研究課題名 ：

リング交換が生む新奇な磁性状態の解明

Official members:

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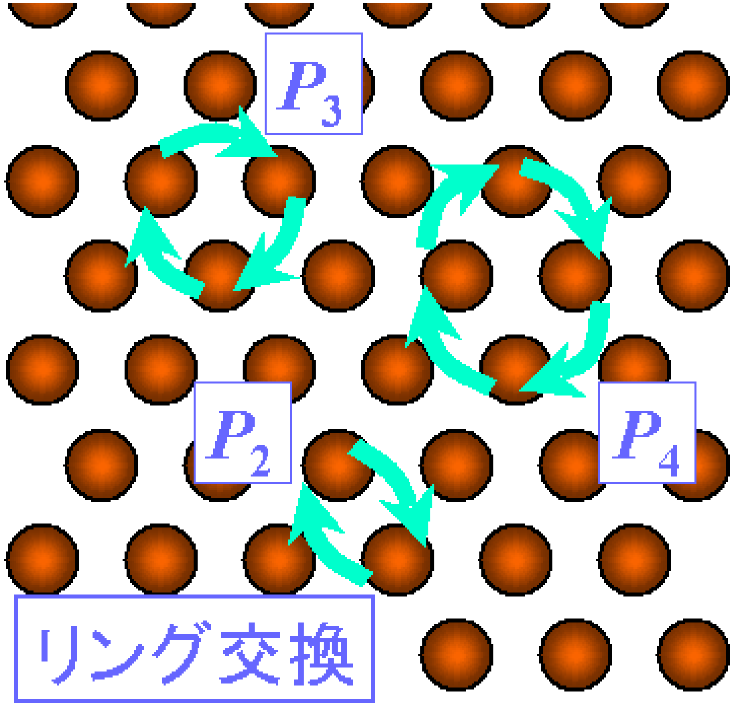
Ring Exchange Interaction

2D Quantum Solids [Solid ^{3}He films, Wigner crystals]



<u>Quantum tunneling process</u>

Cyclic permutation
of *n* particles

↓

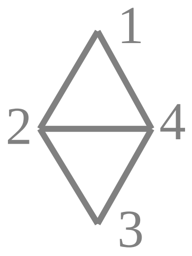
Cyclic permutation
of *n* spins ($\boldsymbol{P_n}$)

n-spin cyclic permutation operators:

$$2P_2 = \vec{\sigma}_i \cdot \vec{\sigma}_j + 1$$

$$P_3 + P_3^{-1} = \vec{\sigma}_i \cdot \vec{\sigma}_j + \vec{\sigma}_j \cdot \vec{\sigma}_k + \vec{\sigma}_k \cdot \vec{\sigma}_i$$

$$4\left(P_4 + P_4^{-1}\right) = \sum_{1 \le i < j \le 4} \vec{\sigma}_i \cdot \vec{\sigma}_j + \left(\vec{\sigma}_1 \cdot \vec{\sigma}_2\right)\left(\vec{\sigma}_3 \cdot \vec{\sigma}_4\right) + \left(\vec{\sigma}_1 \cdot \vec{\sigma}_4\right)\left(\vec{\sigma}_2 \cdot \vec{\sigma}_3\right) - \left(\vec{\sigma}_1 \cdot \vec{\sigma}_3\right)\left(\vec{\sigma}_2 \cdot \vec{\sigma}_4\right)$$



4 spin ring exchange itself has strong spin frustration

Multiple Spin Exchange Model

$$H = -\sum_n (-1)^n J_n \sum P_n, \qquad \left(J_n < 0\right)$$

$$= J \sum_{\text{n.n.}} P_2 + K \sum_{\square} \left(P_4 + P_4^{-1}\right) + \ldots$$

upto $n \le 4$

$$J = 2J_3 - J_2 \ (<0)$$

$$K = -J_4 \ (>0)$$

Competition between FM *J* and AF *K* interactions.

solid ^{3}He films:

	Low density	High density
Estimates of $-K/J$ (> 0)	0.2	0.1
Weiss temp. J_χ	AF	FM

↓

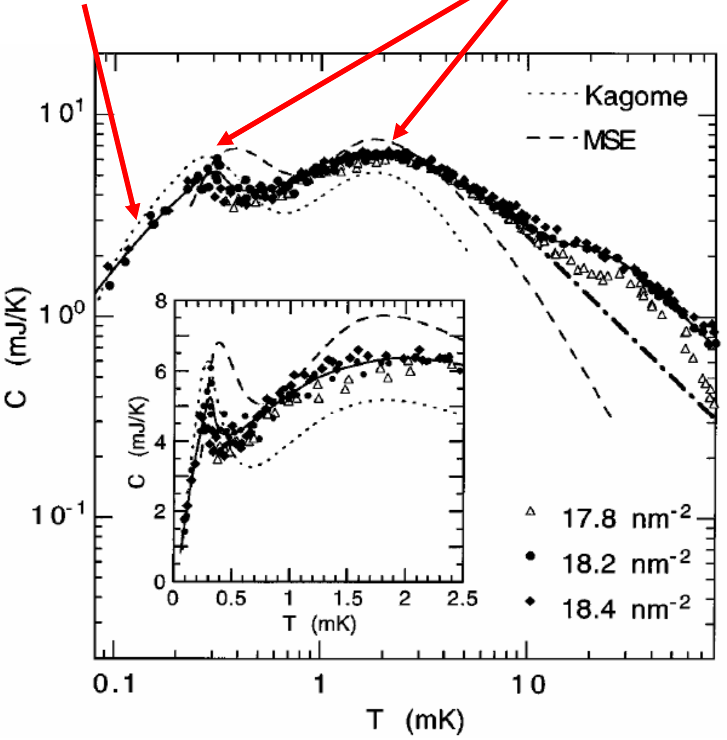
Strong frustration (competition)

Low-density regime in 2nd layer

K. Ishida *et al.*, PRL **79**, 3451 (1997)

linear specific heat
(c.f. 2D FM)

double peak structure



gapless spin liquid

Electron systems

✓ **Copper Oxides** [$La_xCa_{14-x}Cu_{24}O_{41}$, La_2CuO_4]

$$H = J\sum_{\langle i,j\rangle}\vec{S}_i\cdot\vec{S}_j + J_4\sum_{\square}\left(P_4+P_4^{-1}\right)$$

$J_4 / J = 0.05 \sim 0.1$

- **Hubbard model**

$$H = -t\sum_{\langle i,j\rangle}\left(c_i^{\dagger}c_j + h.c.\right) + U\sum_{i} n_{i\uparrow}n_{i\downarrow}$$

M. Takahashi,
J. Phys. C **10, 1289 ('77)**

High-order term derived from the strong coupling expansion

$$H = \frac{t^2}{2U}\sum_{\langle i,j\rangle}\vec{\sigma}_i\cdot\vec{\sigma}_j$$

$$+\frac{5t^4}{2U^3}\sum_{\square:(i,j,k,l)}\underline{\left[\left(\vec{\sigma}_i\cdot\vec{\sigma}_j\right)\left(\vec{\sigma}_k\cdot\vec{\sigma}_l\right)+\left(\vec{\sigma}_i\cdot\vec{\sigma}_l\right)\left(\vec{\sigma}_k\cdot\vec{\sigma}_j\right)-\left(\vec{\sigma}_i\cdot\vec{\sigma}_k\right)\left(\vec{\sigma}_j\cdot\vec{\sigma}_l\right)\right]}$$

$$\boxed{4\left(P_{1234}+P_{1234}{}^{-1}\right)-\sum_{1\le i<j\le 4}\vec{\sigma}_i\cdot\vec{\sigma}_j}$$

- **d-p model**

H.J.Schmidt and Y.Kuramoto

Present understanding

- Magnetism induced by ring exchange is not well understood.
  - ➢ Mean field approximation is not good,

    ---- because of strong quantum fluctuations.
- Ring exchange causes cooperative dynamics of many magnons.

