Novel Order and Dynamics in Frustrated and Random Magnets

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What is frustration?

Everyone is happy
No frustration!

Someone is unhappy
Frustration!
Geometrical frustration (A)

Some spin is “unhappy”
Spin direction is not uniquely determined
→ Nontrivial degeneracy
Enhanced fluctuations
Geometrical frustration (B)

Frustration partially resolved due to the spin canting $\rightarrow$ New symmetry

New degrees of freedom
Chirality

right-handed chirality $\kappa +$

left-handed chirality $\kappa -$
Gemetrically frustrated lattices

- Triangular lattice
- Kagome lattice
- Pyrochlore lattice
Frustrated magnets

“Spins” are located on the lattice interacting with each other, with certain “frustration” in it, often leading to exotic order.

- **Spin**: $Si$
  - Ising spin: $S=S_z$
  - $XY$ spin: $S=(S_x, S_y)$
  - Heisenberg spin: $S=(S_x, S_y, S_z)$
Magneteic phase transition

Energy gain

Entropy gain

Temperature $T$

Ferromagnet

Ordered state

Disordered state (paramagnetic state)

Antiferromagnet (AF)

Transition point $T_c$

Critical behaviors
0. **Introduction**

I. **Spins are ordered**
   - Novel topological excitations and textures
   - Coupling to other degrees of freedom

II. **Spins are frozen**
   - Randomness
   - Spin glass

III. **Spins remain disordered**
   - Quantum fluctuations
   - Quantum spin liquid
I. Spins are ordered but often with some structures in the presence of frustration.

Appearance of chirality on macroscopic scale "spin composite"
Chirality-induced topological excitations in noncollinear magnets

I
Vortex formed by vector chirality
\[ S_i \times S_j \]
\[ \rightarrow \text{Z}_2 \text{ vortex} \]

II
Topologically nontrivial texture formed by scalar chirality
\[ S_i \cdot S_j \times S_k \]
\[ \rightarrow \text{skyrmion} \]
Topological excitation in triangular Heisenberg AF

* Novel type of vortex

--- $Z_2$ vortex

[H.Kawamura & S. Miyashita, ‘84]

No topological distinction between “$R$” & “$L$”
(no “circulation” in the usual sense)

$$V = SO(3) \rightarrow P_1(SO(3)) = Z_2$$
[two-valued topological quantum number]

Vortex formed by chirality vectors
Novel topological transition and dynamics

* Novel finite-temperature topological transition (or a crossover) occurring at $T=T_v$ within the paramagnetic state driven by the $Z_2$-vortex binding-unbinding.

[H. Kawamura and S. Miyashita]

* Novel dynamics borne by free $Z_2$ vortices, leading to “curtain-like” characteristic dispersion spectrum.

(To be examined experimentally)
Skyrmion and skyrmion lattice

Topologically stable spin texture where scalar chiralities wrap the sphere

Manganese oxide forms a lattice

Applying magnetic field

[S. Mori et al.]
Frustration-induced skyrmions

Skyrmions usually induced by antisymmetric Dzialoshinskii-Moriya interaction

Skyrmions induced by frustrated symmetric exchange interaction ever possible?

Yes!

→ Anti-skyrmion

Anti-particle of the skyrmion world

skyrmion chirality +

anti-skyrmion chirality −
Frustration induced skyrmion lattice

$J_1$-$J_3$ ($J_1$-$J_2$) triangular lattice

Heisenberg model in fields

$$\mathcal{H} = -J_1 \sum_{\langle i, j \rangle} S_i \cdot S_j - J_{2,3} \sum_{\langle\langle i, j \rangle\rangle} S_i \cdot S_j - H \sum_i S_{i,z},$$

incommensurate spin order appears

**triple-$q$**

$$\left( H/|J_3| = 2.0, T/|J_3| = 0.30 \right)$$

**Skyrmion**

$$\left( H/|J_3| = 2.0, T/|J_3| = 0.41 \right)$$

**Skyrmion**

Magnetic phase diagram

Energy minima in $q$-space

1st Brillouin zone
Coupled composite I: To lattice — gigantic magnetostriction

\[ \text{CdCr}_2\text{O}_4 \quad (\text{ZnCr}_2\text{O}_4, \text{HgCr}_2\text{O}_4) \]

