



이학박사 학위논문

## Topological Phenomena in Pyrochlore Iridates

파이로클로르 이리듐 산화물의 위상학적 현상

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오태구

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이 논문을 이학박사 학위논문으로 제출함 2022년 5월

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### Abstract

## Topological Phenomena in Pyrochlore Iridates

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Physics in condensed matter is determined by spin-orbit coupling and electronic correlation. When spin-orbit coupling and electronic correlation are comparable to the bandwidth, the correlated topological phases like Weyl semimetal, axion insulator, and topological Mott insulator emerge. Because of their unique physical phenomena, correlated topological phases are getting more attention.

Pyrochlore iridates, whose chemical formula is  $R_2Ir_2O_7$  (R: rare-earth), have a strong correlation and spin-orbit coupling at the same time. Hence, pyrochlore iridates are playgrounds for investigating the correlated topological phases. Especially, pyrochlore iridates are the first candidate that a Weyl semimetal develops when the antiferromagnetic order is developed at a low temperature. The antiferromagnetic order is called all-in-all-out (AIAO) since all spins point from or to the unit cell center.

However, the smoking gun evidence for a magnetic Weyl semimetal is missing so far for two reasons. First, the ground state is mostly an antiferromagnetic insulator, so the window for Weyl semimetal is negligible. Second, although the Weyl semimetal is present in pyrochlore iridates, the signals from the Weyl semimetal are canceled by the cubic symmetry. In order to find Weyl semimetal in pyrochlore iridates, we apply the perturbations to pyrochlore iridates near the transition point from antiferromagnetic insulator to paramagnetic metal. We expect that the perturbation widens the window for Weyl semimetal and breaks the cubic symmetry so that the difficulties above can be overcome. The perturbations used here are magnetic field and strain. Accordingly, the dissertation is divided into two parts.

First, the magnetic field is applied to  $(Nd_xPr_{1-x})_2Ir_2O_7$  single crystals, which is between the antiferromagnetic insulator and paramagnetic metal phase. Then, the experiment shows a variety of topological semimetals. The group theory shows that the quadratic band crossing in paramagnetic metal has a high effective spin J = 3/2, so the magnetic field induces an anisotropic Zeeman term as well as a usual Zeeman term. The interplay of two Zeeman terms and AIAO order gives rise to the various topological semimetals, like the 4-pair Weyl, 2-pair Weyl, double Weyl, and nodal-line semimetals. The magnetic field controls Zeeman terms and AIAO by changing the spin configuration of pyrochlore iridates.

Second, when strain is applied to Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> and Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films, which are the antiferromagnetic insulator and paramagnetic metal, respectively. For Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>, the strain induces the insulator-to-metal transition and anomalous Hall Effect at zero magnetic fields. The model calculation shows that strained Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> is trivial metal with electron and hole pockets. Since the magnetization is zero at zero fields, the origin of the anomalous Hall Effect is neither the magnetization nor the Weyl semimetal. In fact, the strain-induced  $T_1$ -octupole is the origin of the anomalous Hall Effect, since  $T_1$ -octupole has the same symmetry as magnetization. On the other hand, for Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>, the experiments show numerous pieces of evidence for Weyl semimetals, such as anomalous Hall Effect, negative magnetoresistance, and planar Hall Effect. Especially, the planar Hall Effect can be explained by the chiral anomaly of Weyl semimetal and AIAO order. The results in the dissertation provide the reasons why Weyl semimetal emerges in pyrochlore iridates, and the method to find the Weyl semimetal as well. This dissertation facilitates the experimental discovery of novel topological phases in pyrochlore iridates.

**Keywords :** Topological phenomena, Anomalous Hall Effect, Pyrochlore Iridates, Weyl semimetal **Student Number :** 2016-24222

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**Chapter 1** 

#### Introduction

#### 1.1 Correlated Topological Phases



Figure 1.1: The schematics of 2D phase diagram of condensed matter as a function of  $\lambda/t$  and U/t.

Physics in condensed matter physics is determined mostly by the spinorbit coupling (SOC) and electronic correlation [1]. The Hamiltonian in a generic system is written as

$$H = \sum_{\langle ij\rangle,\sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + \lambda \sum_{i} \vec{L}_{i} \cdot \vec{S}_{i} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}, \qquad (1.1)$$

where  $c_{i\sigma}$  is the annihilation operator for an electron at site *i*,  $\lambda$  is the SOC, U is the correlation,  $\vec{L}_i$  is the orbital angular momentum,  $\vec{S}_i$  is the spin

angular momentum, and  $n_{i\sigma}$  is the number operator.

The 2D phase diagram as a function of  $\lambda/t$  and U/t is shown in Fig. 1.1. In the phase diagram, one can see four different regions. When  $\lambda$  and U are small, it is just a simple metal or insulator. When  $\lambda$  gets stronger, the phase becomes topological. When U gets stronger, the phase becomes correlated. When both  $\lambda$  and U are comparable to the bandwidth, the phase gets both topological and correlated features. In this region, the correlated topological phases such as Weyl semimetal, axion insulator, topological Mott insulator, spin liquid, and quadrupolar ordered phases can be obtained.

The correlated topological phases attract much attention from the community since they show unique physical properties. For example, the Weyl semimetal is the phase that the point node crossings between different energy bands are described by the Weyl equation. This is topological because the integration of Berry curvature on the surface surrounding the Weyl node is quantized. Weyl semimetal shows unique physical properties. First, since Weyl nodes are the source of Berry curvature, it can induce the anomalous Hall Effect. Second, two Weyl nodes are connected on the surface by the surface Fermi arc. Third, when the magnetic and electric fields are applied in the same direction, the charge pumping between a pair of Weyl nodes called the chiral anomaly appears. A negative longitudinal magnetoresistance can be induced by the chiral anomaly.

#### **1.2** Pyrochlore Iridates

Pyrochlore iridates have the chemical formula  $R_2Ir_2O_7$ , where R is rare-earth such as lanthanides. Since R is an *f*-electron system and Ir is a *d*-electron system, pyrochlore iridates have strong SOC and strong correlation. Furthermore, the structure of the pyrochlore iridates is frustrating. (See Figs. 1.2a-b.) The unit cell of the pyrochlore iridates is a tetrahedron. Suppose that an antiferromagnetic exchange interaction exists in the system.



Figure 1.2: (a) The crystal structure of pyrochlore iridates. (b) The frustration in the unit cell of pyrochlore iridates. (c) AIAO order of pyrochlore iridates.

Then, we can determine at most two spins as up and down, but the other two spins remain undetermined. The frustrated structure makes the system host various magnetic structures. Thus, pyrochlore iridates are useful playgrounds for studying the various correlated topological phases.

Especially, pyrochlore iridates are the first candidate materials that Weyl semimetal is predicted to exist. At low temperatures, pyrochlore iridates develop an antiferromagnetic order. The antiferromagnetic order is called the all-in-all-out (AIAO) order, since all spins point from or to the center of the unit cell (Fig. 1.2c.) When antiferromagnetic order sets in, time-reversal symmetry is broken, and a magnetic Weyl semimetal can emerge [2].

#### **1.3** Motivation and Outline of the dissertation

However, the smoking gun evidence for the Weyl semimetal in pyrochlore iridates is still missing for two reasons. First, the ground state is either an antiferromagnetic (AFM) insulator or paramagnetic metal. In Fig. 1.3a, the experiment results for various rare-earth ions  $R^{3+}$  are shown, as a function of ionic radius and temperature [1, 3]. For every rare-earth ion except  $Pr^{3+}$ , the ground state is an AIAO insulator. For  $Pr^{3+}$ , the ground



Figure 1.3: (a) 2D phase diagram from the experiments. This figure is revised from one in Ref. 1, 3. (b) 2D phase diagram from theoretical analysis. This figure is revised from one in Ref. 4.

state is a paramagnetic metal.

This can be explained by theoretical analysis [4, 5]. In Fig. 1.3b, the phase diagram as a function of nearest neighbor hopping parameter  $(t_{\sigma})$  and Hubbard repulsion (U) is drawn. The AIAO Weyl semimetal is in a very small region (green area) between the transition from an AIAO insulator and a paramagnetic metal. The window for Weyl semimetal is infinitesimal to be realized.

Furthermore, although the Weyl semimetal is present in pyrochlore iridates, it cannot be observed by the cubic symmetry. When AIAO order in Fig. 1.2c sets in, the system have  $3C_2$ ,  $8C_3$ ,  $6\sigma T$ , and  $6S_4T$ , where  $C_2$  is twofold rotation,  $C_3$  is threefold rotation,  $\sigma$  is the mirror,  $S_4$  is the fourfold roto-inversion, and T is time-reversal symmetry. Accordingly, the signals from Weyl semimetal, for example, anomalous Hall Effect (AHE) must vanish.

In order to the topological semimetals in pyrochlore iridates, one should overcome the two difficulties above. Thus, this dissertation reports the journey for finding topological semimetals in pyrochlore iridates by means of perturbations. The perturbations are applied to pyrochlore iridates near the transition between the AFM insulator and paramagnetic metal. In Fig. 1.3, R = Pr, Nd are close to the transition point. One can expect two effects from the perturbations. Since both valence and conduction bands are close together, perturbations are expected to make the system metallic and make the window for topological semimetal expanded. Moreover, perturbations point in a certain direction, and the cubic symmetry must be broken. Accordingly, signals from the Weyl semimetal such as AHE can be detected.

Since the perturbations applied to the system are the magnetic field and strain, this dissertation is divided into two parts. In the first part, the magnetic-field induced topological semimetals in pyrochlore iridates is discussed. The magnetic field is applied to  $(Nd_xPr_{1-x})_2Ir_2O_7$  single crystals, which is close to the transition point between AFM insulator and paramagnetic metal. Various topological semimetals such as Weyl and nodal-line semimetals are observed. Motivated by the experiment, the generic topological band structure is investigated. In paramagnetic metal, it was observed that quadratic band crossing (QBC) with fourfold degeneracy is at  $\Gamma$  point [6]. Because of the large band degeneracy and strong SOC, the Zeeman field can induce an anisotropic Zeeman effect as well as the conventional isotropic Zeeman effect. The competition of the energy scales of two Zeeman terms and the exchange energy of Ir electrons give rise to the various topological semimetals. Furthermore, we show the three energy scales can be controlled by the modulation of the magnetic structure, which couples to the degenerate states at QBC. The general topological band structure under a magnetic field is proposed.

In the second part, the strain-induced topological semimetals in pyrochlore iridates are discussed. Primarily, we explain that the anomalous Hall Effect in relaxed Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films comes from the AFM domain wall of the system. Since the AIAO order has cubic symmetry, the anomalous Hall Effect must be canceled. However, when the domain wall is generated, twofold rotations are broken and anomalous Hall Effect is allowed. Next, we apply the strain to Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films which is the AFM insulator. In the experiment, the strained Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films show a large enhancement of AHE compared to the relaxed ones. The recent theory proposed that higher-rank magnetic multipoles formed by clusters of spins can generate the AHE without magnetization [7, 8]. We prove that the strain-induced cluster  $T_1$ -octupoles are the only source of observed AHE. Lastly, we apply the strain to  $Pr_2Ir_2O_7$  thin films which is the paramagnetic metal. The anomalous Hall Effect, magnetoresistance, and planar Hall Effect support that the strained  $Pr_2Ir_2O_7$  develops an AIAO order and the Weyl semimetal sets in. With the experimental analysis and renormalization group calculations, we find an interacting Weyl liquid state with logarithmically renormalized Fermi velocity dressed by long-range Coulomb interaction.

This dissertation is based on my publications [9–13], and many contents are overlapped with them. I make an effort to streamline the publications and to carry the significant concepts in this dissertation. This may lead to the omission of references cited previously. The readers can find the original papers and look for the references there.

#### **Chapter 2**

#### Backgrounds

This chapter introduces the physical background of this dissertation. This chapter is composed of three parts. In the first part, the Weyl semimetal and its physical properties are introduced. Next, all physical models we use in this dissertation are described in detail. Lastly, the cluster multipoles which are widely utilized in this dissertation are explained.

#### 2.1 Weyl semimetal

#### 2.1.1 Introduction to Weyl semimetal

A Weyl semimetal is the phase where there are gapless excitations that are protected by topological invariants and symmetry near the Fermi level [14]. The gapless excitations are described by the so-called Weyl equation,

$$H = \pm v \vec{k} \cdot \vec{\sigma},\tag{2.1}$$

where  $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$  are the Pauli matrices. The sign  $\pm$  is the chirality of Weyl fermions. The Weyl semimetal phase appears when time-reversal T or inversion P is broken in 3-dimensional systems. The number of Weyl nodes in the Brillouin zone must be even, because of Nielsen-Ninomiya Theorem.

Before proceeding, we first define the Berry connection and Berry curvature. Suppose that there is a Hamiltonian with periodic potential  $H = H_0 + U(\vec{r})$  where  $U(\vec{r}) = U(\vec{r} + \vec{R})$  ( $\vec{R}$ : Bravais lattice vector). Then, the eigenstates of the Hamiltonian are described by

$$\psi_{\vec{k}}(\vec{r}) = e^{i\vec{k}\cdot\vec{r}}u_{\vec{k}}(\vec{r}), \qquad (2.2)$$

where  $u_{\vec{k}}(\vec{r}) = u_{\vec{k}}(\vec{r}+\vec{R})$ . This is the Bloch Theorem, and  $u_{\vec{k}}(\vec{r}) = \langle \vec{r} | u(\vec{k}) \rangle$ is called Bloch wave functions. Let us denote  $|u_n(\vec{k})\rangle$  is the *n*-th Bloch wavefunction of the Hamiltonian, an Abelian Berry connection is defined as

$$\vec{A}_n(\vec{k}) = -i\langle u_n(\vec{k}) | \nabla_k | u_n(\vec{k}) \rangle, \qquad (2.3)$$

and the Berry curvature is defined as

$$F_n^{ab}(\vec{k}) = \partial_{k_a} A_n^b - \partial_{k_b} A_n^a.$$
(2.4)

The Weyl semimetal is considered a topological phase. This is because the net Berry flux surrounding the gapless excitation is quantized,

$$\int \frac{d^2k}{(2\pi)} F_n^{ab}(\vec{k}) = \pm 1.$$
(2.5)

This can be proved by using Eq. 2.1. When we represent  $\vec{k} = k(\sin\theta\cos\phi)$ ,  $\sin\theta\sin\phi$ ,  $\cos\theta$ ), The eigenstates of the Hamiltonian are given by

$$|u_{\uparrow}(\vec{k})\rangle = [\cos(\theta/2)e^{-i\phi}, \sin(\theta/2)]^{T},$$
  
$$|u_{\downarrow}(\vec{k})\rangle = [\sin(\theta/2)e^{-i\phi}, -\cos(\theta/2)]^{T},$$
 (2.6)

The Berry curvature for the down arrow band is

$$\vec{F}(\vec{k}) = -\frac{1}{2}\frac{\vec{k}}{k^3}$$
(2.7)

Then, the integration of Berry curvature on a shell surrounding the excitation

$$\int \frac{d\vec{S}}{2\pi} \cdot \vec{F}(\vec{k}) = -1.$$
(2.8)

Please note that the Weyl nodes can be regarded as a source or sink of the Berry curvature.

#### 2.1.2 Physical consequences of Weyl semimetal

The topological properties of Weyl semimetal give rise to a myriad of exotic physical consequences. We introduce two representative physical consequences, the anomalous Hall Effect (AHE) and chiral anomaly.

The AHE is the emergence or enhancement of Hall voltage in the timereversal broken systems. There are extrinsic and intrinsic mechanisms for AHE. The extrinsic mechanism is the side-jump and skew-scattering from the magnetic impurities. The intrinsic mechanism, on the other hand, is from the Berry curvature of the electronic energy band. Weyl semimetal gives rise to anomalous Hall Effect by intrinsic effects. The intrinsic anomalous Hall conductivity is given by [15]

$$\sigma_{ab} = \frac{e^2}{\hbar} \int \frac{d^d k}{(2\pi)^d} \sum_n F_n^{ab}(\vec{k}) f(\epsilon_n(\vec{k})), \qquad (2.9)$$

where  $\epsilon_n(\vec{k})$  is the electronic energy,  $f(\epsilon)$  is the Fermi-Dirac distribution, and  $F_n^{ab}$  is the Berry curvature. Because the Weyl nodes are a source or sink of a Berry curvature, Weyl semimetal exhibits the AHE.

For example, let us consider a model with two Weyl nodes. We choose a Hamiltonian like [16].

$$H = [2t_x(\cos k_x - \cos k_0) + m(2 - \cos k_y - \cos k_z)]\sigma_x + 2t_y \sin k_y \sigma_y + 2t_z \sin k_z \sigma_z.$$
(2.10)



Figure 2.1: The schematics of the chiral anomaly in Weyl semimetal

This models includes a pair of Weyl nodes at  $\pm \vec{k}_W = \pm (k_0, 0, 0)$ . If we assume the half-filling, the Weyl nodes are exactly at the Fermi level. By fixing  $k_x$ , we think  $H_{k_x}(k_y, k_z)$  as a 2-dimensional band structure, which are all gapped whenever  $k_x \neq \pm k_W$ . Therefore, the Chern number of each 2D plane can be calculated. When  $k_x \in (-k_0, k_0)$ , C = 1, and otherwise, C = 0. Hence, by using the formula in Eq. 2.9, we find that

$$\sigma_{yz} = \frac{e^2}{2\pi h} (2k_0). \tag{2.11}$$

Note that the anomalous Hall conductivity is proportional to the distance between the pair of Weyl nodes.

Meanwhile, the chiral anomaly is the violence of the preservation of chiral currents under electromagnetic fields. This occurs because the Weyl nodes have chiral zeroth Landau levels. (See Fig. 2.1.) Introducing the magnetic field along z-direction to Eq. 2.1, one easily obtains the *n*th Landau

levels of Weyl fermions are [17]

$$E_n(k_z) = \mp v k_z (n = 0),$$
  

$$sgn(n)v\sqrt{k_z^2 + 2|n|eB} (n \neq 0).$$
(2.12)

Introducing the electric field additionally, the chemical potentials of lefthanded to right-handed Weyl fermions become different. Hence, the number of chiral charges is not conserved anymore. That is,

$$\frac{dn_{R/L}}{dt} = \pm \frac{e^2}{h^2} \vec{B} \cdot \vec{E}, \qquad (2.13)$$

This gives rise to the negative magnetoresistivity or positive magnetoconductivity [18, 19].

$$\sigma(B) = \sigma_0 + \frac{e^4}{4\pi^4 G(\epsilon_F)} B^2 \tau, \qquad (2.14)$$

where  $G(\epsilon_F)$  is the density of states at the Fermi level, and  $\tau$  is the internode scattering time.

#### 2.2 Physical models

This part introduces the models for the theoretical calculation of pyrochlore iridates. There are two kinds of models. The first one is the Hubbard model which describes the itinerant electron systems. The second one is the classical spin model which describes the localized spin systems.