One of our goals:

Novel quantum magnetic phase

- Novel order ?
- Novel excitation ?
- Novel dynamics ?

Exotic Magnetism induced by Ring Exchanges

Tsutomu Momoi (RIKEN)

Contents:

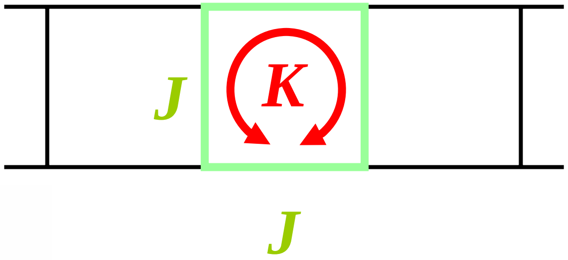
- Ladder Lattice -- chiral order --
- Square Lattice -- nematic order –
- Future prospects

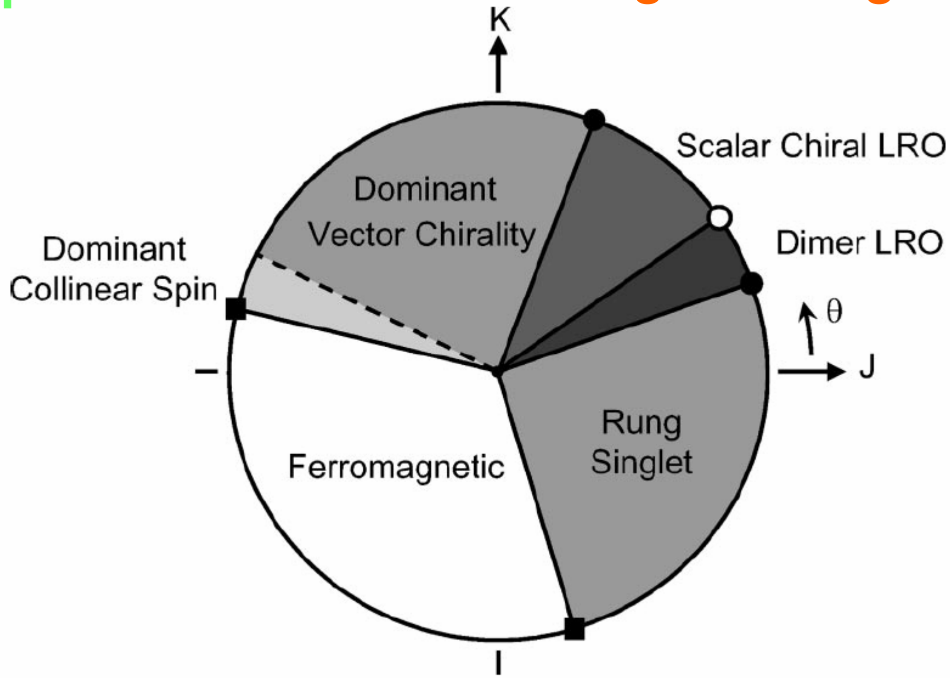
Two-Leg Ladder — Spin chirality and Duality —

Collaborators: T. Hikihara (Hokkaido Univ.), M. Nakamura (Tokyo Sci. Univ.)

Hamiltonian:

$$H = J\underbrace{\sum_l \vec{s}_{1,l}\cdot\vec{s}_{2,l} + J\sum_l\sum_{\mu=1,2}\vec{s}_{\mu,l}\cdot\vec{s}_{\mu,l+1}}_{\text{2-leg spin ladder}} \quad + \quad K\sum_{\square}\underbrace{\left(P_4+P_4^{-1}\right)}_{\text{ring exchange}}$$





A. Läuchli *et al.*, PRB **67**, 100409(R) (2003).

T. Hikihara, TM, and X. Hu, PRL **90**, 087204 (2003).

T. Hikihara, TM, X.Hu, PRL **90** (2003) 087204.
TM, H.Hikihara, M.Nakamura, X,Hu, PRB **67** (2003) 174410.

➢ Duality relation in the two-leg ladder

- Duality transformation:

$$\vec{S}_l \equiv \frac{1}{2}(\vec{s}_{1,l}+\vec{s}_{2,l})+\vec{s}_{1,l}\times\vec{s}_{2,l},$$
$$\vec{T}_l \equiv \frac{1}{2}(\vec{s}_{1,l}+\vec{s}_{2,l})-\vec{s}_{1,l}\times\vec{s}_{2,l}.$$

- Dual operators
  - ✧ Neel-spin and vector chirality.
  - ✧ staggered dimer order and scalar chiral order

- Duality relation in the parameter space

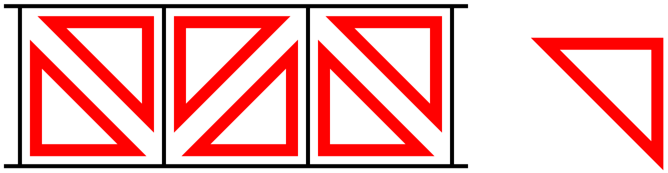
➢ Matrix-product solvable models on the ladder

Matrix-product ansatz + Duality transformation

- **Staggered scalar-chiral state**

$$\left|\Psi_0(u)\right\rangle = \mathrm{Tr}\left\{\tilde{g}_1(u)\tilde{g}_2(-u)\cdots\tilde{g}_{L-1}(u)\tilde{g}_L(-u)\right\},$$