Frustration resolved by lattice distortion

Formation of spin-lattice coupled composite

Spins order due to frustration resolution

Spinel compound: \text{CdCr}_2\text{O}_4

[H. Ueda et al., ‘05]
Coupled composite II: To conduction electrons — Scalar chirality and quantum Berry phase

Phase of conduction electron

Effective flux $\Phi$

$= \frac{1}{2} \times$ scalar chirality $\chi$

Scalar chirality

$\chi = S_1 \cdot S_2 \times S_3$

breaks time-reversal symmetry

Chirality serves as a fictitious magnetic field

Chirality-driven anomalous Hall effect
Novel chiral order probed by the anomalous Hall effect

Metallic pyrochlore \( \text{Pr}_2\text{Ir}_2\text{O}_7 \)

[Y.Machida, S.Nakatsuji et al. ‘10]

Time-reversal symmetry breaking not accompanying the magnetic order

Chiral spin liquid
Skyrmion exhibits chirality-driven anomalous Hall effect

Skyrmion exhibits chirality-driven anomalous Hall effect

Rich electromagnetic response exhibited by skyrmions and antiskyrmions
II. Spins are frozen

Randomness and spin glass (SG)

Canonical SG: CuMn, AuFe, etc.

* Competition between F and AF arising from RKKY interaction
* Alloying randomness

Frustration + Randomness

dilute magnetic alloy

Antiferromagnetic (AF)
Ferromagnetic (F)
What is the fate of the spin in SG?

No periodicity in the system

Does not order?

Slow down gradually toward $T=0$?

A sharp thermodynamic transition!

Experimental finding of the SG transition

[Canella and Mydosh, 1972] $T_{ac}$ susceptibility

$AuFe$

$dc$ susceptibility

[CuMn] $T$

[Nagata, Keesom, Harrison 1979]
In SG, spins are frozen in time, but in a spatially random manner breaks ergodicity. Exibits various intriguing phenomena. Typical test case of "complex" systems.
Canonical SG  CuMn, AuFe, etc.

Heisenberg system with weak random magnetic anisotropy

The true nature of experimental SG transition and SG ordered state is still at issue

Standard scenario assumes:
Weak magnetic anisotropy induces the crossover from the isotropic Heisenberg to the anisotropic Ising behavior.

→ tends to fail in several respects
  e.g., critical properties, phase diagram, absence of crossover etc.
Chirality scenario of SG transition

Chirality is a hidden order parameter of real SG transitions. Experimental SG transition is a “disguized” chiral-glass transition

[Part I]
Isotropic Heisenberg SG in 3D exhibits spin-chirality decoupling, with the chiral-glass ordered phase not accompanying the standard SG order.

[Part II]
The spin is recoupled to the chirality, i.e., mixed into the chirality via the random magnetic anisotropy.
Spin-chirality decoupling in the 3D isotropic Heisenberg SG

How the spin and chiral correlations grow?

\[ T_{CG} > T_{SG} \]

Spin-chirality decoupling
Large-scale Monte Carlo simulations on the SG model

Correlation length ratios $\xi/L$

Chiral critical properties

$\beta \sim 1$  $\gamma \sim 2$

$\nu \sim 1.4$  $\eta \sim 0.6$

[Viet and Kawamura, '09]
Chirality hypothesis

Isotropic (ideal) system

→ “spin-chirality decoupling”

Real system is weakly anisotropic!
Spin is “recoupled” to the chirality due to the weak random magnetic anisotropy $D$.

