A05: Quantum crystal and ring exchange

Novel magnetic states induced by ring exchange

Members:

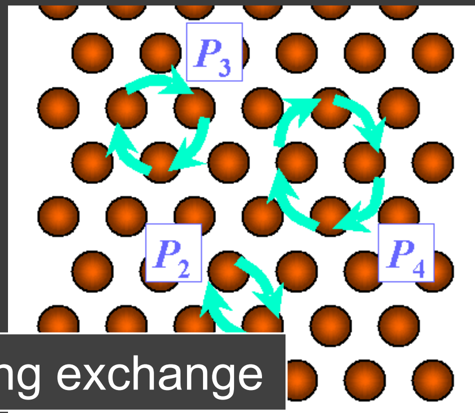
Tsutomu Momoi (RIKEN)
Kenn Kubo (Aoyama Gakuinn Univ.)
Seiji Miyashita (Univ. of Tokyo)
Hirokazu Tsunetsugu (ISSP, Univ. of Tokyo)

Takuma Ohashi (RIKEN → Osaka Univ.)
Masahiro Sato (RIKEN)

- Multiple-spin exchange model on the triangular lattice

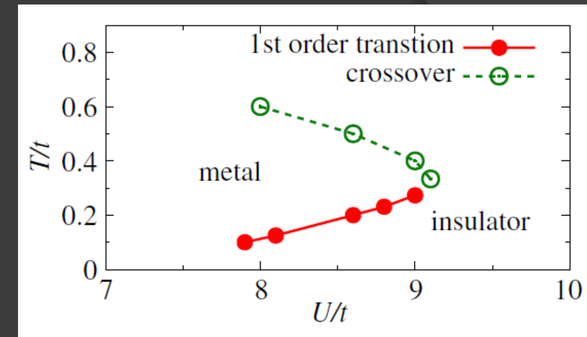
2D solid ^{3}He

(T. Momoi, K. Kubo)



Ring exchange

- Mott transition in frustrated electron systems

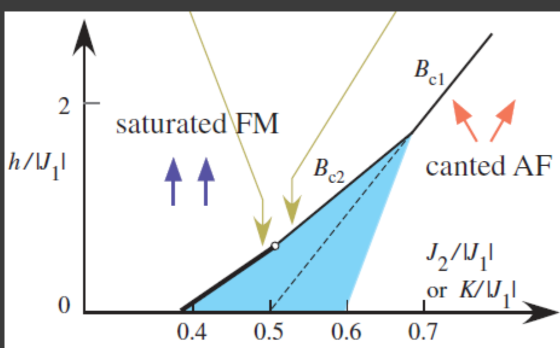


reentrant behavior

(T. Ohashi, T. Momoi, H. Tsunetsugu, N. Kawakami)

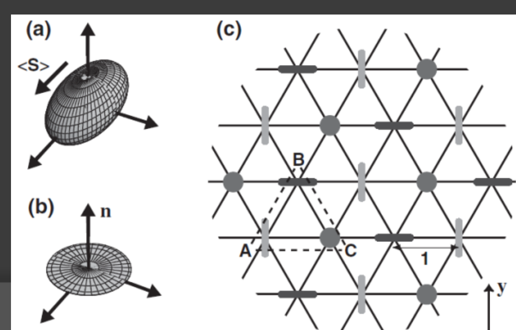
- Spin nematic/quadrupolar phases

S=1/2 frustrated ferromagnets



spin-triplet RVB state (T. Momoi)

S=1 bilinear-biqurdratic model (H. Tsunetsugu)



- Spin dynamics, spin crossover (S. Miyashita)
- Supersolid
- Magnetism in cold atoms (S. Miyashita)

Magnon pairing and crystallization in triangular lattice multiple-spin exchange model

--- SPIN NEMATIC PHASES IN FRUSTRATED MAGNETS ---

Tsutomu Momoi
（RIKEN）

Collaborators:

Philippe Sindzingre (Univ. of P. & M. Curie)
Kenn Kubo (Aoyama Gakuinn Univ.)
Nic Shannon (Bristol Univ.)
Ryuichi Shindou (RIKEN)

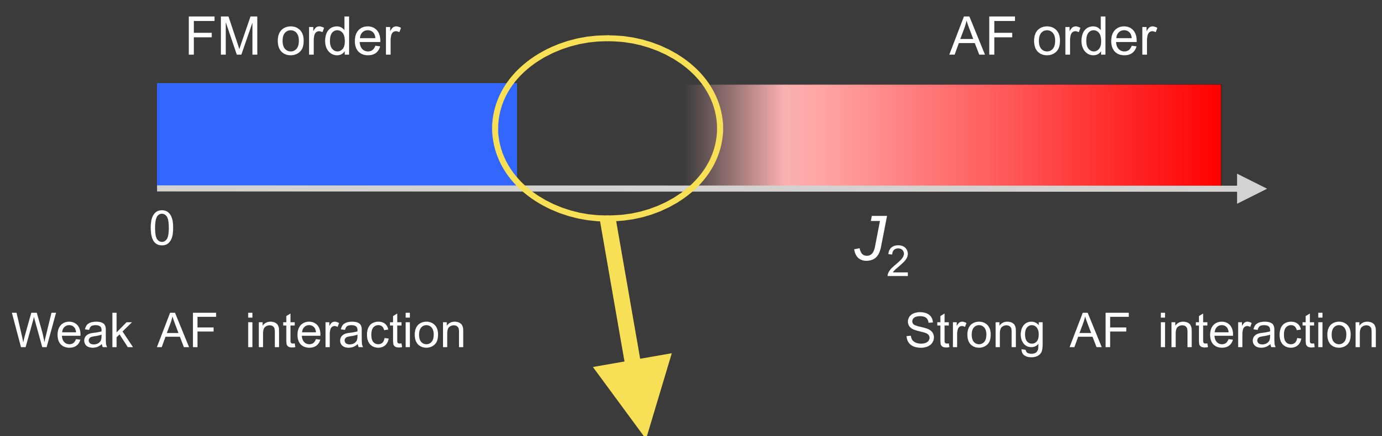
Outline

1. Introduction: Spin nematic order
   BEC of bound magnon pairs
   Spin-triplet RVB state
2. Multiple-spin exchange model: J-J_4-J_5-J_6 model
   Spin nematic phase, 1/2 magnetization plateau
3. Summary

Introduction: Competition between FM and AF orders

Nearest-neighbor FM interaction J_1

+ competing antiferromagnetic interaction J_2

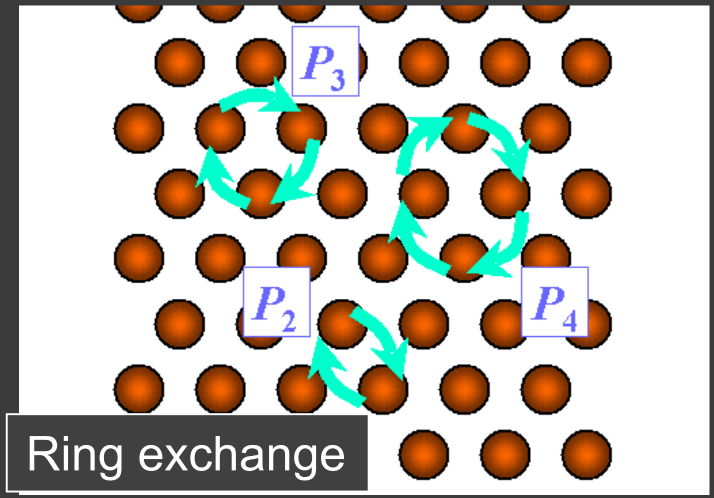


Emergence of new quantum phase

Frustrated magnets with 1st neighbor FM interaction

Triangular lattice

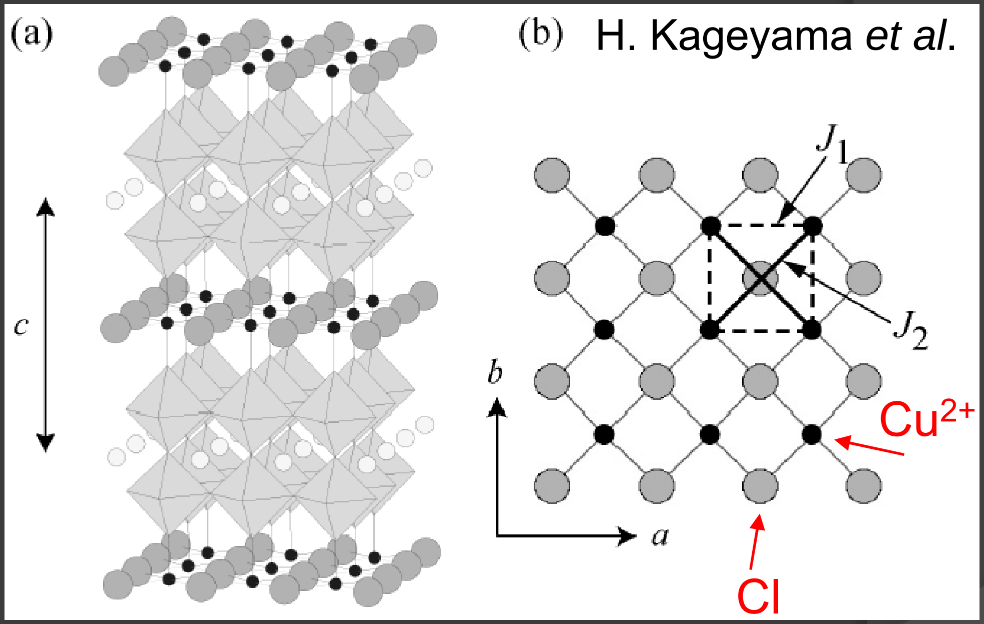
2D solid He3



Square lattice

$Pb_2VO(PO_4)_2$ E. Kaul *et al.*

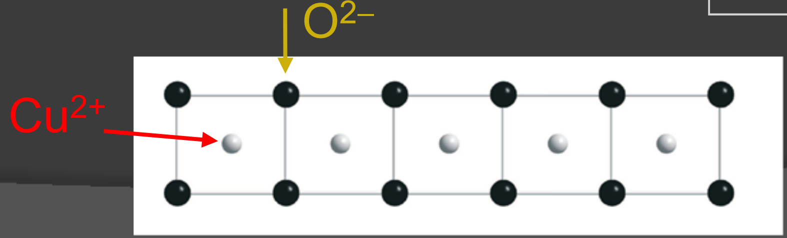
$(CuCl)LaNb_2O_7$, $(CuBr)A_2Nb_3O_{10}$



1D zigzag lattice

edge-sharing chain cuprates

$LiCuVO_4$, $LiCu_2O_2$, $Rb_2Cu_2Mo_3O_{12}$, Li_2ZrCuO_4



Kanamori-Goodenough Rule

FM nearest neighbor J_1

AF next nearest neighbor J_2

Spin nematic phase in between FM and AF phases

J_1-J_2 model

$$H = J_1 \sum_{\text{N.N.}} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\text{N.N.N}} \mathbf{S}_i \cdot \mathbf{S}_j,$$