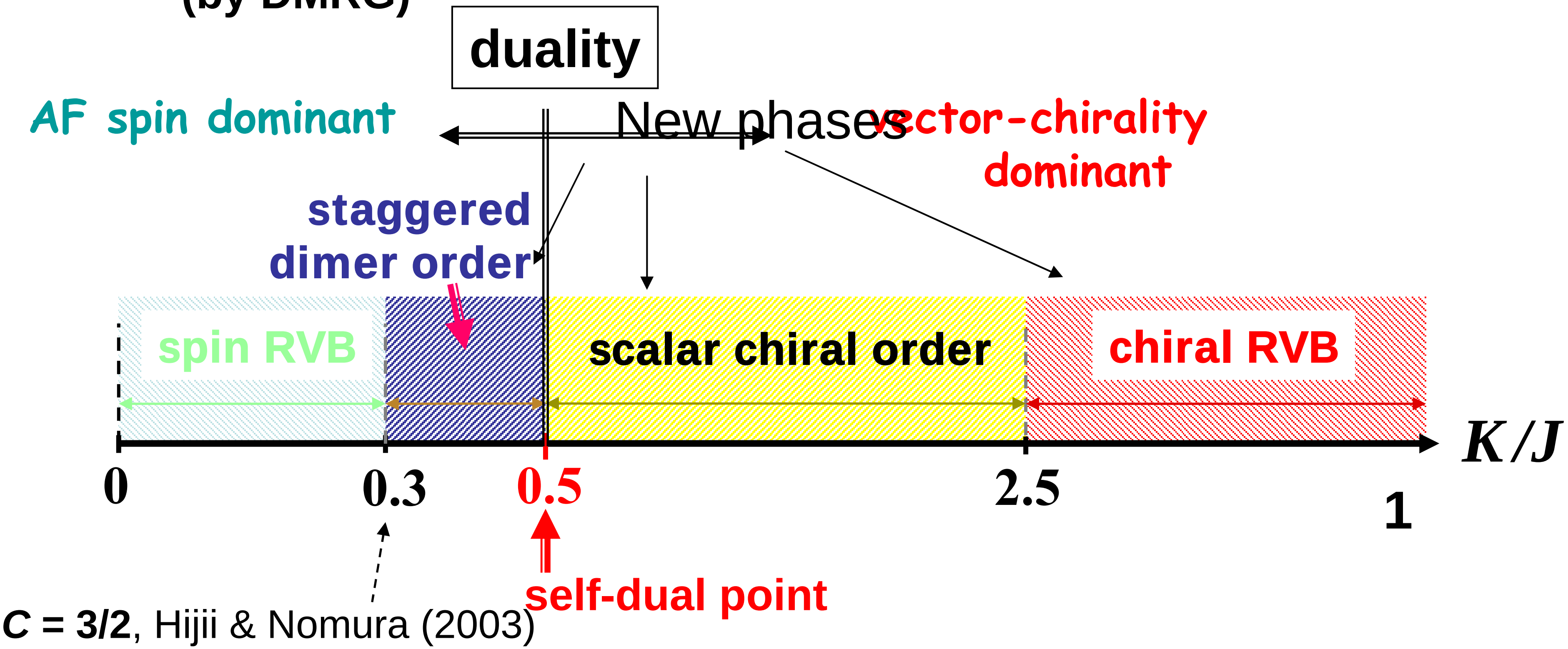
where $\tilde{g}_l(u)=\frac{1}{2}\left\{ iu\mathbf{1}\left|s\right\rangle_l - \sqrt{2}\boldsymbol{\sigma}^+\left|t^{+1}\right\rangle_l + \sqrt{2}\boldsymbol{\sigma}^-\left|t^{-1}\right\rangle_l + \boldsymbol{\sigma}^z\left|t^0\right\rangle_l \right\}.$



$$= \quad \left\langle \vec{s}_{1,l}\cdot\left(\vec{s}_{1,l+1}\times\vec{s}_{2,l+1}\right)\right\rangle > 0$$

Phase diagram:
(by DMRG)

T. Hikihara, TM, X. Hu (2003).
A. Laeuchli, G. Schmid, M. Troyer (2003).



duality

AF spin dominant

New phases

vector-chirality dominant

staggered dimer order

spin RVB

scalar chiral order

chiral RVB

self-dual point

C = 3/2, Hijii & Nomura (2003)

Scalar chiral phase

(1) ground states are doubly degenerate,
(2) there is a finite energy gap,
(3) scalar chiral LRO and short-range spin correlation.

Square lattice

—— Nematic order in frustrated ferromagnets ——

Collaborators:

Nic Shannon (MPI, Germany & Bristol Univ., UK)
Philippe Sindzingre (M. et P. Curie Univ., France)

N. Shannon, TM, and P. Sindzingre, to appear in *Phys. Rev. Lett.*

Frustrated antiferromagnets…… well studied.

AF interaction prefers formation of dimer singlets.

Resonating valence bonds *P. W. Anderson*

- Triangular lattice AF model
- Square lattice AF J1-J2 model
- Kagome lattice

……

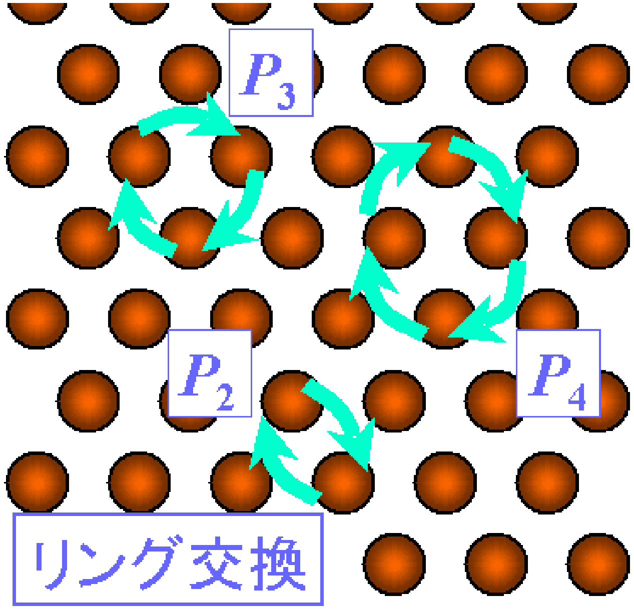
Some of them show spin LRO and some of them show spin gaps…

Frustrated ferromagnets…… not well studied.

Competition between FM and AF interactions.

Frustrated ferromagnets

☐ Solid ^{3}He films on graphite



Competition between

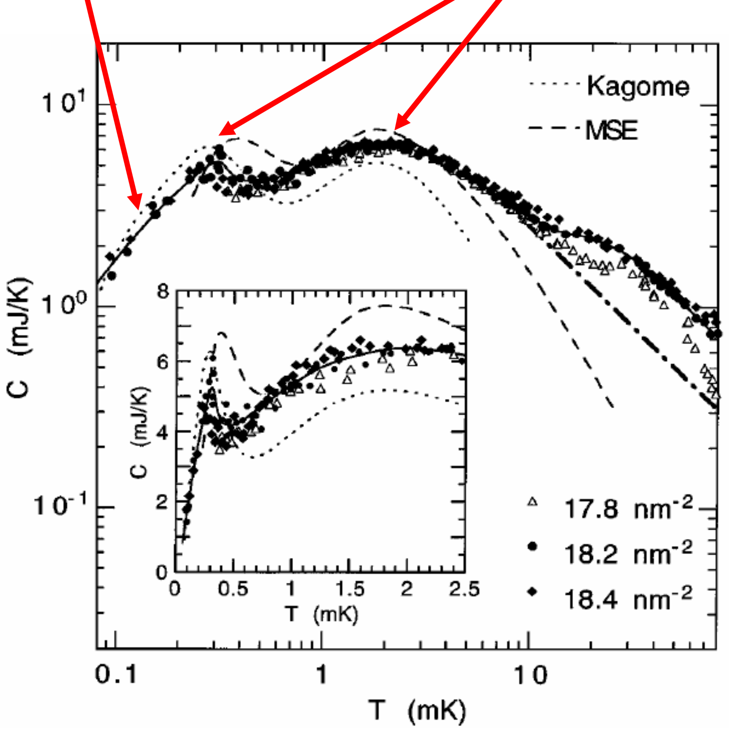
FM 3-spin exchange

and AF 4-spin exchange

$$H = J\sum_{\text{n.n.}} P_2 + K\sum_{\square}\left(P_4 + P_4^{-1}\right),$$

Low-density regime in 2nd layer

K. Ishida *et al.*, PRL **79**, 3451 (1997)



linear specific heat
(c.f. 2D FM)

double peak structure

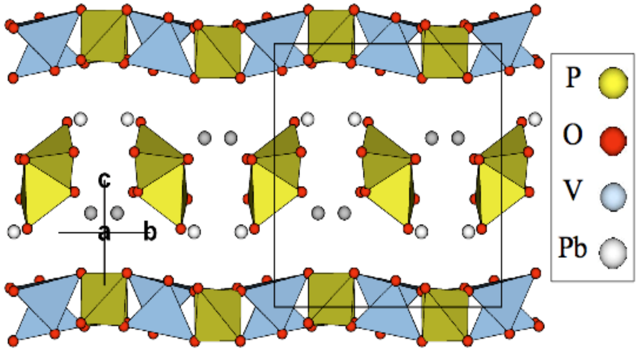
gapless spin liquid

Frustrated ferromagnets   square lattice

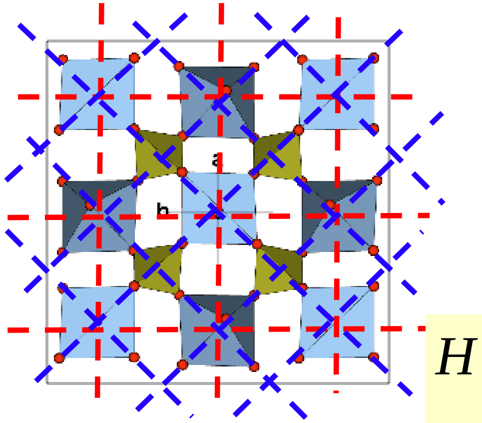
☐ $Pb_2VO(PO_4)_2$ S=1/2 square lattice system