$Z_2 \times SO(3)$

The chiral-glass transition now appears as the SG transition.

“spin-chirality recoupling”

experimental SG exponents = CG exponents

[H. Kawamura ‘92]
### Critical properties of canonical SG

<table>
<thead>
<tr>
<th>Material</th>
<th>(\beta)</th>
<th>(\gamma)</th>
<th>(\nu)</th>
<th>(\eta)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CuMn &amp; AgMn [de Courtenary et al.]</td>
<td>1.0±0.1</td>
<td>2.2±0.1</td>
<td>≈ 1.4</td>
<td>≈ 0.4</td>
</tr>
<tr>
<td>AgMn [Bouchiat]</td>
<td>1.0±0.1</td>
<td>2.2±0.2</td>
<td>≈ 1.4</td>
<td>≈ 0.4</td>
</tr>
<tr>
<td>AgMn [Levy et al.]</td>
<td>0.9±0.2</td>
<td>2.1±0.1</td>
<td>≈ 1.3</td>
<td>≈ 0.4</td>
</tr>
<tr>
<td>CuAlMn [Simpsons]</td>
<td>≈ 1.0</td>
<td>≈ 1.9</td>
<td>≈ 1.3</td>
<td>≈ 0.5</td>
</tr>
<tr>
<td>PdMn [Coles and Williams]</td>
<td>0.9±0.15</td>
<td>2.0±0.2</td>
<td>≈ 1.3</td>
<td>≈ 0.4</td>
</tr>
<tr>
<td>AuFe [Taniguchi &amp; Miyako]</td>
<td>1.0±0.2</td>
<td>2.0±0.2</td>
<td>≈ 1.3</td>
<td>≈ 0.5</td>
</tr>
<tr>
<td>CdCr(<em>{2.08})In(</em>{2.08})S(_4) [Vincent et al.]</td>
<td>0.75±0.10</td>
<td>2.3±0.4</td>
<td>≈ 1.3</td>
<td>≈ 0.2</td>
</tr>
<tr>
<td>(\pm J) Campbell et al.</td>
<td>≈ 0.82</td>
<td>≈ 6.5</td>
<td>2.72±0.08</td>
<td>−0.40±0.04</td>
</tr>
<tr>
<td>(\pm J) Hasenbusch et al.</td>
<td>≈ 0.77</td>
<td>≈ 5.8</td>
<td>2.45±0.15</td>
<td>−0.375±0.010</td>
</tr>
<tr>
<td>Fe(<em>{0.5})Mn(</em>{0.5})Ti(_3) [Gunnarsson et al.]</td>
<td>≈ 0.54</td>
<td>4.0±0.3</td>
<td>≈ 1.7</td>
<td>≈ −0.35</td>
</tr>
</tbody>
</table>

**Chiral (Heisenberg)**

\(\beta\sim 1\)  \(\gamma\sim 2\)  \(\nu\sim 1.4\)  \(\eta\sim 0.6\)
Direct test of the chirality scenario

Chirality directly measured with anomalous Hall effect as a probe!

Chirality exhibits an eminent singularity!

[T.Taniguchi et al ('04)]

[T.Taniguchi et al ('06)]
III. Spins remain disordered

Novel liquid-like quantum spin state without magnetic long-range order?

Quantum spin liquid (QSL)
Quantum fluctuations
antiferromagnetically coupled two spins

* classical spin

* $S = 1/2$ quantum spin

$(1/\sqrt{2})(|\uparrow \downarrow\rangle - |\downarrow \uparrow\rangle)$

$S=0$ non-magnetic state (energy gap to triplet)

RVB state

[P.W. Anderson ('73)]

Realizable in frustrated systems?