#### 2.2.1 Hubbard Model

When we theoretically calculate the conduction properties of pyrochlore iridates, we use a Hubbard model. The Hubbard model is based on Ref. 5,

20,

$$H = H_0 + H_U + H_{fd} + H_B + H_Z \tag{2.15}$$

where  $H_0$  is the hopping of Ir electrons,  $H_U$  is the Hubbard repulsion for Ir electrons,  $H_{fd}$  is the fd-exchange between rare-earth and Ir electrons, and  $H_B$  and  $H_Z$  are the Zeeman effects for Ir and rare-earth, respectively. That is,

$$\begin{split} H_{0} &= \sum_{\langle ab \rangle, \alpha, \beta} c^{\dagger}_{a,\alpha} v_{ab} (t_{1} + it_{2} \vec{d}_{ab} \cdot \sigma_{\alpha\beta}) c_{b,\beta} \\ &+ \sum_{\langle \langle ab \rangle \rangle \alpha\beta} c^{\dagger}_{a,\alpha} (t_{1}' + i(t_{2}' \vec{R}_{ab} + t_{3}' \vec{D}_{ab}) \cdot \sigma_{\alpha\beta}) c_{b,\beta}, \\ H_{U} &= U \sum_{a} n_{a\uparrow} n_{a\downarrow}, \\ H_{fd} &= \sum_{\langle ai \rangle, \mu\nu} \Lambda^{\mu\nu}_{ai} \sigma^{\mu}_{a} \tau^{\nu}_{i}, \\ H_{B} &= \frac{1}{2} \sum_{a} c^{\dagger}_{a,\alpha} (\vec{B} \cdot \sigma_{\alpha\beta}) c_{a,\beta}, \\ H_{Z} &= -\sum_{i} \gamma (\vec{B} \cdot \vec{V}_{i}) \tau^{z}_{i}. \end{split}$$
(2.16)

Here, a, b are the Ir sublattice indices, i is the Nd sublattice index,  $\vec{d}_{ab}$  $(\vec{R}_{ab}, \vec{D}_{ab})$  are the (next-)nearest-neighbor DM vectors,  $\sigma$  are Pauli matrices for Ir spins,  $\alpha, \beta = \uparrow, \downarrow$  are indices for Pauli matrices,  $\tau$  is the number for rare-earth spins,  $\mu, \nu = x, y, z$  are the coordinate indices,  $c_{a,\alpha}$  is the annihilation operator for Ir electrons,  $n_{a\alpha} = c^{\dagger}_{a,\alpha}c_{a,\alpha}$  is the number operator,  $v_{ab}$ is the hopping parameter change by the strain,  $\vec{B}$  is the magnetic field,  $\vec{V}_i$  is the local-z axis for rare-earth spins, and  $\gamma = 5$  is the g-factor for rare-earth spins. We explain the model in detail as follows. Note that we deal with rare-earth spins as an Ising one.

We define the hopping parameters first. The hopping parameters are

from Ref. 5.

$$t_1 = \frac{130}{243} t_{oxy} + \frac{17}{324} t_\sigma - \frac{79}{243} t_\pi,$$
  
$$t_2 = \frac{28}{243} t_{oxy} + \frac{15}{243} t_\sigma - \frac{40}{243} t_\pi,$$

$$t_{1}' = \frac{233}{2916} t_{\sigma}' - \frac{407}{2187} t_{\pi},$$
  

$$t_{2}' = \frac{1}{1458} t_{\sigma}' + \frac{220}{2187} t_{\pi},$$
  

$$t_{3}' = \frac{17}{324} t_{\sigma}' + \frac{460}{2187} t_{\pi},$$
(2.17)

where  $t_{oxy}$  is the hopping through oxygen ions,  $t_{\sigma}(t'_{ma})$  is the hopping through (next-)nearest-neighbor  $\sigma$ -bonding, and  $t_{\pi}(t'_{\pi})$  is the hopping through (next-)nearest-neighbor  $\pi$ -bonding. In the most calculations, We use  $t_{oxy} = 1$ ,  $t_{\sigma} = -0.8$ ,  $t_{\pi} = -2t_{\sigma}/3$ .

We define the DM vectors here. The nearest-neighbor DM vectors are

$$\begin{split} \vec{d}_{ab} &= 2\vec{f}_{ab} \times \vec{b}_{ab}, \\ \vec{f}_{ab} &= \frac{1}{2}(\vec{r}_{a} + \vec{r}_{b}) - \vec{C}, \\ \vec{b}_{ab} &= \vec{r}_{b} - \vec{r}_{a}. \end{split} \tag{2.18}$$

Here,  $\vec{r_a}$  is the *a*-th sublattice position, and  $\vec{C}$  is the unit cell center position. The next-nearest-neighbor DM vectors are

$$\vec{R}_{ab} = \vec{b}_{ac} \times \vec{b}_{cb},$$
  
$$\vec{D}_{ab} = \vec{d}_{ac} \times \vec{d}_{cb}.$$
 (2.19)

Here, c means the common neighbor of a and b sublattices.

Next, we define the parameters in  $H_{fd}$ .

$$(G_{1}^{x}\vec{a}_{i} + G_{2}^{x}\vec{a}_{i}\bar{\times}\vec{d}_{ai}\bar{\times}\vec{d}_{ai}) \cdot \hat{e}_{\mu}(\nu = x),$$

$$\Lambda_{ai}^{\mu\nu} = G^{y}(\vec{a}_{i}\bar{\times}\vec{d}_{ai}\bar{\times}\vec{d}_{ai}]) \cdot \hat{e}_{\mu}(\nu = y),$$

$$(G_{1}^{z}\vec{a}_{i} + G_{2}^{z}\vec{a}_{i}\bar{\times}\vec{d}_{ai}\bar{\times}\vec{d}_{ai}) \cdot \hat{e}_{\mu}(\nu = z).$$
(2.20)

 $\overline{\times}$  is the symmetrized vector product, and  $\vec{a}_i$  is the position of *i* sublattice. The parameters we use here is  $G_1^x = 0, G_2^x = 0.01, G_y = -0.006, G_1^z = -0.06, G_{2z} = -0.02.$ 

Next, we define the hopping change  $v_{ab}$ . For a compressive strain on [111] plane,

$$v_{ab} = 1 - \delta, (ab = 12, 13, 14),$$
  
 $1 + \delta, (ab = 23, 24, 34).$  (2.21)

Lastly, we use the Hartree-Fock mean-field theory on the Hubbard model to calculate the ground state self-consistently.

$$H_U^{MF} = -U(\sum_a 2\langle \vec{m}_a \rangle \cdot \vec{m}_a - \langle \vec{m}_a \rangle^2)$$
(2.22)

where  $\vec{m}_a = \frac{1}{2} \sum_{\alpha\beta} c^{\dagger}_{a\alpha} \sigma_{\alpha\beta} c_{a\beta}$ .

#### 2.2.2 Spin Model

On the other hand, when we theoretically calculate the ground state from the classical spin model of pyrochlore iridates. The spin model is given by

$$H = H_{ex} + H_{DM} + H_{ani} + H_{fd} + H_B + H_Z,$$
(2.23)

where  $H_{ex}$  is the exchange interaction,  $H_{DM}$  is the DM interaction,  $H_{ani}$  is the anisotropic interaction,  $H_{fd}$  is the fd-exchange between Ir and Nd spins, and  $H_B$  and  $H_Z$  are the Zeeman interaction of Ir and rare-earth spins.

$$\begin{split} H_{ex} &= \sum_{\langle ab \rangle, \langle \langle ab \rangle \rangle} J_{ab} \vec{S}_a \cdot \vec{S}_b, \\ H_{DM} &= \sum_{\langle ab \rangle, \langle \langle ab \rangle \rangle} \vec{D}_{ab} \cdot \vec{S}_a \times \vec{S}_b, \\ H_{ani} &= \sum_{\langle ab \rangle, \langle \langle ab \rangle \rangle} \Gamma_{ab}^{\mu\nu} S_a^{\mu} S_b^{\nu}, \end{split}$$

$$H_{fd} = J_K \sum_{\langle ai \rangle} \Lambda_{ai}^{\mu\nu} S_a^{\mu} \tau_i^{\nu},$$
  

$$H_B = -\vec{h} \cdot \sum_a \vec{S}_a,$$
  

$$H_Z = -\gamma \sum_i (\vec{h} \cdot \vec{V}_i) \tau_i^z,$$
(2.24)

Note that  $J_K = 0(1)$  for the absence (existence) of the rare-earth spins.  $\gamma = 5$  is the relative g-factor of rare-earth spins to Ir spins.  $\vec{h}$  is the magnetic field.  $\Lambda_{ai}^{\mu\nu}$  is defined in Eq. 2.20.  $\vec{V}_i$  is the local-z axis for the rare-earth *i* sublattice.  $\vec{S}_i$  is the Ir spin, and  $\vec{\tau}_i$  is the rare-earth spin. We deal with the spins as a number, not an operator since this is the classical theory.

The other parameters we use here are obtained from the perturbation theory of the Hubbard model. The second order perturbation gives

$$J_{ab} = \frac{4}{U} (t_{ab}^2 - \frac{d_{ab}^2}{3}),$$
  
$$\vec{D}_{ab} = \frac{4}{U} (2t_{ab}\vec{d}_{ab}),$$
  
$$\Gamma^{\mu\nu}_{ab} = \frac{4}{U} (2d^{\mu}_{ab}d^{\nu}_{ab} - \delta^{\mu\nu}\frac{d^2_{ab}}{3}),$$
 (2.25)

where  $\vec{d}_{ab}$  is the DM vector, and  $t_{ab}$  is the hopping strength between a and b sublattices.

#### 2.3 Cluster Magnetic Multipole

For the systematic approach to the magnetic order, we use the concept of cluster multipoles [7, 8]. In the magnetic unit cell, the spin cluster is defined as the set of atoms connected by the symmorphic crystalline symmetries. The cluster multipoles of ath order are defined as

$$M_{ab} = \sum_{i=1}^{N_c} \vec{m}_i \cdot \vec{P}_{ab}(\vec{r}_i), \qquad (2.26)$$

where

$$\vec{P}_{ab}(\vec{r}_i) = \sqrt{\frac{4\pi}{2a+1}} \nabla(r^a Y^*_{ab}(\theta_i, \phi_i))$$
(2.27)

Here,  $N_c$  is the number of atoms in the spin cluster,  $\vec{r_i}$  is the position of *i*th atom,  $Y_{ab}(\theta_i, \phi_i)$  is the spherical harmonics,  $\vec{m_i}$  is the magnetic moment at *i*th atom, and  $b \in [-a, a]$  is the magnetic quantum number. The cluster toroidal multipoles can also be defined as follows.

$$T_{ab} = \frac{1}{a+1} \sum_{i=1}^{N_c} (\vec{r}_i \times \vec{m}_i) \cdot \vec{P}_{ab}(\vec{r}_i).$$
(2.28)

Note that cluster multipoles are related to the responses to the magnetic field, while cluster toroidal multipoles are related to the responses to both electric and magnetic fields. Since this dissertation considers the response under the magnetic field, only the cluster multipoles of pyrochlore lattices are described below.

The magnetic unit cell of pyrochlore iridates is a tetrahedron [21]. The point group of the tetrahedron is the  $T_d$  group, which has threefold rota-



Figure 2.2: The cluster multipoles in pyrochlore iridates. (a) Dipoles, (b) Octupoles, and (c) Dotriacontapoles (32-poles).

tions  $C_3$ , twofold rotations  $C_2$ , fourfold roto-inversions  $S_4$ , and diagonal mirrors  $\sigma_d$ . Accordingly,  $T_d$  group has five distinct irreducible representations:  $A_1$ ,  $A_2$ , E,  $T_1$ , and  $T_2$ . By projecting the general spin configuration of pyrochlore lattices, we find a total of 12 kinds of cluster multipoles that form the bases of irreducible representations. The results are shown in Fig. 2.2. Pyrochlore lattices have  $T_1$ -dipoles, an  $A_2$ -octupole,  $T_1$ -octupoles,  $T_2$ -octupoles, E-dotriacontapoles. The dipoles are the ferromagnetic orders, the  $A_2$ -octupole is the AIAO order,  $T_1$ -octupoles are antiferromagnetic orders related to the spin ice,  $T_2$ -octupoles are the Palmer-Chalker phase, and E-dotriacontapoles are the local-XY phase. Note that in the viewpoint of cluster toroidal multipoles,  $T_2$ -octupoles and E-dotriacontapoles are the toroidal quadrupoles.

Lastly, we leave here the equation describing each magnetic multipole.  $\vec{m}_i = (m_{ix}, m_{iy}, m_{iz})$  denote the magnetic moment at *i*th atoms.  $T_1$ -dipoles are

$$M_{x} = \frac{1}{4}(m_{1x} + m_{2x} + m_{3x} + m_{4x}),$$
  

$$M_{y} = \frac{1}{4}(m_{1y} + m_{2y} + m_{3y} + m_{4y}),$$
  

$$M_{z} = \frac{1}{4}(m_{1z} + m_{2z} + m_{3z} + m_{4z}).$$
(2.29)

The  $A_2$ -octupole is

$$A_{2} = \frac{1}{4\sqrt{3}}(m_{1x} + m_{1y} + m_{1z} + m_{2x} - m_{2y} - m_{2z} - m_{3x} + m_{3y} - m_{3z} - m_{4x} - m_{4y} + m_{4z}).$$
(2.30)

The  $T_1$ -octupoles are

$$T_{1x} = \frac{1}{4\sqrt{2}} (-m_{1y} - m_{1z} + m_{2y} + m_{2z} + m_{3y} - m_{3z} - m_{4y} + m_{4z}),$$

$$T_{1y} = \frac{1}{4\sqrt{2}} (-m_{1x} - m_{1z} + m_{2x} - m_{2z} + m_{3x} + m_{3z} - m_{4x} + m_{4z}),$$

$$T_{1z} = \frac{1}{4\sqrt{2}} (-m_{1x} - m_{1y} + m_{2x} - m_{2y} - m_{3x} + m_{3y} + m_{4x} + m_{4y}).$$
(2.31)

The  $T_2$ -octupoles are

$$T_{2x} = \frac{1}{4\sqrt{2}} (-m_{1y} + m_{1z} + m_{2y} - m_{2z} + m_{3y} + m_{3z} - m_{4y} - m_{4z}),$$

$$T_{2y} = \frac{1}{4\sqrt{2}} (m_{1x} - m_{1z} - m_{2x} - m_{2z} - m_{3x} + m_{3z} + m_{4x} + m_{4z}),$$

$$T_{2z} = \frac{1}{4\sqrt{2}} (-m_{1x} + m_{1y} + m_{2x} + m_{2y} - m_{3x} - m_{3y} + m_{4x} - m_{4y}).$$
(2.32)

#### The E-dotriacontapoles are

$$E_{1} = \frac{1}{4\sqrt{6}} (m_{1x} + m_{1y} - 2m_{1z} + m_{2x} - m_{2y} + 2m_{2z} - m_{3x} + m_{3y} + 2m_{3z} - m_{4x} - m_{4y} - 2m_{4z}),$$

$$E_{2} = \frac{1}{4\sqrt{2}} (m_{1x} - m_{1y} + m_{2x} + m_{2y} - m_{3x} - m_{3y} - m_{4x} + m_{4y}).$$
(2.33)

#### **Chapter 3**

# Part I: Magnetic field-induced topological semimetals in pyrochlore iridates

#### 3.1 Diversity of topological phases in $(Nd_xPr_{1-x})_2Ir_2O_7$ single crystals

#### 3.1.1 Introduction

The pyrochlore iridates R<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> are composed of corner-sharing tetrahedrons of rare-earth R and Ir ions. Since the lattice is a geometrically frustrated lattice, pyrochlore iridates are a fertile playground for exotic electromagnetic states [1, 2, 22, 23]. The angle-resolved photoemission spectroscopy shows that Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> is a paramagnetic semimetal with a quadratic band crossing at  $\Gamma$  point [6]. This is an essential ingredient for diverse topological phases. For example, the antiferromagnetic all-in-all-out (AIAO) magnetic order breaks time-reversal symmetry and lifts the band degeneracy, giving rise to linearly dispersed Weyl nodes in three dimensions. Here, we terms Weyl semimetal (WSM (4/0)) [2, 4, 24]. Another unconventional electronic states are comprehensively discussed with different magnetic configurations [20, 25–27]. By the uniaxial magnetic anisotropy along [111] or equivalent directions, various magnetic configurations can be gained under the interplay between exchange interactions and external magnetic field [28]. When a magnetic field applied along  $H \parallel [001] (H \parallel [111])$  is stronger than the exchange interaction, AIAO turns into two (three) magnetic moments direct inwards and the other two (one) direct outwards of the unit cell, forming 2/2 (3/1) configuration.

Another key factor is the electron bandwidth, demonstrating the inverse

of the effective electron correlation (U) [29]. One can tune the bandwidth by applying hydrostatic pressure [30, 31] or substituting R site [3, 32] that can prompt metal-insulator transition (MIT). The Pr compound is a paramagnetic semimetal down to  $120 \ mK$  [33], while the paramagnetic or antiferromagnetic AIAO insulator appears with smaller R ionic radius [34-36], apparently similar to the correlation-induced MIT as observed for 3d-electron systems [31, 37, 38]. At the point of quantum MIT (between R=Nd and Pr), nevertheless, the unconventional magneto-transport was reported, such as anomalous Hall effect! [39, 40], metallic AIAO domain walls [41, 42], and field-induced MIT [20, 27]. This may be related to the predicted topological phases in the system. The quantum MIT including such correlated topological phases may build an ideal stage of a novel quantum criticality [43, 44], but has been little explored up to now. To address this subject, we systematically measure magneto-transport on R=Nd and R=Nd $_{0.5}$ Pr $_{0.5}$ compounds under external hydrostatic pressures (P) and magnetic fields (H). This makes us delicately tune the effective bandwidth and magnetic configuration. We reveal a variety of topological phases as a function of bandwidth and a magnetic field near the quantum critical point.

# **3.1.2** Electromagnetic phase diagram for pyrochlore iridates.

We present the temperature dependence of resistivity under several pressures in Fig. 3.1d-h. The resistivity at ambient pressure increases extremely below  $T_N = 22 K$ , which shows the recent improvement of sample quality. The transition temperature systematically lowers with increasing pressure as in previous studies [30, 31]. Figure 3.1i displays the temperature dependence of longitudinal resistivity for x = 0.5 (R=Nd<sub>1-x</sub>Pr<sub>x</sub>). This also shows a sharp increase below 4 K. We plot the  $T_N$  as a function of pressure P and the portion of Pr x in Fig. 3.1j. We use the empirical equation be-



Figure 3.1: Schematics of magnetic configuration for (a) all-in-all-out, (b) 2-in-2-out, and (c) 3-in-1-out states, respectively. Temperature dependence of longitudinal resistivity for R=Nd (x = 0) at (d) 0 GPa, (e) 1.0 GPa, (f) 1.4 GPa, (g) 1.8 GPa, (h) 2.2 GPa and (i) x = 0.5 for R=Nd<sub>1-x</sub>Pr<sub>x</sub> (effectively 3.3 GPa), respectively. The black lines denote the resistivity of the trained state that the magnetic domain wall contribution is eliminated. The blue lines denote the resistivity under  $H \parallel [111]$  of 14 T and the red ones denote the resistivity under  $H \parallel [001]$ . (j) The change in MIT temperature as a function of pressure (bottom axis) and the portion of Pr x (top axis).  $\Delta_x = 0.1$  effectively corresponds to  $\Delta P = 0.65$  GPa. The circles are the MIT temperature for x = 0, the triangles are that for x = 0.5, and the square is that for x = 1.

tween the chemical and physical pressures [31] that  $\Delta x=0.1$  corresponds to the pressure change  $\Delta P = 0.65 \ GPa$ . From now on, we consider x = 0.5equivalent to the application of  $P = 3.3 \ GPa$  on x = 0. Since the  $T_N$  is almost linearly decreased as pressure increases, we can explore a wide range of effective bandwidths. Note that the AIAO insulator stands up to P = 5.0 $GPa \ (P = 1.7 \ GPa$  on the x = 0.5). The strength of the insulating phase is also previously reported.