E. Kaul *et al.*, JMMM **272-276 (II)**, 922 (2004)

$Pb_2VO(PO_4)_2$: Structure



spin-1/2 V^{4+} in layered pyramids



FM J_1 – AF J_2 model

$J_1 < 0$ FM
$J_2 > 0$ AF

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

☐ $(CuCl)LaNb_2O_7$ —— S=1/2 square lattice system, $\Theta_W > 0$

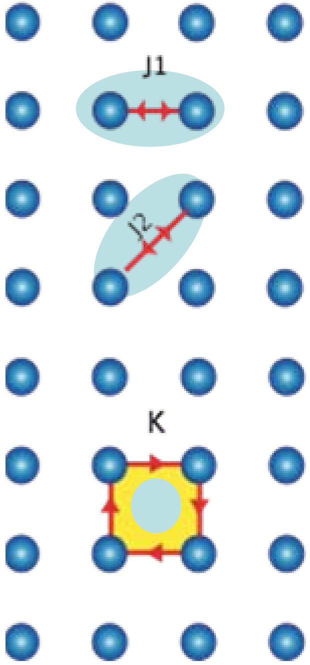
H. Kageyama *et al.*, J. Phys. Soc. Jpn. (2005)

No spin order, spin gap,   S=2 bound states?

the model we consider

extended FM Heisenberg model on square lattice

$$\mathcal{H} = 2J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i \mathbf{S}_j + 2J_2 \sum_{\langle ij \rangle_2} \mathbf{S}_i \mathbf{S}_j + K \sum_{\langle 1234 \rangle} P_{1234} + P_{1234}^{-1}$$



FM n.n. interaction

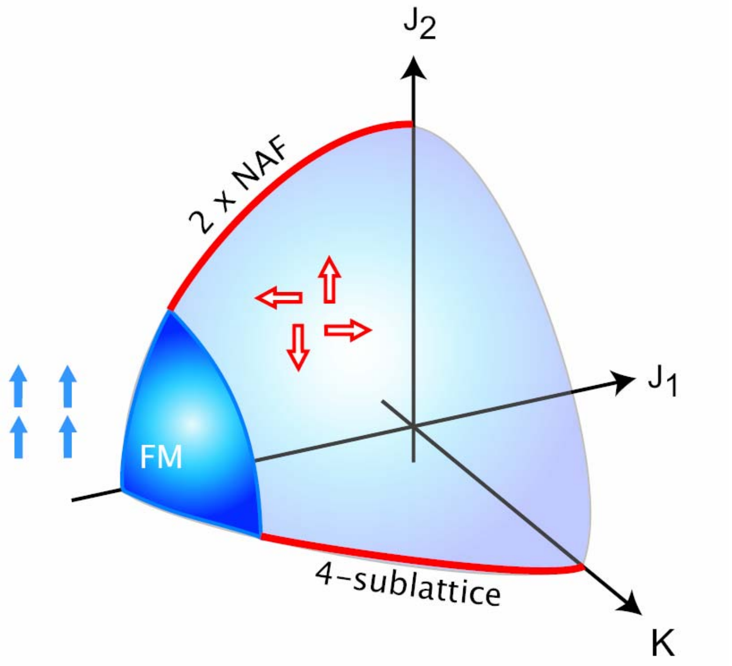
$J_1 < 0$

AF n.n.n. interaction

$J_2 > 0$

AF 4-spin cyclic exchange

$K > 0$



mean field phase diagram

Previous theoretical works

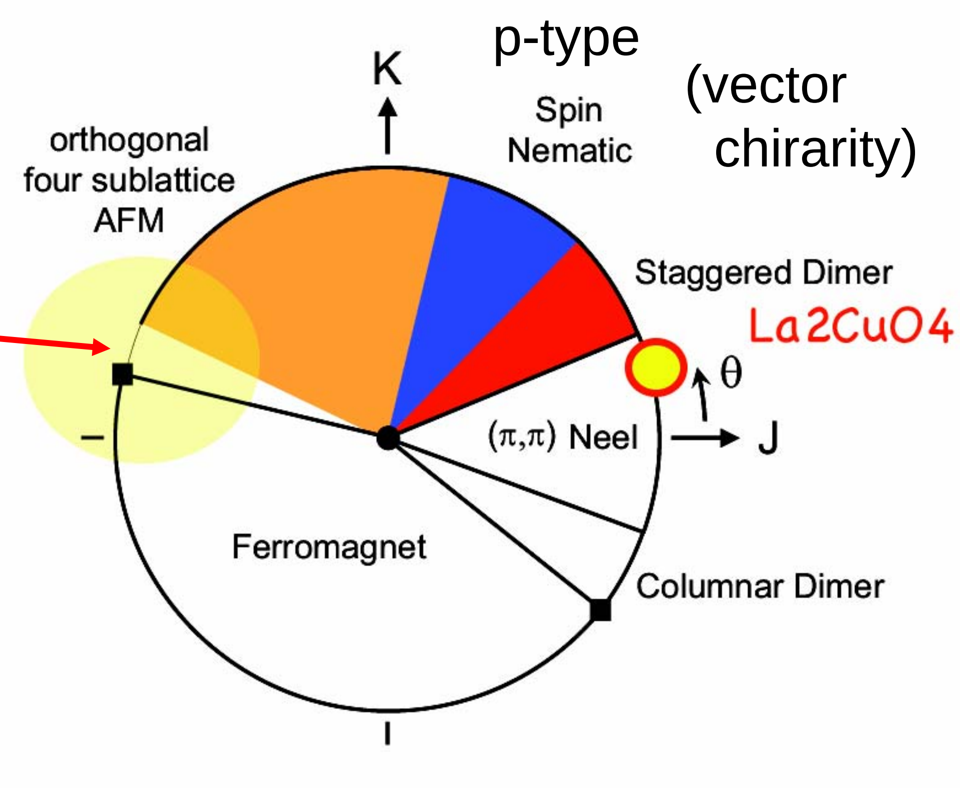
- Multiple-spin exchange model on the square lattice

$$H = J\sum_{\text{n.n.}} P_2 + K\sum_{\square}\left(P_4 + P_4^{-1}\right),$$

$(J_2 = 0)$

A. Laeuchli *et al.*, PRL (2005)

$$J < 0 \ \ (\text{FM}), \qquad K > 0 \ \ (\text{AF})$$



p-type (vector chirarity)

La2CuO4

Unknown phase (spin liquid ?)

