Vector spin chirality in classical & quantum spin systems

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Motivation:

- A recent flurry of activities on the so-called "multiferroic" materials
- As a result, a new type of coupling of spin & lattice came into focus
- The key theoretical concept appears to be:
- Two noncollinear spins form a nonzero vector spin chirality (spin chirality)

 $C_{ij} = \langle S_i \times S_j \rangle$

The spin chirality couples to the lattice displacement or dipole moment P_{ij}

Control of ferroelctricity using magnetism

Nature 426, 55 (2003)
Cited: 248 times as of April, 2007TbMnO3Magnetic control of
polarizationferroelectric

T. Kimura¹*, T. Goto¹, H. Shintani¹, K. Ishizaka¹, T. Arima² & Y. Tokura¹



Mn d orbitals are orbital ordered

Magnetic interaction contain both ferro and antiferro exchanges

Fertile ground for frustrated magnetism

Control of ferroelctricity using magnetic field

Cheong & Mostovoy Review Nature Materials 6, 13 (2007)



At low temperature, both magnetic & dipole order exist

Impossible without the existence of a strong coupling between magnetic order & dipole order

Magnetic field along b switches polarization from c to b axis in TbMnO $_3$ (Kimura)

Connection to Spiral Magnetism

The original work of Kimura demonstrated controllability of FE through applied field

Connection to spiral magnetism made clear by later neutron scattering study of Kenzelman et al.

PRL **95**, 087206 (2005)

PHYSICAL REVIEW LETTERS

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Magnetic Inversion Symmetry Breaking and Ferroelectricity in TbMnO₃

M. Kenzelmann,^{1,2,3} A. B. Harris,⁴ S. Jonas,³ C. Broholm,^{3,5} J. Schefer,² S. B. Kim,⁶ C. L. Zhang,⁶ S.-W. Cheong,⁶ O. P. Vajk,⁵ and J. W. Lynn⁵



Theoretical idea

With spiral magnetism one can define a SENSE of ROTATION (chirality) given by the outer product of two adjacent spins $C_{ii} = \langle S_i \times S_i \rangle$

 C_{ij} is odd under inversion, another order parameter of the same symmetry, P_{ij} (olarization), can couple linearly to it as $C_{ij}*P_{ij}$

For collinear antiferromagnetism the sense of rotation is ill-defined



Spin-polarization coupling



In the DM interaction, P_{ij} is a static vector, in multiferroics, P_{ij} is dynamic.

Katsura, Nagaosa, Balatsky PRL 95, 057205 (2006) Jia, Onoda, Nagaosa, Han arXiv:cond-mat/0701614

Multiferroics with noncollinear magnetic and ferroelectric phase

Material	d-electron	Polarization (µC/m²); Q	Specifics
Tb <mark>Mn</mark> O ₃ (Kimura et al Nature 2003; Kenzelman et al PRL 2005)	d ⁴ (t _{2g}) ³ (e _g) ¹	800; q~0.27	Orbital order
<mark>Ni₃V₂O₈</mark> (Lawes et al PRL 2005)	d ⁸ (t _{2g}) ⁶ (e _g) ²	100; q~0.27	Kagome
Ba _{0.5} Sr _{1.5} Zn ₂ Fe ₁₂ O ₂₂ (Kimura et al PRL 2005)	d ⁵ [(t _{2g}) ³ (e _g) ²]	150 (B=1T); N/A	N/A
<mark>CoCr₂O</mark> 4 (Yamasaki et al. PRL 2006)	Co ²⁺ :d ⁷ (e) ⁴ (t ₂) ³ Cr ³⁺ :d ³ (t _{2g}) ³	2; [qq0] q~0.63	ferrimagnetic
<mark>Mn</mark> WO ₄ (Taniguchi et al. PRL 2006)	d ⁵ (t _{2g}) ³ (e _g) ²	50; q=(214, .5, .457)	N/A
Cu <mark>Fe</mark> O ₂ (Kimura et al PRB 2006)	d ⁵ (t _{2g}) ³ (e _g) ²	400 (B>10T); 1/5 <q<1 4<="" td=""><td>2D triangular; Field-driven</td></q<1>	2D triangular; Field-driven
Li <mark>Cu</mark> VO ₄ (Naito et al cond-mat/0611659)	d ⁹ (t _{2g}) ⁶ (e _g) ³	N/A; q~0.532	1D chain
Li <mark>Cu₂O</mark> 2 (Park et al PRL 2007)	d ⁹ (t _{2g}) ⁶ (e _g) ³	<10; q~0.174	1D chain