Square lattice

N. Shannon, TM, and P. Sindzingre, *PRL* **96**, 027213 (2006).

1D zigzag lattice

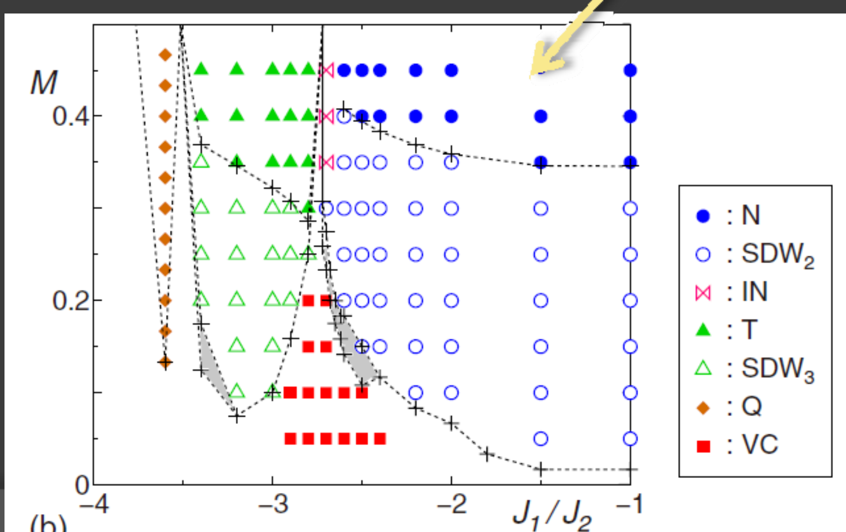
T. Hikihara, L. Kecke, TM, and A. Furusaki, PRB (2008)

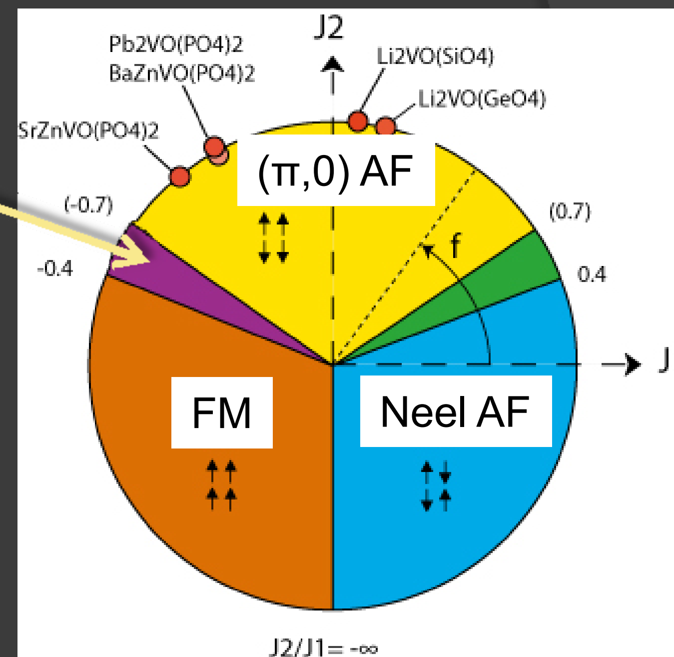
M. Sato, TM, and A. Furusaki, PRB (2009)

Poster P40

Nematic phase

FM J_1, AF J_2





FM nearest neighbor J_1

AF next nearest neighbor J_2

Characteristics of spin nematic order in spin-1/2 frustrated ferromagnets

N. Shannon, TM, and P. Sindzingre, *PRL* **96**, 027213 (2006).

- uniform state, i.e. no crystallization
- no spin order $\langle \vec{S}_i \rangle = 0$ at h=0

  or no transverse spin order $\langle S_i^x \rangle = \langle S_i^y \rangle = 0$ for h>0
- gapless excitations

} spin liquid-like behavior

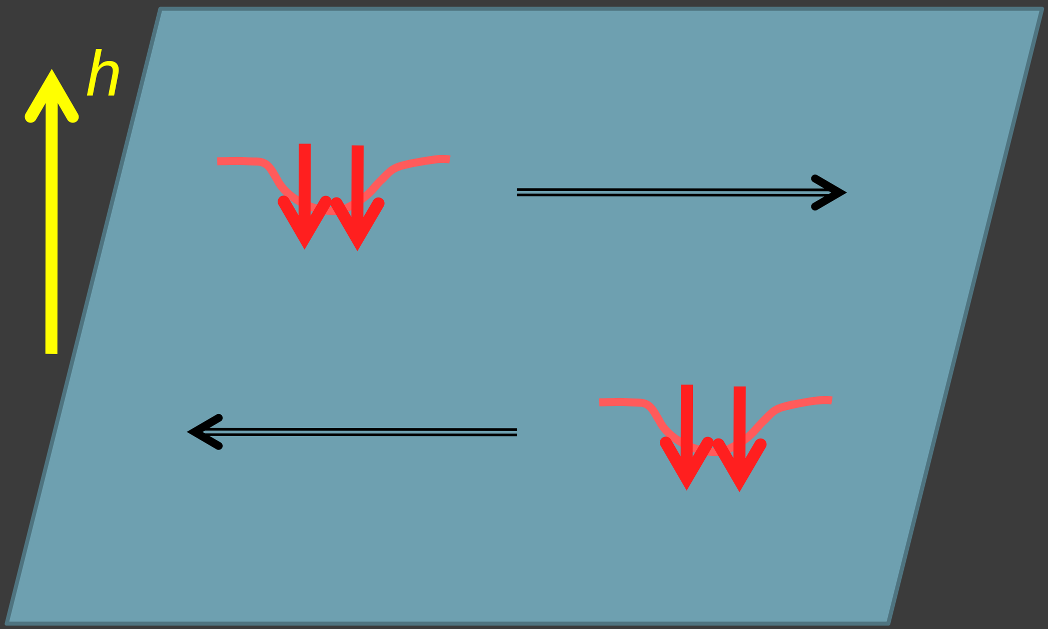
- spin quadrupolar order

$$\langle Q_{ij}^{x^2-y^2} \rangle = \langle S_i^x S_j^x - S_i^y S_j^y \rangle, \qquad \langle Q_{ij}^{xy} \rangle = \langle S_i^x S_j^y + S_i^y S_j^x \rangle$$

Bond-nematic order

Spin nematic order can be regarded as

"BEC of bound magnon pairs with ***k*** =(0,0)"



A. V. Chubukov, PRB (1991)

N. Shannon, TM, and P. Sindzingre, PRL (2006).

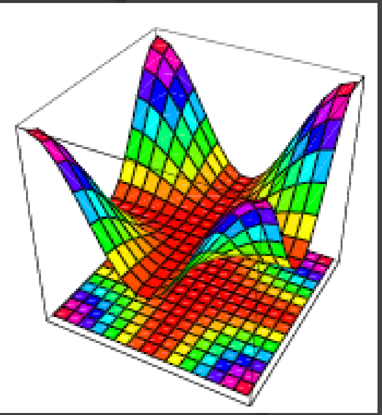
phase coherence $\left\langle S_i^- S_j^- \right\rangle = Qe^{2i\theta}$

spin quadrupolar order

$$\left\langle S_i^- S_j^- \right\rangle = \left\langle S_i^x S_j^x - S_i^y S_j^y \right\rangle - i\left\langle S_i^x S_j^y + S_i^y S_j^x \right\rangle = Qe^{2i\theta}$$

$x^2 - y^2$ $\quad$ xy

Why bound magnon pairs are stable in frustrated FM ?

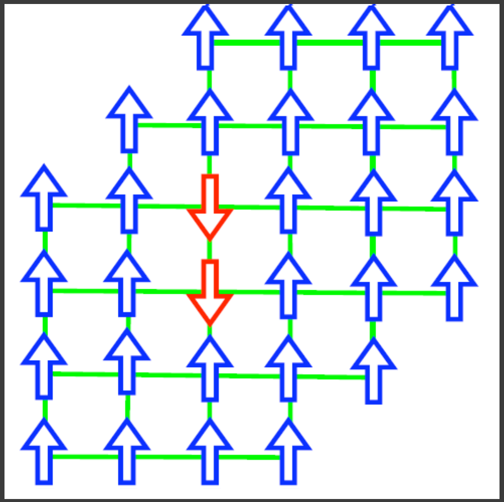


Near saturation field,

1. Individual magnons are nearly localized

In square-lattice J_1-J_2 model,
zero line modes at $J_2/|J_1| = ½$.

2. Two (or three) magnon bound states are mobile and stable



In square-lattice J_1-J_2 model,
d-wave two-magnon bound states
with $\boldsymbol{k}$ =(0,0) are most favored.

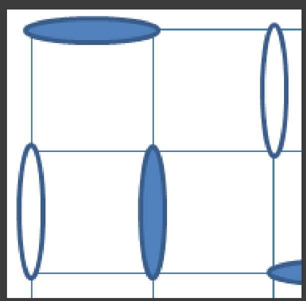
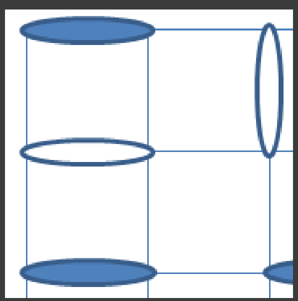
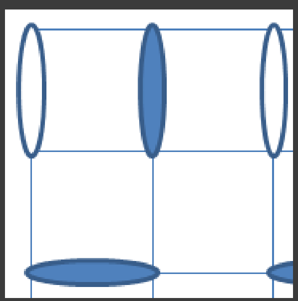
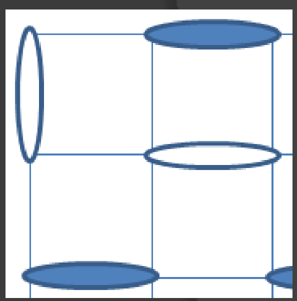
Coherent motion

Bond-nematic ordered state in S=1/2 magnets

Roughly speaking,…..

Linear combination of all possible configurations of $S^z = \pm 1$ dimers

$$\sum_{\substack{\text{dimer}\\ \text{configuration}}} (-1)^{\#\text{ of vertical } S^z=1 \text{ dimers}} \left| \text{dimers with } S^z = \pm 1 \right\rangle$$

=  --  +  -- 

+ ……..

entangled state

cf. Spin quadrupolar order state in S = 1 bilinear-biquadratic model

$$\text{wave function} \approx \otimes_i \left| \phi_i \right\rangle$$

product state

$$\left\langle Q_i^{x^2-y^2} \right\rangle = \left\langle S_i^x S_i^x - S_i^y S_i^y \right\rangle$$

$$\left\langle Q_i^{xy} \right\rangle = \left\langle S_i^x S_i^y + S_i^y S_i^x \right\rangle$$

Site-nematic order

Slave boson formulation of spin nematic states in frustrated ferromagnets

R. Shindou and TM, PRB (2009)

Fermion representation

$$S_j^{\mu} = \frac{1}{2} f_{j\alpha}^{\dagger} \left[\sigma_{\mu} \right]_{\alpha\beta} f_{j\beta} \qquad (\mu = x, y, z)$$

Local constraint $f^{\dagger}_{j,\alpha} f_{j,\alpha} \equiv 1$

$f_{j\uparrow}, \ f_{j\downarrow}$ fermion operators

Using Hubbard-Stratonovich transformation, we can decouple FM interaction into triplet pairing

$$-4\boldsymbol{S}_i \cdot \boldsymbol{S}_j \rightarrow -|\boldsymbol{D}_{ij}|^2 + \sum_{\mu=x,y,z} [\psi_i^{\dagger} U_{ij,\mu} \psi_j \tau_{\mu}^{\ t}], \qquad U_{ij,\mu}^{tri} = \begin{bmatrix} 0 & D_{ij,\mu} \\ -D_{ij,\mu}^{*} & 0 \end{bmatrix}$$

where $\boldsymbol{D}_{ij}$ denote d-vectors of triplet pairing

$$\psi_j = \begin{bmatrix} f_{j,\uparrow} & f_{j,\downarrow} \\ f_{j,\downarrow}^{\dagger} & -f_{j,\uparrow}^{\dagger} \end{bmatrix}$$

$$\hat{\Delta}_{jl} = \begin{pmatrix} \langle f_{j\uparrow} f_{l\uparrow} \rangle & \langle f_{j\uparrow} f_{l\downarrow} \rangle \\ \langle f_{j\downarrow} f_{l\uparrow} \rangle & \langle f_{j\downarrow} f_{l\downarrow} \rangle \end{pmatrix} = \begin{pmatrix} -D_{jl}^{x} + iD_{jl}^{y} & D_{jl}^{z} \\ D_{jl}^{z} & D_{jl}^{x} + iD_{jl}^{y} \end{pmatrix}$$

In mean-field approximation, FM interaction prefers triplet pairing.