Instability in FM state at boundary with AF

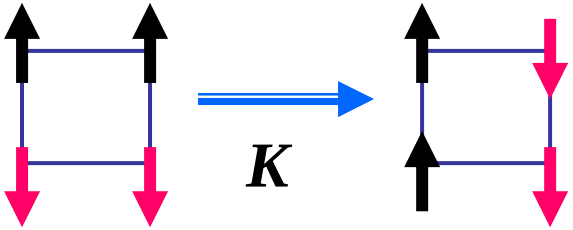
- <u>One-magnon excitation</u>

dispersion: $\varepsilon(\mathbf{k}) = -4(J_1+2K)+2(J_1+2K)\left(\cos k_x + \cos k_y\right)$

At $K/|J_1| = 1/2$, **zero flat mode** (localized magnons!)

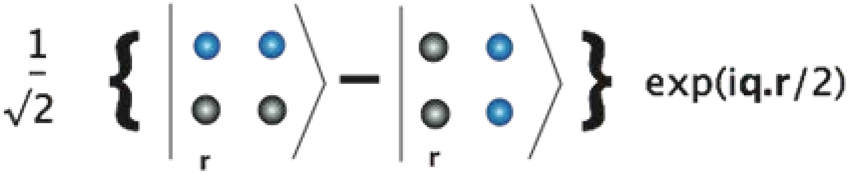
One magnon instability.

- <u>Two-magnon excitation</u> ----exactly solvable----



Cooperative dynamics

d-wave symmetry



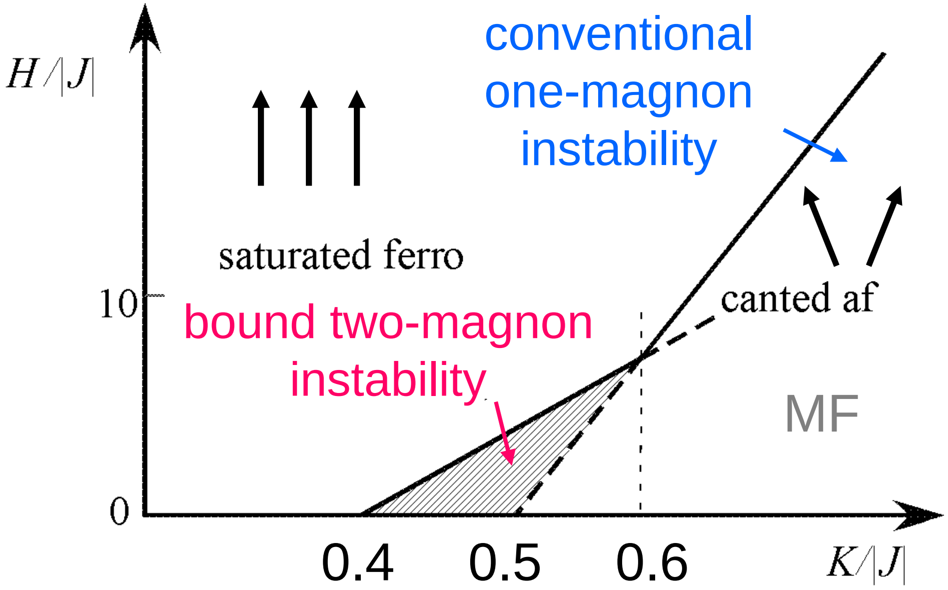
The lowest state has $q = 0$

! Two-magnon bound states have lower energy at one-magnon instability point.

→ New instability

- **In the magnetic field**

Two-magnon instability first appears at H_{c2} before one-magnon instability (H_{c1}).



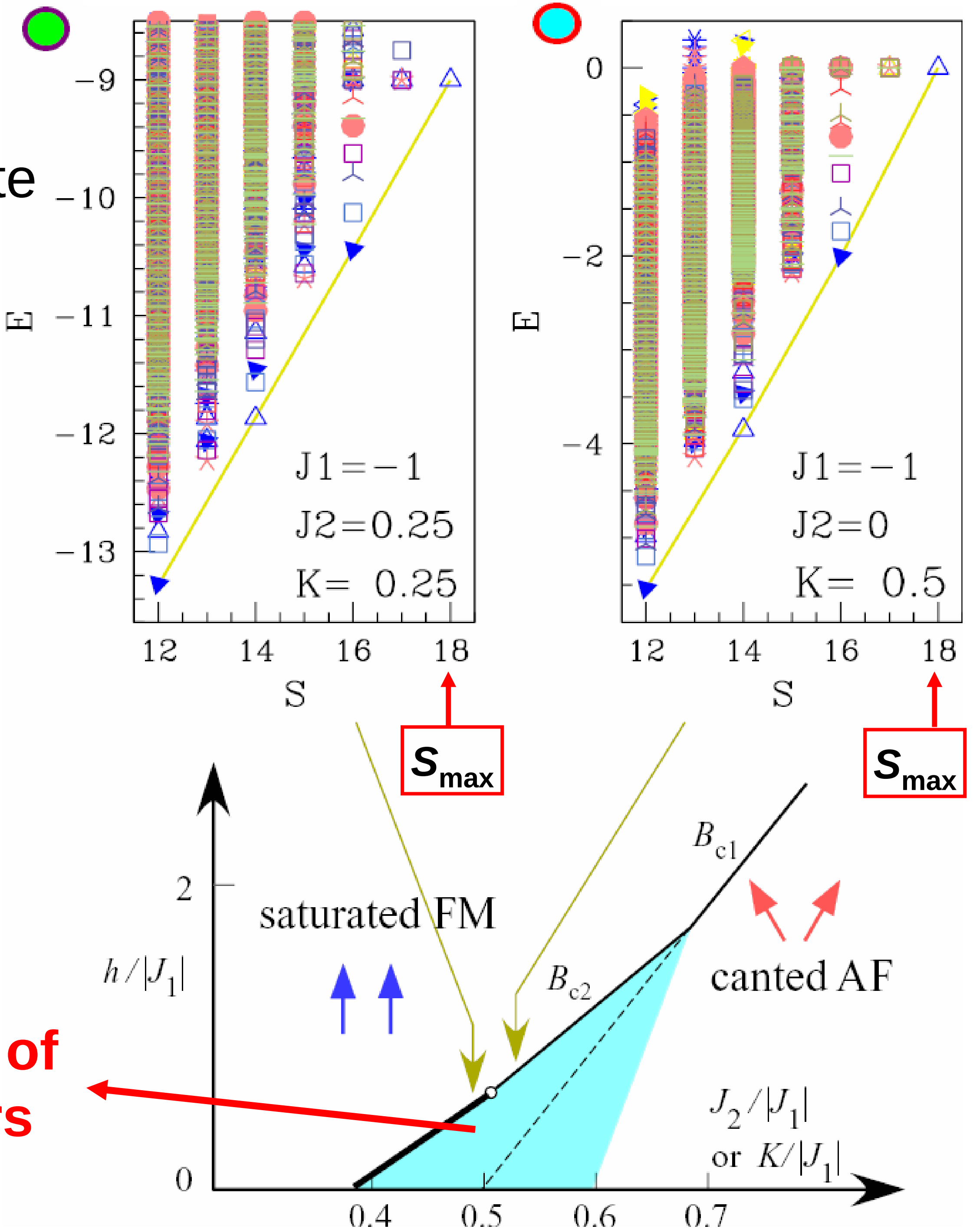
Magnon pairs have

- hopping process.
- repulsive interactions between bound bi-magnons (if $K>0$).

