Between classical order and quantum spin liquids: intermediate quantum effects

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Alois Loidl and group
Augsburg
Quantum Magnetism

- spin ice
- AFs
- skyrmions
- magnetization plateaux
- spin density wave
- spin nematic
- quantum paramagnets
- quantum criticality
- quantum spin liquids

Fully classical
Local quantum fluctuations
Quantum entanglement
quantum-ness
Quantum Magnetism

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Local quantum fluctuations

quantum-ness
Nice example with strong quantum renormalizations

- All phases encountered are ordered, short-range entangled states
- BUT most are different from those of the classical model
- And excitations are highly renormalized from linear spin waves
Results for three leg antiferromagnetic spin tube

Based on combination of DMRG, field theory, 1d methods, and high field expansion

Ru Chen, Hyejin Ju, Hong-Chen Jiang, Oleg Starykh, and LB, 2013
Cone state

Fully consistent with rigid spins
This is the classical ground state *throughout* the phase space

Excitations are gapless spin waves - semiclassical quantization of small oscillations of spins
According to the classical molecular field theory,

22, 23 a transition from a helical spin structure to a fan structure can occur when an external field is applied in the easy plane. The helix-fan transition is accompanied by a jump in magnetization, and not by the plateau. Examples of this include the recently observed phase transition in RbCuCl$_3$ for a magnetic field perpendicular to the c axis.  

24, 26 At low temperatures, RbCuCl$_3$ has a monoclinic structure, which is closely related to the crystal structure of CsCuCl$_3$.  

27, 28 The exchange interaction along the c axis is ferromagnetic, and interactions...
Planar orders

Classical ground state is always umbrella-like, but quantum fluctuations almost completely remove this
SDW states can be considered soliton lattices, and can be understood based on the behavior of spin chains.

Large domain of SDW state means that quasi-1d nature is enhanced by quantum fluctuations.

Non-uniform spin lengths are non-classical!

Excitations are phasons, which do not correspond to small spin rotations.
Quantum Magnetism

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Quantum Magnetism:

- Quantum entanglement
- quantum criticality
- quantum spin liquids
- quantum paramagnets
- Local quantum fluctuations
- Fully classical
Spin nematic

Frustrated ferromagnetic chain

\[ H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - h \sum_i S_i^z. \]

Hikihara et al., 2008

Sudan et al., 2009

LiCuVO\(_4\)
Spin nematic

Frustrated ferromagnetic chain

$$H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - h \sum_i S^z_i.$$
Spin nematic

Frustrated ferromagnetic chain

\[ H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - h \sum_i S_i^z. \]

H/(|J_1|+J_2) vs. J_2/(|J_1|+J_2)

FM

quasi-spin-nematic
Magnon binding

single spin flip
\[ |\psi_1\rangle = \sum_i \psi(i) S_i^- |\text{FM}\rangle \]
\[ E - E_{\text{FM}} = \epsilon_1 + h \]

double spin flip
\[ |\psi_2\rangle = \sum_{i,j} \psi(i,j) S_i^- S_j^- |\text{FM}\rangle \]
\[ E - E_{\text{FM}} = \epsilon_2 + 2h \]

\[ \epsilon_2 < 2 \epsilon_1 : \text{“molecular” bound state} \]

Formation of molecular fluid

For d>1 at T=0 this is a molecular BEC = true spin nematic
Hidden order

No dipolar order
\( \langle S_i^+ \rangle = 0 \)
\( \langle S_i^\pm \rangle = 0 \)
\( \langle S_i^+ S_j^- \rangle \sim e^{-|i-j|/\xi} \)
\( S^z=1 \) gap

Nematic order
\( \langle S_i^+ S_{i+a}^+ \rangle \neq 0 \)
Magnetic quadrupole moment
Symmetry breaking \( U(1) \to Z_2 \)

can think of a fluctuating fan state
Quasi-1d nematic

1d $J_1$-$J_2$ chain is only quasi-spin-nematic power-law correlations

$\Psi \sim (S^+)^2 : \text{spin-nematic}$

$\phi \sim S^z e^{i q x} : \text{SDW}$

$\Psi$ “dominant”

$\phi$ “dominant”
Quasi-1d nematic

1d $J_1$-$J_2$ chain

Interchain coupling

$$\sim J' \phi_y \phi_{y+1} + (J')^2/J \Psi_y \Psi_{y+1}$$

extra suppression of spin-nematic order in quasi-1d limit
Quasi-1d nematic

1d $J_1$-$J_2$ chain

In case of SDW$_2$, LRO of the longitudinal spin correlation is short ranged and decays exponentially with distance. When interchain couplings are not frustrated, as in frustrated systems with weak interchain couplings, the nematic phase region is reduced to a higher-field regime compared to the previous theories for spin-nematic phases. We note that, in interchain couplings, however, the nematic phase region has been identified.