#### 3.1.3 Anomalous magneto-transport near MIT.

Figure 3.1d–i also display the longitudinal resistivity under  $H \parallel [001]$ and [111] of 14 T. For  $H \parallel$  [001], while the resistivity slightly decreases by the magnetic field at ambient pressure, the extreme increase of the resistivity below  $T_N$  is suppressed about  $P \ge 1.0$  GPa. This means that the systematic application of pressure makes the system critical where various electromagnetic phases compete with each other strongly, as the colossal magnetoresistance in perovskite manganites [45]. Similarly, the large magnetoresistance was reported even at ambient pressure [20, 27]. This can be attributed to the off-stoichiometry of the crystal. The iridium deficiency, for example, changes the band filling and effectively pushes the system forward to criticality. On the other hand, the applied magnetic field  $H \parallel [111]$  induces different magnetotransport properties from  $H \parallel [001]$ . The resistivity begins to rise gradually above  $T_N$  and reaches a plateau at the lower temperature. The observed magnetotransport for each field direction is ascribed to the emergence of a novel electronic state induced by  $H \parallel [001] (H \parallel [111])$ , which tunes AIAO to the 2/2 (3/1) configuration in R 4f moments as shown in Fig. 3.1b,c. The saturated magnetization for  $H \parallel [001] (H \parallel [111])$  agrees well with the predicted values in 2/2 (3/1) state. In Nd<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub> without delectrons, where the Nd spins form AIAO order at zero field, also shows 2/2 or 3/1 order under magnetic field [46]. Furthermore, for x = 0.5, the specific heat peak divided by temperature  $(C_V/T)$  moves to a higher temperature, with broadening when we increase  $H \parallel [001] (H \parallel [111])$ . These features implies that the increasing  $H \parallel [001] (H \parallel [111])$  induces the 2/2 (3/1) type magnetic order even at  $T > T_N$  [47, 48]. Because of the coupling between 4f and 5d moments, the magnetic configuration of 5d moments is linked to that of 4f ones, leading to the observed magnetotransport. Also, note that in this experiment, the chiral anomaly effect was removed by applying the magnetic field perpendicular to the current. Chiral anomaly is the negative


Figure 3.2: Magnetotransport of  $(Nd_{1-x}Pr_x)_2Ir_2O_7$  at different pressures. Magnetic field dependence of longitudinal resistivity (a–e) and Hall conductivity (f–j) for  $H \parallel [001]$  at (a,f) 1.0 GPa, (b,g) 1.4 GPa, (c,h) 1.8 GPa, (d,i) 2.2 GPa and (e,j) 3.3 GPa (x = 0.5), respectively. Magnetic field dependence of longitudinal resistivity (k–o) and Hall conductivity (p–t) for  $H \parallel [111]$  at (k,p) 1.0 GPa, (l,q) 1.4 GPa, (m,r) 1.8 GPa, (n,s) 2.2 GPa and (o,t) 3.3 GPa (x = 0.5), respectively. The solid (dotted) lines are the resistivity on increasing (decreasing) field sweep, indicated by black arrows.

magnetoresistance induced in WSM materials when the magnetic field is applied parallel to electric current [49]. Thus, the anisotropic magnetoresistance observed here can only be attributed to the magnetic configuration change.

#### **3.1.4** Magnetotransport properties for $H \parallel [001]$ .

The *H*-dependence of longitudinal resistivity for  $H \parallel [001]$  at various pressures is shown in the top panels of Fig. . The sharp decrease of longitudinal resistivity is followed by a hysteresis between field-increasing and field-decreasing sweeps below  $T_N$ , as observed in previous reports [20, 27]. The Hall conductivity in Fig. f-j provides intuitions into the observed fieldinduced MIT. When  $T > T_N$ , the Hall conductivity is proportional to the magnetic field, which is the normal Hall effect. On the contrary, when  $T < T_N$ , the Hall conductivity shows non-monotonous field dependence. It almost vanishes at low magnetic fields, suddenly rises up at intermediate fields, and eventually decreases towards negative at high fields. This feature is more evident at the low temperature. A sign change of Hall response also appears in the Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> poly-crystals [40]. The complex behavior of the Hall response cannot be explained by either the normal or anomalous Hall resistivity [15]. The contour plots of the longitudinal and Hall conductivity as a function of temperature and magnetic field for  $H \parallel [001]$  at several pressures are exhibited in Fig. 3.3a-j.

Generally, the Hall conductivity depends on the relaxation time. For instance, the zero Hall conductivity at low fields implies the localization of electrons, in agreement with the large value of longitudinal resistivity  $(\rho_{xx}=410 \text{ m}\Omega\text{cm})$ . On the other hand, the Hall conductivity at high fields decreases towards negative, while the longitudinal resistivity saturates around  $\rho_{xx}(B)=0.4 \text{ m}\Omega\text{cm}$  (Fig. 3.1.4a–e). One possible candidate for the electronic phase is the topological semimetal in the 2/2 spin configuration. This phase, which we dubbed line node semimetal (LSM), has a nodal line in the  $k_z = 0$  plane and two Weyl points on the  $k_z$  axis as presented in Fig. 3.4b. Next, the major result of Hall conductivity presented here is a large signal with a positive sign in intermediate fields. When the field increases, the Hall conductivity shows a drastic change including a sign reversal. This



Figure 3.3: Contour plot of conductivity and Hall conductivity under various pressures. (a,f,k,p) 1.0 *GPa*, (b,g,l,q) 1.4 *GPa*, (c,h,m,r) 1.8 *GPa*, (d,i,n,s) 2.2 *GPa*, and (e,j,o,t) 3.3 *GPa*. (a-j)  $H \parallel [001]$ , and (k-t)  $H \parallel [111]$ .

can be explained by the phase transition between the 4/0 WSM (Fig. 3.4a) and 2/2 LSM (Fig. 3.4b). We schematically show the phase transition in Fig. 3.4d,f. Since 4/0 WSM and 2/2 LSM have distinct Fermi surface topology, the phase transition requires a crucial modulation of the energy band near the Fermi level. The modulation is accompanied by the emergence of electron/hole pockets, which can change the Hall conductivity drastically. The competition of the normal and anomalous Hall conductivities are theoretically calculated, which illustrates the non-monotonic field dependence. We show the calculation in Ref. 9.

In contour plots of longitudinal and Hall conductivities in Fig. 3.3a-j,

we can reveal the characteristic relation between the MIT and Hall conductivity for  $H \parallel [001]$ . Both longitudinal and Hall conductivities are small in a low-field and low-temperature region, where the phase is AIAO insulating. When the field increases, the Hall conductivity drastically changes with a sign reversal. This can be attributed to the phase transition between the WSM and LSM, as shown in Fig. 3.4d,f. Previously, the WSM phase was theoretically predicted to exist in a very narrow temperature region at zero field [4, 24], and thus would be difficult to observe the phase by optical [50] and angle-resolved photoemission spectroscopy [51]. Interestingly, the WSM phase is extended by  $H \parallel [001]$ , which deforms the regular 4/0 spin configuration. Moreover, on increasing pressure, both AIAO insulating and WSM phases shrink, while the LSM phase expands toward zero temperature and zero fields. The diverse competing phases come close to each other in free energy and merge at the quantum critical point, such as antiferromagnetic Mott insulator, a paramagnetic semimetal, the topological pseudo-4/0 WSM, and 2/2 LSM.

#### **3.1.5** Unconventional semimetal phases in $H \parallel [111]$ .

Let us consider the magnetotransport for  $H \parallel [111]$  now, shown in Fig. 3.1.4k–t. Right above  $T_N$ , the resistivity is greatly increased by the magnetic field, while the Hall conductivity exhibits a sharp upturn and flips its sign in a higher field (Fig. 3.1.4p–t). There is a report for paramagnetic  $Pr_2Ir_2O_7$  that shows similar magnetotransport properties at a much lower temperature ( $T = 30 \ mK$ ) [52]. Below  $T_N$ , when temperature decreases, a sharp dip is observed around  $H = 3 \ T$  in longitudinal resistivity, which comes from the emergence of metallic domain walls [41, 42]. Remarkably, the longitudinal resistivity shows the unique H dependence followed by a hysteresis between field-increasing and -decreasing sweeps. It is conspicuous at  $T = 9 \ K$  and  $P = 1.0 \ GPa$  as shown in Fig. 3.1.4k. Furthermore,



Figure 3.4: Schematic band structures of (a) WSM (4/0) with small H, (b) LSM (2/2) with  $H \parallel [001]$ , (c) WSM (3/1) with  $H \parallel [111]$ . The schematic phase diagram near quantum critical points (d-e) as a function of U and H, and (f-g) as a function of T and H. (d,f)  $H \parallel [001]$ , and (e,g)  $H \parallel [111]$ .

the longitudinal resistivity saturates around  $\rho_{xx} \approx 7 \text{ m}\Omega \text{cm}$  above H = 9 T where the hysteresis loop ends. This indicates the transition from the 4/0 to the 3/1 magnetic configuration. To observe the evolution of the respective phases, we present the contour plot of longitudinal and Hall conductivities in Fig. 3.3k–t and the schematic phase diagrams in Fig. 3.4e,g. The conductivity is quite small in the high field where the Hall conductivity has a positive sign. This can be attributed to the emergence of the new semimetal phase with the 3/1 magnetic state.

To reveal the electronic energy band in the 3/1 magnetic configuration,

we perform a mean-field calculation. The method for the calculation is in Ref. 9. The outstanding feature in the 3/1 configuration is that only one trigonal axis parallels to  $H \parallel [111]$  exists. This is in contrast to the 4/0 configuration with four trigonal axes. Note that a pair of Weyl nodes are always lying on one of the four trigonal axes in the AIAO configuration. Because each pair of Weyl nodes is confined on a one-dimensional line, pair-annihilation easily occurs at the Brillouin zone boundary when the pair separation increases. In the 3/1 configuration, on the other hand, threefold rotation symmetry breaking allows six Weyl nodes to deviate away from the original one-dimensional subspace. Instead, they move in a two-dimensional mirror plane. The remaining two Weyl nodes on [111] are still confined, but their pair-annihilation gives rise to another WSM with six Weyl points (WSM (3/1)) as described in Fig. 3.4c. When the point nodes move in the two-dimensional plane, it is less likely to collide with the other pair than those moving in a one-dimensional line. Thus, WSM (3/1) has more stability than WSM (4/0) and covers a larger area in the phase diagram. WSM (3/1)phase survives intensively in any H range, while it is stable only within a finite U range.

We suggest a variety of topological phases can emerge as a function of bandwidth or electronic correlation by combining the systematic approach to transport with the theoretical calculations, as shown in Fig. 3.4d–f. An important point is that all topological states merge towards the quantum critical point. In the next section, we theoretically analyze the field-induced topological states near the quantum critical point.

## **3.2** Magnetic field-induced topological phases in pyrochlore iridates

#### **3.2.1** Introduction

Spin-orbit coupling and electronic correlation are two ultimate constituents underlying enormous emergent physical phenomena in condensed matter systems [1, 53]. Particularly, when these two energies are comparable to the electronic bandwidth, diverse correlated phases with new topological properties are predicted to appear generally [2, 49, 54–56]. Pyrochlore iridates whose chemical formula is  $R_2Ir_2O_7$  (R: a rare-earth ion; see Fig. 3.5(a)) are an archetype showing such correlated topological properties that can possibly host various enthralling electronic states [1, 53]. In the paramagnetic metal (PM) phase, these materials are predicted to have a quadratic band crossing (QBC). There are doubly degenerate hole-like and electronlike bands touching at the  $\Gamma$  point [5]. Recently, in Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> [6] an ARPES approach finds electron dispersion which accords to QBC. When a magnetic phase transition takes place below  $T < T_N$ , various interesting electronic states show up through the QBC. For example, it is predicted theoretically that an antiferromagnetic (AF) Weyl semimetal (WSM) phase exist between a PM and an AF insulator (AFI) where all-in all-out (AIAO) magnetic order, as shown in Figs. 3.5(b) and 3.5(c) [2, 4, 5, 21, 34].

Nevertheless, in the real world, the WSM phase only comes out in an infinitesimal window at the boundary between PM and AFI. The only exception is the case  $Pr_2Ir_2O_7$  where the PM prevails down to a few mK. However, by mixing R sites with Pr and Nd ions or applying physical pressure,  $T_N$  is systematically controllable, and the system approaches the quantum critical point (QCP). Near the QCP, a ground state of semimetal with AF ordering may be obtainable [9]. Fascinatingly, in systems near QCP, anomalous transports are observed, such as metallic magnetic domain walls, anomalous Hall effect, and magnetic field induced metal-insulator transi-



Figure 3.5: (a) The lattice structure of pyrochlore iridates. (Red: rare-earth, blue: iridium) (b) Schematics of all-in-all-out (AIAO) magnetic ordering. (c) Positions of Weyl nodes relevant to AIAO ordering.

tions [20, 25, 27, 39–42, 52, 57]. Especially, in the previous section, we discussed that the systematically tuned  $(Nd_{1-x}Pr_x)_2Ir_2O_7$  under pressure reach the QCP, and the unusual magneto-transport properties were found therein. We associated those properties with topological semimetals under a magnetic field emerging near QCP. [9]. By combining previous experimental and theoretical reports, we summarize the schematic phase diagram shown in Fig. 3.6. This implies that the magnetic field in the system near the QCP guarantees to produce various topological semimetals carrying point and line nodes.

The main purpose of this section is to arrange a general theoretical structure to understand the magnetic field-induced topological semimetals near the QCP of pyrochlore iridates. To discuss this issue, we set up the PM phase with QBC, and approach the QCP by including the magnetic

field and AIAO order. The QBC at the  $\Gamma$  point can be considered as the fourfold degenerate states of the total angular momentum J = 3/2. Because of the large J = 3/2 with strong spin-orbit coupling, a nontrivial Zeeman coupling comes out. The Zeeman field H generates an unconventional anisotropic Zeeman coupling ( $\propto \vec{H} \cdot \vec{J}^3$ ) in addition to the usual isotropic Zeeman coupling ( $\propto \vec{H} \cdot \vec{J}$ ). Moreover, the AIAO order plays a role as an additional magnetic energy scale. Because the exchange energy of AIAO ordering and the two different Zeeman couplings are comparable near the QCP, the energy competition creates a number of novel topological semimetal phases in accordance with the low-energy theory. Considering the microscopic degrees of freedom in the lattice, we prove that the transaction between three distinct magnetic energy scales can be compactly depicted by magnetic multipole moments (MMMs) of the spin clusters in the unit cell. Magnetic field-induced tune of MMMs in the unit cell and its coupling to the degenerate states at QBC lie at the most essential part of emergent topological semimetals of pyrochlore iridates near the QCP.

#### 3.2.2 Quadratic band crossing and AIAO ordering

Figure 3.7(a) shows the QBC in the PM phase [4–6, 58]. Since each energy band is doubly degenerate by the time-reversal and inversion symmetries, the  $\Gamma$  point has fourfold degeneracy. The fourfold degeneracy is considered to carry the total angular momentum J = 3/2. The low-energy physics of the QBC can be effectively expressed by the Luttinger Hamiltonian [59],

$$H_0(\vec{k}) = \epsilon_0(\vec{k}) + \sum_{i=1}^5 d_i(\vec{k})\Gamma_i,$$
(3.1)

where  $\epsilon_0(k) = k^2/2m$  and  $\Gamma_i$  are the 4 × 4 matrices obeying the Clifford algebra  $\{i, j\} = 2\delta_{ij}$  (i, j = 1 - 5). A complete set of Hermitian 4 × 4



Figure 3.6: A schematic phase diagram near QCP which summarizes the previous section, as a function of electronic correlation U, temperature T, and magnetic field h for a given electronic bandwidth.

matrices is established by defining 10 Hermitian matrices as  $\Gamma_{ij} = [i, j]/2i$ and the identity matrix. The function  $d_{1-5}(\vec{k})$  is *d*-orbital like second order polynomials.

When the AIAO order develops in Ir spins below  $T_N$ , the QBC is broken into 4 pairs of Weyl nodes (WNs). Each pair is confined at the lines along threefold rotational axes [4]. The WSM with 4 pairs of WPs can be expressed by adding  $H_{AIAO} = -\alpha\Gamma_{45}$  to Eq. 3.1. Note that  $\alpha \propto Um_{AIAO}$ where U is the local Coulomb repulsion and  $m_{AIAO}$  represents the magnetic moment magnitude at each site of the AIAO order. The distance between each WN pair is proportional to  $\sqrt{|\alpha|}$ . Hence, when  $\alpha > \alpha_c$  for some crit-



Figure 3.7: (a) The quadratic band crossing (QBC). (b) The energy level splitting of J = 3/2 eigenstates by isotropic and anisotropic Zeeman terms.

ical value, WN pairs reach the Brillouin zone boundary and pair-annihilate at L points, and the system becomes a trivial insulator. Previous study [5] showed that WN pair creation at  $\Gamma$  point and annihilation at L points finish only within 1 % change of U/t, where t is the nearest-neighbor hopping. Accordingly, the WSM phase emerges in a very small region of the phase diagram, so it is very hard to achieve WSM in experiments.

#### 3.2.3 Topological semimetals induced by Zeeman field

Let us now consider that a magnetic field is applied to the semimetal with QBC. The external Zeeman field  $\vec{H}$  applied for QBC gives the follow-



Figure 3.8: (a) Energy levels of J = 3/2 states as a function of  $\theta = \tan^{-1}(\beta_2/\beta_1)$ , and the associated phase diagram. DWSM (LSM) denotes a pair of double Weyl nodes (a nodal line with a pair of Weyl nodes). (b) Positions of point/line nodes in DWSM and LSM.

ing terms,

$$\mathcal{H}_B = -\beta_1 \vec{B} \cdot \vec{J} - \beta_2 \vec{B} \cdot \vec{J}^3, \qquad (3.2)$$

where  $\vec{J} = (J_x, J_y, J_z)$ ,  $\vec{J}^3 = (J_x^3, J_y^3, J_z^3)$ , and  $\vec{B}(\vec{H}, \vec{M}, ...)$  is the effective Zeeman field as a function of H and the magnetization  $\vec{M}$ .  $\beta_1$  and  $\beta_2$  determine the weight of isotropic and anisotropic Zeeman couplings, respectively. The anisotropic Zeeman coupling with  $\vec{J}^3$  appears because of spin-orbit coupling and the large J = 3/2. Usually, the anisotropic Zeeman term is proportional to spin-orbit coupling and negligibly contributes to Zee-

man splitting [60, 61]. Nonetheless, in pyrochlore iridates, the anisotropic Zeeman term is large enough to control the energy splitting at the  $\Gamma$  point can be controlled.

Generally, the isotropic Zeeman term breaks the degenerate eigenstates at the  $\Gamma$  point into different energy levels. The eigenstates are equally space, as shown in Fig. 3.7(b). Thus, when  $\beta_1 \neq 0$  and  $\beta_2 = 0$ , a level crossing between different  $J_z$  at the point does not occur by  $\vec{H}$ . On the other hand, when both the isotropic and anisotropic Zeeman terms exist at the same time, the level crossing occurs between different  $J_z$  occurs, and the energy sequence is rearranged by  $\beta_2/\beta_1 \equiv \tan \theta$ . Figure 3.8(a) presents the change of energy levels at  $\Gamma$  as a function of  $\theta$  under  $\vec{H} \parallel [001]$ . The level crossing appears at several angles  $\theta_c$ , where phase transitions between different topological semimetals occur. When  $\vec{H} \parallel [001]$ , a double Weyl semimetal (DWSM) where a pair of WNs with the charge  $\pm 2$  at the  $k_z$  axis, or a linenode semimetal (LSM) where a nodal line on the  $k_z = 0$  plane with a pair of WNs with the charge  $\pm 1$  at the  $k_z$  axis appears. This is shown in Fig. 3.8(b). On the other hand, when  $\vec{H} \parallel [111]$ , the system has a lower symmetry than that under  $\vec{H} \parallel [001]$ , the band crossing occurs more simply. The detail is shown in Ref. [10].

