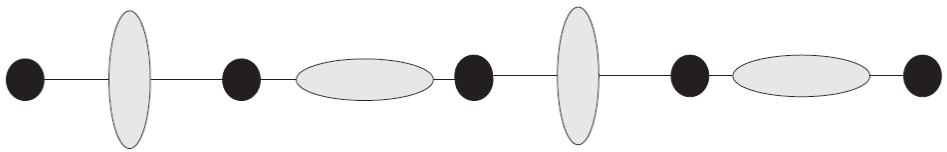
$$|\psi\rangle = \exp(\psi S_Q^-)|0\rangle \qquad \langle \psi | S_i^- | \psi \rangle \propto e^{iQr_i}$$

  - dipolar symmetry $\quad C_\pi^z |\psi\rangle = -|\psi\rangle$

- coherent pair condensate

$$|\Delta\rangle \cong \exp\left(\Delta \sum f_{ij} S_i^- S_j^-\right)|0\rangle$$

  - nematic symmetry $\quad C_\pi^z |\Delta\rangle = |\Delta\rangle$



Spin correlations

- absence of the sub-lattice magnetization $\langle \Delta | S_i^- | \Delta \rangle \equiv 0$

- exponential decay in transverse and longitudinal channels

$$\langle \Delta | S_i^- S_j^+ | \Delta \rangle \approx |\Delta|^2 \sum_l f_{il}^* f_{lj} \qquad \langle \Delta | \delta S_i^z \delta S_j^z | \Delta \rangle` \approx |\Delta|^2 |f_{ij}|^2$$

- quadrupolar (nematic) order parameter

$$Q_{ij}^{\alpha\beta} = \frac{1}{2} \langle S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha \rangle$$

$$Q_{ij}^{xx} + iQ_{ij}^{xy} = \frac{1}{2} \langle S_i^+ S_j^+ \rangle \approx \frac{\Delta}{2} f_{ij}$$

Self-consistent BCS-type theory

- bosonization via Holstein-Primakoff tranformation
- mean-field averages

$$\Delta_{ij} = \langle a_i a_j \rangle \qquad n_r = \langle a_i^+ a_{i+r} \rangle$$

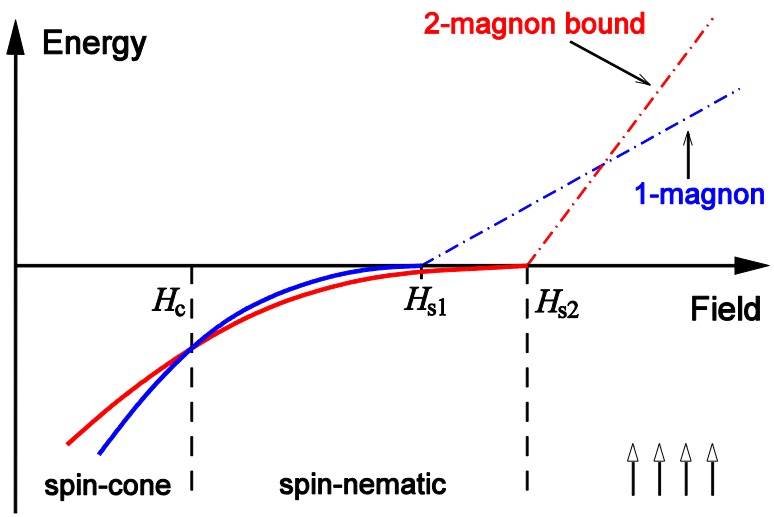
- quasiparticle excitations: unpaired magnons

$$\varepsilon_{k/2+q} = \sqrt{A_q^2 - B_q^2} - \sum_r J(r)\left(\tfrac{1}{2} - n - n_r\right)\sin qr \sin \tfrac{1}{2} kr$$