RED = magnetic ions

Existing experiments

Often, there is the magnetic transition to COLLINEAR spin states, for which no polarization is induced

A second transition at a lower temperature to spiral spin states cause nonzero polarization



Abstracting away

Spiral spin phase supports an additional DISCRETE order parameter having to do with the sense of spin rotation

This OP can couple to uni-directional polarization P

Can we envision a phase without magnetic order, but still has the remnant of chirality?



Chiral spin state in helical magnet?

Recent work: Ginzburg-Landau theory of spin models with SU(2)->O(2) symmetry



Examples of quantum spins with chirality

Hikihara et al. considered a spin chain with nearest and next-nearest neighbour XXZ-like interactions for S=1



 $H_{i,NN} = J_1(S_{ix}S_{i+1,x} + S_{iy}S_{i+1,y} + \Delta S_{iz}S_{i+1,z})$

 $H_{i,NNN} = J_2(S_{ix}S_{i+2,x} + S_{iy}S_{i+2,y} + \Delta S_{iz}S_{i+2,z})$

$$H = \sum_{i} \left(H_{i,NN} + H_{i,NNN} \right)$$

Long-range order of vector spin chirality

Define spin chirality operator $\hat{\kappa}_{ij} = S_i^x S_j^y - S_i^y S_j^x$ DMRG found chiral phase for S=1 when j=J₂/J₁ is sufficiently large

$$\langle \hat{\kappa}_{i,i+1} \hat{\kappa}_{j,j+1} \rangle \rightarrow \kappa^2 \text{ as } |i-j| \rightarrow \infty$$



Zittartz's work

Meanwhile, Zittartz found exact ground state for the class of anisotropic spin interaction models with nearest-neighbor quadratic & biquadratic interactions

$$I_{i,\Delta} = S_{ix}S_{i+1,x} + S_{iy}S_{i+1,y} + \Delta S_{iz}S_{i+1,z}$$
$$H = \sum_{i} \left(J_1 I_{i,\Delta_1} + J_2 I_{i,\Delta_2}^2 \right)$$

Zittartz's ground state is SU(2)->O(2) generalization of Affleck-Kennedy-Lieb-Tesaki (AKLT) state.

Zittartz's ground state does not support spin chirality order

On one hand we have numerical evidence of spin chirality in sufficiently frustrating spin-1 chain system.

On the other hand we do not have an explicit construction of such a state

-> Can we find one?

XXZ Hamiltonian with DM

$$H = J_{1} \sum_{\langle ij \rangle} S_{i}^{z} S_{j}^{z} + J_{2} \sum_{\langle ij \rangle} (S_{i}^{x} S_{j}^{x} + S_{i}^{y} S_{j}^{y})$$

Including Dzyalonshinskii–Moriya (DM) interaction

$$H = J_{1} \sum_{\langle ij \rangle} S_{i}^{z} S_{j}^{z} + J_{2} \sum_{\langle ij \rangle} (S_{i}^{x} S_{j}^{x} + S_{i}^{y} S_{j}^{y}) + J_{3} \sum_{\langle ij \rangle} (S_{i} \times S_{j}) \cdot \hat{z}$$

$$H = J_{1} \sum_{\langle ij \rangle} S_{i}^{z} S_{j}^{z} + (J_{2} + iJ_{3}) \sum_{\langle ij \rangle} S_{i}^{+} S_{j}^{-} + (J_{2} - iJ_{3}) \sum_{\langle ij \rangle} S_{i}^{-} S_{j}^{+}$$

$$= J_{1} \sum_{\langle ij \rangle} S_{i}^{z} S_{j}^{z} + (J_{2}^{2} + J_{3}^{2})^{\frac{1}{2}} \sum_{\langle ij \rangle} (e^{i\theta} S_{i}^{+} S_{j}^{-} + e^{-i\theta} S_{i}^{-} S_{j}^{+})$$