In real materials under the magnetic field, AIAO order and two Zeeman terms exist at the same time. Then, the extended Luttinger model  $H_{extended}$  $\equiv H_0 + H_{AIAO} + H_B$  can capture the most generic low-energy band structure. We obtain the general phase diagram in the two-dimensional  $(\theta, \phi)$ plane because we have three distinct energy scales. Note that  $\phi \equiv \tan^{-1}(B/\alpha)$ is the measure of the relative strength of the Zeeman term to the AIAO ordering. Diverse novel topological semimetal arise by varying  $\theta$  and  $\phi$ , as shown in Fig. 3.9(a)



Figure 3.9: (a) The most general phase diagram under  $B \parallel [001]$  as a function of  $(\theta, \phi)$  from the extended Luttinger model. 4P WSM denotes a WSM with four pairs of Weyl nodes, while 2P WSM indicates two pairs of Weyl nodes. Two types of 2P WSM exist. The solid lines are the trajectory from the mean-field lattice model when the magnetic field changes the spin orientation in a unit cell. (b) Positions of Weyl nodes in T1/T2-2P WSM.

## 3.2.4 Lattice model and cluster magnetic multipole moments

Here we present a microscopic illustration of magnetic field-induced topological semimetals. We adopt a tight-binding Hamiltonian  $H = H_0 + H_U + H_Z + H_{fd}$ , where  $H_U = U \sum_i n_{i\uparrow} n_{i\downarrow}$  is the on-site Hubbard re-

pulsion,  $H_Z = \sum_{i,s} c_{i,s}^{\dagger} \frac{(\vec{H} \cdot \sigma_{ss'})}{2} c_{i,s}$  indicates the Zeeman effect on Ir spins, and  $H_{fd}$  is the coupling between Ir and Nd moments.  $c_{i,s}^{\dagger}(c_{i,s})$  is the creation (annihilation) operator for electrons whose spin  $s = \uparrow, \downarrow$  on the *i*th site, and  $n_{i,s} = c_{i,s}^{\dagger} c^{i,s}$  is the number operator. Here, each Ir ion has an effective spin-1/2 described by the Pauli matrix  $\sigma$ . The hopping between Ir sites is depicted by

$$H_{0} = \sum_{ss'} \sum_{\langle ij \rangle} c^{\dagger}_{i,s} (t_{1} + it_{2}\vec{d}_{ij} \cdot \vec{\sigma}_{ss'}c_{j,s'}) + \sum_{\langle \langle ij \rangle \rangle} c^{\dagger}_{i,s} (t'_{1} + i\{t'_{2}\vec{R}_{ij} + t'_{3}\vec{D}_{ij}\} \cdot \vec{\sigma}_{s,s'})c_{j,s}]$$
(3.3)

where  $t_1(t'_1)$  is the spinless nearest-neighbor (next-nearest-neighbor) hopping, and  $t_2(t'_{2,3})$  implies spinful nearest-neighbor (next-nearest-neighbor) hopping. The hopping defined above includes both the oxygen-mediated hopping  $t_{oxy}$  and the direct hopping between Ir ions [4, 5]. We apply meanfield theory to the Hubbard repulsion,  $(H_U \approx H_U^{MF})$ . The local order parameters adopted here are  $m_{\alpha} \equiv \frac{1}{2N} \sum_k \langle c^{\dagger}_{\alpha,s}(\vec{k}) \vec{\sigma}_{s,s'} c_{\alpha,s'}(\vec{k}) \rangle$  where  $\alpha = 1, 2, 3, 4$  is the sublattice site in a unit cell. For  $H_{fd}$ , Nd moments are considered classically. The detail is in Ref. [10].

Figure 3.10(a) shows the energy band of PM from  $H_0$ . Just as the Luttinger Hamiltonian described above, there is a QBC at the  $\Gamma$  point. We depict the spin configurations of four degenerate wavefunctions at the QBC in Fig. 3.10(a), to understand their nature. One interesting property of four degenerate wavefunctions is that the wavefunctions carry cluster magnetic multipole moments (CMMMs) intrinsically. Namely, the two states with  $J_z = \pm 3/2$  have dipolar moments whereas the other two states with  $J_z = \pm 1/2$  have the linear combination of dipolar and octupolar moments. Because of this intrinsic CMMM, those four states can naturally couple to magnetic orders.

The MMMs for a spin cluster were introduced in Ref. 7. Similar to the



Figure 3.10: (a) Depiction of the eigenfunctions for the four degenerate states at QBC which carry the linear combination of dipolar and octupolar orders. (b) A general spin configuration under  $\vec{H} \parallel [001]$  is divided into magnetic multipolar components. (c) The amplitude change of each multipolar component as a function of rotational angle  $\zeta$ . The spin configuration in a unit cell is changed from AI to 2I2O under  $\vec{H} \parallel [001]$ .

multipole moments of an atom [62], one can define the rank-p MMM of a given cluster  $\mu$  as  $M_{pq}^{\mu} = \sqrt{\frac{4\pi}{2p+1}} \sum_{i=1}^{N_c} \vec{m}_i \cdot \nabla_i (|R_i|^p Y_{pq}^*(\theta_i, \phi_i))$  where  $N_c$  is the number of atoms in  $\mu$  cluster,  $q \in [-p, p]$  is the magnetic quantum



Figure 3.11: Phase diagrams from self-consistent mean-field calculations under  $H \parallel [001]$ . For (a), Ir is an Ising spin, while for (b), Ir is a Heisenberg spin. The brown (green) solid line in (a) [(b)] matches to the brown (green) curve in Fig. 3.9(a) by the spin configuration. We use the projection matrix in Eq. 3.5 to obtain  $\alpha$ ,  $\beta_1$  and  $\beta_2$  of extended Luttinger model.

number,  $(R_i, \theta_i, \phi_i)$  are the spherical coordinates of the *i*th atom,  $\vec{m}_i$  is the magnetic moment on atom *i* of  $\mu$  cluster, and  $Y_{pq}(\theta_i, \phi_i)$  is the spherical harmonics of rank *p*. By summing for all clusters in the magnetic unit cell, one acquires the *p*-th order of the CMMM.

We analyze the CMMM of a tetrahedral unit cell as the following.

Since there are four atoms in the unit cell, we classify a total of 12 independent spin degrees of freedom in a unit cell by using group theory. The symmetrized spin configurations of CMMMs are considered as a basis. The general spin configuration can be represented by the linear combination of the basis. For example, the most general spin configuration under  $\vec{H} \parallel [001]$  in a unit cell satisfies the crystalline symmetry  $C_{2z}$  and  $\sigma_d T$ . This can be written as

$$|\psi\rangle_{[001]} = a_D |D\rangle + a_{T_1} |T_1\rangle + a_{A_2} |A_2\rangle, \tag{3.4}$$

where  $|D\rangle$ ,  $|T_1\rangle$ , and  $|A_2\rangle$  are the states having magnetic dipolar,  $T_1$ -octupolar, and  $A_2$ -octupolar moments, and  $a_D$ ,  $a_{T_1}$ , and  $a_{T_2}$  are their amplitudes. (See Fig. 3.10(b).)  $a_D$ ,  $a_{T_1}$ , and  $a_{T_2}$  varies continuously when spin configurations are modulated, as shown in Fig. 3.10(c).

Now we illustrate how the intrinsic CMMMs of the QBC states couple to the CMMM of magnetic order. In fact, the CMMM of a lattice system and the three energy scales  $\alpha$ ,  $\beta_1$ ,  $\beta_2$  of the extended Luttinger model has a close relation. The relation can be revealed when one project the effective Zeeman term in the lattice,  $H_B = \frac{1}{2} \sum_{i,s} \vec{B}_{\text{eff},i} \cdot [c_{i,s}^{\dagger} \vec{\sigma}_{s,s'} c_{i,s'}]$ , to the subspace spanned by the QBC wavefunctions. Here, the effective magnetic field at sublattice i,  $\vec{B}_{\text{eff},i}$ , includes all interactions in the mean-field theory.  $\vec{B}_{\text{eff},i}$  is decided self-consistently for given  $\vec{H}, U, J_{fd}$  and hopping parameters. The projection operator is defined as  $\hat{P}_J = \sum_{J_z} |J_z\rangle \langle J_z|$ , where  $|J_z\rangle$ are the QBC states with  $J_z = \pm 3/2$  and  $\pm 1/2$ , then

$$\hat{P}_J H_B \hat{P}_J = M_{A_2} \Gamma_{45} + \left[\frac{2}{3}M_D - \frac{9}{4}M_{T_1}\right] + M_{T_1} J_z^3.$$
(3.5)

for  $\vec{H} \parallel [001]$ . Here,  $M_{A_2}$ ,  $M_D$ ,  $M_{T_1}$  denote the  $A_2$ -octupole (or AIAO order parameter), the magnetic dipole, and the  $T_1$ -octupole each. It is noteworthy that  $M_D$  and  $M_{T_1}$  decide the relative strength between the isotropic and anisotropic Zeeman couplings. Because the linear combination of CMMMs gives the three energy scales  $\alpha$ ,  $\beta_1$ , and  $\beta_2$ , diverse topological semimetals expected in the extended Luttinger model can simply come out by changing the spin configurations to control the CMMMs.

To describe this idea, we fix  $\alpha$ ,  $\beta_1$ , and  $\beta_2$  by projecting the lattice model for distinct spin orientation changes. We plotted the associated trajectories in Fig. 3.9(a). The red (blue) line in Fig. 3.9(a) denotes the trajectory of the process that spins in a unit cell continuously from the AIAO configuration to the 2-in 2-out state by a magnetic field. For the distinct spin orientation change processes, the CMMM and  $\alpha$ ,  $\beta_1$ , and  $\beta_2$  change differently, resulting in distinct trajectories and topological semimetals.

In real materials, the spin orientation change process under a magnetic field depends on the microscopic parameters like H, U, and  $J_{fd}$  in self-consistent mean-field theory. Figure 3.11 shows two different phase diagrams in the (H, U) plane from self-consistent calculations. Whether Ir spins are considered as an Ising or a Heisenberg spin, we obtain disparate topological phase diagrams. Although topological phase diagrams are disparate, the origin of emergent topological semimetals can be understood based on Fig. 3.9(a) for both cases. For instance, the projected meanfield Hamiltonian along the brown (green) line in the left (right) panel in Fig. 3.11 corresponds to the brown (green) trajectory in Fig. 3.9(a), manifesting the origin of topological semimetals. The emergence of diverse topological semi-metals can be well described by the QBC coupled to three distinct energy scales  $\alpha$ ,  $\beta_1$ , and  $\beta_2$  in the extended Luttinger Hamiltonian.

So far, we consider only a Kramers Nd ion for fd-exchange coupling. On the other hand, the role of the non-Kramers Pr ion should be also considered to apply our theory to  $(Nd_{1-x}Pr_x)_2Ir_2O_7$ . Because of the distinct symmetries of Nd and Pr spins, the form of fd-exchange is also quite disparate [62]. For example, the local in-plane components of the spin operators are electric quadrupoles for  $Pr^{3+}$  while they are magnetic dipoleoctupoles for Nd<sup>3+</sup> [63]. As a result, Pr in-plane spins couple to Ir charge density rather than Ir spin density [26]. Nevertheless, such a change in the fd-exchange coupling can just modify the trajectory in the phase diagram but not the phase diagram itself in Fig. 3.9. This can be apprehended by the change of  $\alpha$ , $\beta_1$ , $\beta_2$  in our extended Luttinger Hamiltonian.

#### 3.2.5 Conclusion

Near the QCP of pyrochlore iridates, the magnetic field-induced topological semimetals are appreciated through the energy band near the  $\Gamma$  point. However, in systems far from the QCP, accidental band crossings away from the  $\Gamma$  point might appear, which changes the total number of WNs. We deal with this problem in Ref. [10].

The QBC near the Fermi level is the key factor for the magnetic fieldinduced topological semimetals in Fig. 3.9(a). Hence, a broad class of materials carrying a similar low-energy band structure, like HgTe [64] or GdPtBi [58] can show similar behavior. Yet, it is noteworthy that in pyrochlore iridates, the non-coplanar magnetic order strengthens the anisotropic Zeeman term in the effective Hamiltonian. This is because the anisotropic Zeeman term depends on the cluster magnetic  $T_1$ -octupolar moment, as shown in Eq. 3.5. The conventional Zeeman term dominates over the anisotropic one in HgTe and GdPtBi, because HgTe is paramagnetic and GdPtBi is collinearly antiferromagnetic. Thus, for these materials, the eligible accessible topological semimetal phases would be limited.

From now on, we discuss the effect of disorder on Fig. 3.9(a), since the Pr doping imports the weak disorder effect in the system. All topological semimetals in Fig. 3.9(a) create small electron or hole pockets surrounding their nodal points or lines deviated away from the Fermi level since the magnetic field breaks the crystalline symmetry. As it is well acknowledged in conventional metals, the weak disorder is an irrelevant perturbation, and

the effect of the disorder is ignored. Although the Weyl points are located exactly at the Fermi level, the weak disorder is still marginally irrelevant in the renormalization group theory [43, 65–67]. The disorder effect in a nodal line semimetal is more delicate [68]. Because the energy of a nodal line is not at the Fermi level at the same time and small Fermi surfaces from Weyl nodes exist, the weak disorder is irrelevant in the LSM phase as well. We believe that what we propose remains true even with the weak disorder.

Lastly, we discuss the magnetic fluctuation near the QCP [44, 69]. It is well known that the modest screening of the Coulomb interaction in QBC induces non-Fermi liquid and unconventional magnetic quantum criticality. However, the magnetic field breaks cubic lattice symmetry allowing the system to have electron or hole pockets near the Fermi level. All topological semimetals in Fig. 3.9(a) have a Fermi surface with nodal points or lines near the Fermi level. In this case, the magnetic phase transition of the AIAO order is expressed by the conventional Hertz-Millis type theory.

## **Chapter 4**

# Part II: Strain-induced topological semimetals in pyrochlore iridates

## 4.1 Domain wall-induced anomalous Hall Effect in pyrochlore iridates

#### 4.1.1 Introduction

The magnetic domain wall (DW) modifies the symmetry and magnetic order, creating new topological properties [70–75]. The research on conductive DWs in ferroelectric thin films helped to understand the fundamental mechanisms, and the DW-based device manufacture [70–73]. The DWs of some ferromagnets show magnetic skyrmion-based topological Hall Effect [74, 75]. However, for DWs of antiferromagnets, topological properties have never been observed. Theoretically, it was expected that the anomalous Hall effect (AHE) can emerge from antiferromagnetic (AFM) DWs [76], but only a few experimental attempts are performed.

After the Weyl semimetal is predicted to exist in AFM  $Y_2Ir_2O_7$  [2], the family of pyrochlore iridates has captivated a great interest from the community [1]. Especially, Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> (NIO) has been frequently studied because it is close to the metal-insulator transition (MIT) [3, 77] and the DW is conductive [42]. Let us discuss the AFM spin configuration of NIO in detail. NIO is composed of two distinct chains of tetrahedra, which are occupied by Ir and Nd ions, respectively. (See Fig. 4.1(a).) All Ir spins in a tetrahedron point inward, and all Ir spins in the neighboring tetrahedron point outward at the same time. (This is not shown in the figure.) Thus, it is called all-in–all-out (AIAO) ordering, which we denote as Ir:AIAO. Simultaneously, Nd spins in



Figure 4.1: Schematics of domain wall structures in Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films. (a) When H = 0, Nd spins form AOAI. By fd-exchange, Ir spins form AIAO. (b) When  $H \parallel [111]$ , Nd spins become 3I1O by the Zeeman effect. Accordingly, Ir spins become tilted AOAI through fd-exchange. (c) Schematics of the AOAI / AIAO domain wall. (d) The enlarged part of the black box in (c). The red arrows are the local magnetic moments. On the domain wall (purple region), finite magnetization arises (purple arrows).

a tetrahedron that is adjacent to Ir one have a similar spin configuration that is a time-reversal partner of AIAO. We denote it as Nd:AOAI. The physical properties of pyrochlore iridates come from the intriguing magnetic orders and coupling between two distinct sublattices [1–3, 25, 42, 63, 77].

When we apply an external magnetic field H, the domain switching

between AIAO and AOAI occurs. Let us understand the DW formation process during the domain switching in an NIO sample. Since Nd ions have a stronger magnetic moment than Ir ones, the Zeeman energy of Nd is also larger than that of Ir. When  $H \parallel [111]$  is applied and increases, Nd spins are flipped and the order changes to a 3-in-1-out (311O) configuration [27]. When we increase H further, Nd spins can flip the Ir spins to form an AOAI configuration through fd-exchange interaction. (See Fig. 4.2(b).) During the spin-flip process, Ir:AIAO and Ir:AOAI exist simultaneously, so the DW forms as shown in Fig. 4.1(c).

Here, we suggest that an NIO epitaxial thin film is an ideal playground to realize antiferromagnetic DW-based topological responses. We assume that the electronic energy band structure of  $Nd_2Ir_2O_7$  is similar to a Weyl semimetal [2, 50]. The Weyl semimetal-like band structure gives rise to a colossal DW conductance [27, 42]. If an NIO is composed of either AIAO or AOAI domain solely, AHE is canceled out by the cubic symmetry of the system [7, 78]. However, if DWs exist, twofold rotation symmetries are broken, and magnetic moments near DWs give finite magnetization. Accordingly, a finite anomalous Hall conductivity arises in the plane made out of highly conducting DW. To find DW-based unconventional magnetotransport, we maximize the effect of DW by searching the low-dimensional NIO thin films [79].

#### 4.1.2 Experiments

#### 4.1.2.1 Deposition of Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films

Our experimental team prepares for the NIO (111) thin films on Ystabilized ZrO2 (YSZ) (111) substrates by pulsed laser deposition with a post-annealing process. They illuminate an NIO polycrystalline target with a KrF excimer laser with  $\lambda = 248$  nm. Its laser fluence and frequency are 4.5 J/cm<sup>2</sup> and 3 Hz each. The distance between the target and the substrate is



Figure 4.2: (a) X-ray Diffraction data of  $Nd_2Ir_2O_7$  thin film (red lines) with YSZ (111) substrate (black lines). (b) (222) diffraction pattern of  $Nd_2Ir_2O_7$  thin film. (c) Rocking curve of  $Nd_2Ir_2O_7$  and the YSZ substrate. (d) The reciprocal map of  $Nd_2Ir_2O_7$  (662).

50 mm. Pyrochlore iridate thin films are infamously difficult to deposit since the iridium is volatile [80–82]. Hence, to form the pyrochlore phase, high oxygen pressure and temperature during the deposition process are compulsory [83]. During the deposition, since the IrO3 gas is prone to form and evaporate, in-situ growth is notoriously difficult. They deposit stoichiometric amorphous Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films at 600  $^{o}C$  firstly. After then, they postanneal the thin films in an electrical box furnace at 1000  $^{o}C$  for 1 hour [84].



Figure 4.3: (a) The Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin film on YSZ (111) substrate is caught in a STEM image from  $[2\bar{1}\bar{1}]$  direction. Bright gray is for Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>, while dark gray is for the YSZ substrate. The fast Fourier transform of the red squares (b) in Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin film and (c) in YSZ substrate.

