Topological and Multipolar Magnets and Spintronics

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Plan

- Multipole Physics on Correlated Electron Systems
- Topological States in Magnetic Systems
- Physics of Antiferromagnetic Weyl Semimetals
- Physics of Multipolar Kondo Lattice Systems
Lecture 4

- Multipole Physics on Correlated Electron Systems
- Topological States in Magnetic Systems
- Physics of Magnetic Weyl Semimetals
- Physics of Multipolar Kondo Lattice Systems
A pair of Weyl points

To satisfy the Gauss’s theorem,

\[ C = \begin{cases} 1 & (-k_0 < k_Z < k_0) \\ 0 & (k_Z < -k_0, k_0 < k_Z) \end{cases} \]

\( k_x - k_y \) plane at \(-k_0 < k_Z < k_0\) can be regarded as the quantum Hall system.

- Hall conductivity

\[ \sigma_{xy} = -\frac{e^2}{(2\pi)^2\hbar} \int_{-k_0}^{k_0} 1 \, dk_z = -\frac{e^2}{(2\pi)^2\hbar} (2k_0) \]
Weyl semimetals with large fictitious field in the $k$-space

Inversion or Time-reversal symmetry breaking

Berry curvature $\Omega(k)$

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Weyl magnets
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Magnetic structure allows to control the distribution of Weyl points

- Large transverse response derived from $\Omega(k)$

Weyl semimetals with large fictitious field in the $k$-space

Inversion or Time-reversal symmetry breaking

Dirac

Weyl

Berry curvature $\Omega(k)$

Topological magnets

Ferromagnets (FMs)

Antiferromagnets (AFMs)

Weyl Magnets: Functional Magnets

Anomalous Hall Effect

Anomalous Nernst Effect

Non-volatile Memory

Thermoelectric Conversion

Large responses are obtained irrespective of size of $M$. 

Enhancement of ANE using topological band structures

\[ S_{\text{ANE}} = \rho \left( -S_{\text{SE}} \sigma_{yx} + \alpha_{yx} \right) \]

Hall conductivity
\[ \sigma_{\text{int}}^{yx} = e \int_{k} \left( e^2 / h \right) \left( 2\pi \right)^{-3} \sum_{n} \Omega_{n,z}(k) f(\epsilon_{n,k}) d\epsilon \]

Transverse TE conductivity
\[ \alpha_{yx} = \frac{k_B}{e} \int_{k} \epsilon_{xyz} \sum_{n,k} \left( \Omega_{n,z}(k) \delta(\epsilon - \epsilon_{n,k}) \right) s(\epsilon, T) d\epsilon \]

Berry curvature
\[ \Omega_{n,z}(k) = -2\text{Im} \sum_{m \neq n} v_{nm,x}(k) v_{mn,y}(k) / \left( \epsilon_m(k) - \epsilon_n(k) \right)^2 \]

Weyl AFMs

Weyl FMs
- UCo$_{0.8}$Ru$_{0.2}$Al: Asaba et al., Sci. Adv. 7, eabf1467 (2021).

Nodal-web/-plane FMs

~10 times larger $S_{\text{ANE}}$ than that of conventional FMs
Enhancement of ANE using topological band structures

\[ S_{\text{ANE}} = \rho \left( -S_{\text{SE}} \sigma_{yx} + \alpha_{yx} \right) \]

Hall conductivity

\[ \sigma_{\text{int}}^{yx} = \epsilon_{\text{xyz}} \left( \frac{e^2}{\hbar} \right) \int (2\pi)^{-3} \sum_n \Omega_{n,z}(k) f(\epsilon_{n,k}) \, dk \]

Transverse TE conductivity

\[ \alpha_{yx} = \frac{k_B}{e} \int_{\epsilon} \epsilon_{\text{xyz}} \sum_{n,k} \{ \Omega_{n,z}(k) \delta(\epsilon - \epsilon_{n,k}) \} \, s(\epsilon, T) \, d\epsilon \]

Berry curvature

\[ \Omega_{n,z}(k) = -2\text{Im} \sum_{m \neq n} v_{nm,x}(k)v_{mn,y}(k) \left( \epsilon_m(k) - \epsilon_n(k) \right)^2 \]

Weyl AFMs


Weyl FMs

UCO0.8Ru0.2Al: Asaba et al., Sci. Adv. 7, eabf1467 (2021).