DM interaction -> phase angle, flux

Staggered DM, arbitrary DM



Staggered oxygen shifts gives rise to

"staggered" DM interaction -> "staggered" phase angle, "staggered" flux

We can consider the most general case of arbitrary phase angles:

$$H = J_1 \sum_{\langle ij \rangle} S_i^z S_j^z + J_2 \sum_{\langle ij \rangle} (e^{i\theta_{ij}} S_i^+ S_j^- + e^{-i\theta_{ij}} S_i^- S_j^+)$$

Connecting non-chiral & chiral Hamiltonians

Define the model on a ring with N sites:

$$H[\{\theta_{ij}\}] = J_1 \sum_{\langle ij \rangle} S_i^z S_j^z + J_2 \sum_{\langle ij \rangle} (e^{i\theta_{ij}} S_i^+ S_j^- + e^{-i\theta_{ij}} S_i^- S_j^+)$$

Carry out unitary rotations on spins

$$S_i^+ \to U_i^+ S_i U_i = S_i^+ e^{-i\alpha_i}$$

Choose angles such that $e^{i[\alpha_i - \alpha_j]} \equiv e^{i\theta_{ij}}$

This is possible provided

$$e^{i\sum_{\langle ij \rangle} \theta_{ij}} = 1$$

Hamiltonian is rotated back to XXZ:

$$U^{\dagger}[\{\theta_{ij}\}]H[\{\theta_{ij}\}]U = H_{\mathsf{X}\mathsf{X}\mathsf{Z}}$$

Connecting non-chiral & chiral Hamiltonians



Dillenschneider, Kim, Han arXiv: 0705.3993 [cond-mat.str-el] DM interaction and its connection to gauge transformation was noted earlier.

Shekhtman, Entin-Wohlman, Aharony PRL 69, 839 (1992)

Connecting non-chiral & chiral eigenstates

Correlation functions are also connected. In particular,

$$\langle \{\theta_{ij}\} | S_i^- S_j^+ | \{\theta_{ij}\} \rangle = e^{i[\alpha_i - \alpha_j]} \langle \mathsf{XXZ} | S_i^- S_j^+ | \mathsf{XXZ} \rangle$$

Since
$$\langle S_i^- S_j^+ \rangle = \langle S_i^x S_j^x + S_i^y S_j^y \rangle + i \langle S_i^x S_j^y - S_i^y S_j^x \rangle$$

and
 $\langle X X Z | S_i^- S_j^+ | X X Z \rangle = \langle X X Z | S_i^x S_j^x + S_i^y S_j^y | X X Z \rangle$

It follows that a non-zero spin chirality must exist in $|\{\theta_{ij}\}\rangle$

$$\langle \{\theta_{ij}\} | S_i^x S_j^y - S_i^y S_j^x | \{\theta_{ij}\} \rangle = \langle \mathsf{XXZ} | S_i^- S_j^+ | \mathsf{XXZ} \rangle \sin[\alpha_i - \alpha_j]$$

Eigenstates of $H[\{\theta_{ij}\}]$ are generally chiral.

Analogy with persistent current

For S=1/2, Jordan–Wigner mapping gives $J_2 \sum \left(e^{i\theta_{ij}} S_i^+ S_j^- + e^{-i\theta_{ij}} S_j^+ S_i^- \right)$ $\langle ij \rangle$ $e^{i\Phi} = e^{i\sum_i \theta_{i,i-1}}$ $J_2 \sum \left(e^{i\theta_{ij}} f_i^+ f_j + e^{-i\theta_{ij}} f_j^+ f_i \right)$ $\langle ij \rangle$ Spin chirality maps onto bond current $i\langle S_i^+S_j^--S_j^+S_i^-\rangle$ $i\langle f_i^+f_j-f_j^+f_i\rangle$

When the flux is integral, there will be no persistent current !? The gauge-invariant definition of fermion current is

$$i\langle e^{i\theta_{ij}}f_i^+f_j - e^{-i\theta_{ij}}f_j^+f_i\rangle \square i\langle e^{i\theta_{ij}}S_i^+S_j^- - e^{-i\theta_{ij}}S_j^+S_i^-\rangle$$