Theoretical description of bond-nematic states

When triplet pairing appears, spin space becomes anisotropic.

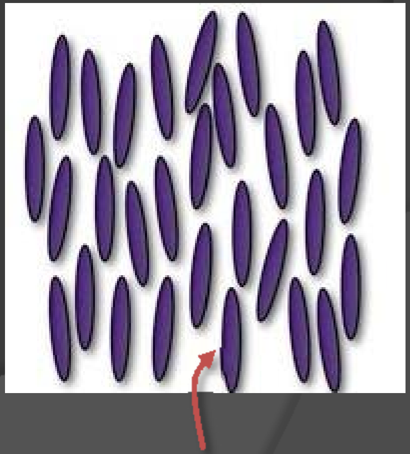
Quadrupolar order parameter in mean-field approximation

$$-2Q_{jl}^{\mu\nu} = D_{jl}^{\mu} D_{jl}^{\nu} - \frac{\delta_{\mu\nu}}{3} \left| \boldsymbol{D}_{jl} \right|^2 + \text{H.c.}$$

For example, $\left\langle S_j^- S_l^- \right\rangle = \left\langle f_{j\downarrow}^{\dagger} f_{j\uparrow} f_{l\downarrow}^{\dagger} f_{l\uparrow} \right\rangle = \left\langle f_{j\uparrow} f_{l\uparrow} \right\rangle^* \left\langle f_{j\downarrow} f_{l\downarrow} \right\rangle = -\left(D_{jl}^x\right)^2 + \left(D_{jl}^y\right)^2 - \mathrm{i}\left(D_{jl}^x D_{jl}^y + D_{jl}^y D_{jl}^x\right)$

cf. nematic order in liquid crystals, $\boldsymbol{d}\,(\boldsymbol{r})$: director vectors

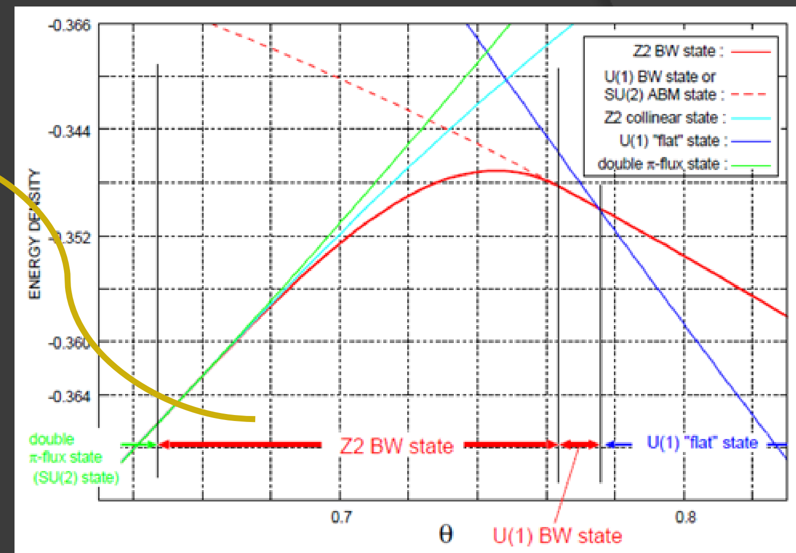
$$Q^{\mu\nu}(r) = d^{\mu}(r) d^{\nu}(r) - \frac{\delta_{\mu\nu}}{3} \left| \boldsymbol{d}(r) \right|$$



director – D-vector correspondence

Mean-field approximation of square lattice J_1-J_2 model

$$(J_1, J_2) \equiv J(\sin\theta, \cos\theta)$$



New phase

triplet-pairing on FM interactions and

hopping amplitude on AF interactions

spin-triple resonating valence bond state

(spin-triplet RVB state)

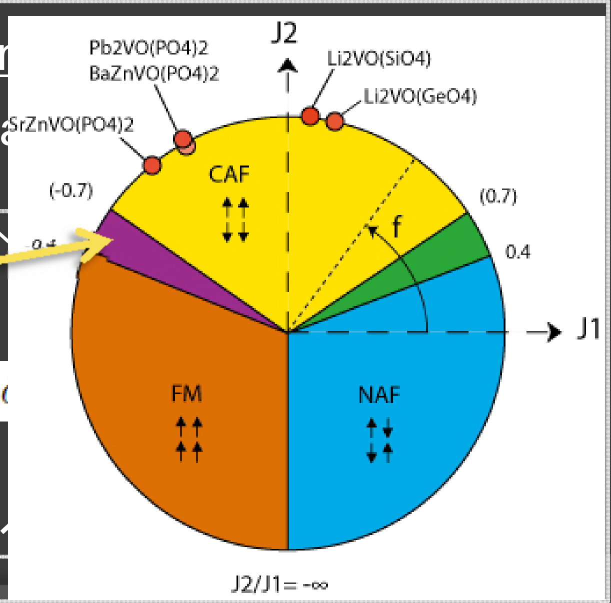
NN bon

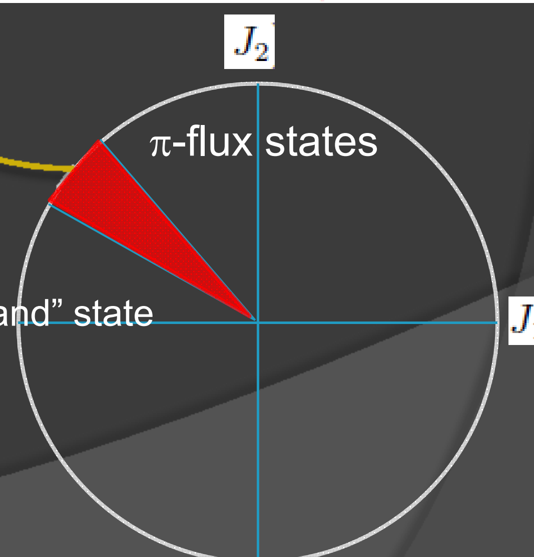
✓ Balia

$-iD_y$

Nematic phase

FM J_1, AF J_2

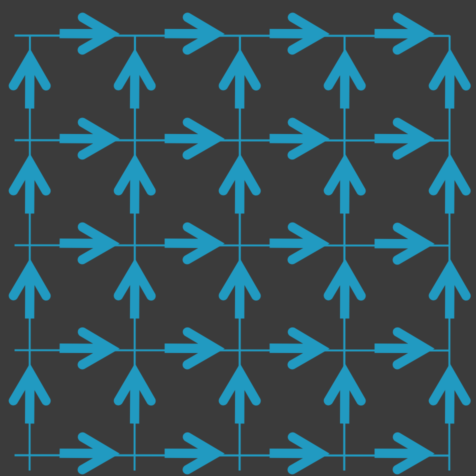




N. Shannon, TM and P. Sindzingre ('06)

This mean-field solution has the same magnetic structure as d-wave bond nematic state.

BW state $\mathbf{d}(k) \equiv \hat{x}\sin k_x + \hat{y}\sin k_y.$

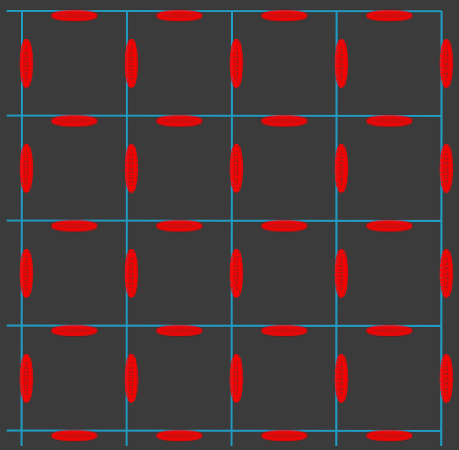


$$D^x_{jl} = \mathrm{i}\left(\delta_{j,l+e_x} - \delta_{j,l-e_x}\right)$$

$$D^y_{jl} = \mathrm{i}\left(\delta_{j,l+e_y} - \delta_{j,l-e_y}\right)$$

d-wave bond nematic state

N. Shannon, TM and P. Sindzingre (‘06)



$Q_{xx} - Q_{yy} > 0$

$Q_{xx} - Q_{yy} < 0$

Low energy excitations around the BW state

- Spin fluctuation has gapless Nambu-Goldstone modes
- Individual spinon excitations have a full gap

$$2E_{\pm} \equiv \pm\sqrt{J_1^2 D^2(\sin^2 k_x + \sin^2 k_y) + 4J_2^2\left(\chi^2 \cos^2 k_x \cos^2 k_y + \eta^2 \sin^2 k_x \sin^2 k_y\right)}.$$

- Gauge fluctuation also has a gap. *(a gapped Z_2 state)*

Perspectives

Variational Monte Carlo simulation

Magnetism of two-dimensional solid ^{3}He on graphite

4/7 phase in 2nd layer of 2D solid ^{3}He on graphite

gapless spin liquid

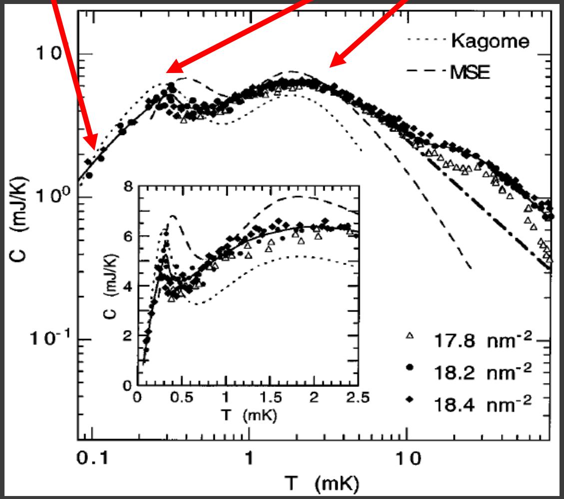
magnetization plateau at 1/2

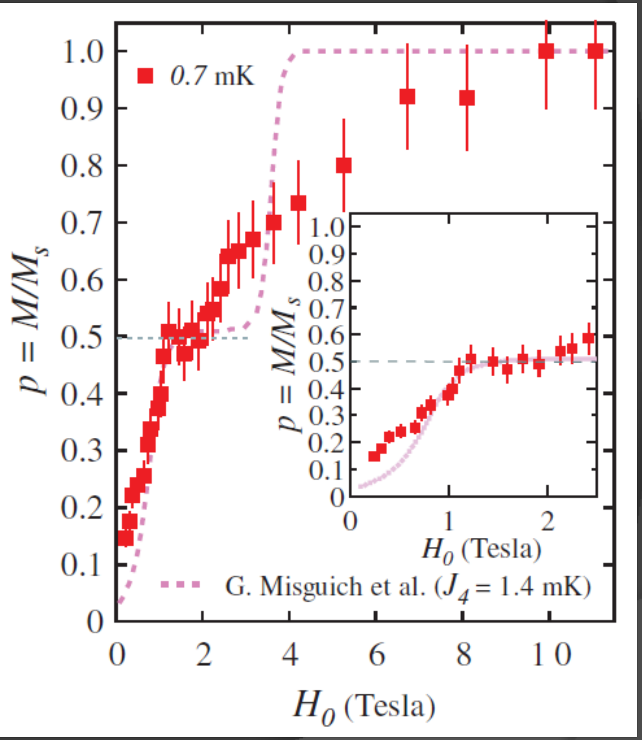
✓ specific heat

K. Ishida, M. Morishita, H. Fukuyama, PRL (1997)

linear specific heat (cf. 2D FM)