Nodal-web/-plane FMs


~10-100 times larger \( S_{\text{ANE}} \) than that of conventional FM

\[ \alpha \approx \frac{1}{\hbar} \text{Im} \left[ \int \epsilon_{\text{xyz}} \sum_{n,k} \{ \Omega_{n,z}(k) \delta(\epsilon - \epsilon_{n,k}) \} \right] \]

\[ \sigma_{\text{int}}^{yx} = \epsilon_{\text{xyz}} \left( \frac{e^2}{\hbar} \right) \int (2\pi)^{-3} \sum_n \Omega_{n,z}(k) f(\epsilon_{n,k}) \, dk \]

\[ \alpha_{yx} = \frac{k_B}{e} \int_{\epsilon} \epsilon_{\text{xyz}} \sum_{n,k} \{ \Omega_{n,z}(k) \delta(\epsilon - \epsilon_{n,k}) \} \, s(\epsilon, T) \, d\epsilon \]

\[ \Omega_{n,z}(k) = -2\text{Im} \sum_{m \neq n} v_{nm,x}(k)v_{mn,y}(k) \left( \epsilon_m(k) - \epsilon_n(k) \right)^2 \]

Topological (Weyl) AFM Mn₃Sn

Mn₃Sn : Chiral antiferromagnetic order \((T_N \sim 430 \text{ K})\)

Order parameter :
Cluster magnetic octupole
Suzuki et al., PRB 95, 094406 (2017).

[Berry curvature]
\(\Omega(k)\)

[Real space]
Magnetic structure

[01\bar{1}0]
[\bar{2}110]
[0001]

Weyl points

Antiferromagnets exhibiting large transverse responses

Anomalous Hall effect


[Real space]
Magnetic structure

Large transverse responses of Weyl AFM Mn$_3$Sn

*Anomalous Nernst effect*

*Anomalous Hall effect*

*Magneto-optical Kerr effect*


**M independent ANE of Weyl AFM Mn$_3$Sn**

\[ S_{(A)NE} = Q_0 B + Q_S \mu_0 M + S_{ANE} \]

\[ S_{ANE} = \rho \left( \alpha_{yx} - S_{SE} \sigma_{yx} \right) \sim -0.5 \mu V/K \]

\[ \rho S_{SE} \sigma_{zx} \sim -0.1 \mu V/K \]

Large spontaneous ANE at room temperature

\[ S_{ANE} = Q_0 B + Q_S \mu_0 M + S_{ANE} \]

\[ S_{ANE} \sim 0.005 \mu_B \]

\[ S_{ANE} \sim 0.002 \mu V/K \]

\[ M \text{ independent ANE} \propto \Omega(k) \sim 100 T \]

\[ \Omega(k) \text{ around } E_F \]

\[ \Omega(k) \text{ below } E_F \]

\[ \equiv \text{ FM metals} \]

ANE induced by large $\Omega(k)$ from topological band structures

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**Ikhlas, Tomita et al., Nat. Phys. 13, 1085 (2017).**
Topological (Weyl) ferromagnet \( \text{Co}_2\text{MnGa} \)

\[ \delta_{\mathrm{yr}} = \rho_{\mathrm{xx}y} (\alpha_{\mathrm{yx}} - S_{\mathrm{yy}y} \sigma_{\mathrm{yx}}) \]

around \( E_F \) below \( E_F \)

Largest ANE @ \( T \geq RT \) (6 \( \mu \)V/K @ RT, 8 \( \mu \)V/K @ 400 K)

\[ \delta_{\mathrm{yr}} = \rho_{\mathrm{xx}y} (\alpha_{\mathrm{yx}} - S_{\mathrm{yy}y} \sigma_{\mathrm{yx}}) \]

around \( E_F \) below \( E_F \)
Topological band structure of Co$_2$MnGa

Weyl points

Lifshitz transition


Figs. Courtesy H. Nakamura

Type-I Weyl

Critical point

Type-II Weyl

Diminished DOS

Large DOS (Flat band) Lifshitz transition

Finite DOS

< 20K
\[ \alpha_{yx} \approx -\frac{\pi^2 k_B^2 T}{3} |e| \frac{\partial \sigma_{yx}}{\partial E_F} \]
\[ \Rightarrow \frac{\alpha_{yx}}{T} = \text{Constant (Mott relation)} \]

\[ \alpha_{yx} \approx \frac{\alpha_{yx, \text{max}}}{T_0} \log \left( \frac{|\mu - E_0|}{k_B T_0} \right) \]
\[ \Rightarrow \frac{\alpha_{yx}}{T} = -\log T \]

suggests quantum Lifshitz transition

Large $\Omega(k)$ at Weyl points & DOS due to quantum Lifshitz transition

\documentclass{article}
\usepackage{amsmath}
\begin{document}
\begin{equation}
\alpha_{yx} \approx -\frac{\pi^2 k_B^2 T}{3} |e| \frac{\partial \sigma_{yx}}{\partial E_F}
\end{equation}
\end{document}
Nodal-web ferromagnet $D0_3$-$Fe_3X$ ($X = Ga, Al$)

