A05: Quantum crystal and ring exchange

Novel magnetic states induced by ring exchange

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- Spin nematic/quadrupolar phases
  - S=1/2 frustrated ferromagnets
  - Spins triplet RVB state
    (T. Momoi)

- Multiple-spin exchange model on the triangular lattice
  - 2D solid $^3$He
    (T. Momoi, K. Kubo)

- Spin dynamics, spin crossover
  (S. Miyashita)

- Supersolid

- Mott transition in frustrated electron systems
  - Reentrant behavior
    (T. Ohashi, T. Momoi, H. Tsunetsugu, N. Kawakami)

- 2D solid $^3$He

- Ring exchange

- Spin dynamics, spin crossover (S. Miyashita)

- Supersolid

- Magnetism in cold atoms (S. Miyashita)
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## Outline

1. **Introduction:**
   - Spin nematic order
   - BEC of bound magnon pairs
   - Spin-triplet RVB state

2. **Multiple-spin exchange model: J-J₄-J₅-J₆ 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

**Triangles lattice**
- 2D solid He3
- Ring exchange

**Square lattice**
- Pb$_2$VO(PO$_4$)$_2$ (E. Kaul et al.)
- (CuCl)LaNb$_2$O$_7$, (CuBr)A$_2$Nb$_3$O$_{10}$

**1D zigzag lattice**
- edge-sharing chain cuprates
  - LiCuVO$_4$, LiCu$_2$O$_2$, Rb$_2$Cu$_2$Mo$_3$O$_{12}$, Li$_2$ZrCuO$_4$

**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

$$H = J_1 \sum_{\text{N.N.}} S_i \cdot S_j + J_2 \sum_{\text{N.N.N.}} S_i \cdot S_j,$$

Square lattice

1D zigzag lattice

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

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

- 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 quadrupolar order

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

Spin nematic order can be regarded as

“BEC of bound magnon pairs with $k=(0,0)$”

A. V. Chubukov, PRB (1991)

$$\langle S^-_i S^-_j \rangle = Q e^{2i\theta}$$

Spin quadrupolar order
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 $k = (0,0)$ 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

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

\[ = \quad -\quad + \quad - \quad + \quad \ldots \ldots \quad \text{entangled state} \]

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

wave function $\approx \bigotimes_i |\phi_i> = \left< Q_i^{x^2-y^2} \right> = \left< S_i^x S_i^x - S_i^y S_i^y \right>$

product state $\left< Q_i^{xy} \right> = \left< S_i^x S_i^y + S_i^y S_i^x \right>$
Slave boson formulation of spin nematic states in frustrated ferromagnets

R. Shindou and TM, PRB (2009)

Fermion representation

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

Local constraint

\[ f_{j,\alpha}^{\dagger} f_{j,\alpha} = 1 \]

\[ f_{j\uparrow}, f_{j\downarrow} \quad \text{fermion operators} \]

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

\[ -4S_i \cdot S_j \rightarrow -|D_{ij}|^2 + \sum_{\mu=x,y,z} [\psi_i^{\dagger} U_{ij,\mu} \psi_j \tau_\mu^{\dagger}] \]

where \( D_{ij} \) denote d-vectors of triplet pairing

\[ \hat{\Delta}_{jj} = \left( \begin{array}{cc} \langle f_{j\uparrow}^{\dagger} f_{i\uparrow} \rangle & \langle f_{j\uparrow}^{\dagger} f_{i\downarrow} \rangle \\ \langle f_{j\downarrow}^{\dagger} f_{i\uparrow} \rangle & \langle f_{j\downarrow}^{\dagger} f_{i\downarrow} \rangle \end{array} \right) = \left( \begin{array}{cc} -D_{z}^x + iD_{z}^y & D_{z}^2 \\ D_{z}^2 & D_{z}^x + iD_{z}^y \end{array} \right) \]

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

\[ -2Q_{ij}^{\mu\nu} = D_{ij}^{\mu} D_{ij}^{\nu} - \frac{\delta_{ij}}{3} |D_{ij}|^2 + \text{H.c.} \]

For example,

\[ \langle S_i^- S_j^- \rangle = \langle f_{j\uparrow} f_{j\uparrow}^\dagger f_{i\downarrow}^\dagger f_{i\downarrow} \rangle = \langle f_{j\uparrow} f_{j\uparrow}^\dagger \rangle^* \langle f_{i\downarrow} f_{i\downarrow} \rangle = - (D_{ij}^x)^2 + (D_{ij}^y)^2 - i(D_{ij}^x D_{ij}^y + D_{ij}^y D_{ij}^x) \]

\[ Q_{ij}^{\mu\nu}(r) = d_{ij}^{\mu}(r) d_{ij}^{\nu}(r) - \frac{\delta_{ij}}{3} |d(r)| \]

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

director – D-vector correspondence
Mean-field approximation of square lattice $J_1-J_2$ model

New phase

triplet-pairing on FM interactions and hopping amplitude on AF interactions

spin-triple resonating valence bond state

(spinn-triplet RVB state)
This mean-field solution has the same magnetic structure as d-wave bond nematic state.