The X-ray diffraction (XRD)  $\theta - 2\theta$  scans of the NIO thin film on the YSZ substrate is shown in Fig. 4.2(a). In this figure, only peaks from NIO (111) and YSZ (111) are present, implying that it is in a single phase, Figure 4.2(b) presents the enlarged XRD peak near NIO (222). Figure 4.2(c)



Figure 4.4: (a) The longitudinal resistivity  $\rho_{xx}$  of Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> single crystal (blue dots) [27] and thin films (a red line) as a function of temperature T. (b) The magnetoresistance (MR) of Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin film for diverse  $H \parallel [111]$  from 0 to 14 T, as a function of temperature.  $T_N^{Nd} = 15$  K and  $T_N^{Ir} = 30$  K indicates the ordering temperature of Nd and Ir magnetic moments. (c) The Normalized MR values as a function of temperature show that the sign change of derivative of MR occurs for H > 7 T at  $T_N^{Nd}$ . (d) Normalized MR at 2 K under  $H \parallel [111]$  from a single crystal [27] (blue dots) and thin films (a red line)

displays that the width of the rocking curve is  $\sim 0.05^{\circ}$ , which implies that the NIO sample is highly crystalline. We map X-ray diffraction into the reciprocal space to compare the in-plane lattice constant of NIO film with that of YSZ substrate. Figure 4.2(d) offers the reciprocal map around the YSZ (662) and NIO (331) Bragg reflections. The  $Q_x$  and  $Q_z$  of the NIO (662) Bragg peak are smaller than those of the YSZ (331) one. We determine the lattice constants of NIO thin film to be a = b = 10.380 Å, that is similar to those of NIO polycrystals (a = b = c = 10.375 Å). Accordingly, the experimental team raises the NIO thin films with high structural quality.

### 4.1.2.2 Scanning transmission electron microscopy measurements

We probe the micro-structure of our NIO thin films by scanning transmission electron microscopy (STEM). Figure 4.3(a) displays a STEM image of the NIO thin film on a YSZ (111) substrate from  $[2\overline{1}\overline{1}]$  crystalline direction. A blurry area near the interface of thin film and substrate comes from the structural defects or the damage by the electron beam of STEM measurements. We take the red box regions from NIO thin films and YSZ substrates in Fig. 4.3 and perform fast Fourier transforms (FFTs). which are in Figs. 4.3(b-c). The epitaxial relationship between NIO and the YSZ substrate can be determined by NIO [111] || YSZ [111] and Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> [011] || YSZ [011].

#### 4.1.2.3 Magnetotransport measurements

The dc magnetotransport properties are measured. During these magnetotransport measurements, the current is applied along  $[1\bar{1}0]$ , and H is applied along [111] that is perpendicular to the current. Figure 4.4(a) shows that the NIO thin film displays a MIT near  $T_N^{Ir} \sim 30$  K. However,  $\rho(2K)/\rho(300K) \sim 10$  is much smaller than the single crystal value  $\rho(2K)/\rho(300K) \sim 1000$ . Note that  $\rho(2K)/\rho(300K)$  in single crystals also varies in a vast range of 10 - 1000 as well [51]. Therefore, the NIO thin films and single crystals have similar sample quality [51].

NIO thin films are cooled down to 2 K without field, and the mag-



Figure 4.5: (a) Normalized MR under  $H \parallel [111]$  at diverse temperatures from 2 to 40 K. (b) The asymmetric components of increasing sweep MR at 1.8 K (H > 0). Region (I) is where Ir is AIAO and Nd is AOAI, Region (II) is where Ir is AIAO and Nd is 311O, and Region (III) is where Ir is AOAI and Nd is 311O. The red (blue) and orange lines are the hysteresis of Ir (Nd) domain switching and of the total, respectively. (c) The fit parameters *a* from Eq. 4.2. Red circles (blue diamonds) are the hysteresis from Ir (Nd) domain switching. (d) The temperature dependence of coercive fields of the Ir (red circles) and Nd (blue diamonds) spins from MR data.

netoresistance is measured under H while T is raised up to 300 K. Figure 4.4(b) shows the change of resistance as a function of T under various  $H \parallel [111]$  from 0 to 14 T. The magnitude of MR gets increases as the temperature decreases. Figure 4.4(c) shows normalized MR curves as a function

of temperature at various H values. The MR peaks appear at  $\sim 15$  K which is close to  $T_N^{Nd}$  [34].

#### 4.1.3 Antiferromagnetic domain switching

#### 4.1.3.1 Magnetoresistance Hysteresis

Figure 4.4(d) exhibits the normalized MR curve as a function of H at 2 K compared with that of the single crystal [27]. The normalized MR curve of thin films exhibits a hysteresis with wider MR dips near  $\pm 3$  T than that of single crystals. Note that 3 T is near the coercive field  $H_C$  of the single crystal [27]. The dip structure comes from structural defects in the thin film and the fixation of DWs on such defects. On the other hand, the hysteretic behavior is the same as that of the single crystal (blue squares), implying that it comes from the DW switching.

Hysteresis appears below 30 K =  $T_N^{Ir}$ . In Fig. 4.5(a), we present normalized MR curves under  $H \parallel [111]$  at diverse temperatures. The MR changes its sign from positive to negative when the temperature decreases near  $T_N^{Nd} \sim 15$  K [34]. (See Fig. 4(c).) The dip structures are easily seen from 5 to 15 K, and near  $H_C = 3$  T of single crystal as mentioned above [27]. It is already reported that the hysteresis comes from AFM domain switching. NIO thin film data can be also explained in terms of the AIAO and AOAI domain switching.

## 4.1.3.2 Asymmetric analysis on hysteretic magnetoresistance curves

The asymmetric part of MR is known as containing magnetic domain switching information [84, 85]. The asymmetric part of the normalized MR curves is defined as

$$MR_{Asym}(H) = \frac{\rho_{xx}(H) - \rho_{xx}(-H)}{2\rho_{xx}(0)}$$
(4.1)

Figure 4.5(b) exhibits the  $MR_{Asym}(H)$  curve (black dots) at 1.8 K the increasing field sweep from 0 to 30 T. Note that the  $MR_{Asym}(H)$  curve has a peak and a dip in sequence. Furthermore,  $MR_{Asym}(H)$  changes its sign around 3.0 T, which has never been observed in other magnetic materials.

By analyzing the asymmetric MR curve, we can get an intuition of magnetic order and DW dynamics. In most ferroic materials, domain switching occurs at the coercive field  $H_C$ . When the domain switching is widened by fixing DWs to defects, asymmetric MR near  $H_C$  follows a Gaussian function [86, 87]. That is, by considering the asymmetry, we use

$$MR_{Asym,FIT}(H) = a(e^{-b(H-H_c)^2} - e^{-b(H+H_c)^2}).$$
 (4.2)

where a and b are arbitrary constants, which represent the magnitude and width of the hysteresis, respectively.

However, the sign change in  $MR_{Asym}(H)$  in Fig. 4.5(b) cannot be explained by a single Gaussian function in Eq. 4.2. This means that the hysteresis comes from the magnetic domain switching of both Ir and Nd sublattices. Thus, we introduce two domain switching functions to fit the experimental  $MR_{Asym}(H)$  curves. We show the red (blue) line for Ir (Nd) domain switching in Fig. 4.5(b). Both Ir and Nd spins play a role in magnetotransport in our NIO thin films.

Here is the reason that the red line is related to Ir and the blue line is related to Nd domain switching. To fit the MR, we use  $MR_{Ir}+MR_{Nd}$ .  $MR_{Ir}$ ( $MR_{Nd}$ ) is the red (blue) line calculated by Eq. 4.2 with  $a = a_{Ir}$  ( $a = a_{Nd}$ ). Figure 4.5(c) shows  $a_{Ir}$  and  $a_{Nd}$  as a function of temperature. When the temperature decreases,  $a_{Ir}$  appears near  $T_N^{Ir}$ , and  $a_{Nd}$  appears near  $T_N^{Nd}$ . Note that  $a_{Ir} > a_{Nd}$  for all available temperatures since the conduction of NIO is dominated by the Ir-O network [1]. Hence, we conclude that both Ir and Nd come together to determine the magnetotransport.

#### **4.1.3.3** Domain switching caused by the *fd*-exchange

Figure 4.5(b) indicates that when H increases, Nd domain switching occurs earlier than Ir one. According to this, we summarize the process of domain switching as follows. First, when H turns on, the Ir-AIAO and Nd-AOAI dominate. Second, as H increases, Nd spins flip but Ir spins do not because of the Zeeman energy difference, Nd-311O and Ir-AIAO are formed Lastly, at strong H, as Nd spins flip the Ir spins, Ir-AOAI domain is formed. This switching process is in Fig. 4.1.

Figure 4.5(b) also implies that Ir and Nd spins are strongly coupled by fd-exchange. We determine the coercive fields where each Ir and Nd spin are switched by  $MR_{Asym}(H)$  curve, Figure 4.5(d) shows the fitted values for  $\mu_0 H_c^{Nd}$  (blue diamonds) and  $\mu_0 H_c^{Ir}$  (red circles). Nd spin switching is absent for  $T > T_N^{Nd}$ . When  $T < T_N^{Nd}$ , on the other hand,  $H_c^{Nd} \sim 2T$  appears. Ir-AIAO to AOAI domain switching appears when  $T < T_N^{Ir}$ , around  $H_C^{Ir} sim7$  T at 20 K. When  $T < T_N^{Nd}$ ,  $H_C^{Ir}$  decreases down to 3 T. The colossal decrease of  $H_C^{Ir}$  around  $T_N^{Nd}$  can only be explained by two sublattice systems and emphasizes the role of the fd-exchange interaction on Ir domain switching.

## 4.1.4 Anomalous Hall Effect caused by antiferromagnetic domain walls

#### **4.1.4.1** AHEs observed in Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films

We measure Hall resistivity  $\rho_{xy}$  of the NIO thin film under  $H \parallel [111]$ Figure 4.6(a) shows the  $\rho_{xy}$  curves as a function of  $\mu_0 H$  at various temperatures. When  $T < T_N^{Ir} = 30$  K, a hysteretic behavior with a hump is observed, which differs from the conventional Hall effect. The behavior becomes more pronounced as  $T < T_N^{Nd}$ . In ferromagnetic DWs, the same behavior has been observed, which is the topological Hall effect coming from magnetic skyrmions [74, 75].

We present the anomalous part of  $\rho_{xy}$  (AHE) at 2 K as black circles in Fig. 4.6(b). In the figure, the peaks appear ~ 2–3 T, which is close to the  $\mu_0 H_c^{Ir}$  values from the previous section. That is, the DWs play an important role in the AHE. Additionally, the experimental AHE saturates above 5 T, implying that the AHE comes from other origins than DWs, like the scalar spin chirality in the bulk. We hereafter denote the contribution to AHE from scalar spin chirality and DW as bulk AHE and DW AHE, respectively.

## 4.1.4.2 Bulk anomalous Hall Effect: Scalar spin chirality

The AHE in non-coplanar AFM is explained in terms of the scalar spin chirality. If there are three spins  $\vec{S}_i, \vec{S}_j$ , and  $\vec{S}_k$ , the scalar spin chirality is defined as  $\vec{S}_i \cdot \vec{S}_j \times \vec{S}_k$  [88], which is the solid angle subtended by the three spins. The AHE comes from the fictitious magnetic flux coming from the real space Berry phase which is proportional to the scalar spin chirality [15, 39, 89–94].

When the magnetic field is absent, both magnetization and scalar spin chirality are zero inside NIO with a single domain. However, when Nd-3I1O order and Ir-canted-AOAI are set in by a strong magnetic field as shown in Fig. 4.1(b) both magnetization and scalar spin chirality arises and the bulk NIO gives rise to the AHE. We here estimate  $\rho_{xy}$  values in such a case. Ir-AOAI (AIAO) is canted by the Nd-3I1O (1I3O) through *fd*-exchange. Therefore, Ir-AOAI generates scalar spin chirality which leads to a fictitious magnetic field parallel to the magnetization [95]. Furthermore, Nd has much larger magnetic moments (~ 2.4  $\mu_0 B/Nd$ ) than (~ 0.2 $\mu_0 B/Ir$ ) [27]. Ir spins do not contribute to magnetization as much as Nd ones.

Since the AHE depends on the number of Nd-311O mostly, we will focus on the Nd spin configuration. We quantify Nd-311O order by computing the magnetization per unit cell. After then, we infer the bulk AHE by the portion of the Nd-311O order. Let us consider the Hamiltonian that describes a single Nd unit cell under  $H \parallel [111]$ ,

$$H = -J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - \mu \vec{B} \cdot \sum_i \vec{S}_i - K \Phi B_x B_y B_z$$
(4.3)

where J is the exchange interaction,  $\mu$  is the Nd magnetic moment strength,  $\vec{B}$  is a magnetic field, and K is the coupling of AIAO order to the field, and  $\Phi$  is the AIAO order parameter. Note that

$$\Phi = \frac{1}{4\sqrt{3}} (S_{1x} + S_{1y} + S_{1z} + S_{2x} - S_{2y} - S_{2z} - S_{3x} + S_{3y} - S_{3z} - S_{4x} - S_{4y} + S_{4z}).$$
(4.4)

Here, since Nd spins have anisotropy, we consider Nd spin as an Ising one. Then, the Nd spins are given by  $\vec{S}_1 = \frac{\sigma_1}{\sqrt{3}}(1,1,1), \vec{S}_2 = \frac{\sigma_2}{\sqrt{3}}(1,-1,-1), \vec{S}_3 = \frac{\sigma_3}{\sqrt{3}}(-1,1,-1)$ , and  $\vec{S}_4 = \frac{\sigma_4}{\sqrt{3}}(-1,-1,1)$ . ( $\sigma_i = \pm 1$ .) Then, Eq.4.3 becomes

$$H = -J \sum_{\langle ij \rangle} \sigma_i \sigma_j - \mu \vec{B} \cdot \sum_i \vec{S}_i - K \Phi B_x B_y B_z \tag{4.5}$$

When J > 0 and  $\vec{B} = 0$ , the ground state is either AIAO ( $\sigma_i = 1, \forall i$ ) or AOAI ( $\sigma_i = -1, \forall i$ ). Then, the partition function is given by

$$Z = \sum_{\sigma_i = \pm 1} \exp(-\beta H), \tag{4.6}$$



Figure 4.6: (a) The Hall resistivity  $\rho_{xy}$  as a function of H under  $H \parallel [111]$  at diverse temperatures. The black arrows are the sweep directions. (b) Anomalous Hall resistivity, which is the total Hall subtracted by the normal Hall at 2 K. The black circles are the experimental data. The blue lines are the bulk anomalous Hall Effect. The red circles are the domain wall Hall Effect. (c) Anomalous Hall resistivity as a function of T and  $\mu_0 H$ . Red circles are the coercive field  $H_c^{Ir}$  where Ir domain switching occurs according to MR data.

and the expectation value of magnetization parallel to [111] is

$$\langle M \rangle = \frac{1}{Z} \sum_{\sigma_i = \pm 1} (\sigma_1 - \frac{1}{3} (\sigma_2 + \sigma_3 + \sigma_4)) \exp(-\beta H).$$
 (4.7)

We value the bulk AHE of NIO by assuming that the bulk AHE is proportional to the Nd magnetization. All DWs disappear when H is suf-
ficiently large, then the bulk AHE dominates over the DW AHE. We use the saturated Hall resistivity (~  $8\mu$  cm) at ~ 2 T ( $\mu_0 H_c^{Nd}$ ) in Fig. 4.6(b) in the experiment, to estimate the bulk AHE contribution analytically. The estimated bulk AHE is represented as the blue line in Fig. 4.6(b). We note here that the bulk AHE cannot explain the experiment solely, and DW AHE is necessary.

## 4.1.4.3 Domain wall anomalous Hall Effect: Symmetry analysis

Let us consider the emergence of AHE by momentum space Berry curvature through symmetry analysis. First, the anomalous Hall Effect vanishes in a single AIAO or AOAI domain of pyrochlore iridates [78]. The anomalous Hall conductivity is obtained by integrating the Berry curvature of the occupied energy bands.

$$\sigma_{\alpha\beta} = \frac{e^2}{\hbar} \int_{BZ} \frac{d^3k}{(2\pi)^3} \sum_n f(\epsilon_n(\vec{k}) - \mu) F_{\alpha\beta}(\vec{k})$$
(4.8)

where  $f(\epsilon_n(\vec{k}) - )$  is the Fermi-Dirac distribution, and  $F_{\alpha\beta}$  is the Berry curvature. For convenience, let us denote  $F_x(\vec{k}) = F_{yz}(\vec{k}), F_y(\vec{k}) = F_{zx}(\vec{k})$ , and  $F_z(\vec{k}) = F_{xy}(\vec{k})$ . When twofold rotation symmetry  $C_{2z}$  exists,

$$F_x(-k_x, -k_y, k_z) = -F_x(k_x, k_y, k_z),$$
  

$$F_y(-k_x, -k_y, k_z) = -F_y(k_x, k_y, k_z),$$
  

$$F_z(-k_x, -k_y, k_z) = F_z(k_x, k_y, k_z).$$
(4.9)

 $\sigma_{yz}$ ,  $\sigma_{zx}$  vanishes by the integration while  $\sigma_{xy}$  can be finite. In a single domain of AIAO or AOAI order, AHE vanishes for all directions since pyrochlore iridates are invariant under  $C_{2x}$ ,  $C_{2y}$ , and  $C_{2z}$  symmetries.

However, when DW is present, AHE can arise from the net Berry cur-



Figure 4.7: A domain wall of Ir pyrochlore lattice. Domain wall breaks the twofold rotation symmetries, giving rise to the anomalous Hall Effect. The direction of the anomalous Hall is parallel to the domain wall.

vature effect. The DW breaks all twofold rotation symmetries but keeps a threefold rotational symmetry about the axis perpendicular to the DW (see Fig. 4.7). Let the threefold rotation axis as [111]. Then, the Berry curvature changes as

$$F_{x}(k_{y}, k_{z}, k_{x}) = F_{y}(k_{x}, k_{y}, k_{z}),$$

$$F_{y}(k_{y}, k_{z}, k_{x}) = F_{z}(k_{x}, k_{y}, k_{z}),$$

$$F_{z}(k_{y}, k_{z}, k_{x}) = F_{x}(k_{x}, k_{y}, k_{z}).$$
(4.10)

Therefore, the Berry curvature components along [111] satisfy

$$F^{[111]}(\vec{k}) + F^{[111]}(C_3\vec{k}) + F^{[111]}(C_3^{-1}\vec{k}) \neq 0,$$
(4.11)

where  $C_3$  is the threefold rotation operator. This gives  $\sigma^{[111]} \neq 0$ , which

means the AHE occurs in the plane parallel to the DW.

# 4.1.4.4 Unconventional AHE at the antiferromagnetic domain walls of Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films

When  $H \parallel [111]$ , DWs are predicted to form in the (111) plane since it lowers the frustration [96, 97]. We provide the detail in Ref. 11. Moreover, in Cd<sub>2</sub>Os<sub>2</sub>O<sub>7</sub> single crystal whose ground state is also AIAO, the DW formation prefers to orient normal to  $\vec{H}$  [97]. Accordingly, in our experimental geometry, DWs in the (111) plane can play an important role.

Figure 4.6(b) shows anomalous part of  $\rho_{xy}$  at 2 K as a function of H. As aforementioned, we cannot interpret the exotic behavior of  $\rho_{xy}$  in terms of the bulk AHE solely. The bulk AHE is shown as the blue curve in Sec. 4.1.4.2. The red circles in Fig. 4.6(b) are the data that is not explained by the bulk AHE.