$D0_3$-$Fe_3X$ ($X = Ga, Al$)

Calc. for $\sim$1300 samples using MI

<table>
<thead>
<tr>
<th>Formula</th>
<th>Space group</th>
<th>$\sigma_{\text{max}} (A K^{-1} m^{-1})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe$_3$Pt</td>
<td>$Pm3m$</td>
<td>6.2</td>
</tr>
<tr>
<td>Fe$_3$Ga</td>
<td>$Pm3m$</td>
<td>3.0</td>
</tr>
<tr>
<td>Fe$_3$Al</td>
<td>$Pm3m$</td>
<td>2.7</td>
</tr>
</tbody>
</table>

$\bullet$ Giant ANE comparable to Co$_2$MnGa ($S_{\text{ANE}} \sim 5.5 \mu V/K$ @ RT)

$\bullet$ Binary systems consisting of safe & inexpensive elements

[Bulk & Film ($D0_3$)] Sakai†, …, TH† et al., Nature 581, 53 (2020).

S. Minami et al., PRB 102, 205128 (2020).

Zhou, Sakuraba, APEX 13, 043001 (2020).
Heat flux sensor

Heat flux $q = \kappa(T_1 - T_2)/L$

Sensitivity (1 x 1 cm²)

- 10 μV/(W/m²) (100 mV/W)

Visualizing the heat flow

- Heat dissipation/reception around an engine
- Abnormal heat generation in electronics
- Thermal conductivity (insulation)
- Health Care (deep body temperature)
Flexible heat flow sensor using thin-film fabrication

**Price:**

- SE $500 → ANE $1-10

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**Sensitivity**

- **ANE-type heat flux sensor**
  - Heat Flux
  - 2D

**Flexible, Simple, Large area**

**Sensitivity**

- 0.001 - 0.01 mV/W · m⁻²

<table>
<thead>
<tr>
<th>Material</th>
<th>Sensitivity (μV/K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mn₃Sn</td>
<td>0.35</td>
</tr>
<tr>
<td>Fe₃Ga</td>
<td>~6</td>
</tr>
</tbody>
</table>

**Seebeck effect**

- 3D

**Sensitivity**

- 0.01 mV/W · m⁻²

**Denso HP**

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**E.g., Zhou & Sakuraba, APEX 13, 043001 (2020); TH et al., Adv. Funct. Mater. 31, 2008971 (2021)...**

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**Flexible heat flow sensor using thin-film fabrication**

**Price:**

- SE $500 → ANE $1-10
Flexible, Simple, Large area

**Sensitivity**

0.001 - 0.01 mV/W \cdot m^{-2}

- \text{Mn}_3\text{Sn} : 0.35 \mu V/K
- \text{Fe}_3\text{Ga} : \sim 6 \mu V/K

**Problem:** Shape anisotropy

- No ANE
- Finite ANE

**Magnetic hardness parameter** $\kappa$

$$\kappa = \left(\frac{K}{\mu_0 M^2}\right)^{1/2}$$

Large $\kappa \Rightarrow$ ideal arrangement

- e.g., Zhou & Sakuraba, APEX 13, 043001 (2020); TH et al., Adv. Funct. Mater. 31, 2008971 (2021)…

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**Flexible heat flow sensor using thin-film fabrication**

**Price:** SE $500 \rightarrow$ ANE $1-10$
Collaboration work with Nitto Denko Corp.

Roll to Roll fabrication

PET film

Fe₃Ga target

Material for electrode

100°C

Sheet Sample

Lithography-1

Lithography-2

Su-8 coating

Sensor image

Direct sensing of perpendicular heat flux

\[ V_y^{SE} + V_{ANE} \]

[Diagram showing the direct sensing of perpendicular heat flux]

\[ S_{SE}^{Magnet} = S_{SE}^{Electrode} \]

[Schematic representation of the electrode and magnet setup]

No offset

large offset


H. Tanaka Y. Nakanishi H. Machinaga

Mass producible flexible sensor for perpendicular heat flux sensing
Plan

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Multipolar phenomena in Ce$^{3+}$-based systems

La-doped CeB$_6$ : $B$-$T$ phase diagram featuring dipolar, quadrupolar, and octupolar orders

Ce$_3$Pd$_{20}$Si$_6$ : Two electron localization transitions driven by dipolar and quadrupolar d.o.f.