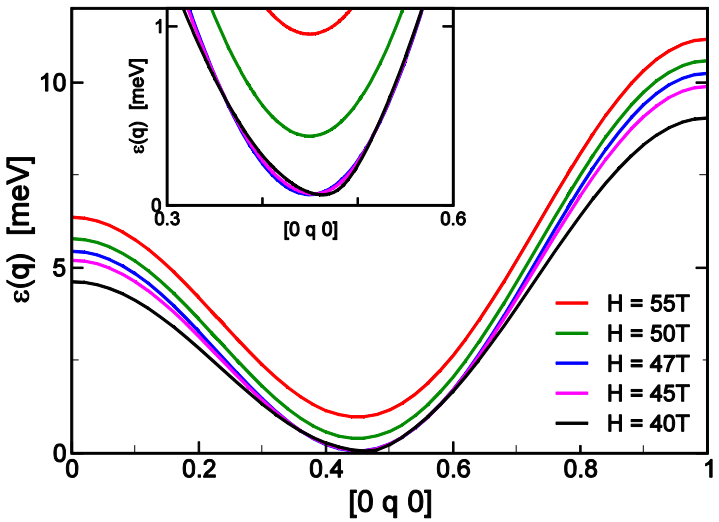
$$A_q = H - \sum J(r)\left(\tfrac{1}{2} - n - n_r\right)\cos qr \cos \tfrac{1}{2} kr \qquad B_q = \sum J(r)\Delta_r \cos qr$$

Self-consistentctheory: results

- energy-field diagram



- magnon excitations



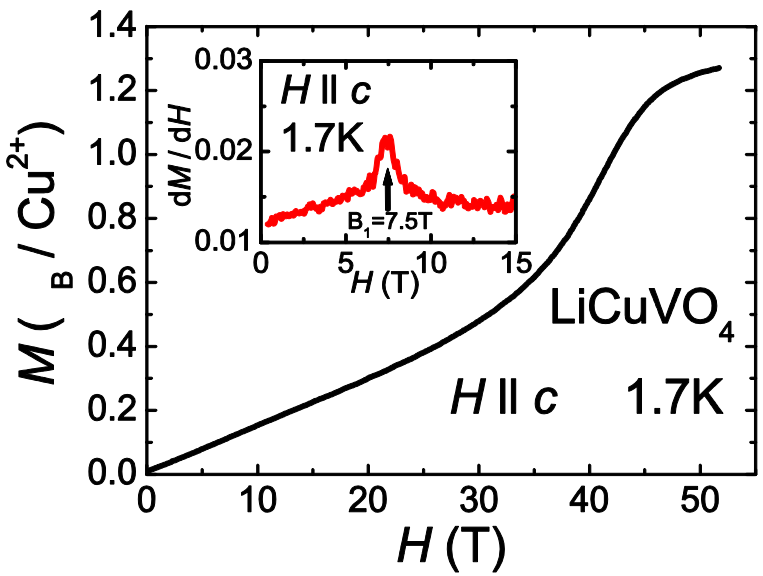
► for $LiCuVO_4$: $H_{s2} = 47T$ and $H_c = 44T$

Tsunetsugu & MZ, EPL **92**, 37001 (2010)

New ultra-high-field phase in $LiCuVO_4$

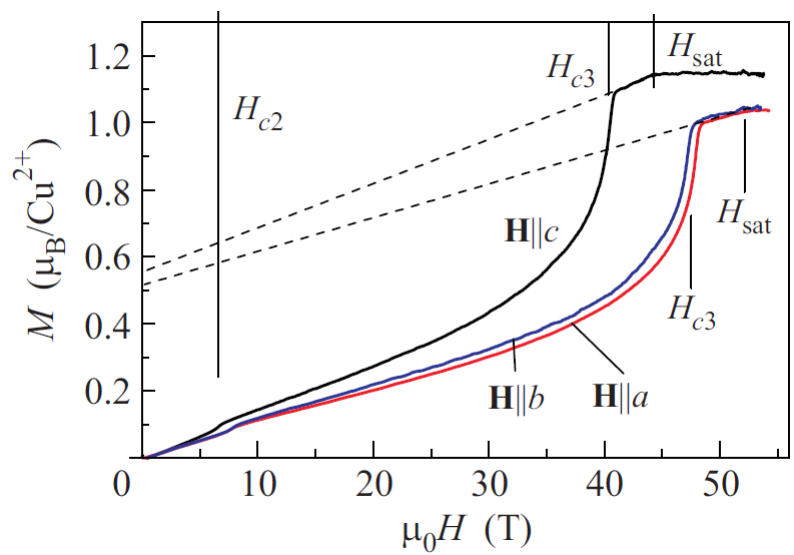
- pulsed field experiments

Banks et al (2006)



- ultra-high field phase

Svistov et al (2011)