Given a Hamiltonian with non-chiral eigenstates, a new Hamiltonian with chiral eigenstates will be generated with non-uniform U(1) rotations:

$$S_i^+ \to U_i S_i U_i^\dagger = S_i^+ e^{i\alpha_i}$$
$$U[\{\alpha_i\}] H_{\text{non}-\chi} U^\dagger [\{\alpha_i\}] = H_\chi$$
$$U[\{\alpha_i\}] = \prod_i U_i$$

AKLT Primer (in Schwinger bosonese)

Well-known Affleck-Kennedy-Lieb-Tasaki (AKLT) ground states and parent Hamiltonians can be generalized in a similar way

$$H^{\mathsf{AKLT}} = \sum_{\langle ij \rangle} S_i \cdot S_j \left(1 + \frac{1}{3} S_i \cdot S_j \right)$$

Using Schwinger boson singlet operators

$$A_{ij}^{+} = a_i^{+}b_j^{+} - b_i^{+}a_j^{+}$$

$$H^{\mathsf{AKLT}} = \frac{1}{24} \sum_{\langle ij \rangle} (6 - A_{ij}^{\dagger} A_{ij}) (4 - A_{ij}^{\dagger} A_{ij})$$
$$A_{ij}^{\dagger} A_{ij} = 2(1 - S_i^z S_j^z - S_i^{\dagger} S_j^{-} - S_i^{-} S_j^{+})$$

AKLT ground state is $|AKLT\rangle = \prod_{\langle ij\rangle} A^+_{ij} |0\rangle$

Arovas, Auerbach, Haldane PRL 60, 531 (1988)

From AKLT to chiral-AKLT

Aforementioned U(1) rotations correspond to

$$a_i^+ \to a_i^+ e^{i\theta_i/2}, \quad b_i^+ \to b_i^+ e^{-i\theta_i/2}$$

$$A_{ij}^{+} = a_i^{+}b_j^{+} - b_i^{+}a_j^{+} \to A_{ij}^{+}[\theta_{ij}] = e^{i\theta_{ij}/2}a_i^{+}b_j^{+} - e^{-i\theta_{ij}/2}b_i^{+}a_j^{+}$$

$$H^{\chi \mathsf{AKLT}} = \frac{1}{24} \sum_{\langle ij \rangle} (6 - A^{\dagger}_{ij} [\theta_{ij}] A_{ij} [\theta_{ij}]) (4 - A^{\dagger}_{ij} [\theta_{ij}] A_{ij} [\theta_{ij}])$$

$$A_{ij}^{+}A_{ij} = 2(1 - S_i^z S_j^z - e^{i\theta_{ij}} S_i^{+} S_j^{-} - e^{-i\theta_{ij}} S_i^{-} S_j^{+})$$

Chiral-AKLT ground state is $|\chi AKLT\rangle = \prod_{\langle ij \rangle} A^+_{ij}[\theta_{ij}]|0\rangle$

Correlations in chiral-AKLT states

Equal-time correlations of chiral-AKLT states easily obtained as chiral rotations of known correlations of AKLT states:

With AKLT:
$$\langle S_i \cdot S_j \rangle_0 = 2\delta_{ij} + 4(1 - \delta_{ij})(-1/3)^{|i-j|}$$

With chiral-AKLT:

$$\langle \{\theta_i\} | S_i^x S_j^x | \{\theta_i\} \rangle = \langle \{\theta_i\} | S_i^y S_j^y | \{\theta_i\} \rangle = \frac{1}{3} \langle S_i \cdot S_j \rangle_0 \times \cos \theta_{ij}$$
$$\langle \{\theta_i\} | S_i^x S_j^y - S_i^y S_j^x | \{\theta_i\} \rangle = \frac{2}{3} \langle S_i \cdot S_j \rangle_0 \times \sin \theta_{ij}$$
$$\langle \{\theta_i\} | S_i^z S_j^z | \{\theta_i\} \rangle = \frac{1}{3} \langle S_i \cdot S_j \rangle_0$$

Excitation energies in SMA

Calculate excited state energies in single-mode approximation (SMA) for uniformly chiral AKLT state: $\theta_{ij} = \theta$