double peak structure





✓ No drop of susceptibility down to 10μK

R. Masutomi, Y. Karaki, and H. Ishimoto, PRL (2004).

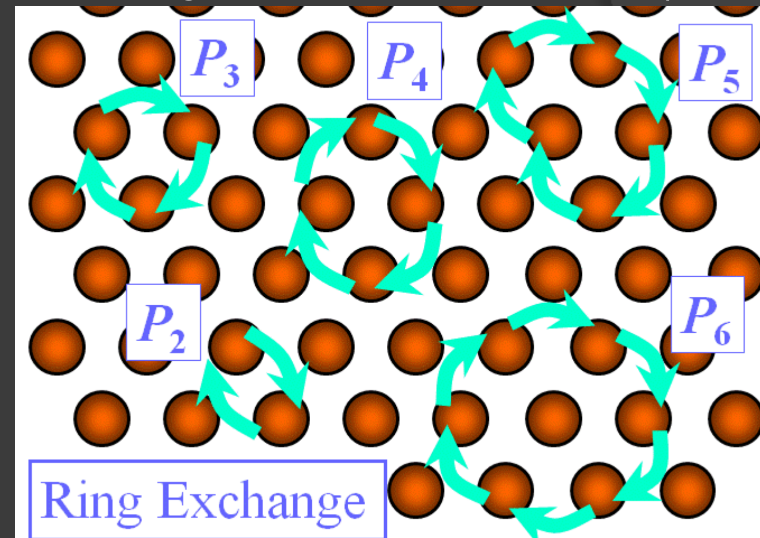
H. Nema, A. Yamaguchi, T. Hayakawa, and H. Ishimoto, *PRL* (2009).

Theoretical model: multiple-spin exchange model

Ring-exchange interactions

Dirac,
Roger, Hetherington, Delrieu, RMP **55**, 1 (1983)

$$\mathcal{H} = J_2 \sum P_2 + \sum_{n>2} (-1)^n J_n \sum (P_n + P_n^{-1})$$
$$= J \sum P_2 + J_4 \sum (P_4 + P_4^{-1})$$
$$- J_5 \sum (P_5 + P_5^{-1}) + J_6 \sum (P_6 + P_6^{-1})$$



Three spin exchange is dominant and ferromagnetic

$$P_3 + P_3^{-1} = P_2(i,j) + P_2(j,k) + P_2(k,i)$$

→ effective two spin exchange is ferromagnetic
($J=J_2-2J_3$)

"Frustrated ferromagnet"

Parameter fitting   Collin et al., PRL **86**, 2447 (2001).

$J=-2.8$, $J_4=1.4$, $J_5=0.45$, $J_6=1.25$ (mK)

In case of two- and four-spin exchange model (*J*-J_4 model)

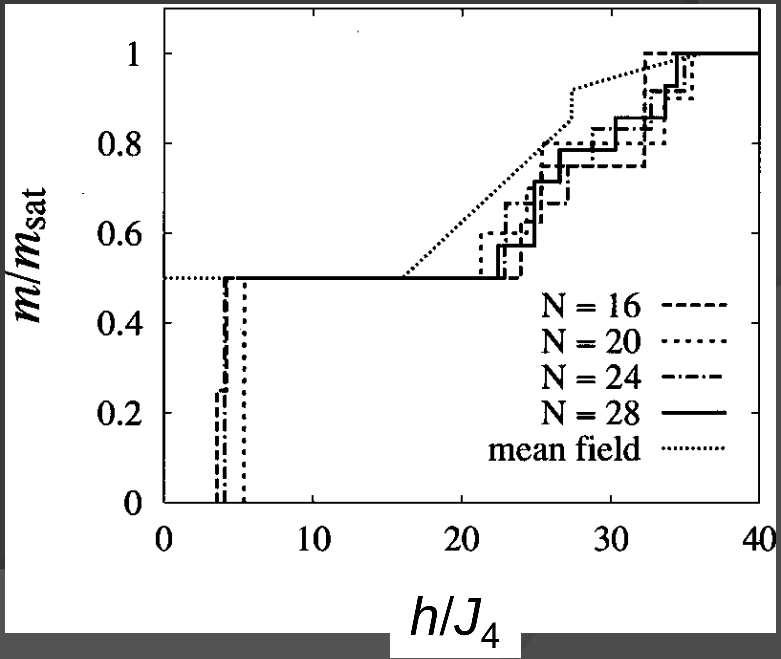
In a strong J_4 regime $J_4/|J| = ½$

- ☐ At zero field, the ground state doesn't have any order and it has a large spin gap.

  G.Misguich, B.Bernu, C.Lhuillier, and C.Waldtmann, PRL (1998)

- ☐ Magnetization process has a wide plateau at m/m_{sat} = ½, which comes from *uuud* spin-density wave structure

  TM, H. Sakamoto, and K.Kubo, PRB (1999)



In case of two- and four-spin exchange model (J-J_4 model)

Near the border of FM phase <u>*0.24 < K/|J| < 0.28*</u>

TM, P. Sindzingre, N. Shannon, PRL (2006)

- $m > 0$,

  condensation of 3 magnon bound states

  → "Triatic order" (octupolar order)

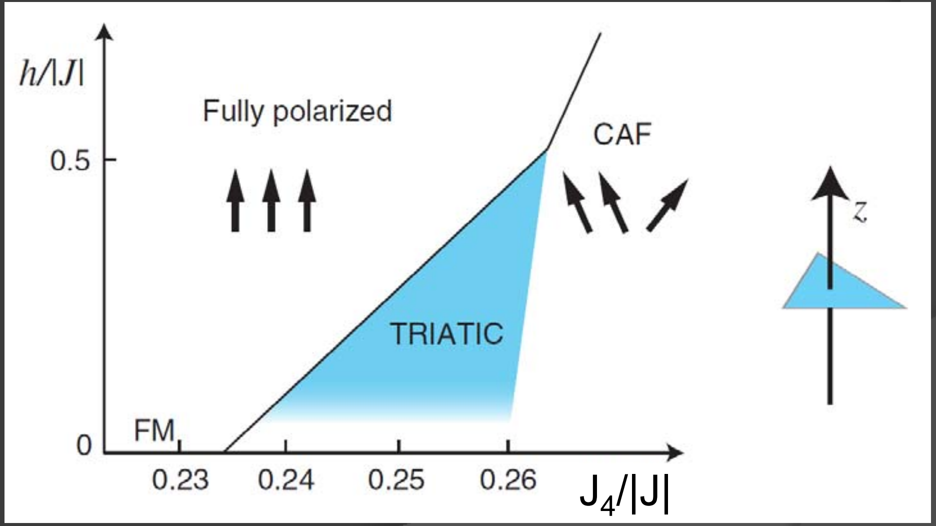
$$\left\langle S_i^- S_{i+e_1}^- S_{i+e_2}^- \right\rangle = \varphi e^{3i\vartheta}$$

$$\left\langle S_i^x \right\rangle = \left\langle S_i^y \right\rangle = 0$$

- $m = 0$,

  strong competition between

  nematic and triatic correlations



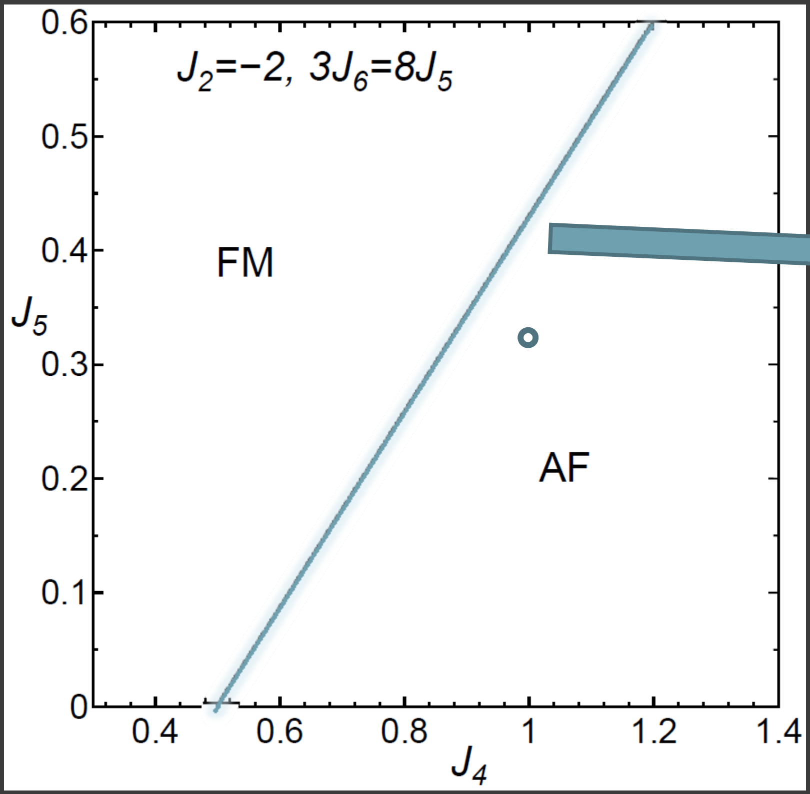
cf. $J_4/|J|=0.5$, $J_5/|J|=0.16$, $J_6/|J|=0.44$ Collin et al.

J-J_4-J_5-J_6 ring-exchange model

We aim at giving a quantitative comparison with experiments.

- In the classical limit (S→ ∞)

Mean-field phase diagram



- In the quantum case (S=1/2)

One magnon excitations

$$\varepsilon(k) = h - 2\left(J_2 + 4J_4 - 10J_5 + 2J_6\right) \times \left\{3 - \cos\mathbf{k}\cdot\mathbf{e}_1 - \cos\mathbf{k}\cdot\mathbf{e}_2 - \cos\mathbf{k}\cdot\mathbf{e}_3\right\}$$

have zero flat mode

at mean-field phase boundary.

Individual magnons are localized !