⇓

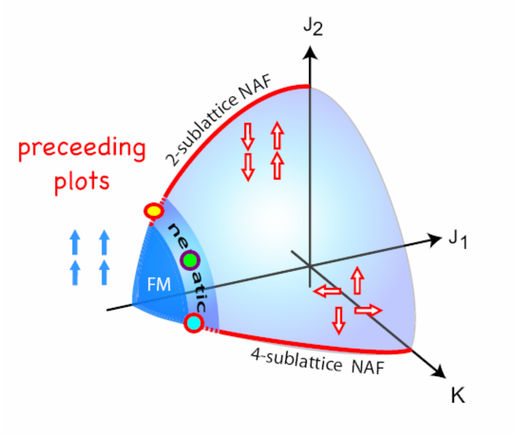
2nd order transition at saturation field

Energy spectrum
near fully porlarized state



S_{max}

S_{max}



Condensation (BEC) of d-wave magnon pairs

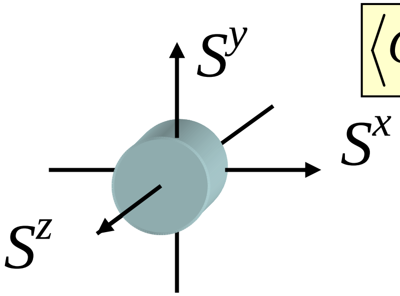
What is the nature of this phase?

- Spin nematic operator (n-type) —— quadrupole order

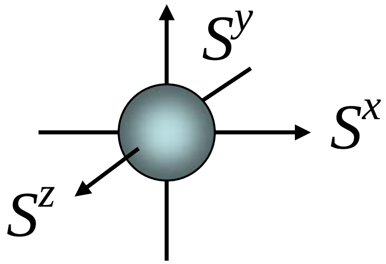
$$Q^{\alpha\beta}\left(r_i,r_j\right)=\frac{1}{2}\left(S_i^{\alpha}S_j^{\beta}+S_i^{\beta}S_j^{\alpha}\right)-\frac{1}{3}\delta_{\alpha\beta}\left\langle\vec{S}_i\cdot\vec{S}_j\right\rangle$$

nematic order

$$\left\langle Q^{xx}\right\rangle=\left\langle Q^{yy}\right\rangle>\left\langle Q^{zz}\right\rangle$$



disorder (isotropic)



- Magnon pairing operator

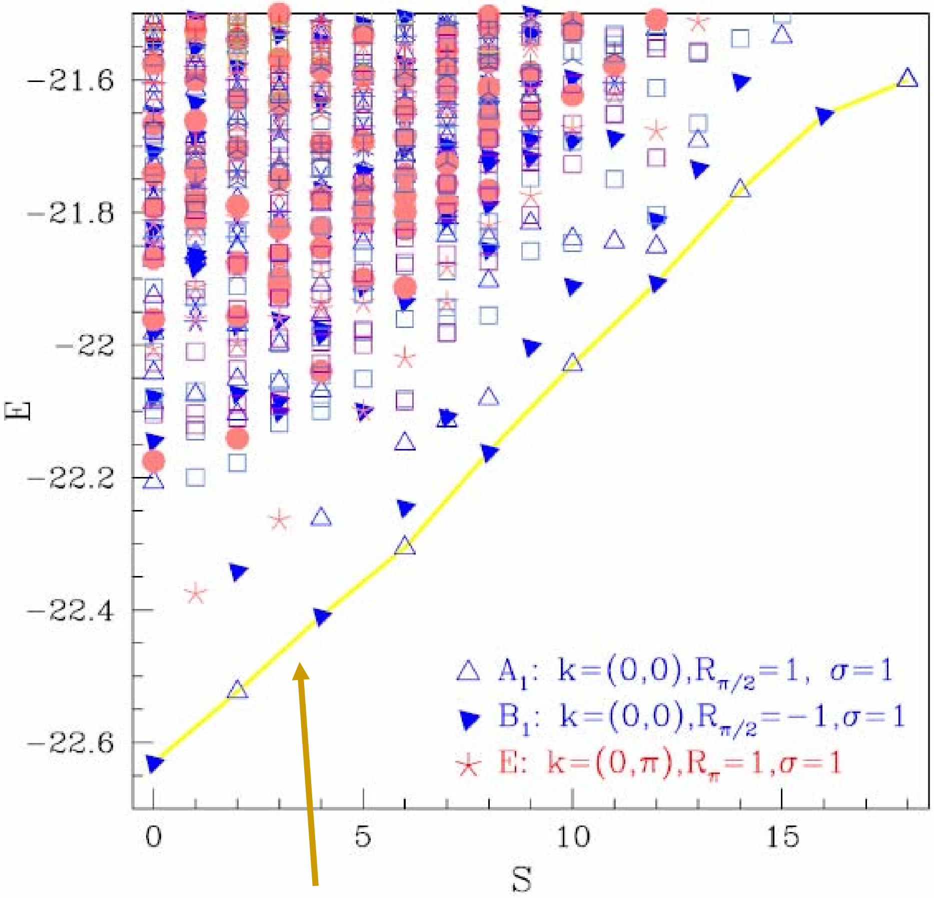
$$S_i^- S_j^-$$

Relation between the magnon pairing and the nematic operators

$$S_i^- S_j^- = Q^{xx} - Q^{yy} - 2iQ^{xy}$$

BEC of bound magnon pairs = Spin nematic LRO

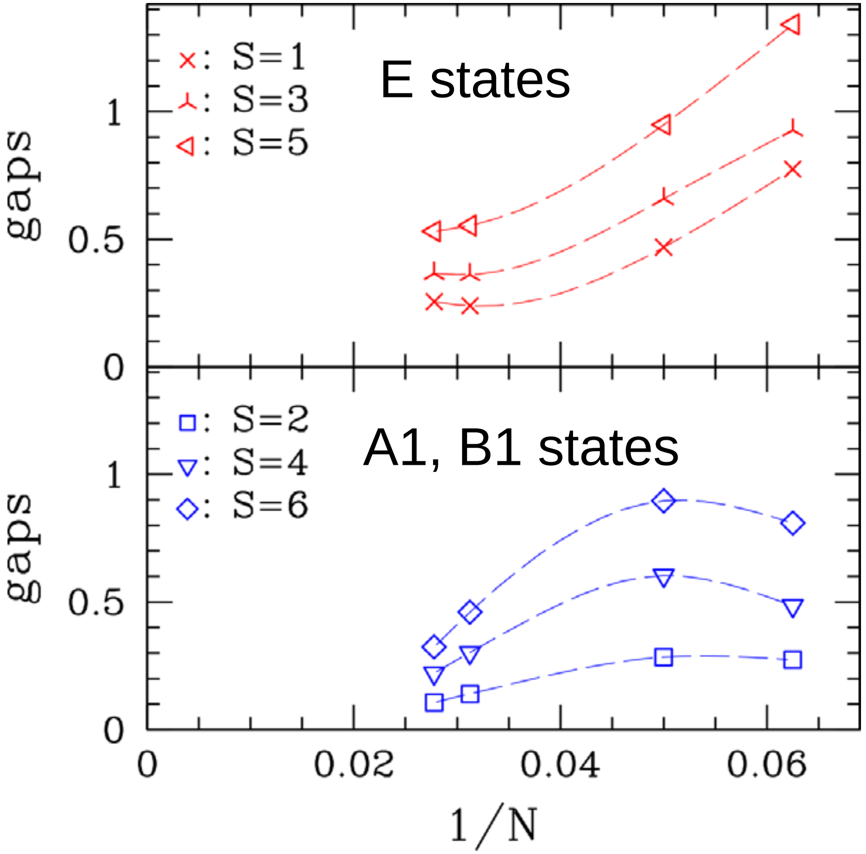
Full spectrum: J_1-J_2 model on square lattice J_1=-1, J_2=0.4



Only even spin sectors contribute to ground state.