BW state

\[ d(k) \equiv \hat{x} \sin k_x + \hat{y} \sin k_y. \]

\[
D^x_{j'l} = i \left( \delta_{j',l+e_x} - \delta_{j',l-e_x} \right)
\]

\[
D^y_{j'l} = i \left( \delta_{j',l+e_y} - \delta_{j',l-e_y} \right)
\]

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

d-wave bond nematic state

\[ Q_{xx} - Q_{yy} > 0 \]

\[ Q_{xx} - Q_{yy} < 0 \]
Low energy excitations around the BW state

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

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

- 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 2$^{\text{nd}}$ layer of 2D solid $^3$He on graphite

- gapless spin liquid
  - specific heat
  - linear specific heat
    - (cf. 2D FM)
  - double peak structure
- magnetization plateau at 1/2

- No drop of susceptibility down to 10$\mu$K
Theoretical model: multiple-spin exchange model

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

\( J = J_2 - 2J_3 \)

Parameter fitting  

\( J = 2.8, \quad J_4 = 1.4, \quad J_5 = 0.45, \quad J_6 = 1.25 \text{ (mK)} \)
In case of two- and four-spin exchange model \((J-J_4)\) model

**In a strong \(J_4\) regime \(J_4/|J| = 1/2\)**

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

- Magnetization process has a wide plateau at \(m/m_{\text{sat}} = 1/2\), 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 \((J-J_4 \text{ model})\)

**Near the border of FM phase** \(0.24 < K/|J| < 0.28\)

- \(m > 0\),
  - condensation of 3 magnon bound states
  - “Triatic order” (octupolar order)
  
- \(m = 0\),
  - strong competition between nematic and triatic correlations

\[
\langle S_i^- S_{i+e_1}^- S_{i+e_2}^- \rangle = \phi e^{3i\theta}
\]

\[
\langle S_i^x \rangle = \langle S_i^y \rangle = 0
\]

\(\theta = \phi - \pi \)

**Images**: Fully polarized triangle, CAF, TRIATIC

- \(J_4/|J|=0.5\), \(J_5/|J|=0.16\), \(J_6/|J|=0.44\)

Collin et al.
In the classical limit \((S \to \infty)\)

Mean-field phase diagram

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

One magnon excitations

\[ \varepsilon(k) = h - 2\left(J_2 + 4J_4 - 10J_5 + 2J_6\right) \times \left\{3 - \cos k \cdot e_1 - \cos k \cdot e_2 - \cos k \cdot e_3\right\} \]

have zero flat mode at mean-field phase boundary.

*Individual magnons are localized!*
Magnon instability to the FM (fully polarized) state at saturation field

$J_2 - J_4$ model

(case of $J_5 = J_6 = 0$)

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

Antiferro-triatic state

space rotation $R_{\pi/3} \rightarrow \exp(\pm i2\pi/3)$

$d + id$ wave

Chiral (?) nematic state

3 sublattice structure

Canted AF

$J_2 = -2, 3J_6 = 8J_5$

$J_5$

$J_4$

FM

2 mag.

1 mag.

3 mag. (F)

3 mag. (AF)
Numerical results

Instability at saturation

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

magnetization process

$\Delta S^z = 2$

$\Delta S^z = 3$
Condensation of bosons with two spices

d\pm i\sigma\text{-wave magnon pairs}

\[ j = \exp\left( \pm i \frac{2\pi}{3} \right) \quad (\pm: \text{chirality}) \]

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

density imbalance \( n_+ > n_- \)

chiral nematic order

non-chiral nematic order

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

\[ O_{d+id} - O_{d-id} \]
Chiral symmetry breaking?

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

Answer: No.

However, some of them are not degenerate → no chiral symmetry breaking.
Possible nematic orders induced by d+id-wave magnon pairs

(a) Chiral nematic order

\[
S = \frac{N}{2} - 2(3\pi - 2)
\]

Chiral nematic order

\[
R_{2\pi/3} = j, j^2, R_\pi = 1
\]

(b) Non-chiral nematic orders

\[
S = \frac{N}{2} - 2(3\pi - 1)
\]

Non-chiral nematic order \((Q_+ + Q_-)\)

\[
R_{2\pi/3} = j, j^2, R_\pi = 1
\]

Non-chiral nematic order \((Q_+ - Q_-)\)

\[
R_{2\pi/3} = j, j^2, R_\pi = 1
\]
Magnetization plateau at $m/m_{\text{sat}} = 1/2$

Symmetries are consistent with BEC of bound magnon pairs

No magnon bound state
Crossover from FM interaction dominant system to AF ring exchange dominant system

\[ \frac{m}{m_{sat}} = \frac{1}{2} \]

SDW (uuud structure)

\[ J_2 = -2, \, 3J_6 = 8J_5 \]

\[ J_{eff} = J - 10J_5 + 2J_6 \]
Phase diagram

- Still large size dependence remains
- Too large $J_6$?
Another magnetization plateau?

SDW at $m/m_{\text{sat}} = 5/9$

9-fold degeneracy

- Unit vectors $(3, 0)$, $(3/2, 3\sqrt{3}/2)$
- Reciprocal vectors $(2\pi/3, 2\pi/3\sqrt{3})$, $(0, 4\pi/3\sqrt{3})$

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

Unusual magnon excitations in $S(k, \omega)$

**1d case**

$h \parallel z$

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