We reveal here that the red circles are related to DW AHE. The maximum value of red circles is  $\rho_{xy} \sim 15 \ \mu\Omega$  cm at 2 T which is twice as the saturated  $\rho_{xy} \sim 8 \ \mu\Omega$  cm. The maximum field 2 T agrees with the coercive field of Ir  $H_c^{Ir} \sim 3$  T from MR where the DWs play a significant role. The red circles tend to be the Gaussian functions which are also used in the fitting of MR<sub>Asym</sub>(H) [see Eq. 4.2]. This indicates that a broad DW switching occurs, which matches our MR analysis. Moreover,  $\rho_{xy} \propto \rho_{xx}$  [11] indicates that the AHE at  $H_c^{Ir} \sim 3$  T is intrinsic.

Furthermore, a contour plot of the anomalous part of  $\rho_{xy}$  in Fig. 4.6(c) as a function of T and H supports our argument as well. This supports a large enhancement of DW AHE. For a given T, the H value where  $\rho_{xy}$ is maximized is relevant to  $H_C^{Ir}$  from MR (red circles). The high density of DWs near  $H_C^{Ir}$  gives rise to a maximum DW AHE. The peak value of AHE is also greatly raised below  $T_N^{Nd}$ , This supports that the strong coupling between Ir and Nd through fd-exchange affects the magnetotransport. Experimental observation already reveals that the DWs of NIO are highly conductive than bulk NIO. The conductivity is an order of magnitude higher in DWs [20, 27]. In comparison, on  $Sm_2Ir_2O_7$  thin films where the DW conductance is lower [31], the hysteresis in AHE is absent. To sum up, the hysteretic AHE with humps observed in NIO thin films originates from DW.

In summary, we observe a large AHE in an NIO thin film and reveal that the AHE is attributed to the AFM DWs. The strong fd-exchange effectively lowers the energy barrier for Ir domain switching and makes it occur at the weak H-field. Ir-domain switching induces the net Berry curvature effect at the DW, resulting in a large DW AHE  $\sim 15\mu$  cm at 2 K.

# 4.2 Anomalous Hall Effect in Strained Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films

#### 4.2.1 Introduction

The anomalous Hall Effect (AHE) is a universal transport property that appears in a wide range of time-reversal symmetry broken systems. AHE can arise from either extrinsic or intrinsic ways [15]. The extrinsic mechanism denotes skew scattering or side jump because of magnetic impurities, while the intrinsic mechanism originates from Berry curvature in momentum space. Since the topology of electronic energy eigenstates is ciphered in the Berry curvature, AHE is an effective tool to examine the topological effects of condensed matter [98, 99]. The AHE is also widely utilized in applications like memory devices. [100]. From a conventional viewpoint, itinerant ferromagnets are the major source to observe the AHE. The anomalous Hall conductivity is known to be proportional to the magnetization [101], which is an order parameter of time-reversal symmetry breaking. However, in recent reports, the non-collinear antiferromagnets like Mn<sub>3</sub>X (X=Sn,Ge) and GdPtBi can exhibit a large AHE [94, 102-104]in spite of negligible magnetization. The report of large AHE in antiferromagnets implies that ferromagnetism is not the only necessary condition for AHE. An alternative source for AHE other than ferromagnetism is proposed in Ref. 7. The theoretical analysis showed that higher-rank magnetic multipole formed in a cluster of spins (so-called cluster multipoles) in antiferromagnet can induce the AHE when the multipole has the same symmetry as ferromagnetic order. Subsequently, the octupoles in spin clusters also give rise to the anomalous Nernst [105] and magneto-optical Kerr effects [106] in Mn<sub>3</sub>Sn. However, since antiferromagnets are not directly coupled to the electromagnetic fields [107], the cluster multipoles can hardly be manipulated. This forces strong limitations on experiments for antiferromagnetic AHE. Here, we describe that we apply a strain on antiferromagnetic Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> (NIO) thin film,

and show that the strain can create the AHE by producing the higher-rank cluster multipoles. We investigate that the biaxial strain on pyrochlore iridates tunes the spin structure and induces a certain type of cluster octupoles. Since the induced cluster octupoles have the same symmetry to ferromagnetic order, generating the net Berry curvature effect and thus a finite AHE as well. We anticipate that the strain-induced AHE can be applied to a wide range of spin-orbit–coupled topological magnets [7] and antiferromagnetic spintronics [100, 108].

# 4.2.2 Strain-induced cluster multipoles in a pyrochlore lattice

Pyrochlore iridates family has the chemical formula  $R_2Ir_2O_7$  (R: rareearth). This is a geometrically frustrated antiferromagnet with complex lattice structures. In Fig. 4.8A, we show that pyrochlore iridates are composed of corner-sharing tetrahedrons whose vertices are occupied by R and Ir. In  $R_2Ir_2O_7$ , both electronic correlations and spin-orbit coupling are large enough to induce a unique antiferromagnetic spin structure, so-called all-inall-out (AIAO) order [2, 3]. In the circle of Fig. 4.8B, all spins in a tetrahedron point either inward or outward simultaneously. The Néel temperatures where the AIAO order sets in are  $T_{Ir}^N$  30 K [3] for Ir and  $T_{Nd}^N$  15 K [20] for Nd, respectively. By breaking the time-reversal symmetry, the AIAO order induces correlated topological phases [10, 23] such as a Weyl semimetal.

Nevertheless, the AIAO ordering preserves the cubic symmetries, the net Berry curvature effect, and the integration of Berry curvature over the Brillouin zone (BZ), vanishes. If the cubic crystalline symmetries are kept, AHE does not appear in the system. There are many efforts to break the cubic symmetries by a magnetic field, in the pressured NIO and Pr-doped NIO single crystal samples [9, 10, 109]. However, the spin configuration tuned by the magnetic field is weak and readily turns back when the mag-



Figure 4.8: (A) The lattice structure of pyrochlore iridates. Yellow dots are Nd ions and red dots are the Ir ions. (B) Schematics of strained Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films on YSZ (111) substrate. The deformed lattice structure and spin configuration are shown in the circle. (C) The spin configuration without strain. AIAO order is expressed by an  $A_2$ -octupole solely. (D) The spin configuration under strain. The deformed AIAO order by strain is decomposed into an  $A_2$ -octupole, a dipole, and a  $T_1$ -octupole.

netic field is off. Hence, we highly desire a stable method to break the cubic symmetries. Here, we utilize strain engineering and investigate the transport phenomenon.

In Fig. 4.8B, we show that the biaxial strain deforms the unit cell along the [111] direction. This will break the cubic symmetries like twofold rotations and fourfold roto-inversions. Since the deformation tunes magnetic anisotropy of each spin [78], the orientation of Ir spins is adjusted. We adopt cluster multipole theory to describe the orientation of Ir spins systematically. Note that since Ir d-electrons are the main source of conduction, we consider Ir only [2]. Since there are 4 atoms in the unit cell, all Ir spin configurations can be classified into 12 distinct cluster multipoles under 5 different irreducible representations. The detail is in Ref. 10.

In a usual NIO single crystal (bulk), the ground state is the AIAO order that corresponds to an  $A_2$ -octupole (see Fig. 4.8C). Since the  $A_2$ -octupole keeps most cubic symmetries like twofold rotations, AHE is canceled out. However, in a strained NIO (*s*-NIO), the ground state is tuned from the AIAO order. We denote the tuned spin configuration by strained AIAO (*s*-AIAO). *s*-AIAO is expressed by a linear combination of a dipole, an  $A_2$ octupole, and a  $T_1$ -octupole in Fig. 4.8D. The dipole is the ferromagnetic order, while the  $T_1$ -octupole is a type of antiferromagnetic order different from AIAO. We here maintain that the  $T_1$ -octupole can induce the AHE in our system.

#### **4.2.3** Characterizations of *r* and *s*-NIO thin films

Here the sample preparation of our experimental group is explained. They prepare two different NIO thin films on the YSZ (111) substrates: relaxed (*r*-NIO) and fully strained (*s*-NIO) films. Since the lattice constants of NIO and YSZ mismatch, the biaxial strain arises as shown in Fig. 4.8B [16, 110]. Since the twice of the lattice parameter of YSZ is smaller than that of NIO, the strain is compressive. The strain is estimated as  $\epsilon = (2a_{YSZ} a_{NIO})/a_{NIO} \approx 0.96\%$ , where  $a_{NIO} = 10.38$ Å and  $a_{YSZ} = 5.14$ Å.

Numerous efforts [81, 82, 84] showed that the *in-situ* growth of pure NIO thin film is extremely challenging. Although the proper crystalline growth conditions are imposed on pyrochlore oxides [83], NIO is unsettled



Figure 4.9: (A) The X-ray diffraction data of Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> film on a YSZ (111) substrate. (B) Reciprocal map around YSZ (331) and NIO (662) peaks. Since  $Q_x$  values of two peaks are the same, the compressive strain of 1% is applied to Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films. (C) STEM image from [110] crystalline direction shows the clear interface between Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin film and substrate. The colored dots denote the position of Nd and Ir ions. The fast Fourier transform of the green box is in (D) and that of the light blue box is in (E). (D-E) indicate that the lattice constants of Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> and YSZ are the same.

because of the production of  $IrO_3$  gas [111]. To remove this instability, the "solid-phase epitaxy (SPE)" [81, 84] method has been used. In SPE, amor-



Figure 4.10: (A) The longitudinal resistivity  $\rho_{xx}$  as a function of temperature T. The orange line (black dotted line) is from s-NIO (r-NIO). The energy band structure in (B) r-NIO and (C) s-NIO. r-NIO is a trivial insulator and s-NIO is a trivial semimetal, which explains the change of  $\rho_{xx}$  in (A). (D) The anomalous Hall conductivity  $\sigma_{xy}^A$  of s-NIO (orange dots) and r-NIO (black dotted lines) thin films. s-NIO has an order larger  $\sigma_{xy}^A$  than r-NIO. Arrows are the sweep directions. Anti-hysteresis is observed. (E) The Berry curvature along high-symmetry lines of Brillouin zone of r-NIO (upper panel) and s-NIO (lower panel). The Berry curvature is larger in r-NIO but the net contribution vanishes by high symmetry. (F) The Brillouin zone of pyrochlore iridates.

phous NIO films are grown at a lower temperature (T) initially, and then they thermally annealed ex-situ in a sealed tube. SPE is known to produce r-NIO thin films [81, 82]. To apply the strain, our experimental team invent an unprecedented in-situ growth method, the so-called repeated rapid hightemperature synthesis epitaxy (RRHSE) [112]. The detail of this method is in Ref. 12. By the RRHSE method, the experimental team succeeds to grow the fully *s*-NIO films on YSZ (111) substrates. The success is evident as shown in Fig. 4.9A, an X-ray diffraction data with the NIO (III) and YSZ (III) (I: integer) peaks. This implies the epitaxial growth of the NIO single phase. Especially, the satellite peaks or "thickness fringes" near the NIO (222) peaks are found. These peaks mean that a high-quality sharp interface is formed between NIO and YSZ. Figure 4.9B is the reciprocal space mapping near the NIO (662) and YSZ (331) Bragg peaks. Note that the lattice parameter d(662) of bulk NIO is 1.19 Å, while that of YSZ is 1.18 Å. Since the NIO thin film (662) Bragg peak has the same  $Q_x$  value as the YSZ (331) peak, the NIO thin film is fully strained at about 1 %.

Figure 4.9C shows a STEM image of the NIO thin film on the YSZ substrate. The NIO pyrochlore phase is high-quality with few structural disorders. Figures 4.9D-E displays fast Fourier transform of the green and light blue boxes in Fig. 4.9C. The NIO film has the identical inverse lattice constant to the YSZ substrate. This shows us that the NIO film is strained as well.

### **4.2.4** Electronic structures of *r* and *s*-NIO thin films

Here, let us compare the *r*-NIO and *s*-NIO thin films. Figure 4.10A shows the longitudinal resistivity  $\rho(T)$  of *r* (black dotted line) and *s*-NIO (orange line) as a function of temperature *T*. The *r*-NIO film is metallic at high *T*, and metal-to-insulator transition occurs at 30 K. The sudden upturn below  $T_{Ir}^N = 30$  K is attributed to the insulating nature. Hence, the  $\rho(T)$  of *r*-NIO film is the Arrhenius plot at low *T*. This result agrees with the single crystal results [20, 27]. On the other hand,  $\rho(T)$  of *s*-NIO films is smaller than that of *r*-NIO films by an order of magnitude. The  $\rho(T)$  of *s*-NIO film is semimetallic. This is because we found  $\partial \rho / \partial T > 0$  at most *T* and the conductivity  $\sigma_{xx} \sim 1600 \ \Omega^{1} \ \mathrm{cm}^{1}$  at 2 K. The small upturn below 10 K



Figure 4.11: (A)  $\sigma_{xy}^A$  as a function of  $\mu_0 H$  at 3 K. The orange dots are the measured data, the green (blue) lines are the fitting curve attributed to Ir (Nd), and the black lines are the total fitting curve. (B)  $\sigma_{xy}^A$  as a function of  $\mu_0 H$  at various temperatures. Near  $T_N^{Ir} = 30$  K, the anomalous Hall conductivity begins to appear, which means Ir ordering begins to play a role. Near  $T_N^{Nd} = 15$  K, the hysteresis occurs, which means Nd ordering begins to play a role via fd-exchange. (C) The temperature dependence of the Ir and Nd contribution to anomalous Hall Effect. Green (blue) dots are  $\sigma_{xy}^{Ir} (\sigma_{xy}^{Nd})$  at 9 T. The red dots are the anomalous Hall conductivity at 0 T, (spontaneous Hall conductivity) which comes out below 15 K.

comes from disorders.

We perform the mean-field calculation of Hubbard Hamiltonian to understand the transport and electronic energy band change by the strain. The detail of the model is in Sec. 2.2. Note that the valence and conduction bands near the Fermi level come mostly from Ir 5d-electrons [2]. The calculated electronic energy band for r-NIO thin film is in Fig. 4.10B. The energy gap between valence and conduction bands is about 13 meV, which indicates the insulating nature [51]. The calculated energy band for s-NIO thin film is in Fig. 4.10C. The compressive strain of about 1 % modifies the valence and conduction bands to touch the Fermi level. As small electron and hole pockets emerge near L points, the system becomes semimetallic. The calculations explain the semimetallic s-NIO film and the insulating r-NIO film.

### 4.2.5 Large AHE in *s*-NIO thin films

In Fig. 4.10D, the anomalous Hall conductivity (AHC;  $\sigma_{xy}^A(H)$ ) as a function of H at 2 K is present. The r-NIO AHC is in the black dotted line and s-NIO AHC is in the orange line. The s-NIO film exhibits a stronger AHC than the r-NIO film. At 9 T, AHC of s-NIO is 2.4  $\Omega^{-1}$  cm<sup>-1</sup> while that of r-NIO is 0.2  $\Omega^{-1}$  cm<sup>-1</sup>. The spontaneous Hall conductivity (SHC; AHC at zero fields  $\sigma_{xy}^A(0)$ ) of the s-NIO films is 1.04  $\Omega^{-1}$  cm<sup>-1</sup>, an order stronger than that of the r-NIO film.

In the previous section, we claim that the *r*-NIO film can induce small AHC and SHC because of the AIAO domain walls [11]. However, the domain walls are insufficient to explain the large AHC and SHC in the *s*-NIO film. We think the net Berry curvature effect induced by the strain is attributable. We cross-check the argument by comparing *s*-NIO magnetotransport with those of typical ferromagnets. For instance, in ferromagnets like (Ga, Mn)As [113] and CuCr<sub>2</sub>Se<sub>4-x</sub>Br<sub>x</sub> [114],  $\sigma_{xx}(H = 0) \sim 1000$  $\Omega^{-1}$  cm<sup>-1</sup> while  $\sigma_{xy}^A(0) \sim 1$  to  $10 \Omega^{-1}$  cm<sup>-1</sup>. They obey a scaling relationship  $\sigma_{xy}^A \propto \sigma_{xx}^{1.6}$ , which indicates the intrinsic AHE of the systems [101]. The scaling relationship for the *s*-NIO film is the same, so our *s*-NIO thin film is also related to a net Berry curvature effect. (The detail is in Ref. 12.)

We calculate the Berry curvature effect on AHC in the mean-field calculations of Hubbard Hamiltonian mentioned above (see Sec. 2.2.) The formula for AHC is given by

$$\sigma_{xy}^A = \frac{e^2}{\hbar} \sum_n \int \frac{d^3k}{(2\pi)^3} f(\epsilon_n(\vec{k}) - \mu) \Omega_{xy}(\vec{k}) \tag{4.12}$$

where  $f(\epsilon_n(\vec{k}) \ \mu)$  is the Fermi-Dirac function,  $\mu$  is the Fermi level, and  $\Omega_{xy}$  is the Berry curvature. Figure 4.10E shows the calculated  $\Omega_{[111]}(\vec{k})$  along the high-symmetry lines of BZ for both r and s-NIOs.  $\Omega_{[111]}(\vec{k})$  is large at L points (see Fig. 4.10F.) for both the r- and s-NIOs. The Berry curvature at each L-point is larger in r-NIO than in s-NIO. However,  $\sigma_{xy}^A$  vanish only in the r-NIO, because the integration of  $\Omega_{[111]}$  cancels out by the cubic crystalline symmetries of r-NIO.

Here we discuss why  $\sigma_{xy}^A$  vanishes. When there are twofold rotation symmetries  $C_2$  about *a*-axis,

$$\Omega_{ab}(k_a, k_b, k_c) = -\Omega_{ab}(k_a, -k_b, -k_c),$$
  

$$\Omega_{ac}(k_a, k_b, k_c) = -\Omega_{ac}(k_a, -k_b, -k_c)$$
(4.13)

Hence, when we integrate for all BZ,  $\sigma_{ab}^A$  and  $\sigma_{ac}^A$  vanish. In *r*-NIO, twofold rotation exists for *x*, *y*, and *z*-axes, so the net Berry curvature effect must be hidden [7]. On the contrary, the *s*-NIO breaks  $C_2$  symmetries and the net Berry curvature effect arises. Thus, the large AHE is generated in the *s*-NIO films.

#### 4.2.6 Antihysteresis of AHC

Remarkably, the  $\sigma_{xy}^A$ (H) curve of *s*-NIO shows an antihysteresis, as shown in Fig. 4.10D. During the increasing (decreasing) sweep process,  $\sigma_{xy}^A$ changes its sign about `1 (1) T. The sign change of  $\sigma_{xy}^A$  contrasts a typical hysteresis in a usual ferroic material, where the sign change occurs H > 0for increasing sweep and H < 0 for decreasing sweep. The antihystersis was reported in the previous studies, but it was not fully examined [109].



Figure 4.12: (A) The M-H curve (purple squares) and  $\sigma_{xy}^A$  (orange dots) of s-NIO films at 3 K. M = 0 but  $\sigma_{xy}^A \neq 0$  at 0 T, which implies an alternative cause of the anomalous Hall Effect. (B)  $\sigma_{xy}^A$  at 3 K. The orange dots are the experimental data, and the green (blue) line is the Ir (Nd) contribution to anomalous Hall conductivity. The blue arrows indicate the sweep directions. The numerical calculations of dipole M and  $T_1$ -octupole  $\omega$  as a function of effective Zeeman field h, (C) without strain and (D) under strain. The straininduced  $T_1$ -octupole is the origin of the spontaneous Hall Effect in s-NIO films.