Long quest ...
Ground state of the simplest nearest-neighbor bilinear Heisenberg model

Theory suggests

Triangular lattice

AF magnetic LRO at $T=0$
even for $S=1/2$

Kagome lattice

Liquid-like ground state without magnetic
long-range order nor the spin freezing
Experimental discovery of the “quantum spin liquid” state

Quantum spin liquid states observed in certain $S=1/2$ frustrated AFs

**Triangular lattice**

- $\kappa$-(ET)$_2$Cu$_2$(CN)$_3$
- EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

$S=1/2$ organic salts
Mott insulator


**Kagome lattice**

herbersmithite: ZnCu$_3$(OH)$_6$Cl$_2$

[D.G. Nocera et al]

What is its nature?

→ still remains controversial
$S = \frac{1}{2}$ organic triangular AF

$\kappa-(ET)_2Cu_2(CN)_3$

NMR spectrum
[Y. Shimizu, K. Kanoda et al '03]

Specific heat
[S. Yamashita et al '08]

Gapless spin liquid behavior!

EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

NMR spectrum
Role of possible randomness?

Randomness in the interaction might induce “Anderson-localization” of singlets.

“RVB state”

In spin and molecular glasses, the $T$–linear low-$T$ specific heat prevails, reflecting its local low-energy collective excitations.
Randomness really exists? → Likely!

Charge inhomogeneity in triangular organic salts
QSL in the vicinity of charge (ferroelectric) order

Domain growth associated with the CO QCP hindered
→ CO (dipole-glass) state?

Critical slowing down associated with a CO QCP?
Randomness in the anion layer plays a role of “seed”?
Strong coupling between the spin and the charge (polarization)

\[ \kappa -(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3 \]

Intradimer charge imbalance?

Random freezing of electric polarization at low \( T \)

\[ \text{AC dielectric constant} \]

[Exp. \( \kappa -\text{ET} \)]

Effective randomness in the exchange interaction between spins

Bond-random $S=1/2$ AF Heisenberg model on the triangular lattice

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_{ij} \hat{S}_i \cdot \hat{S}_j \quad (0 \leq J(1-\Delta) \leq J_{ij} \leq J(1+\Delta))$$

$\Delta$: randomness parameter ($0 \leq \Delta \leq 1$)

- $\Delta = 0$: no randomness
- $\Delta = 1$: maximal randomness

Exact numerical calculations on finite spin clusters of $N<32$

[K. Watanabe, H. Kawamura et al., '14]

Transition from the AF state to the nonmagnetic (random-singlet) state at $\Delta \sim 0.6$. 
Computed physical quantities

Specific heat

- $T$ - Linear

$C = \gamma T + cT^3$

$\gamma$ term: calc. $\sim 20 \text{ mJK}^{-2}$
exp. $\kappa \cdot ET \sim 12, \text{ dmit } \sim 20 \text{ mJK}^{-2}$

[S. Yamashita et al '08]

NMR relaxation rate

$T^{-1}$

[S. Itou et al '08, '10]
What about kagome?

Randomness due to $\text{Zn}^{2+}$ replacement by $\text{Cu}^{2+}$

A phase transition at $\triangle \sim 0.4$ between the randomness-irrelevant non-magnetic state and the randomness-relevant random-singlet state.
Computed physical quantities

Magnetization curve

\[ M(T) \]

[Exp.] Magnetization curve

\[ M(H) \]

[T. Shimokawa et al., '15]

Dynamical structure factor \( \Delta = 1 \)

[Exp.] Dynamical structure factor

\[ S(q, \omega) \]

[H. Kawamura et al., '14]

Neutron inelastic

[Exp.] Neutron inelastic

\[ S_{mag}(Q, \omega) \]

[T. H. Han, et al., '12]

\[ H \]

[T]
Frustration + Quantum fluctuations + Randomness

= Random singlet state

QSL-like gapless non-magnetic state
Summary

Frustration, together with randomness and quantum fluctuations, gives rise to a variety of novel order, new dynamics and exotic thermodynamic states, developing new states of matter, new concepts, and potential new applications.