In AF ordered phase, A1, B1, and E states must be gapless,

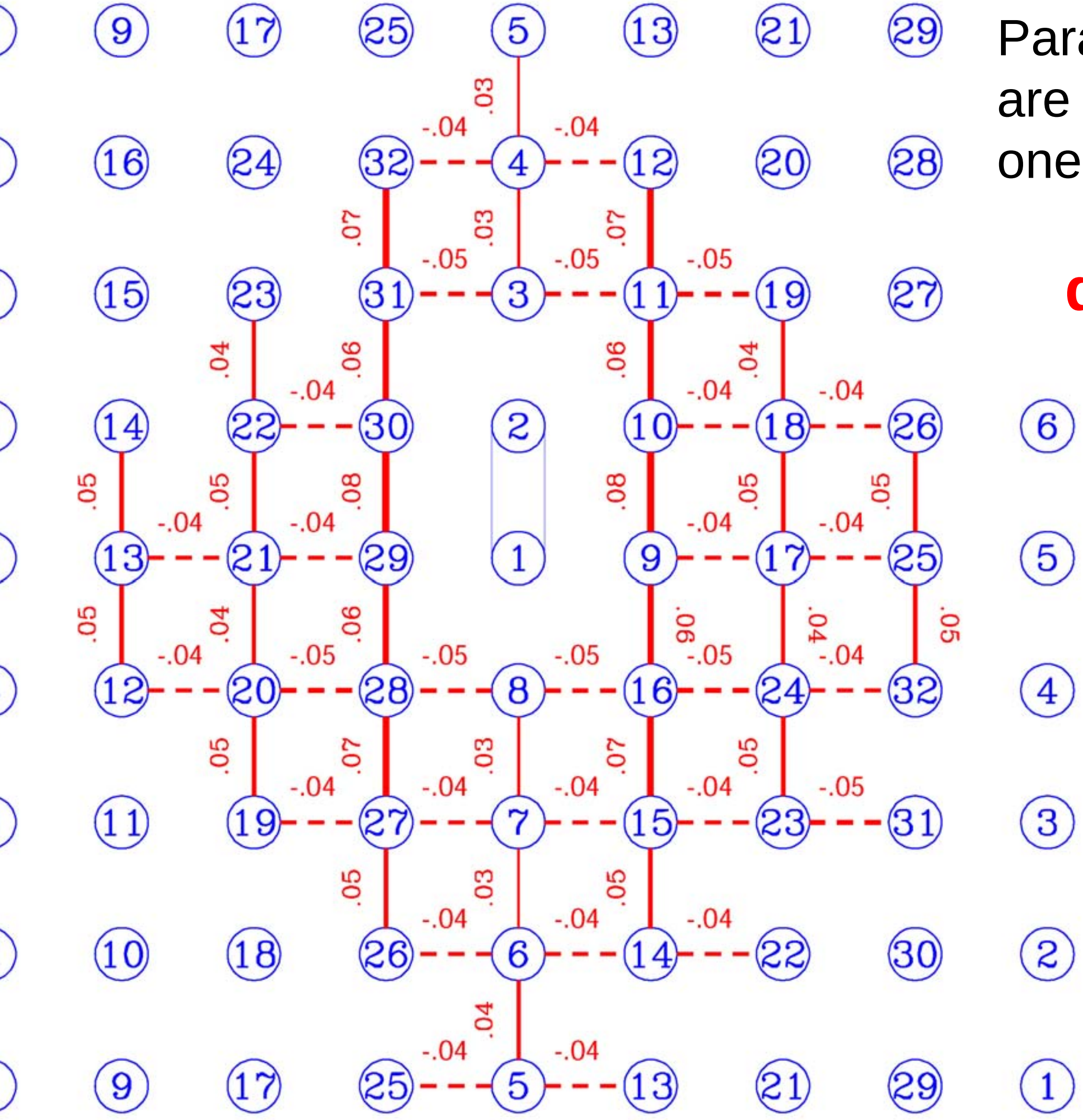
but



No spin order [**Spin liquid**]

Condensation (BEC) of d-wave magnon pairs

- Long-range nematic correlation in S=0 ground state

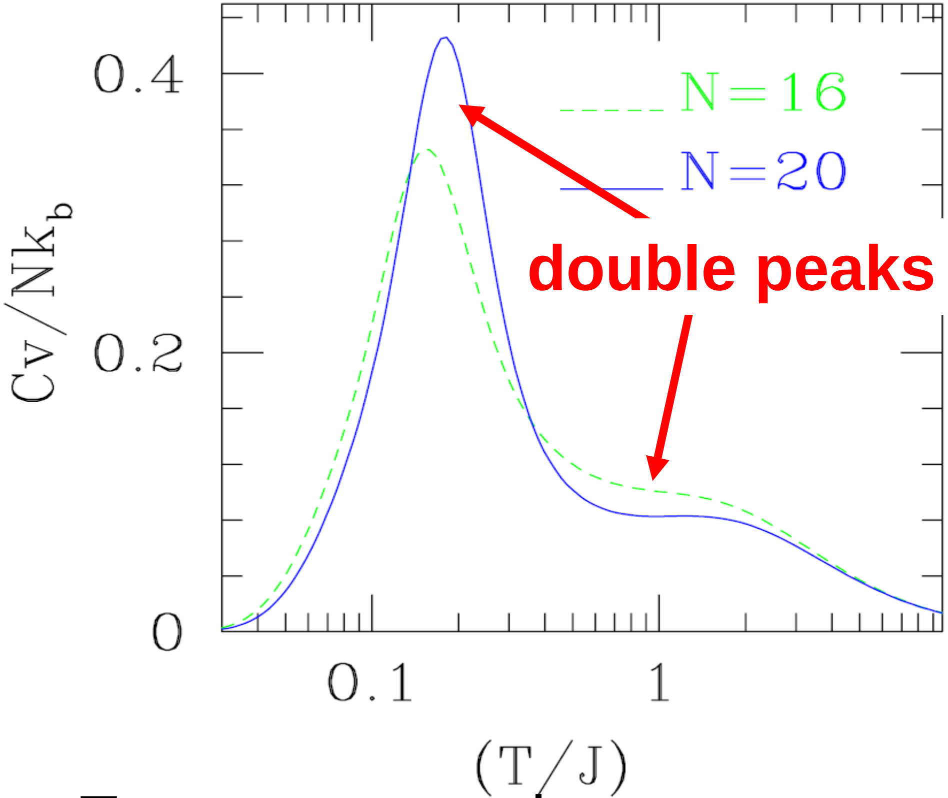


Parallel bond correlations are positive, but perpendicular ones are negative.

d-wave symmetry.

J_1-J_2 model: $J_1=-1$, $J_2=0.4$

Heat capacity



double peaks

Two energy scales

- High temperature peak: $T \sim J_1$

  formation of spin 1 objects due to short-range spin correlation.

