Title: Anomalous transport property in the nodal metallic spin ice Pr2Ir2O7

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Abstract: Pyrochlore Pr2Ir2O7 is a rare material with various unique properties such as geometrical frustration, c-f hybridization and Fermi node in the band structure. Although Pr3+ carries the effective moment of ~3μB with Curie-Weiss temperature θ ± ~ θC-20 K, no long-range order is observed down to the partial freezing at Tf ~ 0.3 K, suggesting the geometrical frustration [1]. Magnetic Grüneisen ratio diverges γmag ~ T-3/2 without tuning any parameter, indicating the zero-field quantum criticality [2]. Besides, recent angle-resolved photoemission spectroscopy (ARPES) measurement reveals the Fermi node at Γ point in Pr2Ir2O7, which can be an origin of the various topological phases such as topological insulator and Weyl semimetal [3]. One of the most interesting and striking properties of Pr2Ir2O7 is non-trivial anomalous Hall effect: spontaneous Hall effect appears even in the absence of any spin freezing, which is attributed to the chiral spin liquid state [4]. In this presentation, we will discuss the recent results for the anomalous Hall effect for various samples of Pr2Ir2O7.
Anomalous transport property in the nodal metallic spin ice Pr$_2$Ir$_2$O$_7$

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Pr-based pyrochlore oxides $\text{Pr}_2T_2\text{O}_7 (T = \text{Ir, Zr})$

Geometrical frustration on pyrochlore lattice $\Rightarrow$ Nontrivial ground state.

$$H = \sum_{<i,j>} J_{\parallel} S_i^z S_j^z,$$

(i) Ferro $J_{\parallel} > 0$

Spin ice state (2-in-2-out)

macroscopic degeneracy $\Rightarrow$ Residual entropy $\frac{1}{2} R \ln \frac{3}{2}$

$\text{Dy}_2\text{Ti}_2\text{O}_7$

$\text{Ho}_2\text{Ti}_2\text{O}_7$ $\mu_{\text{eff}} \sim 10 \mu_\text{B}$

$\text{Pr}_2\text{Ir}_2\text{O}_7$ $\mu_{\text{eff}} \sim 2.5 \mu_\text{B}$

$\text{Pr}_2\text{Zr}_2\text{O}_7$
Quadratic Fermi-node at the $\Gamma$ point in Pr$_2$Ir$_2$O$_7$


$\mathcal{W} \sim 40$ meV
Quadratic band touching: Luttinger liquid

Quadratic band touching at the $\Gamma$ point

$H = \frac{k^2}{2M_0} + \frac{\left(\frac{5}{4k^2} - (k \cdot J)^2\right)}{2m} - \sum_{i=x,y,z} \frac{k_i^2 J_i^2}{2M_c}$

- Non-Fermi liquid due to the strong interaction.
- Touching points are not topologically protected.
- Parent states to a number of topological phases.
- Dielectric constant can be greatly enhanced.

Anomalous Hall effect in Pr$_2$Ir$_2$O$_7$

- spontaneous AHE at $B = M = 0$
- Anisotropic $\rho_{xy}$ and $\rho_{xx}$:
  1. No spontaneous Hall in 3-in-1-out state.
  2. Large MR and SdH oscillation only in 3-in-1-out state.

Zero field quantum criticality in Pr$_2$Ir$_2$O$_7$

Magnetic Grüneisen ratio $\Rightarrow$ divergence at field tuned QCP

$$\Gamma_H = -\frac{(\partial M/\partial T)_H}{C} = -\frac{1}{T} \frac{(\partial S/\partial H)_T}{(\partial S/\partial T)_H} = \frac{1}{T} \left( \frac{\partial T}{\partial H} \right)_S$$

$= \text{magnetocaloric effect}$

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= magnetocaloric effect


- Diverging $\Gamma_H$ @ $H \to 0$ down to 0.4 K as $\Gamma_H \propto HT^{-3/2}$
- Scaling behavior in $T/H^{4/3}$ without critical field. $\rightarrow$ zero field quantum critical point
**Spontaneous Hall effect in thin film**

- Pr$_2$Ir$_2$O$_7$ thin film
- YSZ(111) substrate

Lattice mismatch 1.15% $\rightarrow$ tensile strain along [111] 2.31%

■ (111)-oriented epitaxial thin film of Pr$_2$Ir$_2$O$_7$ on YSZ (Y-ZrO$_2$) substrate.