The multipolar orders are “hidden” under the dipolar (spin) order.

Let’s design a new material platform to explore pure multipolar phenomena.

D. Jang et al., npj Quantum Mater (2017)
V. Martellia et al., PNAS (2019)
Cubic Pr$^{3+}$ systems: Ideal platform for multipolar physics

**4f Kramers doublet (e.g., Ce$^{3+}$, Yb$^{3+}$)**
- Odd number of f electrons
- Half-integer $J$
- Kramer’s theory: double degeneracy protected by time-reversal symmetry

**Ce$^{3+}$ ($4f^1$) $J = 7/2$**
- SOC + Cubic CEF
- $\Gamma_8 \rightarrow \Gamma_7$
- Always carry magnetic dipoles
- Degeneracy is robust against structural disorder

**4f non-Kramers doublet (e.g., Pr$^{3+}$)**
- Even number of f electrons
- Integer $J$
- Double degeneracy is not protected by time-reversal symmetry but by the local symmetry

**Pr$^{3+}$ ($4f^2$) $J = 5$**
- SOC + Cubic CEF
- $\Gamma_1 \rightarrow \Gamma_5 \rightarrow \Gamma_4 \rightarrow \Gamma_3$
- No magnetic dipoles but high-rank multipoles
- Degeneracy can be lifted by structural disorder
Cubic Pr$^{3+}$ systems: Ideal platform for multipolar physics

**Pr (TM)$_2$Al$_{20}$**

Frank-Kasper cages of 16 Al surrounding the Pr ion → strong c-f hybridization

**CEF scheme of Pr$^{3+}$ in local cubic environment**

Pr$^{3+}$ $4f^2$

J=4

SOC

CEF with a point group symmetry $T_d$

$|\Gamma_3^+\rangle = \frac{1}{2} \sqrt{7} (|+4\rangle + |-4\rangle) - \frac{1}{2} \sqrt{5} |0\rangle$

$|\Gamma_3^-\rangle = \frac{1}{2} \sqrt{5} (-|+2\rangle + |-2\rangle) + \frac{1}{2} \sqrt{3} |0\rangle$

Well-isolated non-Kramers doublet ground state

Pr$\text{V}_2\text{Al}_{20}$ ($T_d$) $\Gamma_1$ 156 K

Pr$\text{T}i_2\text{Al}_{20}$ ($T_d$) $\Gamma_5$ 107 K

$\Gamma_4 = 65$ K

$\Gamma_5 \approx 40$ K

$\Delta$
Cubic Pr$^{3+}$ systems: Ideal platform for multipolar physics

Frank-Kasper cages of 16 Al surrounding the Pr ion → strong c-f hybridization

Pr$^3+$ in local cubic environment

CEF scheme with point group symmetry $T_d$

Pr$V_2Al_{20}$

Pr$Ti_2Al_{20}$

Non-magnetic!
How do multipoles modify quantum phenomena?

**Magnetic Kondo effect**

Single-channel Kondo model ($k = 1$) and exact screening

$f$ electrons become itinerant and enter the Fermi surface in the heavy-fermion Fermi liquid (FL) ground state

$$\rho \sim AT^2$$

$$C/T \sim \frac{m^*}{m_0} \gamma_0$$

**Quadrupolar Kondo effect**

Two-channel Kondo model ($k = 2$) and over-screening

Residual entropy $S_0 = \frac{1}{2} R \ln 2$ leads to a non-Fermi liquid (NFL) ground state

$$\rho \sim T^{1/2}, \ C/T \sim - \ln T,$$

$$\chi \sim T^{1/2} \ \text{or} \sim - \ln T$$

The multipolar Kondo effect represents an alternative route to novel NFLs, distinct from quantum criticality. The NFL is intrinsic to the multipolar Kondo interaction and thus does not require fine-tuning of parameters.

\[ \rho \sim AT^2, \quad C/T \sim \frac{m^*}{m_0} \gamma_0 \]

\[ \chi \sim T^{1/2} \text{ or } -\ln T \]
Multipolar RKKY vs. Multipolar Kondo effect?