Magnon instability to the FM (fully polarized) state at saturation field

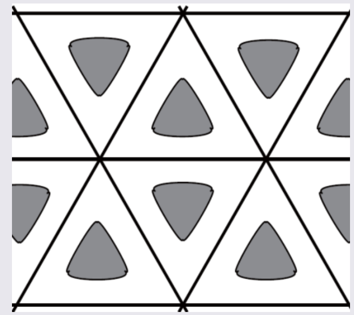
J-J_4- J_2-J_4 model

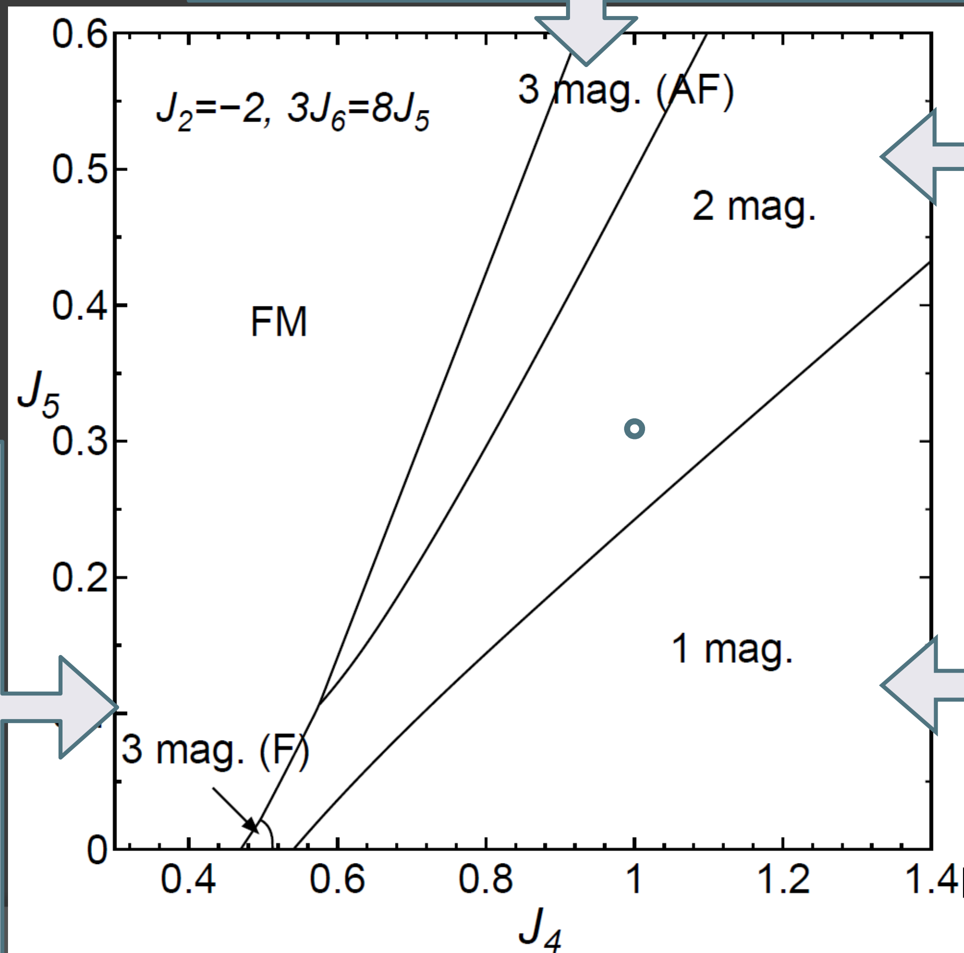
(case of $J_5=J_6=0$) $J_2=-2$

space rotation

$R_{\pi/3} \rightarrow -1$

Antiferro-triatic state





space rotation

$R_{\pi/3} \rightarrow \exp(\pm i 2\pi / 3)$

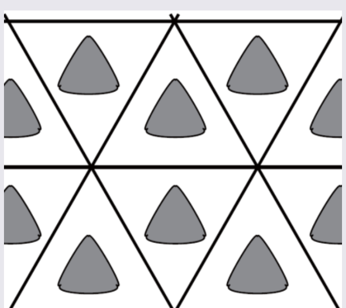
d+i*d* *wave*

Chiral (?) nematic state

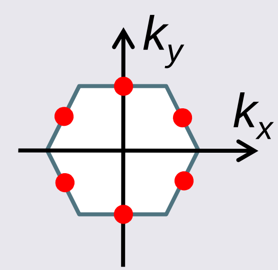
space rotation

$R_{\pi/3} \rightarrow 1$

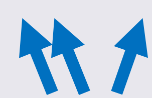
Ferro-triatic state



3 sublattice structure

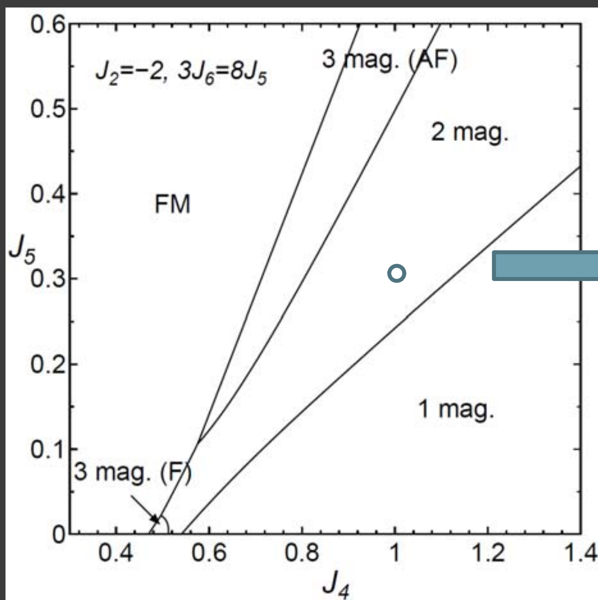


Canted AF

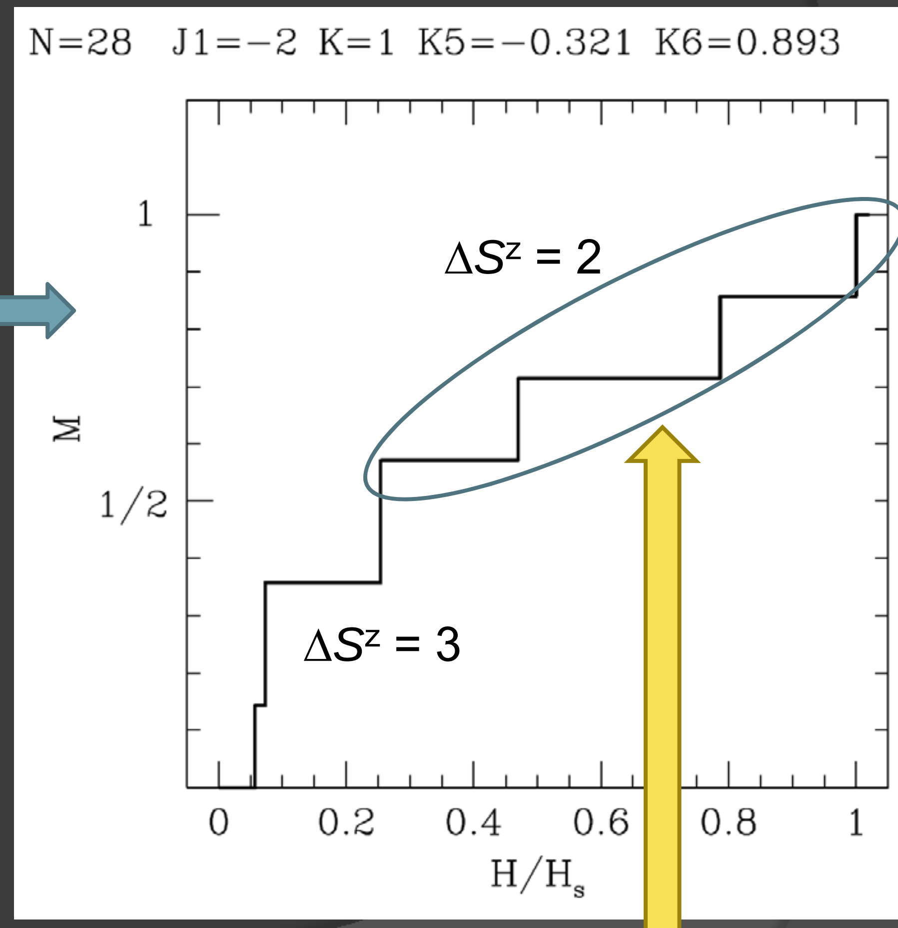


Numerical results

Instability at saturation



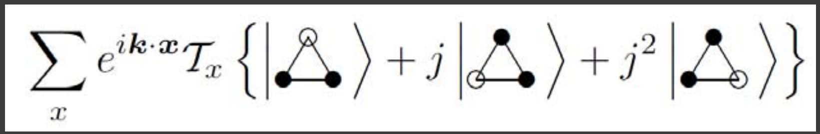
magnetization process



Condensation of *d*+i*d*-wave magnon pairs （BEC）

Condensation of bosons with two spices

d±i*d*-wave magnon pairs

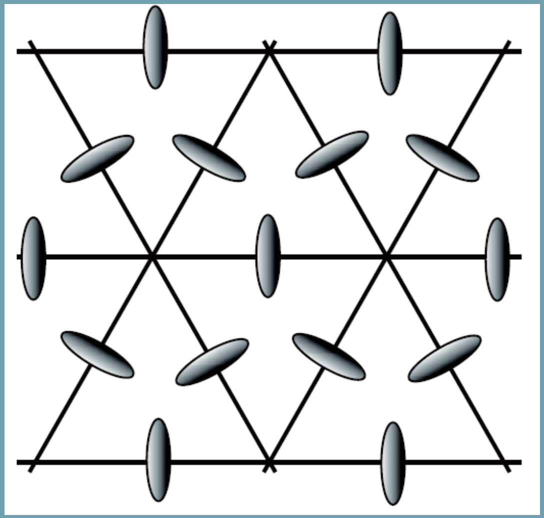


$j = \exp\left(\pm i \frac{2\pi}{3}\right)$ (±: chirality)

wave number ***k*** = (0,0)
double-fold degeneracy with chirality

density imbalance $n_+ > n_-$

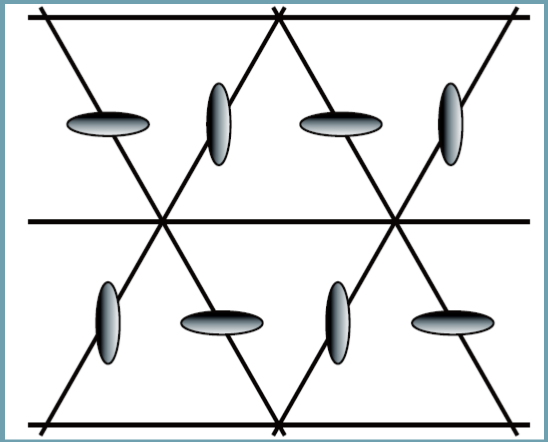
chiral nematic order



$$O_{d+id} = \sum_i \left(S_i^- S_{i+e_1}^- + j S_i^- S_{i+e_2}^- + j^2 S_i^- S_{i+e_3}^- \right)$$