With AKLT:
$$E(k) = \frac{\langle S_{-k}^x H S_k^x \rangle}{\langle S_{-k}^x S_k^x \rangle} = \frac{f(k)}{s(k)}$$

 $f(k) = (10/27)(1 - \cos k), \ s(k) = 2(1 - \cos k)/(5 + 3\cos k)$

With chiral-AKLT: $E_x(k) = \frac{\langle S_{-k}^x H S_k^x \rangle}{\langle S_{-k}^x S_k^x \rangle} = \frac{f(k+\theta) + f(k-\theta)}{s(k+\theta) + s(k-\theta)}$

$$E_z(k) = \frac{f(k)}{s(k)}$$

Excitation energies in SMA



Higher-dimensional chiral AKLT

$$|\chi\rangle = \prod_{\langle ij\rangle} \left(A^+_{ij}[\theta_{ij}]\right)^M$$

M = 2S/(coordination number)

For chiral Hamiltonian, replace

$$S_i^+S_j^- + S_i^-S_j^+ \to e^{i\theta_{ij}}S_i^+S_j^- + e^{-i\theta_{ij}}S_i^-S_j^+$$

In the original AKLT Hamiltonian (For proof, see our paper arXiv:0705.3993)

In higher dimensions, it is not always possible to associate $\theta_{ij} = \theta_i - \theta_j$

A place to look? (perhaps $LaCu_2O_4$)



Coffey, Rice, Zhang PRB 44, 10112 (1991)

A place to look? (perhaps LaCu₂O₄)

Effects of Dsyaloshinskii-Moriya interaction



Weak ferromagnetism (LaCu204)



Cheong, Thompson, Fisk PRB 39, 4395 (1989)

Quantum mechanical spin chirality is readily embodied by the DM interaction

What is condensed by DM is more like the chiral solid, not chiral liquid

Search for vector spin chiral liquid will be interesting

Spin chirality in classical XY Model

The spins along a given axis rotates either clockwise (+1) or counterclockwise (-1). Ising variable can be associated.



Magnetic order implies chirality ordering.

But can chirality order first before magnetism does? If so, one has the classical chiral spin liquid phase.

We consider the modification of the antiferromagnetic XY model on the triangular lattice

$$E = J_1 \sum_{\langle ij \rangle} \cos[\theta_i - \theta_j] + J_2 \sum_{\langle ij \rangle} \cos[2\theta_i - 2\theta_j]$$

For $J_2=0$ this is AFXY on triangular lattice (long history associated with chirality transition)



We consider the modification of the antiferromagnetic XY model on the triangular lattice

$$E = J_1 \sum_{\langle ij \rangle} \cos[\theta_i - \theta_j] + J_2 \sum_{\langle ij \rangle} \cos[2\theta_i - 2\theta_j]$$

For J₁=0 this model supports nematic spin state due to equivalence of $\theta_i \& \theta_i + \pi$ (macroscopic degeneracy)



(Classical) statistical properties of J_2 -only model is identical to those of J_1 -only

$$Z_{2} = \int_{0}^{2\pi} \prod_{i} d\theta_{i} \exp\left(-\beta \sum_{\langle ij \rangle} \cos[2\theta_{i} - 2\theta_{j}]\right)$$
$$= 2^{N} \int_{0}^{2\pi} \prod_{i} d\theta_{i} \exp\left(-\beta \sum_{\langle ij \rangle} \cos[\theta_{i} - \theta_{j}]\right) = 2^{N} Z_{1}$$

Two types of the transition found on triangular lattice

- 1. Kosterlitz-Thouless(KT) transition
- 2. Chirality transition

XY model on triangular lattice



The separation of the phase transition temperatures is extremely small. The chirality-ordered phase is not well-defined.

> Sooyeul Lee and Koo-Chul Lee, Phys. Rev. B 57, 8472 (1998)

We find wide temperature region where chirality is condensed, when J_1 and J_2 co-exist, and $J_2 >> J_1$



Specific heat



Magnetic order undergoes transitions at lower critical temperature.

A scaling plot of M_1 using the Ising exponents $\beta = 1/8$, $\nu = 1$, and $T_c = 0.177$ works very well.