We use a phenomenological model to understand our anti-hysteresis curve.

$$\sigma_{xy}^{H} = f(H) + g(H). \tag{4.14}$$

The model has two distinct tangent-hyperbolic functions f(H) and g(H). f(H) is hysteretic (blue line) and g(H) is non-hysteretic (green line) as displayed in Fig. 4.11A. The experimental data agrees well with f(H) + g(H)(black line). Thus, we must have two distinct origins of AHC to explain its antihysteretic curve.

To specify the origins, we present  $\sigma_{xy}^A(H)$  curves of *s*-NIO films at various *T* below 40 K in Fig. 4.11B. When the system is paramagnetic T > 30 K,  $\sigma_{xy}^A$  vanishes.  $\sigma_{xy}^A$  begins to arise below 30 K and gets stronger when T decreases. Between 15 K and 30 K,  $\sigma_{xy}^A$  has no hysteresis. However, the anti-hysteresis begins to arise below 15 K. We succeed to match all experimental data with the fitting function in Eq. 4.14. Considering that  $T_{Nd}^N \sim 15$ K and  $T_{Ir}^N \sim 30$  K [20], the hysteretic f(H) and non-hysteretic g(H) develops because of Nd and Ir spins, respectively. We name  $f(H) = \sigma_{xy}^{Nd}$  and  $g(H) = \sigma_{xy}^{Ir}$  from now on.

The argument can be supported by the summary of the AHC fitting at H = -9 T in Fig. 4.11. The dotted lines denote  $T_{Nd}^N$  and  $T_{Ir}^N$  values. The transport in the NIO is dominated by the Ir *d*-electrons near the Fermi level. When the transport occurs, the carriers are influenced by the Ir and Nd spins. The non-hysteretic  $\sigma_{xy}^{Ir}(H = -9 \text{ T})$  emerges below  $T_{Ir}^N$ . On the other hand, the hysteretic  $\sigma_{xy}^{Nd}(H = -9 \text{ T})$  emerges below  $T_{Nd}^N$ . Ir spins give the non-hysteretic contribution because the Ir domain switching is absent by its weak Zeeman coupling. Meanwhile, Nd spins give the hysteretic contribution because the Nd domain can be switched by strong Zeeman coupling and transfer the switching to the Ir domain by *fd*-exchange. The hysteresis gives rise to finite SHC as shown in red squares of Fig. 4.11C.

## **4.2.7 AHE from strain-induced** *T*<sub>1</sub>**-octupoles**

The conventional understanding of AHE teaches us that the SHC is proportional to the magnetization M [101]. Accordingly, we compare M

with  $\sigma_{xy}^A$  as functions of H at T = 3 K in Fig. 4.12A, and present  $\sigma_{xy}^{Ir}$  and  $\sigma_{xy}^{Nd}$  in Fig. 4.12B. Surprisingly, the conventional understanding,  $\sigma_{xy}^A(H = 0) \propto M(H = 0)$ , does not hold for our *s*-NIO films. Whereas the *s*-NIO film has a large SHC signal (orange lines), M = 0 at 3 K at H = 0 (purple squares).

Thus, we should consider the alternative origin of SHC. In Fig. 4.8D, we show that the biaxial strain induces three distinct cluster multipoles, a dipole, an  $A_2$ -octupole, and a  $T_1$ -octupole. Since we have zero magnetization, the dipole is crossed out. Since the  $A_2$ -octupole does not give SHC because of its cubic symmetry, it is crossed out as well. The  $T_1$ -octupole is antiferromagnetic, but it can induce AHE since it has the same symmetry to the ferromagnetic order. Therefore, the strain-induced  $T_1$ -octupole generate the SHC without any magnetization.

We illustrate how the strain induces  $T_1$ -octupole by the spin model calculation. The spin model (in Sec. 2.2) includes the various exchange interactions of Ir spins, fd-exchange between Ir and Nd spins, and the Zeeman energy for Ir and Nd spins. We calculate the ground state spin structure and obtain cluster multipoles which are defined in Sec. 2.3. We show the calculated dipole (M) and  $T_1$ -octupole ( $\omega$ ) as a function of effective Zeeman field h in Figs. 4.12C-D. Note that M is in green circles,  $\omega$  is in blue circles, and Figure 4.12C (D) is for r-NIO (s-NIO). In r-NIO, both M and  $\omega$ vanish at h = 0, which explains its small SHC. In s-NIO, on the other hand, both M and  $\omega$  are finite even at h = 0. Intriguingly,  $\omega$  looks alike to  $\sigma_{xy}^{Nd}$ in Fig. 4.12B. We think this is because fd-exchange strengthens  $\omega$ . To sum up, we conclude that the strain-induced  $T_1$ -octupole gives rise to AHE in the s-NIO film.

We here demonstrate that the strain-engineering in an AFM can generate topological phenomena by tuning the cluster multipoles. In our case, we emphasize that strain-induced  $T_1$ -octupole is closely linked to the topological transport in an NIO. We expect that the strain-engineering can be extended to investigate the other topological phenomena in correlated magnetic systems. For instance, we apply the strain onto other members of pyrochlore iridates to realize the correlated topological phases like a Weyl semimetal, a nodal line semimetal, an axion insulator, and a topological insulator [10, 23, 109]. In the next section, accordingly, we describe how we find the correlated topological phases in  $Pr_2Ir_2O_7$ .

## 4.3 Correlated Weyl semimetal in Strained $Pr_2Ir_2O_7$ thin films

#### 4.3.1 Introduction

In modern condensed matter, correlation-driven and topological-driven physics compose central themes [1] It has been thoroughly investigated for several years that electronic correlation denoted by U plays a key role in the metal-to-insulator transition, giant magnetoresistance, and superconductivity [115–117]. Meanwhile, topological-driven physics was developed since a topological insulator (TI) with sizable spin-orbit coupling ( $\lambda$ ) [118] has been discovered, and stretched out swiftly toward Weyl (or Dirac) [2, 55, 56, 119, 120] and nodal-line semimetals [121, 122] as well. Nowadays, the voyage of interest advances toward the reciprocity of electronic correlation and topological-driven physics [1, 22, 122, 123], which gives rise to the correlated topological semimetals (CTPs) [4]. The typical example of CTPs is a correlated Weyl semimetal (WSM), a topological Mott insulator, and an axion insulator. Especially, WSMs attract numerous activities in society, which is characterized by Weyl fermions and surface Fermi arc [2, 55, 56, 119]. WSMs show intriguing physical properties, like phonon-electron fluid [124], the nonlinear Hall effect [42], magnetotransport induced by chiral anomaly [18, 49, 125], anomalous Hall effect (AHE) [94], and anomalous Nernst effect [105]. Nonetheless, the electronic correlation in WSMs is too restricted to be realized in experiments, because candidate materials are rare in nature [119, 123].

Here, we use pyrochlore iridates whose chemical formula is  $R_2Ir_2O_7$ . Pyrochlore iridates have large  $\lambda$  and intermediate U since they are members of 5d transition metal oxides. Hence, pyrochlore iridates is a playground to realize the topological CTPs [1, 23, 57, 69, 78]. The crystal structure of  $R_2Ir_2O_7$  is shown in Fig. 4.13a, and the symmetry of  $R_2Ir_2O_7$  is the same as that of the diamond lattice. Furthermore, pyrochlore iridates are the first candidate that Weyl fermions can emerge in the real material [2].

Most pyrochlore iridates have an all-in-all-out (AIAO) antiferromagnetic order as shown in Fig. 4.13b. It is predicted that the AIAO order induces the WSM state with 4 pairs of Weyl nodes by breaking time-reversal symmetry (TRS) [2]. Nevertheless, as the AIAO order pushes the Weyl nodes toward the Brillouin zone boundary, the Weyl nodes are annihilated by pairs, and the charge gap is opened at the Fermi level. The WSM state, therefore, appears only at the very verge of the gapped state and exists within an infinitesimal temperature range [109]. In almost every compound except R = Pr, the emergence of AIAO order at  $T_N$  accompanies a metal-to-insulator transition as shown in Fig. 4.13c [2]. Despite the abundant theoretical predictions [1, 23, 57, 69, 78], thus, experimental realization of WSM states in pyrochlore iridates remains elusive.

The  $Pr_2Ir_2O_7$  single crystal (bulk) is close to the critical point that metal-to-insulator and paramagnetic-to-antiferromagnetic transitions of pyrochlore iridates [3]. This is paramagnetic and metallic down to a few mK and the resistivity does not increase significantly [33]. The electronic band structure forms a Luttinger semimetal (LSM) state where a quadratic band crossing is across the Fermi level at  $\Gamma$  point [6] This quadratic band crossing can break into diverse topological semimetal phases by symmetry-breaking perturbations [6, 10, 126]. Here, our experimental team succeeds to fabricate pure strained PIO thin films and utilize the strain as a tuning perturbation to find the magnetic WSM phase. The AHE observation evinces the Weyl nodes that give rise to Berry curvature. Magnetotransport results deliver a compelling piece of evidence that chiral anomaly emerges from the Weyl nodes. The most important clue for a correlated WSM phase is the T-dependence of the planar Hall Effect. Considering all transport measurements and theoretical predictions, we report that strained PIO thin film is the magnetic WSM dressed by long-range Coulomb interaction.



Figure 4.13: (a) The lattice structure of pyrochlore iridates, where red spheres are Ir and blue spheres are rare-earth ions. (b) Two distinct domains of pyrochlore iridates, A (AIAO; left) and B (AOAI; right) domains. (c) The phase diagram of pyrochlore iridates family as a function of rare-earth ionic radius and temperature [1, 3]. (d) The mean-field calculated exotic phase diagram of pyrochlore iridates as a function of compressive strain  $\delta$  and Hubbard repulsion U.  $\delta$  is the ratio change of hoppings by the strain and U is normalized by  $t_{oxy}$  which is the oxygen-mediated nearest-neighbor hopping term. Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> are an LSM at the red line in absence of the strain. (e) Schematics of the energy band renormalization in Weyl semimetal because of the correlation effect.

# 4.3.2 Topological Phase Diagram and Epitaxial Growth of PIO

We use the Hubbard model to calculate the ground state of strained PIO by mean-field theory [5, 10, 59]. We draw the phase diagram in terms of Uand  $\delta$  (the ratio of hopping parameter change by the strain) in Fig. 4.13d, based on the ground state we calculate. Note that  $\delta$  indicates the effect of strain in the calculation, and  $\delta > 0$  means that the strain is compressive [12]. We provide the detail in Ref. 13. We observe that the strain applied to LSM produces three fascinating topological phases, a magnetic WSM, an axion insulator, and a TI. Notably, the AIAO order which is absent in the unstrained bulk PIO emerges from a weak strain in the film ( $\delta < 0.3\%$  for U = 1.39). The AIAO order induces a magnetic WSM phase containing 4 pairs of Weyl nodes near the Fermi level. When the strain increases to an intermediate value ( $0.3\% < \delta < 0.7\%$ ), all Weyl nodes are pair-annihilated at  $\Gamma$  point as AIAO order decreases, and the band gap is opened at the Fermi level. Since the time-reversal symmetry is broken, we observe an axion insulator phase where the Fu-Kane  $\mathbb{Z}_4$  index value is 2. When the strain gets increased to  $\delta > 0.7\%$ , the AIAO order disappears, and the system turns into a paramagnetic TI [23]. We concentrate on the magnetic WSM in the weak strain region  $\delta < 0.3\%$ . When the electronic correlation between Ir-5d electrons is effective, the renormalization of Weyl nodes occurs, and we obtain a correlated magnetic WSM phase as shown in Fig. 4.13e.

Our experimental team probes the correlated magnetic WSM by fabricating a weakly strained high-quality PIO thin film. They use a modified solid-phase epitaxy (SPE) method, whose detail is in Ref. 13, 112. We present the X-ray diffraction of PIO (111) film on YSZ (111) substrate in Fig 4.14a, with a bare YSZ substrate as a comparison. The pyrochlore phase crystallization is excellent which is demonstrated by the strong odd-pair peaks. The magnified version of X-ray diffraction is shown in Fig. 4.14b. This indicates that the PIO film is strained since its (222) peak is shifted away from the bulk one. The reciprocal space map in Fig. 4.14c exhibits that the compressive strain  $\sim 0.2\%$  is applied in the (111) plane of PIO thin film. The shift of  $Q_z$  implies that the lattice is elongated along [111] in accordance with the (222) peak shift in Fig. 4.14b. The weak strain in our PIO film is accountable for the emergence of AIAO order and associated magnetic WSM phase as we predict in the mean-field calculations.(See Fig. 4.13d).



Figure 4.14: (a) The X-ray diffraction peaks of  $Pr_2Ir_2O_7$  thin films on the YSZ (111) substrate (a red line) compared with a bare YSZ (a blue line). (b) The enlarged X-ray diffraction near YSZ (111) peak. The green dashed line is the position of the bulk  $Pr_2Ir_2O_7$  (222) peak. (c) The reciprocal map of  $Pr_2Ir_2O_7$  thin films on YSZ substrate near YSZ (331) peaks. \* is the peak position of  $Pr_2Ir_2O_7$  thin film.

## 4.3.3 AHE in Strained PIO Film

We display the temperature-dependent longitudinal resistivity  $\rho_{xx}(T)$ of the PIO films in the absence of a magnetic field in Fig. 4.15a. A Hallbar geometry is utilized to measure the transport as shown in the inset of Fig. 4.15a. When the temperature decreases, both the strained films and the single crystal present a metallic nature until the resistivity reaches the minimum. After then, there is a small upturn at the lower temperature. The resistivity minimum temperature in the strained PIO film (~ 50 K) is slightly higher than that of single crystal PIO (~ 40 K). The resistivity of the strained film enhances by ~ 3-10 times that of a single crystal below 300 K because of the compressive strain. The compressive strain makes the oxygen octahedron surrounding Ir ions more distorted trigonally, leading to the reduction of Ir-O orbital overlapping and the associated suppression of Ir electron conduction [1]. It is similar to the fact that the resistivity of strained Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> (NIO) film is greatly raised from that of bulk NIO [12]. Our experimental team fabricates more samples with a larger compressive strain and confirms



Figure 4.15: (a) The longitudinal resistivity for PIO thin film (a red line) compared with single crystal [33] (a blue line). The inset is the schematics of the experimental geometry. (b) The Hall resistivity as a function of B at various temperatures. (c) The comparison between Hall resistivity of PIO thin film at 2 K (a red line) and PIO single crystal [52] (a blue line). The arrows are the sweep directions.

that the resistivity gets increased when the strain gets stronger. (See Ref. 13 for the detail.) The tendency that the compressive strain induces insulating behavior is consistent with the theoretical calculation in Fig. 4.13d. To confirm the axion insulator or TI, one needs more exhaustive efforts on these samples in the future.

An intriguing phenomenon in the strained PIO is shown in Fig. 4.15b. That is, the AHE appears and persists up to 30 K when  $B \parallel [111]$  is applied in the strained PIO film. The temperature is much higher than the single crystal samples ( $\sim 1.5$  K) [39, 52, 127] Above 30 K, the Hall response is linear to B, which implies that the AHE disappears at high temperatures. Furthermore, what is important is that the Hall resistivity of strained PIO thin film at 2 K presents a hysteretic behavior and a finite value at B = 0. This is the so-called spontaneous Hall Effect (SHE). The single crystal sample also shows SHE but at the lower temperature  $\sim 0.5$  K, as shown in Fig. 4.15c. The SHE in the PIO thin film is attributed to the TRS and cubic crystalline symmetry breaking [12]. The TRS is broken by the strain-

induced AIAO order, and the cubic crystalline symmetry is broken by the applied compressive strain. It is noteworthy that the AHE in the PIO film is unconventional, in that the anomalous Hall resistivity is irrelevant to the magnetization (M) [15]. In conventional viewpoint, the Hall resistivity is phenomenologically described as  $\rho_{xy} = R_0B + 4\pi R_sM$ . The first term is the normal Hall Effect while the other term is the AHE. Hence, in the ferromagnetic systems, the magnetization and spin-orbit coupling give rise to the AHE [15]. Nevertheless, the AHE in our strained PIO with induced AIAO order is attributed to the non-trivial Berry curvature ( $\Omega_{i,k}$ ) produced by the Weyl nodes [128].

#### 4.3.4 Magnetotransport in Strained PIO Film

To access more visions onto Weyl fermions, our experimental team measures the magnetotransport on the strained PIO film. They apply the magnetic field in two directions, perpendicular and parallel to the current I directions. The single crystal PIO sample, previously, shows a giant positive transverse magnetoresistivity (TMR) when  $B \parallel [111] \perp I$  is applied [52]. The positive TMR is attributed to the electron localization by the Lorentz force, which is common in a usual metal.

In our strained PIO system, however, we observe a negative TMR at 2 K with an upturn at a strong field (See Fig. 4.16a.) This feature strongly contrasts the state of strained PIO to that of bulk PIO, a paramagnetic LSM. Remarkably, the upturn at the high field in strained PIO film reminds the TMR in NIO single crystal under pressure [109]. NIO single crystal develops AIAO order at low temperature T < 30 K [3, 109]. The upturn in the TMR in NIO was illustrated by the domain switching from Ir-AIAO (4/0) to Ir-3I1O (3/1) state [109]. Such domain switching happens for the strained PIO film as well. Our previous studies [10] described that the band structure of pyrochlore iridates depends strongly on the spin configuration.

When  $B \parallel [111]$  is applied, the spin configuration of pyrochlore iridates varies from 4/0 to 3/1 state. Accordingly, the domain switching makes the energy band structure near the Fermi energy fluctuate significantly. Theoretically, the WSM in the 4/0 spin configuration has 4 pairs of Weyl nodes. This changes into the WSM with 3 pairs of Weyl nodes by 3/1 spin configuration. The modulation of energy band structure, hence, decreases the density of states near Fermi energy and induces the upturn of TMR, as same as the NIO case [109]

Moreover, similar hysteretic properties to NIO [109] appear in our strained PIO film in Fig. 4.16a. The hysteresis vanishes and the TMR is positive and linear to B at T > 15 K as shown in Fig. 4.16b. In the hysteresis curve of Fig. 4.16a,  $\rho_{xx}$  for decreasing field sweep (blue lines) is larger than that for increasing field sweep when B > 0, and vice versa. The hysteretic curve can be described by the domain wall dynamics of AIAO order in pyrochlore iridates. In the absence of a magnetic field, two degenerate magnetic domains are present, such as the AIAO (A domain) and AOAI (B domain). The domains are time-reversal symmetry partners [84]. Under a weak field, two domains simultaneously exist. When B increases, these two domains can be switched by either  $B \parallel [111]$  or  $B \parallel [\overline{1}\overline{1}\overline{1}]$  [84, 109]. The hysteresis appears during the domain switching process. We present the detail of the hysteresis loop at 5 K in Fig. 4.16c. The hysteresis within  $\pm 4.5$ T comes from the coexistence of the A and B domains. Beyond  $\pm 4.5$  T, the difference between increasing and decreasing sweep vanishes as the multidomain state turns into a single domain state. When B increases further, the spins begin to flip and the system turns into a 3/1 spin configuration. Hence, the hysteresis loop in TMR implies that strained PIO film has an AIAO order below 15 K, in accordance with our theoretical calculations. Note that there also was a report about the detection of local Ir moments in the film sample [129]. We also comment that we cannot use neutron scattering experiments to detect Ir moments, since our PIO thin film is only 40 nm-thick



Figure 4.16: (a,b) Transverse magnetoresistance as a function of B at various temperatures.  $T_O = 15$  K,  $T^* = 30$  K, (a)  $T < T_O$  and (b)  $T_O \leq T \leq T^*$ . The red and blue lines are the increasing and decreasing field sweep, respectively. The inset in (b) is the experimental geometry of TMR. (c) The enlarged TMR measured at 5 K in (a). (d) Longitudinal magnetoresistance as a function of B at various temperatures. The inset is the experimental geometry of LMR. (e) The values of TMR and LMR under B = 9 T at various temperatures. (f) The phase diagram of PIO thin films as a function of temperature. -, +, w., and wo. mean negative components, positive components, with hysteresis, and without hysteresis for TMR, respectively.

that does not meet their requirements [130].