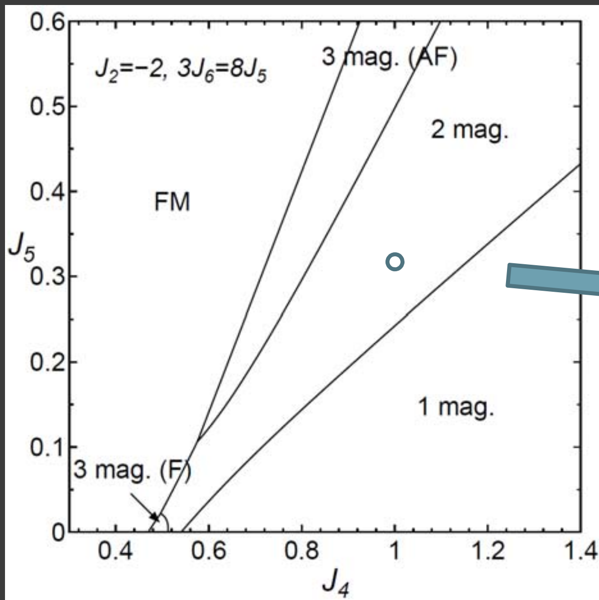
equal density $n_+ = n_-$

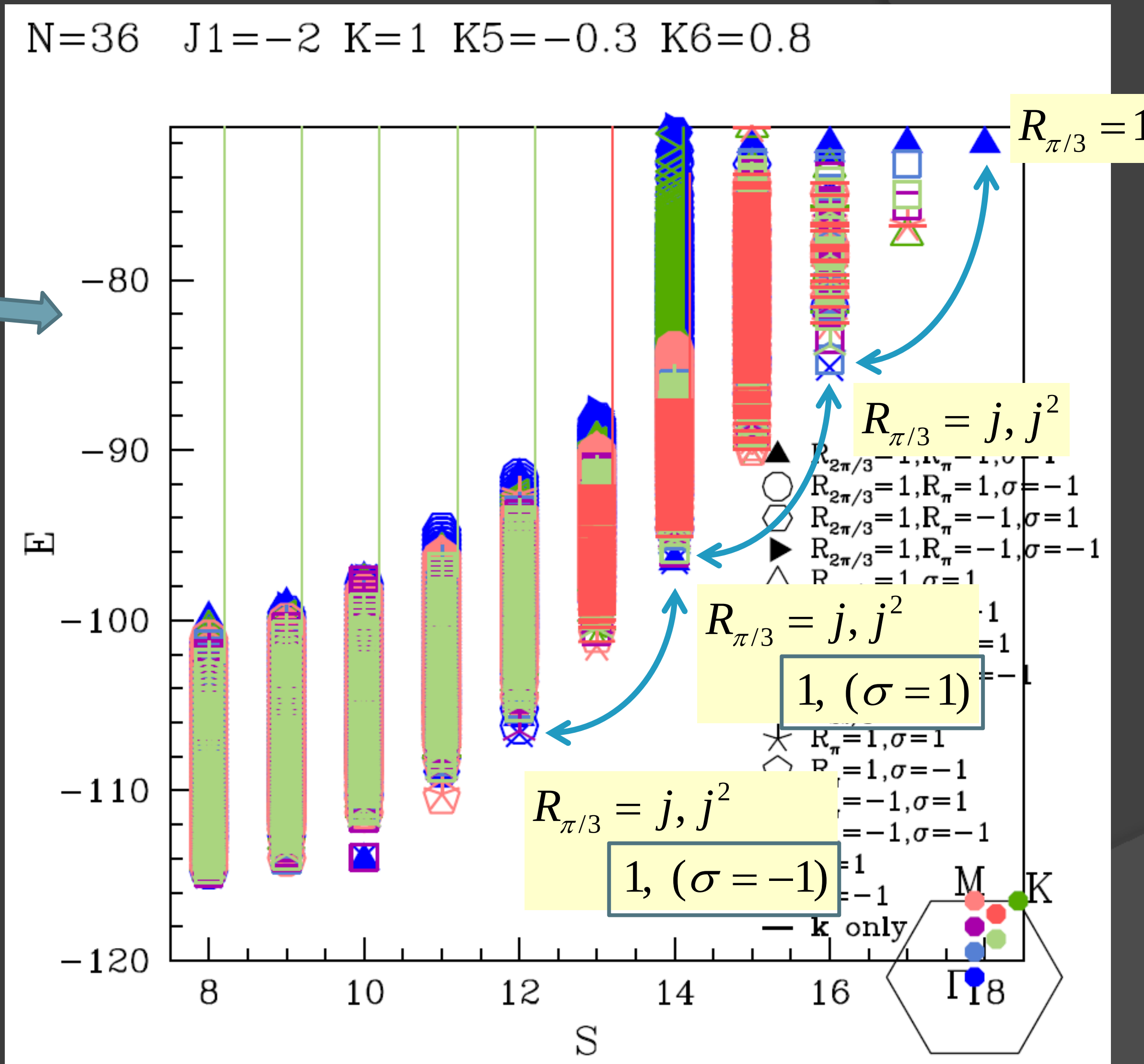
non-chiral nematic order



$$O_{d+id} - O_{d-id}$$

Chiral symmetry breaking ?





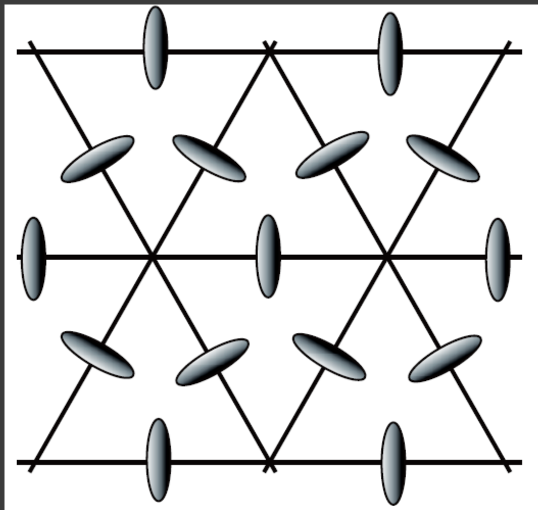
Chiral symmetry breaking acquires double-fold degeneracy in the low-lying states.

<u>Answer: No.</u>

However, some of them are not degenerate
→ no chiral symmetry breaking

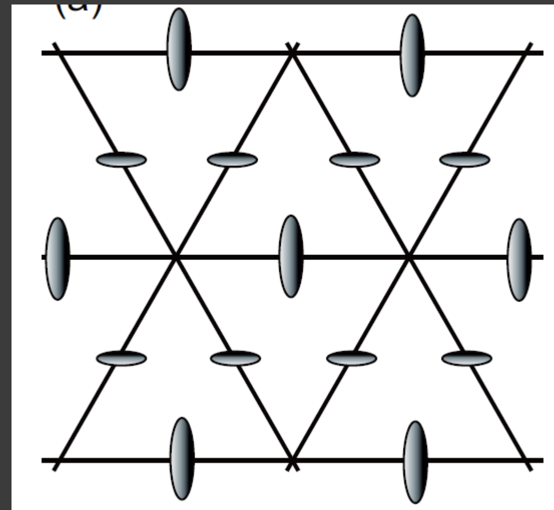
Possible nematic orders induced by d+id-wave magnon pairs

Chiral nematic state



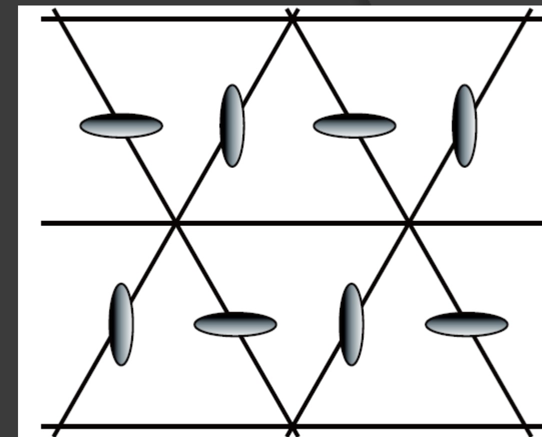
order parameters: Q_+, Q_-

Non-chiral nematic state I



order parameter: $Q_+ + Q_-$

Non-chiral nematic state II



order parameter: $Q_+ - Q_-$

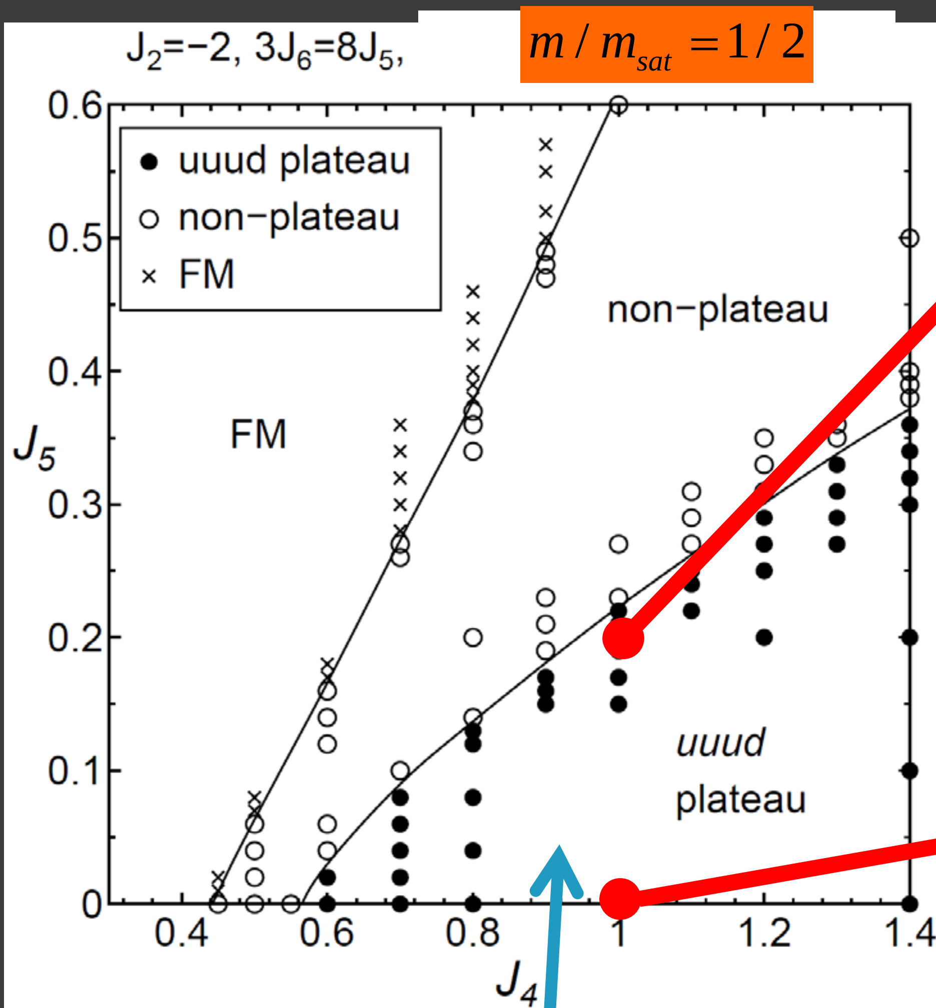
(a) Chiral nematic order

	$S = N/2 - 2(3n-2)$	$S = N/2 - 2(3n-1)$	$S = N/2 - 6n$
Chiral nematic order	$R_{2\pi/3} = j, j^2,\ R_\pi = 1$	$R_{2\pi/3} = j, j^2,\ R_\pi = 1$	$R_{2\pi/3} = 1,\ R_\pi = 1,\ \sigma = \pm 1$