Nematic order



Helicity modulus



Critical phase for nematic order below T_{KT}



Chiral order



Z₂ (chiral) symmetry in nematic & magnetic phase



Degenerate states are chiral counterparts

Magnetic states: $\chi_{ij}^M = (S_i \times S_j \cdot \hat{z}) = \sin[\theta_i - \theta_j]$ are opposite

Nematic states: $\chi_{ij}^N = (S_i \times S_j \cdot \hat{z})^2 = \sin[2(\theta_i - \theta_j)]$ are opposite

Magnetic order IMPLIES chirality symmetry breaking in χ^M_{ij} Nematic order IMPLIES chirality symmetry breaking in χ^N_{ij}

Does nematic order IMPLY chirality symmetry breaking in χ^M_{ij} ??

I can give an argument that in fact it does.

In the presence of nematic ordering, each triangle supports the following configurations



J₂ interaction vanishes within the nematic manifold

 J_1 interaction lifts the degeneracy, results in the effective Hamiltonian valid within the nematic manifold

$$E = -J_1 \sum_{\langle ij \rangle} \sigma_i \sigma_j \qquad \chi \sim \sum_{\langle ij \rangle} \sigma_i \sigma_j$$

At any finite temperature the average of the energy is negative ->
the chirality must be positive!!

Phase diagram: $J_1 - J_2 XY$ model

$$H = J_1 \sum_{\langle ij \rangle} \cos[\theta_i - \theta_j] + J_2 \sum_{\langle ij \rangle} \cos[2(\theta_i - \theta_j)]$$



Quantum version ?

- The nematic order already breaks the chiral symmetry and opens up the possibility of the chirality condensation well above the magnetic transition.
- We are trying to see if the same mechanism will result in chiralitycondensed phase in quantum spin models on triangular lattice:

$$H = J_1 \sum_{\langle ij \rangle} (S_i \cdot S_j)_{\Delta} + J_2 \sum_{\langle ij \rangle} (S_i \cdot S_j)_{\Delta}^2$$
$$(S_i \cdot S_j)_{\Delta} = S_i^x S_j^x + S_i^y S_j^y + \Delta(S_i^z S_j^z)$$

Appendix



J₁-J₂ XY Model (classical)

We consider the modification of the antiferromagnetic XY model on the triangular lattice

$$E = J_1 \sum_{\langle ij \rangle} \cos[\theta_i - \theta_j] + J_2 \sum_{\langle ij \rangle} \cos[2\theta_i - 2\theta_j]$$

In the spin language this is equivalent to putting a bi-quadratic interaction, and $\Delta=0$ in Zittartz' model

$$H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j + J_2 \sum_{\langle ij \rangle} (S_i \cdot S_j)^2$$

We did Monte Carlo (MC) on the classical $J_1 - J_2$ model

Nematic vs. Magnetic Ordering in Λ Lattice



Z₂ (chiral) symmetry in nematic & magnetic phase



Degenerate states are chiral counterparts

Magnetic states:
$$\chi_{ij}^M = (S_i \times S_j \cdot \hat{z}) = \sin[\theta_i - \theta_j]$$
 are opposite

Nematic states: $\chi_{ij}^N = (S_i \times S_j \cdot \hat{z})^2 = \sin[2(\theta_i - \theta_j)]$ are opposite

Degenerate states are chiral counterparts

Magnetic states:

are opposite

Nematic states:

are opposite

Order Parameters

Order parameters referring to magnetic, nematic, and chiral orders are defined

$$M = \frac{3}{L^2} \left| \sum_{i} e^{i\theta_i} \right| \qquad N = \frac{3}{L^2} \left| \sum_{i} e^{2i\theta_i} \right|$$

$$\chi = \frac{1}{L^2} \sum_{\Delta_i} (\sin[\theta_{i1} - \theta_{i2}] + \sin[\theta_{i2} - \theta_{i3}] + \sin[\theta_{i3} - \theta_{i1}])$$

Magnetic, Nematic, Chiral





Phase Diagram

The interaction strengths are parameterized as follows

$$J_1 = 1 - x, J_2 = x (0 < x < 1)$$

Qualitatively the phase diagram looks like



Nematic phase also appears to be chiral

Perhaps a spin system with a sufficiently large frustrating interaction will support a chiral phase, and hence a ferroelectricity, without having the magnetic order