The transition temperature of each order is obtained from Eqs. (9) and (12), where a new phase denoted as the 3D nematic ordered phase has been observed at $T/J \approx -0.5$. When interchain couplings are not frustrated, as in frustrated systems with weak interchain couplings, the nematic phase region is reduced to a higher-field regime compared to the previous theories for spin-nematic phases. We note that, in interchain couplings, however, the nematic phase region has been identified.

The former is a good analogue for classical spin system frustrated magnets with an arbitrary combination of frustrated magnet, which is given by:

$$ T/J \approx -0.5 $$

We note that, in interchain couplings, however, the nematic phase region has been identified.

In both states of $J_1/J_2 = 0.01$, $J_1/J_2 = 0.005$, and $J_1/J_2 = 0.005$, the SDW and nematic ordered phases appear at sufficiently low temperature.

From Eqs. (9) and (12), we find that the ordering wave vector is given by:

$$ k = \frac{\pi}{L} $$

We consider is a part of the spin-phonon coupling interaction, which consists of weakly coupled spin-nematic ordered phases in quasi-1D magnets.
Excitations

- How to diagnose nematic vs. SDW?
- problem: disorder pins SDW, smearing elastic peak, while nematic has no elastic peak

SDW

nematic

details on polarization etc. -- see O. Starykh + LB, 2014
Frustrated ferromagnetic chain

\[
H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - h \sum_i S_i^z.
\]

Multipolar phases

Frustrated ferromagnetic chain
Multipolar phases

Frustrated ferromagnetic chain

\[ H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - h \sum_i S_i^z. \]

amazing progression of successive multipolar phases on approaching the zero field FM critical point!

Is it an infinite progression? Universality?
Lifshitz Point

- Nature of QCP is strongly constrained by exactness of FM ground state
- Effective action - NLσM

\[ S = \int dx d\tau \left\{ i s A_B [\hat{m}] + \delta |\partial_x \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h \hat{m}_z \right\} \]

- Berry phase term
- tunes QCP
- two symmetry allowed interactions at O(q^4)

\[ A_B = \frac{\hat{m}_1 \partial_x \hat{m}_2 - \hat{m}_2 \partial_x \hat{m}_1}{1 + \hat{m}_3}. \]

All properties near Lifshitz point obey “one parameter universality” dependent upon u/K ratio
Lifshitz Point

\[ S = \int dx d\tau \left\{ isA_B[\hat{m}] + \delta |\partial_x \hat{m}|^2 + K |\partial_x^2 \hat{m}|^2 + u |\partial_x \hat{m}|^4 - h\hat{m}_z \right\} \]

- Intuition: behavior near the Lifshitz point should be semi-classical, since “close” to FM state which is classical

\[ x \rightarrow \sqrt{\frac{K}{|\delta|}} x \quad \tau \rightarrow \frac{K}{\delta^2} \tau \]

\[ S = \sqrt{\frac{K}{\delta}} \int dx d\tau \left\{ isA_B[\hat{m}] + \text{sgn}(\delta) |\partial_x \hat{m}|^2 + |\partial_x^2 \hat{m}|^2 + v |\partial_x \hat{m}|^4 - \bar{h}\hat{m}_z \right\} \]

Large parameter: saddle point!

\[ v = \frac{u}{K} \quad \bar{h} = \frac{hK}{\delta^2} \]
Saddle point

\[ S = \sqrt{\frac{K}{\delta}} \int dx d\tau \left\{ isA_B[\hat{m}] + \text{sgn}(\delta)|\partial_x \hat{m}|^2 + |\partial_x^2 \hat{m}|^2 + v|\partial_x \hat{m}|^4 - \overline{h} \hat{m}_z \right\} \]

\( v \) derives from quantum fluctuations

By a spin wave analysis, one finds \( v \sim O(1/S) < 0 \)

\[ h_c = \frac{\delta^2}{8K\sqrt{v}(1-\sqrt{v})}, \quad -1 < v < -\frac{1}{4} \]

local instability of FM state
Multipolar phases

Frustrated ferromagnetic chain

First order metamagnetic transition near Lifshitz point

Open question: can we connect multipolar phases to the metamagnetism?