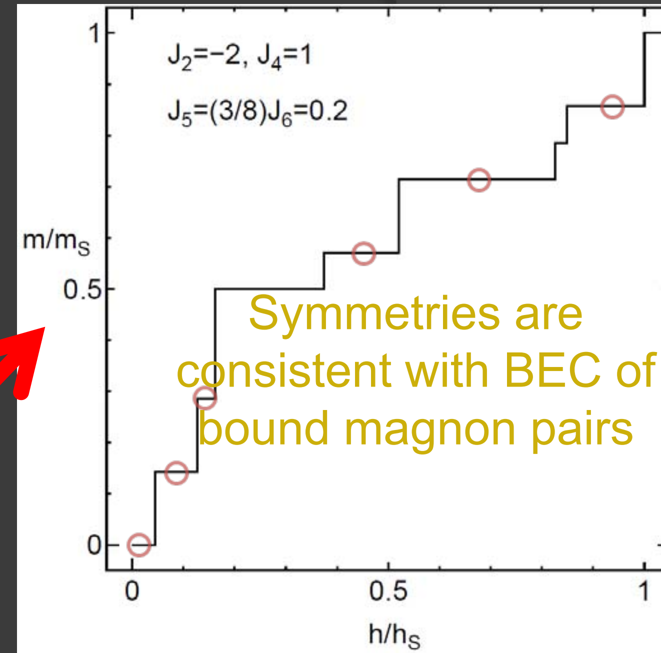
(b) Non-chiral nematic orders

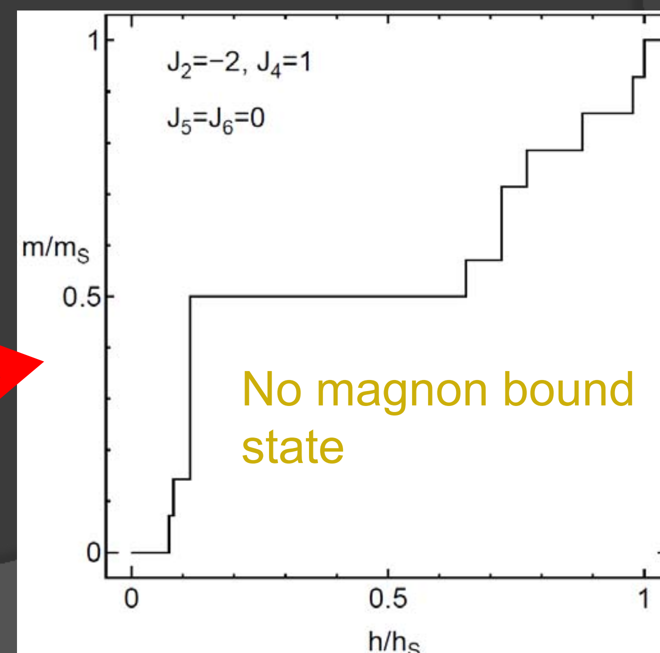
	$S = N/2 - 2$	$S = N/2 - 4n$	$S = N/2 - 2(2n+1)$
Non-chiral nematic order ($Q_+ + Q_-$)	$R_{2\pi/3} = j, j^2,\ R_\pi = 1$	$R_{2\pi/3} = j, j^2,\ R_\pi = 1$ $R_{2\pi/3} = 1,\ R_\pi = 1,\ \sigma = 1$	$R_{2\pi/3} = j, j^2,\ R_\pi = 1$ $R_{2\pi/3} = 1,\ R_\pi = 1,\ \sigma = 1$
Non-chiral nematic order ($Q_+ - Q_-$)	$R_{2\pi/3} = j, j^2,\ R_\pi = 1$	$R_{2\pi/3} = j, j^2,\ R_\pi = 1$ $R_{2\pi/3} = 1,\ R_\pi = 1,\ \sigma = 1$	$R_{2\pi/3} = j, j^2,\ R_\pi = 1$ $R_{2\pi/3} = 1,\ R_\pi = 1,\ \sigma = -1$

Magnetization plateau at m/m_{sat}=1/2

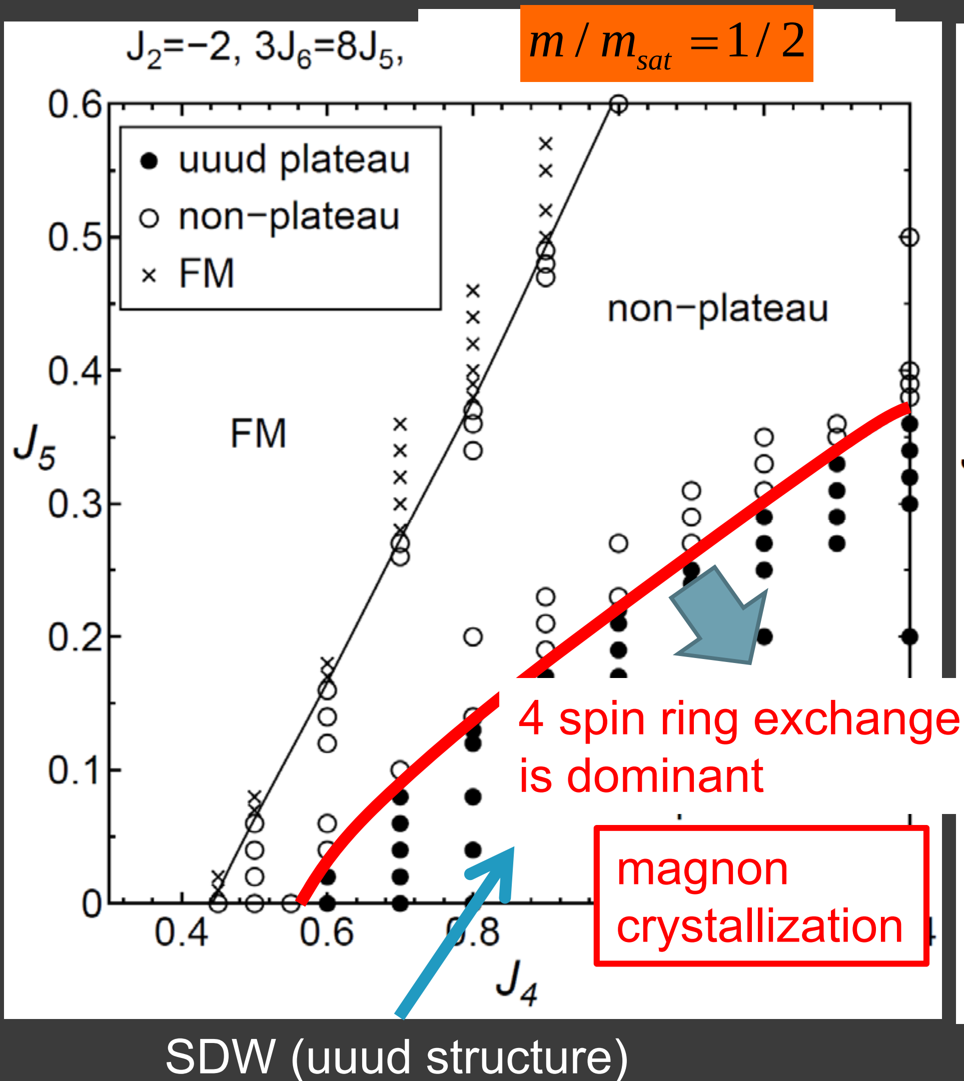


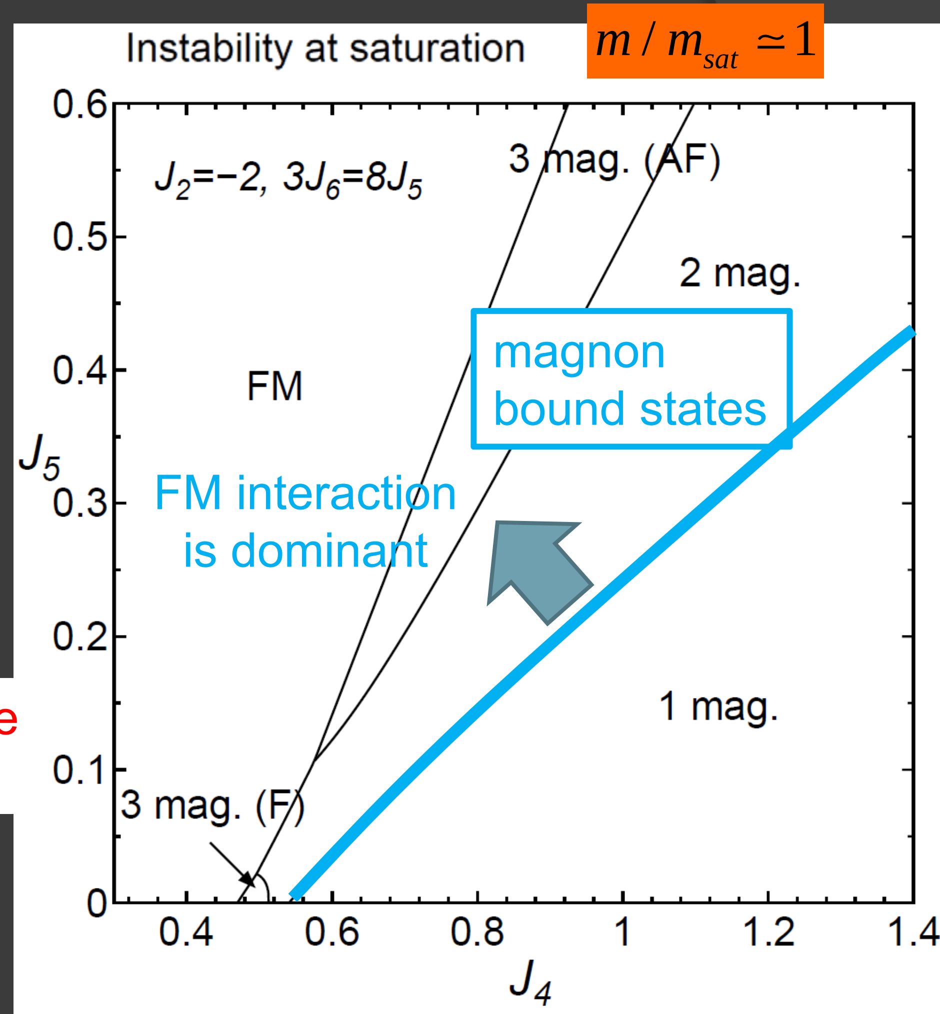
SDW (uuud structure) at m/m_{sat}=1/2





Crossover from FM interaction dominant system to AF ring exchange dominant system