Modulated AFQ order in PrPb$_3$

Single-site multipolar Kondo effect in Y$_{1-x}$Pr$_x$Ir$_2$Zn$_{20}$

T. Ominaru et al., PRL94, 197201 (2005)

Y. Yamane et al., PRL121, 077206 (2018)
Multipolar RKKY vs. Multipolar Kondo effect?

Modulated AFQ order in PrPb₃

Can we find a way to **tune** the competition between the multipolar Kondo effect and RKKY-type multipolar interaction in a lattice system?

Single-site multipolar Kondo effect in Y₁₋ₓPrₓIr₂Zn₂₀

PrIr₂Zn₂₀ (x = 1)

AFQ order

Dilute

Y₁₋ₓPrₓIr₂Zn₂₀

Non-Fermi liquid

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T. Ominaru *et al.*, PRL 94, 197201 (2005)

Y. Yamane *et al.*, PRL 121, 077206 (2018)
Unifying themes of strongly correlated matters

High-$T_c$ cuprate

Universal properties among various material classes?

Fe-based SC

Organic SC

MATBG

Quantum critical non-Fermi-liquid (NFL)

Ordered state (intertwined d.o.f)

Disordered state

QCP
Unifying themes of strongly correlated matters

High-\(T_c\) cuprate

Universal properties among various material classes?

Fe-based SC

Organic SC

MATBG

Quantum critical non-Fermi-liquid (NFL)

Magnetic quantum criticality has been extensively studied, but quantum criticality driven purely by orbital fluctuations is unexplored.
How do multipoles modify quantum phenomena?

Tuning a multipolar Kondo system to a QCP

Will the resultant phase diagram different from the Doniach phase diagram?

Novel quantum critical phenomena and superconductivity?
Pr(Ti, V)\textsubscript{2}Al\textsubscript{20}: Multipolar order, NFL, and quantum criticality

**Long-range multipolar order:**
- PrTi\textsubscript{2}Al\textsubscript{20}: Ferroquadrupolar (FQ) order at $T_Q \sim 2$K
- PrV\textsubscript{2}Al\textsubscript{20}: Two-stage transitions at $T_Q \sim 0.75$K (AFQ) and $T^* \sim 0.65$K (octupolar order?)

A. Sakai and S. Nakatsuji, JPSJ 80, 063701 (2011)
Pr(Ti, V)$_2$Al$_{20}$: Multipolar order, NFL, and quantum criticality

Long-range multipolar order:
- PrTi$_2$Al$_{20}$: Ferroquadrupolar (FQ) order at $T_Q \sim 2K$
- PrV$_2$Al$_{20}$: Two-stage transitions at $T_Q \sim 0.75K$ (AFQ) and $T^* \sim 0.65K$ (octupolar order?)

A. Sakai and S. Nakatsuji, JPSJ 80, 063701 (2011)

Heavy fermion superconductivity:
large $\gamma$ and $dB_{c2}/dT|_{T=T_c}$ $m^*/m_0 \sim 20$, (Ti), 150 (V).

M. Tsujimoto et al., PRL 113, 267001 (2014)
Pr(Ti, V)$_2$Al$_{20}$: Multipolar order, NFL, and quantum criticality

Kondo resonant peak in PrTi$_2$Al$_{20}$ → substantial c-f hybridization

M. Matsunami et al., PRB 84, 193101 (2011)

Tuning chemical pressure: Volume PrTi$_2$Al$_{20}$ > PrV$_2$Al$_{20}$

NFL behavior $\rho \sim \sqrt{T}$ in PrV$_2$Al$_{20}$ due to stronger hybridization

M. Tsujimoto et al., PRL 113, 267001 (2014)

A. Sakai and S. Nakatsuji, JPSJ 80, 063701 (2011)
The $-\ln T$ behavior driven by the magnetic Kondo effect increases in magnitude as $c$-$f$ hybridization enhances under pressure.

Resistivity becomes incoherent near $P_c \sim 11$ Gpa.
Pr(Ti, V)$_2$Al$_{20}$: Multipolar order, NFL, and quantum criticality

- Pronounced enhancement of $T_c$ and effective mass $m^*$ on approaching $P_c \sim 11$ GPa;
- Two SC domes extending to 16 GPa
- Robust NFL behavior covering a wide parameter range; FL phase does not recover under high pressures
Topological and Multipolar Magnets and Spintronics

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