■ Spontaneous $\rho_H$ at $T < 50$ K without $M$.

$\Rightarrow$ Ir-5$d$ electrons breaks the TRS? (Too high for Pr moments.)

**Quadratic band touching** $\Rightarrow$ **Weyl semimetal**

Macroscopic TRS breaking ($\Rightarrow$ all-in-all-out Ir) Cubic symmetry breaking ($\Rightarrow$ strain along [111])
Weyl metallic phases in $(\text{Pr}_{0.5}\text{Nd}_{0.5})_2\text{Ir}_2\text{O}_7$


Anisotropic and nonmonotonic $\sigma_{xx}$ and $\sigma_{xy}$ due to the magnetic configuration.
Proximity to the QCP of Ising order

D. E. MacLaughlin et al., PRB 92, 054432 (2015).

Off-stoichiometric poly crystal shows sharp peak in $C$ at $T = 0.9$ K.

2-in 2-out LRO in polycrystal Pr$_2$Ir$_2$O$_7$. 
Proximity to the QCP of Ising order

D. E. MacLaughlin et al., PRB 92, 054432 (2015).

Gan Chen PRB 94, 205107 (2016).

Off-stoichiometric poly crystal shows sharp peak in $C$ at $T = 0.9$ K.

2-in 2-out LRO in polycrystal $\text{Pr}_2\text{Ir}_2\text{O}_7$.

Possible quantum critical point in spin ice.
Open question

1. Quantum spin liquid
2. QCP of the 2-in 2-out long range order
3. Luttinger liquid (quadratic band touching + strong correlation.)

Anomalous behaviors ($\sigma_H$, zero-field QCP etc.)
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2. QCP of the 2-in 2-out long range order
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Anomalous behaviors ($\sigma_H$, zero-field QCP etc.)

How can we understand the anomalous behaviors in $\text{Pr}_2\text{Ir}_2\text{O}_7$?
Open question

1. Quantum spin liquid
2. QCP of the 2-in 2-out long range order
3. Luttinger liquid (quadratic band touching + strong correlation.)

Anomalous behaviors ($\sigma_H$, zero-field QCP etc.)

How can we understand the anomalous behaviors in Pr$_2$Ir$_2$O$_7$?

✓ Magnetocaloric effect measurement of insulating Pr$_2$Zr$_2$O$_7$
✓ Hall effect measurement of Pr$_{2+x}$Ir$_{2-x}$O$_{7+\delta}$
Magnetic Grüneisen ratio of Pr$_2$Zr$_2$O$_7$
Quality check

Non-Kramers Pr-based compounds

⇒ Sensitive to the sample quality

Sharp metamagnetic transition is one of the sign for the good quality.

K. Kimura et al., preprint
Quality check

Non-Kramers Pr-based compounds
➜ Sensitive to the sample quality

Sharp metamagnetic transition is one of the sign for the good quality.

Larger anomaly due to the metamagnetic transition at ~ 2.3 T in #2.
➜ Better quality for #2
$T$ dependence of $\Gamma_{\text{mag}}/B$

sample #1 (#H315s 1.38 mg)

sample #2 (#NO3B 1.26 mg)

$\Gamma_{\text{mag}}$ diverges at low field as $\Gamma_{\text{mag}} \sim T^{-3/2}$ above $T_m \sim 0.4$ K similar to Pr$_2$Ir$_2$O$_7$. 
The same scaling relation as Pr$_2$Ir$_2$O$_7$ holds in both samples.