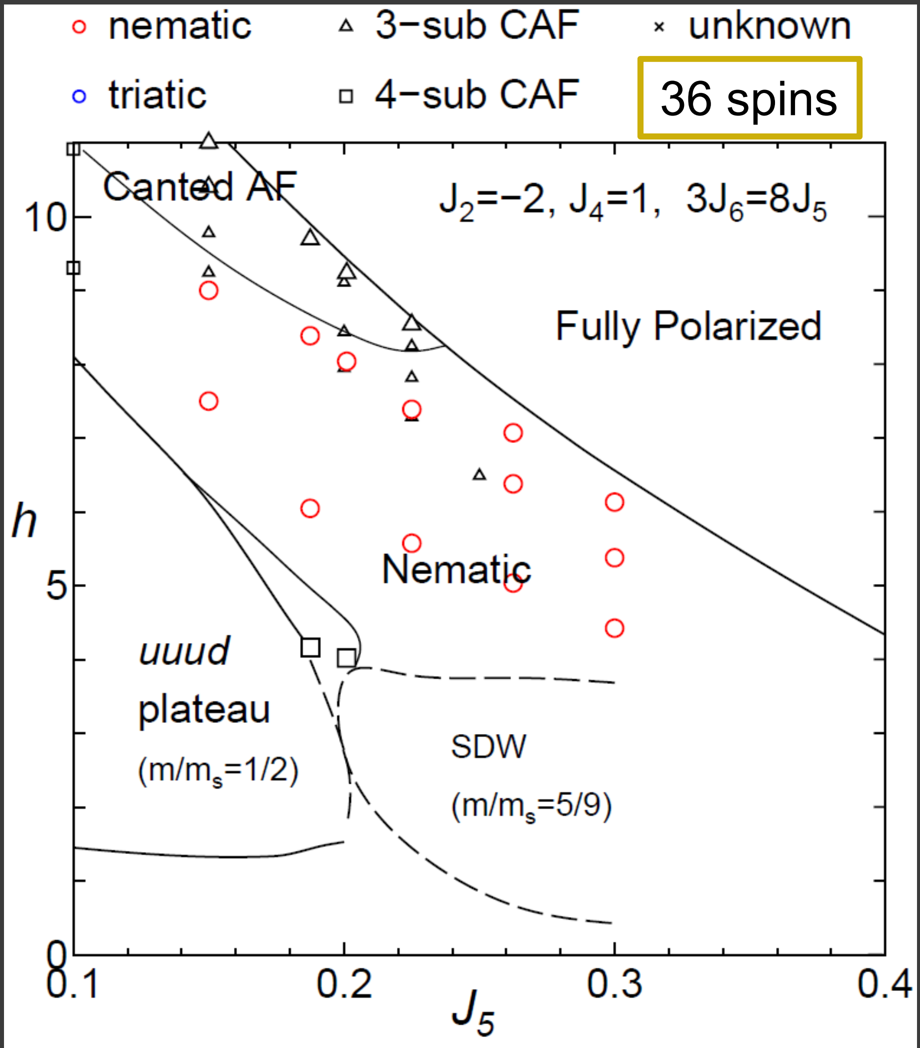


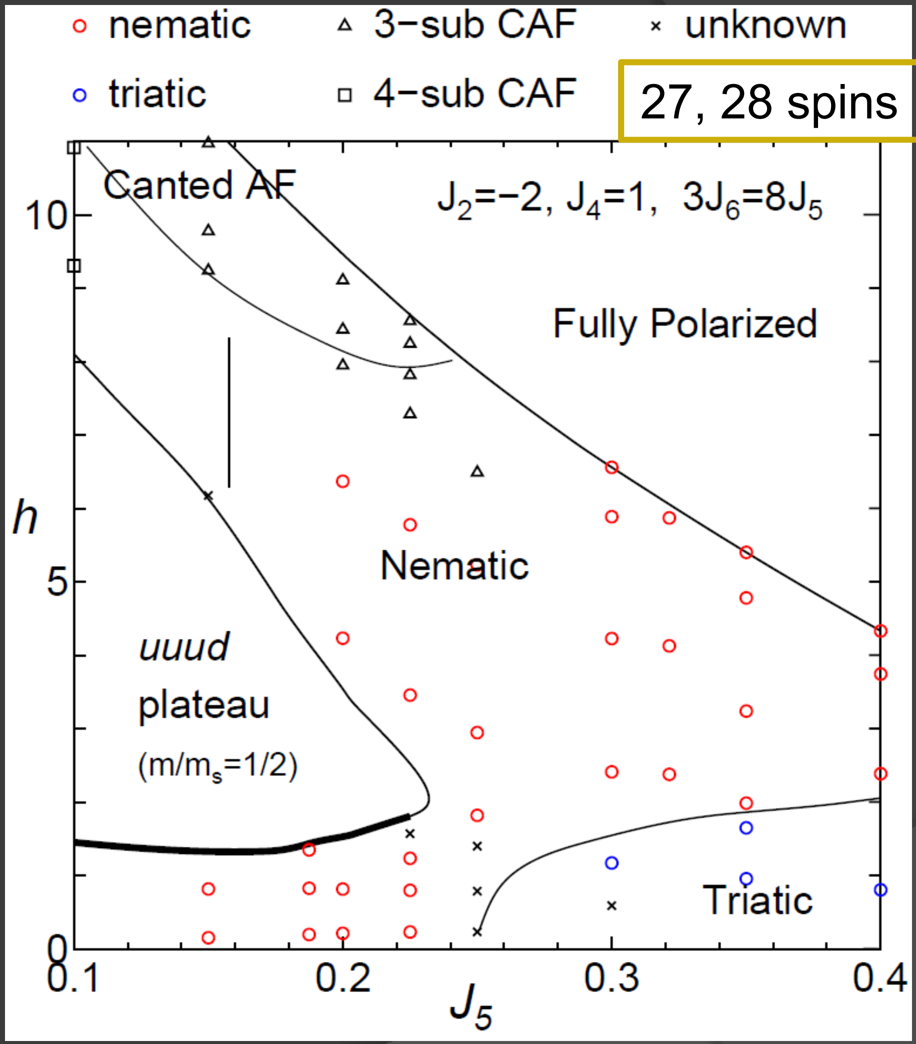


cf. Effective two spin exchange is renormalized by J_5, J_6

$$J_{eff} = J - 10J_5 + 2J_6$$

Phase diagram

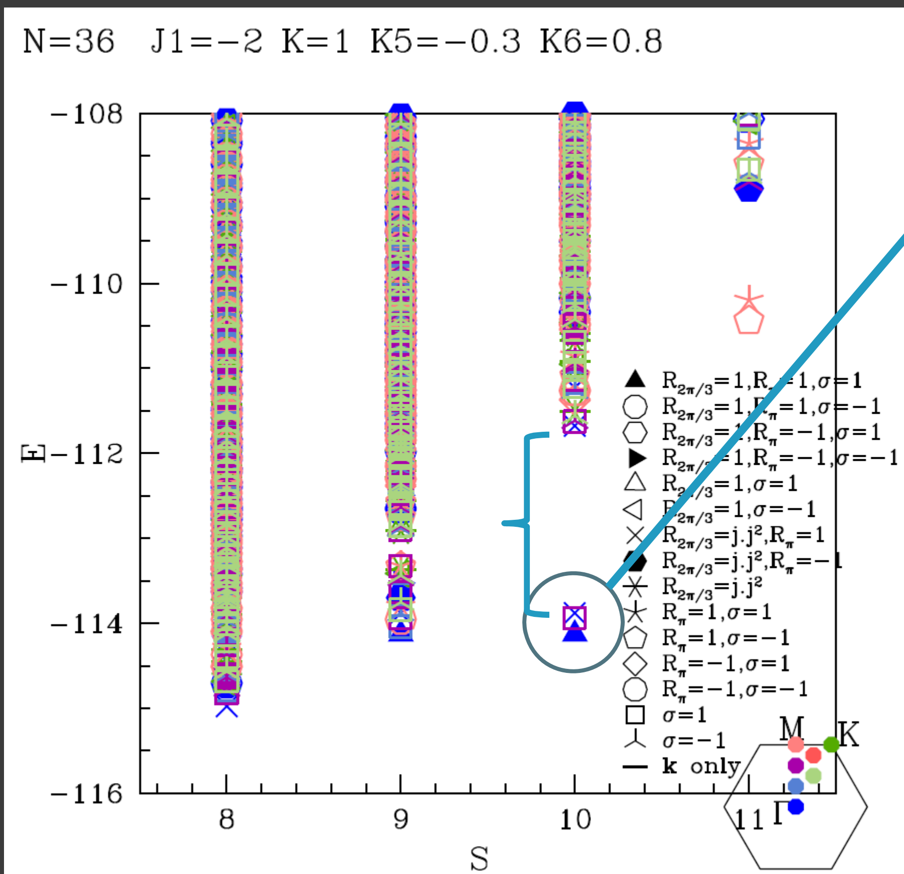




✓ still large size dependence remains
✓ too large J_6 ?

Another magnetization plateau ?

SDW at m/m_{sat}=5/9

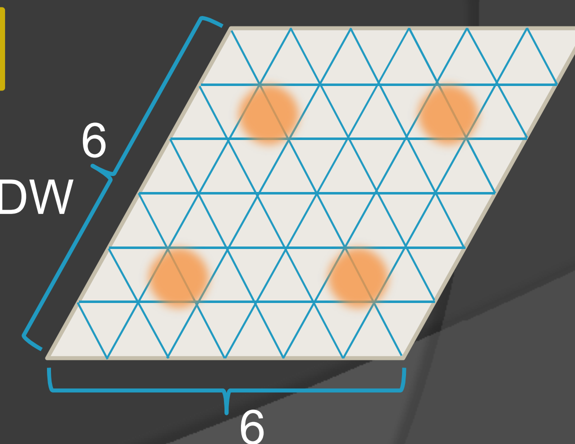


9-fold degeneracy

- Unit vectors
  (3, 0), (3/2, 3*sqrt{3}/2)
- Reciprocal vectors
  (2π/3, 2π/3*sqrt{3}), (0,4π/3*sqrt{3})

36 spins

9 sublattice SDW



“particle” = two magnon bound state

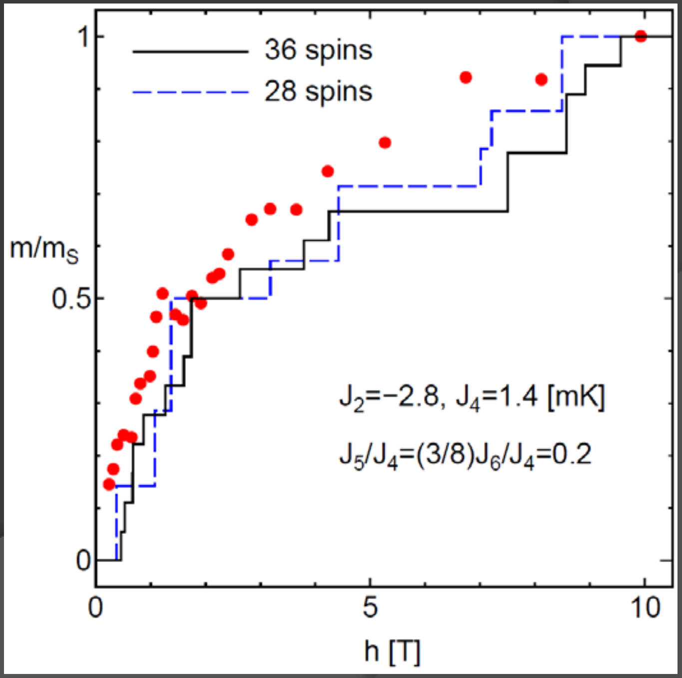
Conclusions

Spin nematic phase appears in spin-1/2 frustrated ferromagnets

- BEC of bound magnon pairs
- spin-triplet RVB state

Multiple-spin exchange model on the triangular lattice

- The 4/7 phase of solid ^{3}He film is in the proximity to the edge of 1/2-plateau.
- Non-plateau states show condensation of *d*+*id* wave magnon pairs, which leads to a non-chiral nematic phase
- Low magnetization region seems to support magnon pairing, but there are still large finite-size effects…

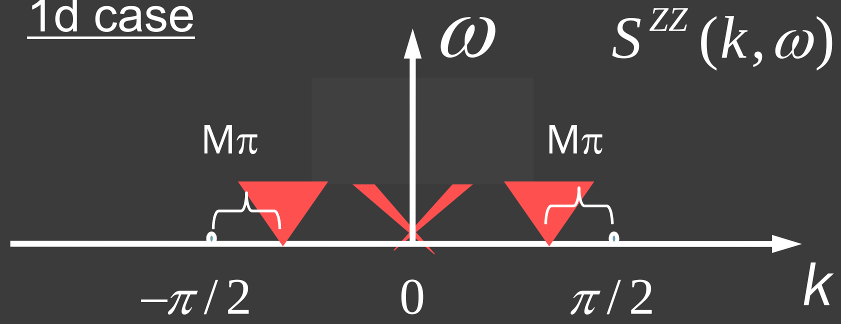


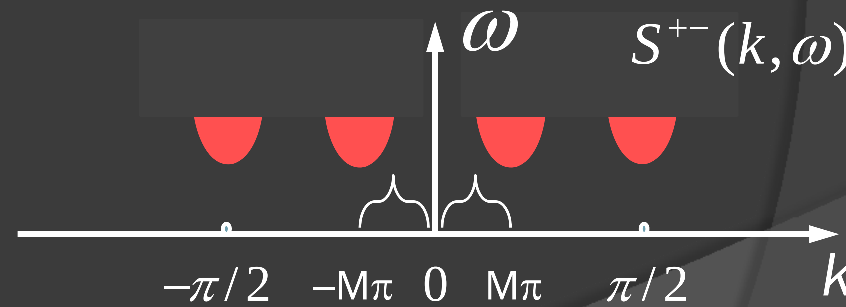
How it looks in experiments.

- uniform ↔ no lattice distortion
- no spin order ↔ no Bragg peak in Neutron scattering
- gapless excitations ↔ specific heat
  -- possibly double peak structure --
- magnon pairing (spin-triplet pairing)
- finite susceptibility

Unusual magnon excitations in $S(k,\omega)$ $\quad$ *h // z*

1d case





→ rapid decay of NMR relaxation rate $1/T_1$

P. 40 M. Sato, TM, and A. Furusaki, PRB **80**, 064410 (2009)