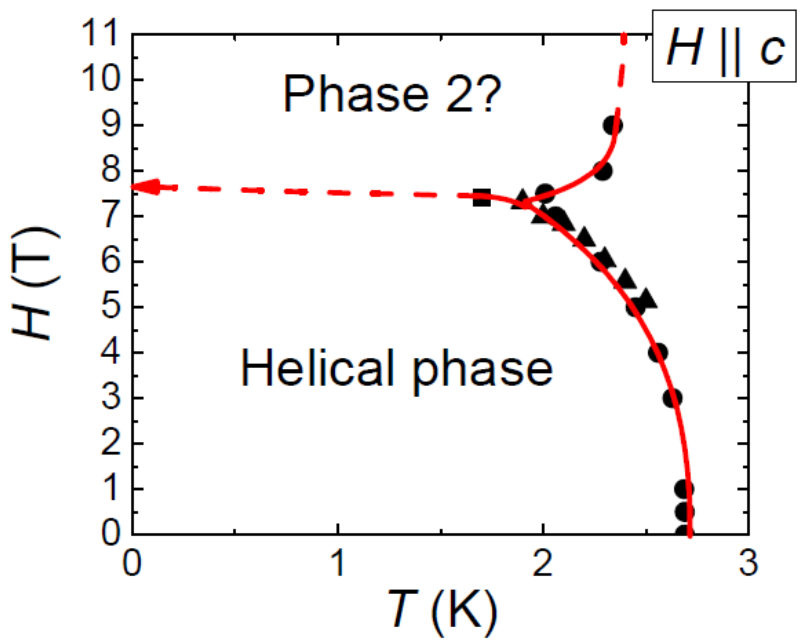


► **the ultra high-field phase is a transverse spin-nematic state**

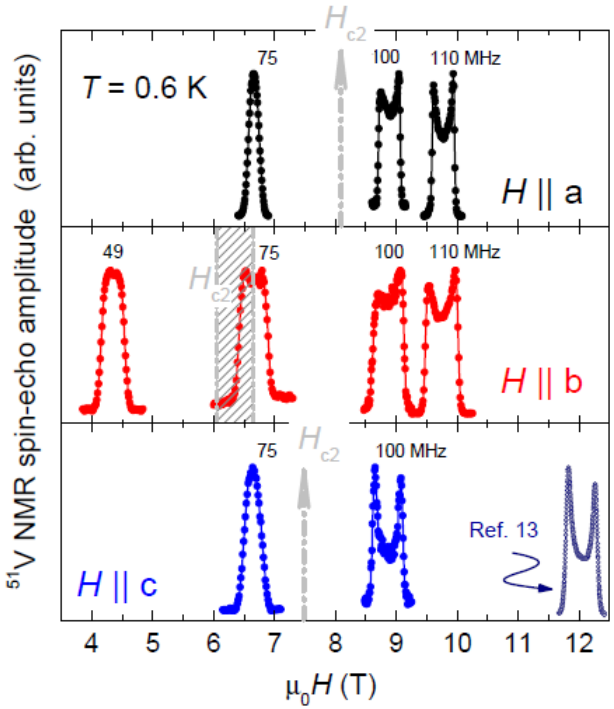
New high-field **phases** in $LiCuVO_4$

- ‘not so’ high-field phase: longitudinal SDW in isotropic AFM?

Banks et al (2006)



Buetgen et al (2008,10)