- Low temperature peak: $T \sim 0.1 J_1$

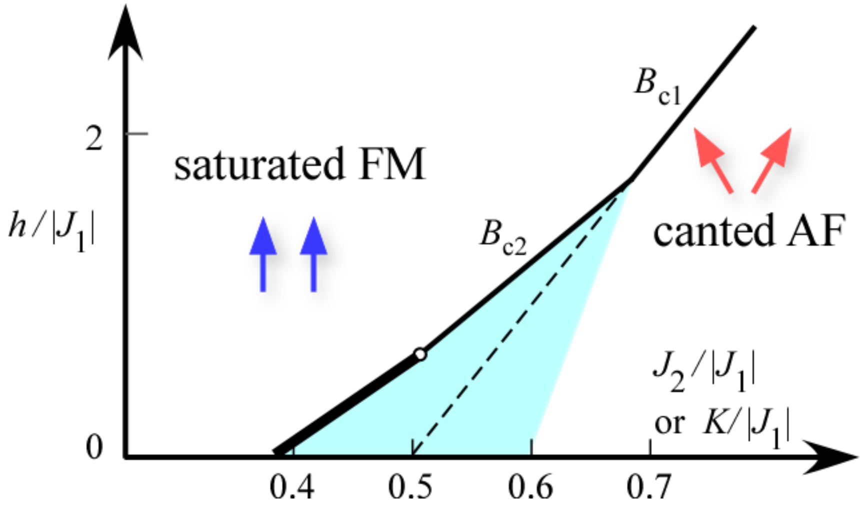
  short-range nematic correlation between spin 1 degrees of freedom.

Square lattice frustrated ferromagnets

(J_1-J_2 model, J-K model)

- Near saturation field, nematic order is induce by two magnon instability.

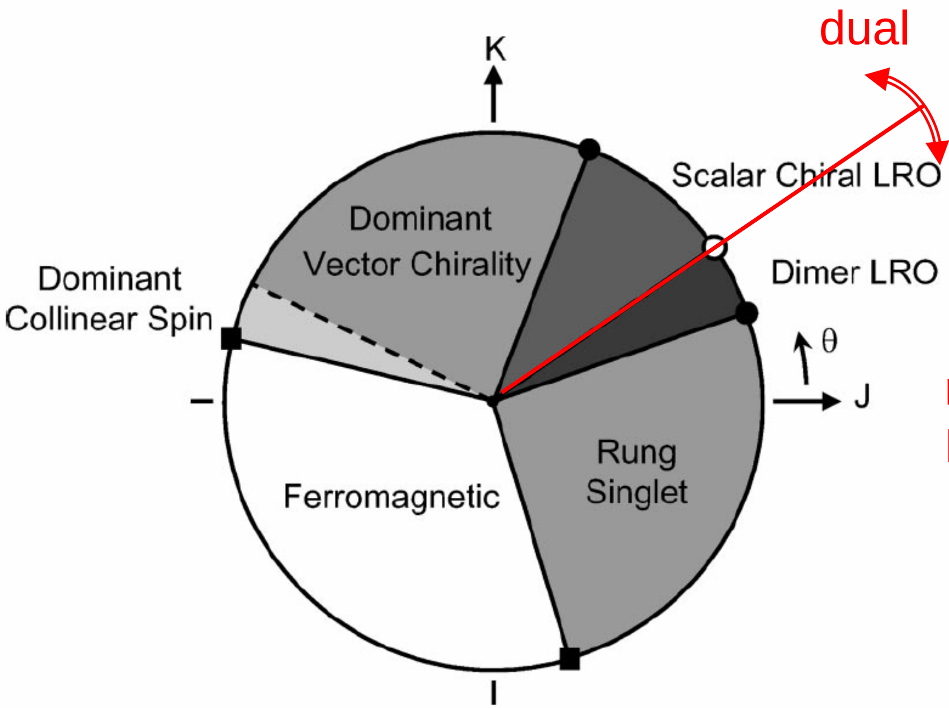
- Down to zero field, there is spin nematic order with d-wave symmetry.



MF

N. Shannon, TM, and P. Sindzingre, to appear in *Phys. Rev. Lett.*

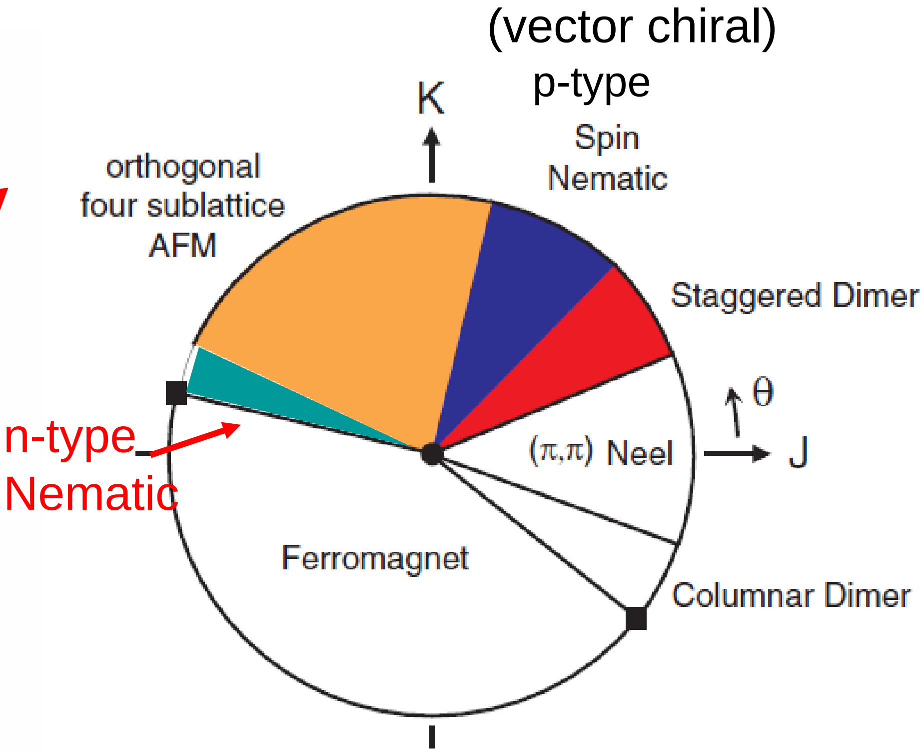
Ladder Lattice



A. Läuchli *et al.*,
PRB **67**, 100409(R) (2003).

T. Hikihara, TM, and X. Hu,
PRL **90**, 087204 (2003).

Square Lattice



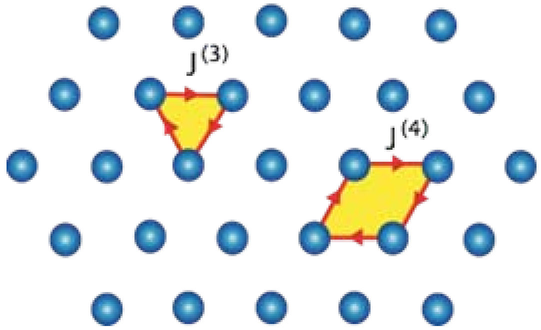
A. Läuchli *et al.*,
PRL **95**, 137206 (2005).

N. Shannon, TM, and P. Sindzingre,
PRL, *In press.*

future prospects

- contact with experiment -

Multiple spin exchange model on a triangular lattice -- application to gapless spin liquid in solid ^{3}He on graphite



$$\mathcal{H} = 2J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i \mathbf{S}_j + K \sum_{\langle 1234 \rangle} P_{1234} + P_{1234}^{-1}$$

☐ Ring exchange induced spin liquid ?

☐ Frustrated ferromagnet ?

→ Nematic order ?