Frustrated Pr moments are the origin of the diverging magnetic Grüneisen ratio.
Hall effect of Pr_{2+x}Ir_{2-x}O_{7+\delta}
Quality check

Single crystal growth

1. \((1/3)\text{Pr}_6\text{O}_{11} + 2\text{IrO}_2 \rightarrow \text{Pr}_2\text{Ir}_2\text{O}_7(\text{poly}) + (2/3)\text{O}_2\)

2. \(\text{Pr}_2\text{Ir}_2\text{O}_7(\text{poly}) : \text{KF flux} = 1 : 200 \rightarrow \text{Pr}_2\text{Ir}_2\text{O}_7(\text{single})\)

Resistivity

<table>
<thead>
<tr>
<th>#</th>
<th>(\alpha) (Å)</th>
<th>Pr : Ir (EDX)</th>
<th>(\rho_{0.1K}/\rho_{\text{min}})</th>
<th>(T_{\text{min}}) (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>10.39</td>
<td>2 : 1.96</td>
<td>1.06</td>
<td>45</td>
</tr>
<tr>
<td>B</td>
<td>10.409</td>
<td>2 : 1.88</td>
<td>1.03</td>
<td>10</td>
</tr>
<tr>
<td>Poly</td>
<td>10.672*</td>
<td>2 : 1.29*</td>
<td>1.03</td>
<td>15</td>
</tr>
</tbody>
</table>

* D. E. MacLaughlin et al., PRB 92, 054432 (2015).

quality

\(\#A > \#B > \text{poly}\)
Specific heat

- Sharp transition at 0.9 K in poly crystal
  consistent with the q = (100)
  order observed in neutron scattering.
- Long-range order still remains in #B.
- Nuclear contribution systematically changes.

4f moments are more dynamic in better sample.
Sharper upturn due to the metamagnetic transition at $B \sim 2.5$ T and larger MR for the better sample.
Anomalous Hall effect in $\Pr_{2+x} \text{Ir}_{2-x} \text{O}_{7+\delta}$

$\Pr_2\text{Ir}_2\text{O}_7$
$T = 30$ mK

$B \parallel [111]$
$I \parallel [110]$

Large spontaneous AHE disappears in the bad sample.
Anomalous Hall effect in Pr$_{2+x}$Ir$_{2-x}$O$_{7+\delta}$

Pr$_2$Ir$_2$O$_7$
$T = 30$ mK

$\sigma_H$ (Ω$^{-1}$ cm$^{-1}$)

$B \parallel [111]$
$I \parallel [110]$

Peak in $\sigma_H$ ➔ crossover from spin ice to Kagome ice.
Peak in $\sigma_H$ becomes more prominent for the good crystals with the small damping rate $1/\tau$.

$\tau$: lifetime of electrons

Large spontaneous AHE disappears in the bad sample.
Anomalous Hall effect in $\text{Pr}_{2+x}\text{Ir}_{2-x}\text{O}_{7+\delta}$

**Spin-ice** | **Kagome-ice** | **3-in 1-out (1-in 3-out)**

$\text{Pr}_2\text{Ir}_2\text{O}_7$
$T = 30\;\text{mK}$

$B \parallel [111]$
$I \parallel [110]$  

Peak in $\sigma_H$ crossover from spin ice to Kagome ice. Peak in $\sigma_H$ becomes more prominent for the good crystals with the small damping rate $1/\tau$.

$\tau$: lifetime of electrons


Large spontaneous AHE disappears in the bad sample.
Spontaneous AHE

- The spontaneous AHE without $M$ at $T < 1.7$ K disappears in #B.

- Spontaneous $\rho_H$ below $T_f$, $\sim 0.4$ K with $M$ is observed in both samples.

$\Rightarrow$ AHE due to the partial spin freezing.
Spontaneous AHE

- The spontaneous AHE without $M$ at $T < 1.7$ K disappears in #B.

- Spontaneous $\rho_H$ below $T_f$, $\sim 0.4$ K with $M$ is observed in both samples.

  AHE due to the partial spin freezing.
Summary

- The same scaling relation of $\Gamma_{\text{mag}}$ without $B_c$ as $\text{Pr}_2\text{Ir}_2\text{O}_7$ also holds in the insulating analog $\text{Pr}_2\text{Zr}_2\text{O}_7$.
- Frustrated Pr moments must be the origin of the diverging $\Gamma_{\text{mag}}$.
- Anomalous spontaneous $\sigma_H$ in bulk sample is highly related to the Pr-local moment; it disappears in the bad sample with LRO.

![Diagram showing $T$ vs $x$ for $\text{Pr}_{2+x}\text{Ir}_{2-x}\text{O}_{7+\delta}$, indicating chiral spin liquid, partial freezing, and long range order at $q = (100)$.]