Higher dimensions?
Quantum Magnetism

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- Spin ice
- Skyrmions
- Quantum criticality
- Quantum entanglement
- Quantum paramagnets
- Spin density wave
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- AFs
- Local quantum fluctuations
- Fully classical

quantum-ness
Spin-orbital entanglement

- Can occur in quantum magnets with strong spin-orbit coupling and orbital degeneracy
- My favorite example: FeSc$_2$S$_4$ (3d diamond lattice)

Fe$^{2+}$: S=2 but 2 orbital states

d orbitals

tetrahedral coordination
Spin-orbital entanglement

• Spin and orbital states couple and can

\[ H_{SOC} = \lambda \mathbf{L} \cdot \mathbf{S} \]

t\text{\textsubscript{2g}} degeneracy

\[ H_{SOC} = -\lambda \left( \frac{1}{\sqrt{3}} \tau^x \left[ S_x^2 - S_y^2 \right] + \tau^z \left[ S_z^2 - \frac{S(S + 1)}{3} \right] \right) \]

e\text{\textsubscript{g}} degeneracy

• Ground state of \( H_{SOC} \) is entangled

\[ |\psi\rangle = \frac{1}{\sqrt{2}} |S_z = 0\rangle |\begin{array}{c}
\text{blue} \end{array}\rangle - \frac{1}{2} (|S_z = 2\rangle + |S_z = -2\rangle) |\begin{array}{c}
\text{red} \end{array}\rangle \]

entanglement of spin+orbitals: \( S(A) = \ln 2 \)
Entanglement versus Order

- Exchange competes with SOC

forms magnetic order
forms local singlets

\[ H = J \sum_{\langle ij \rangle} S_i \cdot S_j - \lambda \sum_i \hat{h}_{\text{SOC}}(i) \]

similar to Néel-dimer
Quantum Critical Point

- SOC competes with exchange

\[ \langle \Phi_Q \rangle \neq 0 \]

FeSc$_2$S$_4$

SO singlet

antiferromagnet

Gang Chen, LB, and Andreas Schnyder, 2009
Quantum Critical Point

- SOC competes with exchange

Described by multicomponent $\phi^4$ theory in $D=3+1=4$ dimensions

In its upper critical dimension but still has the main aspects of such QCPs
QCP

• INS
  
  soft triplon excitation of proximate AF order

  gap at \( k=(1/2,0,0) \)

  \( \sim 1-2K \)

• consistency with heat capacity and NMR \( 1/T_1 \)


bandwidth \( \sim \) few meV
Optics

- New measurements see both SOC-split “excitonic” states and quantum critical continuum (2-triplons)

L. Mittelstädt, M. Schmidt, Zhe Wang, F. Mayr, V. Tsurkan, P. Lunkenheimer, J. Deisenhofer, D. Ish, L. Balents, and A. Loidl· under review
Optics

- New measurements see both SOC-split "excitonic" states and quantum critical continuum (2-triplons)

triplon is strongly damped in the QC regime
Background

- Continuum absorption arises from two-triplon excitations

Polarization
\[ \vec{P} \sim Q_\mu \Phi_Q \times \partial_\mu \Phi_Q \]

Dielectric constant
\[ \epsilon''(\omega) \sim \omega^2 \coth\left(\frac{\omega}{4k_BT}\right) \]

in quantum critical regime
Excitons?

Possible second excitation?

Fe\textsuperscript{2+} SOC ion has internal excitonic level structure

**Fig. 3.** Allowed transitions (arrows) among the spin-orbit levels of the $^5E$ term of Fe\textsuperscript{2+} ions in ZnS permitted by the selection rules for electric- and magnetic-dipole processes. Transitions between levels not joined by an arrow in the figure are forbidden.
• Also important in many 4d+5d double perovskites

• Same mechanism for QCPs invoked recently by Khaliullin for 4d$^4$ and 5d$^4$ systems, e.g. Ir$^{5+}$

• SO-entangled states common in iridates, c.f. Wednesday session
Summary

• There are a range of interesting “semi-exotic” quantum states between classical ordered phases and the extreme example of a quantum spin liquid

• Magnetization plateaux, SDW states, spin nematics, quantum paramagnetic and quantum critical states

• Explored Lifshitz point as a “parent” for multipolar states and metamagnetism

• New evidence for quantum criticality from optics