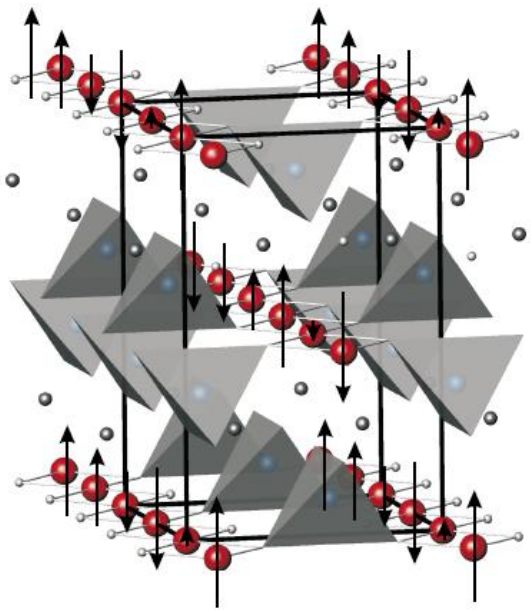


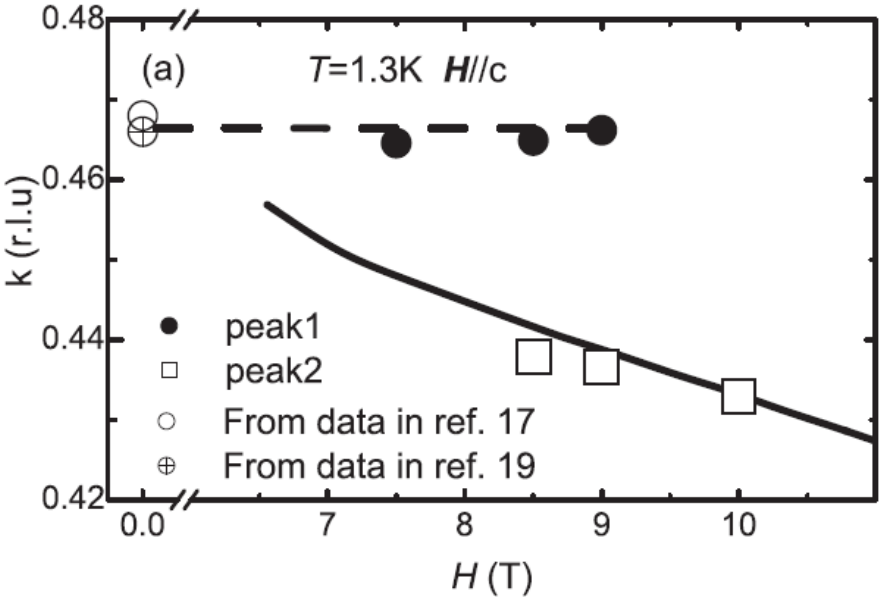
► **relation to the transverse spin-nematic at higher fields?**

Longitudinal SDW in $LiCuVO_4$

- neutron diffraction

Masuda et al (2011)





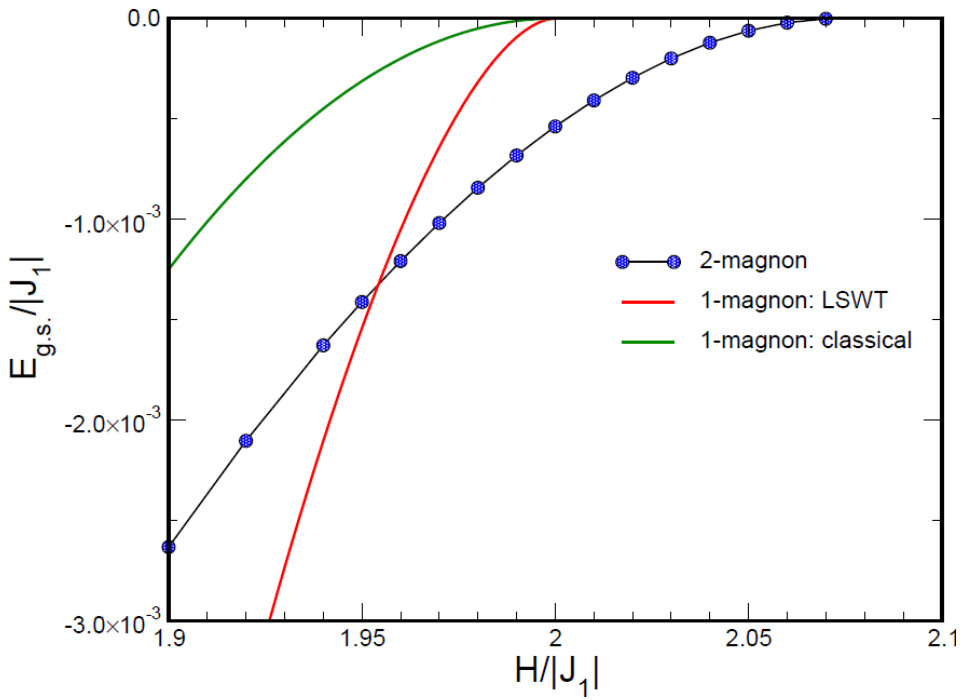
$$Q = \tfrac{1}{2}(1 - 2m^z)$$

► **long-range SDW order?**

2D spin-nematic?

- spin-nematic phase in frustrated SAFM $BaCdVO(PO_4)_2$?

Tsirlin et al (2008)



the high-field region is easily achievable in a Lab: H_s=4.2 T; various experimental techniques may be applied

Summary

- magnon pair condensate = spin nematic (quadrupole) state

- smoking gun for spin-nematic:

  hidden-order transitions, i.e. without evident order parameters

- dynamical properties: collective quadrupole mode in longitudinal channel?

- instabilities in the spin nematic phase: longitudinal IC state in $LiCuVO_4$

- nematic LRO in $BaCdVO(PO_4)_2$ and other frustrated 2D FMs

Thank you!