Mean-field calculation shows that when the LSM meets the AIAO order, it is broken into the magnetic WSM by TRS breaking. Our experimental team measures the longitudinal magnetoresistivity (LMR:  $B \parallel I$ ) to detect the chiral anomaly from the WSM. The chiral anomaly from the WSM is that the charge pumping between a pair of Weyl nodes by their chiral zero Landau level [18, 49, 131]. Hence, the chiral anomaly gives rise to negative LMR.

The non-hysteretic negative LMR at T < 30 K is observed, as shown in Fig. 4.16d. In a weak field, the LMR is weakly positive because of the weak anti-localization in topological matters [18, 49]. We summarize the TMR and LMR at B = 9 T in Fig. 4.16e. When  $T < T^* = 30$  K, the LMR at 9 T is negative while the TMR at 9 T changes its sign from positive to negative at 5 K. The peak of TMR at 9 T is at  $T_O = 15$  K. Such puzzling behavior of TMR could be explained by the competition of AIAO order (negative) and Lorentz force (positive).

When  $T_O < T < T^*$ , TMR at 9 T increases by lowering the temperature, which shows that the system is paramagnetic where the Lorentz force contribution dominates over that of the AIAO order. When  $T < T_O$ , TMR at 9 T decreases by lowering temperature as the AIAO order develops. When T < 7 K, TMR shows a hysteretic behavior, and TMR at 9 T is negative, which shows that the system is antiferromagnetic where AIAO contribution dominates over that of the Lorentz force. As result, the range between 7 to 15 K exhibits the concurrence of paramagnetic and antiferromagnetic phases. More pieces of evidence are in Ref. [13].

Based on the TMR measurements, we draw schematics for phase transition in Fig. 4.16f The quantum oscillation is not observed in the strained PIO film, since the mobility is very low [127]. In the single crystal, the mobility is  $\approx 10 \text{ cm}^2 \text{ V}^{1} \text{ s}^{1}$ , while in our PIO film, it is  $\approx 0.74 \text{ cm}^2 \text{ V}^{1} \text{ s}^{1}$ ). The mobility is far below compared to that in TaAs [49] or SrRuO<sub>3</sub> [132]. Note that the mobility should be above  $103 \text{ cm}^2 \text{ V}^{1} \text{ s}^{1}$ ) to observe quantum oscillation within a few Teslas [133, 134]. Furthermore, as Pr 4*f* electrons strongly hybridize with Ir 5*d* electrons [33], the effective mass is too large to satisfy the condition for quantum oscillations [134]. To observe quantum transport, one needs to improve the film quality in the future.

### 4.3.5 PHE in Strained PIO Film

The chiral anomaly is not the only cause of negative LMR. Since the current jetting also induces the negative LMR [135], we need further evidence of chiral anomaly. Accordingly, we measure PHE in strained PIO film to provide evidence that chiral anomaly and AIAO order exist [18, 136–140]. In Fig. 4.17a, we draw schematics of PHE that the Hall voltage emerges when the magnetic field and current are applied in the same plane, and  $\phi$  is the angle between *B* and *I*. The planar Hall conductivity (PHC) is divided into two parts in our system,

$$\sigma_{xy}^{PHE} = \sigma_{xy}^{chiral} + \sigma_{xy}^{AIAO}, \qquad (4.15)$$

where  $\sigma_{xy}^{chiral}$  and  $\sigma_{xy}^{AIAO}$  are the PHCs from the chiral anomaly and AIAO order, respectively. In previous studies [125], The chiral anomaly PHC is given by

$$\sigma_{xy}^{chiral}(\phi) = \Delta \sigma_{chiral} \sin \phi \cos \phi, \qquad (4.16)$$

where  $\Delta \sigma_{chiral} = \sigma_{\parallel} \sigma_{\perp}$  is the anisotropy of magnetoconductivity made by chiral anomaly.  $\sigma_{\perp}$  and  $\sigma_{\parallel}$  are the conductivities under  $B \perp I$  and  $B \parallel I$ each.

In the gray dots of Figs. 4.17b-d, we present the PHC at different temperatures (2, 4, and 15 K). At 15 K, where both paramagnetic and antiferromagnetic states exist, the PHC can be explained by chiral anomaly solely. However, at 2 and 4 K, the PHC cannot be explained by the chiral anomaly only, which has a sin  $2\phi$  dependence in the blue dashed line of Fig. 4.17b. In this temperature range, our magnetotransport measurements support that AIAO order emerges and prevails. AIAO order corresponds to a higher-rank magnetic multipole called an  $A_2$ -octupole ( $\omega_{xyz}$ ) [12, 141]. Although there are no dipolar moments in AIAO order, an  $A_2$ -octupole can induce an or-



Figure 4.17: (a) Schematics of the experimental geometry of planar Hall Effect measurement. (b-d) The planar Hall conductivity  $\sigma_{xy}^{PHE}$  as a function of rotation angle  $\phi$  under B = 9 T at T = 2, 4, and 15 K, respectively. We use Eqs. 4.16 and 4.18. (e) The PHE coefficients  $\alpha$  and  $\beta$  in Eq. 4.18 as a function of temperature, which is attributed to chiral anomaly and AIAO order, respectively. (f) The renormalization group calculation for the temperature dependence of  $\alpha$  by the electronic correlation effect. The fitting function is  $f(T) = AT + B \ln T + C$ , where A, B, and C are constants. The inset shows the  $\ln T$  dependence of  $\alpha$ .

thogonal magnetization  $M_{\perp}$  by its coupling with B [141]. The "orthogonal magnetization" is named after the fact that its direction is orthogonal to B. The detail is in Ref. 13.

The in-plane orthogonal magnetization contributes to PHE as well as a

chiral anomaly. To sum up, the PHC is given by

$$\sigma_{xy}^{PHE}(\phi) = \sigma_3 B^2 \sin 2\phi + \sigma_4 \frac{\omega_{xyz}^2}{12} B^4 \sin 4\phi$$
 (4.17)

where  $\sigma_3$  and  $\sigma_4$  are constants. The first term is from the chiral anomaly, and the second one is from orthogonal magnetization induced by an AIAO order. The first term is derived from the chiral anomaly, while the second originates from the AIAO ordering. We set two coefficients  $\alpha = \sigma_3 H^2$  and  $\beta = \sigma_4 \omega_{xuz}^2 H^4/12$ , and the above equation becomes

$$\sigma_{xy}^{PHE} = \alpha \sin 2\phi + \beta \sin 4\phi. \tag{4.18}$$

 $\alpha$  and  $\beta$  are the PHC magnitudes induced by the chiral anomaly and  $A_2$ octupole, respectively. We sum up the change of  $\alpha$  and  $\beta$  as a function of
temperature in Fig. 4.17e.

The fitting results of Eq. 4.18 is shown in the red lines of Figs. 4.17bd. Below  $T < T_O$ , the two-origin model in Eq. 4.18 is much improved than chiral anomaly model in Eq. 4.16. This evinces the existence of AIAO order as well. The small variance of fitting and data might come from the spin fluctuation. Meanwhile,  $T_0 < T < T^*$ , the AIAO order vanishes since  $\beta$  approaches 0 in the blue dots of Fig. 4.17e. This is consistent with the conclusion from TMR data in Fig. 4.16f. The chiral anomaly exists up to  $T^*$ since  $\alpha$  is still finite in the red dots of Fig. 4.17e. This indicates that a WSM phase emerges by *B* [142]. Our experimental team checks the reproduction of our experimental results by fabrication of another reference sample. A coherent conclusion is drawn from the reference sample as well. The detail is in Ref. 13. To summarize this part, we confirm the existence of chiral anomaly and AIAO order while excluding the current jetting by the negative LMR and PHE measurements.

Lastly, we discuss here the correlation effect of the WSM phase by

investigating the temperature dependence of PHC magnitude  $\alpha$  [143]. Dimensional analysis shows that

$$\sigma_3 \propto \frac{e^2}{\hbar} \frac{v_F^3 e^2}{\mu_2} \tau, \tag{4.19}$$

where  $\hbar$  is the Planck constant,  $\tau$  is the scattering time, e is the electron unit charge,  $\mu$  is the chemical potential, and  $v_F$  is the Fermi velocity. The theoretical prediction of chiral anomaly gives the same results [18]. The temperature dependence of  $\sigma_3$  and  $\alpha$  can be studied by that of  $v_F$ ,  $\mu$ , and  $\tau$ . We obtain those from a one-loop calculation in a WSM phase dressed by a long-range Coulomb repulsion [143].

$$v_F(T) = v_{F0} (1 + \frac{N_w + 2}{3\pi} \alpha_0 \ln \frac{\hbar\Lambda}{k_B T})^{2/(N_w + 2)},$$
  
$$\mu \propto (1 + \frac{\alpha_0}{\pi\mu_0} T \ln \frac{\hbar\Lambda}{k_B T}),$$
 (4.20)

where  $N_w$  is the number of Weyl nodes,  $\Lambda$  is a cutoff momentum, and  $\alpha_0$  is a dimensionless coupling constant without interaction. At low T, the scattering time of a WSM dressed by a long-range Coulomb repulsion is [144],

$$au \propto 1 + \frac{2\pi^2}{3} (\frac{T}{T_F})^2,$$
(4.21)

where  $T_F$  is the Fermi temperature. Therefore, for a small coupling and a low temperature,  $\alpha = \frac{\sigma_3 H^2}{2}$  as a function of T is approximated,

$$\alpha \propto A T + B \ln T + C, \tag{4.22}$$

where A, B, and C are constants, and the  $\ln T$  and T terms arise from the renormalization of  $v_F$  and  $\mu$ , respectively. The detail is in Ref. 13.

In Fig. 4.17f, the experimental change of  $\alpha$  is in red dots, and the fitting of Eq. 4.22 is shown in a blue line. The experimental data show great accor-

dance with the fitting function from the renormalization group calculation. This demonstrates that the strained PIO system shows the renormalization of Weyl nodes by electronic correlation. In addition, the  $\ln T$  component in  $\alpha$  is very strong as shown in the inset of Fig. 4.17f. This indicates the Fermi velocity is greatly enhanced by the long-range Coulomb repulsion [145]. Only the graphene which is a 2D Dirac semimetal was reported to show an analogous renormalization effect [122, 145, 146]. Our results imply that strained PIO film shows a correlated WSM phase whose Fermi velocity is renormalized logarithmically by long-range Coulomb interaction.

### 4.3.6 Conclusion

We discover a correlated magnetic WSM phase in the strained PIO film which is predicted in the theoretical calculation. The emergence of AHE, negative TMR with the upturn, negative LMR, and PHE evinces the magnetic WSM phase in our system. In addition, the analysis of chiral anomalyinduced PHE  $\alpha$  shows that the electronic correlation effect renormalizes the Fermi velocity of Weyl nodes. We pave a new path of strain-engineering of CTPs by finding the "bulk-absent" AIAO magnetic order in PIO film. PIO is a favorable playground for CTPs since it is expected to host an axion insulator and a TI by applying a larger strain. So far, our works emphasize the reciprocity of perturbations, magnetism, and topological phases in a correlated spin-orbit coupled system, and encourage future research on correlated topological phases in new condensed matter systems.

## **Chapter 5**

## **Conclusion and Outlook**

This dissertation studies the topological semimetals in pyrochlore iridates near the transition point by applying a magnetic field and strain. Here, we summarize the main results and address the outlooks from the results.

## 5.1 Summary

In Chap. 3, the magnetic-field induced topological semimetals in pyrochlore iridates are discussed. First, the experimental results show that the magnetic field induces topological semimetals, such as Weyl semimetal (4/0, 3/1) and nodal-line semimetal (2/2) in  $(Nd_xPr_{1-x})_2Ir_2O_7$  single crystals. Motivated by the results, the generic topological band structure under a magnetic field is studied. In paramagnetic metal, QBC with fourfold degeneracy exists. Because of the large degeneracy and strong SOC, the Zeeman field on QBC can induce both anisotropic and isotropic Zeeman terms. The interplay of Zeeman terms and the exchange energy of Ir electrons leads to the various topological semimetals in pyrochlore iridates. The control of three energy scales is controlled by the modulation of spin configuration by the magnetic field.

In Chap. 4, the strain-induced topological semimetals in pyrochlore iridates are discussed. First, in relaxed Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films, the anomalous Hall Effect comes from the antiferromagnetic domain wall structure. Then, the strained Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films show the large enhancement of the anomalous Hall Effect. This is from the strain-induced cluster  $T_1$ -octupoles that has the same symmetry as the magnetization. Lastly, the strained Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films become the correlated Weyl semimetal. The experiment results support that the AIAO order sets in and time-reversal symmetry is broken. Further theoretical analysis of experimental results shows that they are the interacting Weyl liquid with logarithmically renormalized Fermi velocity dressed by long-range Coulomb interaction.

# 5.2 Perpendicular magnetization in antiferromagnets with higher-rank magnetic multipoles

Some antiferromagnets including pyrochlore iridates develop the magnetization perpendicular to the applied magnetic field. Two sources of perpendicular magnetization (PM) are known. One is the transverse magnetization (TM) that comes from the spin canting, and the other is the orthogonal magnetization (OM) that comes from the coupling of field to the cluster multipoles in the Landau theory. We suggest a unified viewpoint for PM. First, we explain PM by symmetry. PM appears only when all crystalline symmetries are broken but  $C_2T$ ,  $\sigma T$ , and P. Also, one can determine whether PM is an even or odd function by symmetry analysis. Then, PM is divided into OM, spin, and orbital parts. We discuss the origin of spin and orbital PM. The spin PM comes from spin anisotropy. The orbital PM comes from the magnetic energy band change. Then, we explain all numerical and experimental observations. Lastly, we show that PM contributes to the anomalous planar Hall Effect. We build a new guideline for understanding the magnetic responses of the antiferromagnets with complex structures.

## **5.3** Planar Hall Effect in Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> in thin films

In the previous work, the strained  $Pr_2Ir_2O_7$ , which is originally paramagnetic metal, is known to host the planar Hall Effect. The planar Hall Effect is decomposed into twofold and fourfold harmonics as a function of the angle between  $\vec{E}$  and  $\vec{B}$ . This can be explained by the chiral anomaly from Weyl semimetal and OM from strain-induced AIAO order. Here, in
the strained  $Nd_2Ir_2O_7$ , which is an originally antiferromagnetic insulator, the planar Hall Effect has sixfold harmonics in addition to twofold and fourfold ones. This is because of the OM from strain-induced  $T_1$ -octupole. We show the close relationship between the magnetic structure and transport phenomena in an antiferromagnet.

## **Bibliography**

- [1] W. Witczak-Krempa, G. Chen, Y. B. Kim, and L. Balents, Annu. Rev. Condens. Matter Phys. 5, 57 (2014).
- [2] X. Wan, A. M. Turner, A. Vishwanath, and S. Y. Savrasov, Physical Review B 83, 205101 (2011).
- [3] K. Matsuhira, M. Wakeshima, Y. Hinatsu, and S. Takagi, Journal of the Physical Society of Japan 80, 094701 (2011).
- [4] W. Witczak-Krempa and Y. B. Kim, Physical Review B 85, 045124 (2012).
- [5] W. Witczak-Krempa, A. Go, and Y. B. Kim, Physical Review B 87, 155101 (2013).
- [6] T. Kondo, M. Nakayama, R. Chen, J. Ishikawa, E.-G. Moon, T. Yamamoto, Y. Ota, W. Malaeb, H. Kanai, Y. Nakashima, *et al.*, Nature communications 6, 1 (2015).
- [7] M.-T. Suzuki, T. Koretsune, M. Ochi, and R. Arita, Physical Review B 95, 094406 (2017).
- [8] M.-T. Suzuki, T. Nomoto, R. Arita, Y. Yanagi, S. Hayami, and H. Kusunose, Physical Review B 99, 174407 (2019).
- [9] K. Ueda, T. Oh, B.-J. Yang, R. Kaneko, J. Fujioka, N. Nagaosa, and Y. Tokura, Nature communications 8, 1 (2017).
- [10] T. Oh, H. Ishizuka, and B.-J. Yang, Physical Review B 98, 144409 (2018).
- [11] W. J. Kim, J. H. Gruenewald, T. Oh, S. Cheon, B. Kim, O. B. Korneta, H. Cho, D. Lee, Y. Kim, M. Kim, *et al.*, Physical Review B 98, 125103 (2018).

- [12] W. J. Kim, T. Oh, J. Song, E. K. Ko, Y. Li, J. Mun, B. Kim, J. Son, Z. Yang, Y. Kohama, *et al.*, y advances **6**, eabb1539 (2020).
- [13] Y. Li, T. Oh, J. Son, J. Song, M. K. Kim, D. Song, S. Kim, S. H. Chang, C. Kim, B.-J. Yang, *et al.*, Advanced Materials **33**, 2008528 (2021).
- [14] N. Armitage, E. Mele, and A. Vishwanath, Reviews of Modern Physics 90, 015001 (2018).
- [15] N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald, and N. P. Ong, Reviews of modern physics 82, 1539 (2010).
- [16] K.-Y. Yang, Y.-M. Lu, and Y. Ran, Physical Review B 84, 075129 (2011).
- [17] X. Yuan, Z. Yan, C. Song, M. Zhang, Z. Li, C. Zhang, Y. Liu, W. Wang, M. Zhao, Z. Lin, *et al.*, Nature communications 9, 1 (2018).
- [18] D. Son and B. Spivak, Physical Review B 88, 104412 (2013).
- [19] A. Burkov, Journal of Physics: Condensed Matter 27, 113201 (2015).
- [20] Z. Tian, Y. Kohama, T. Tomita, H. Ishizuka, T. H. Hsieh, J. J. Ishikawa, K. Kindo, L. Balents, and S. Nakatsuji, Nature Physics 12, 134 (2016).
- [21] M. Kurita, Y. Yamaji, and M. Imada, Journal of the Physical Society of Japan 80, 044708 (2011).
- [22] D. Pesin and L. Balents, Nature Physics 6, 376 (2010).
- [23] B.-J. Yang and Y. B. Kim, Physical Review B 82, 085111 (2010).
- [24] Y. Yamaji and M. Imada, Physical Review X 4, 021035 (2014).
- [25] G. Chen and M. Hermele, Physical Review B 86, 235129 (2012).
- [26] S. Lee, A. Paramekanti, and Y. B. Kim, Physical review letters 111, 196601 (2013).

- [27] K. Ueda, J. Fujioka, B.-J. Yang, J. Shiogai, A. Tsukazaki, S. Nakamura, S. Awaji, N. Nagaosa, and Y. Tokura, Physical review letters 115, 056402 (2015).
- [28] S. Bramwell and M. Harris, Journal of Physics: Condensed Matter 10, L215 (1998).
- [29] M. Imada, A. Fujimori, and Y. Tokura, Reviews of modern physics 70, 1039 (1998).
- [30] M. Sakata, T. Kagayama, K. Shimizu, K. Matsuhira, S. Takagi, M. Wakeshima, and Y. Hinatsu, Physical Review B 83, 041102 (2011).
- [31] K. Ueda, J. Fujioka, C. Terakura, and Y. Tokura, Physical Review B 92, 121110 (2015).
- [32] K. Ueda, J. Fujioka, and Y. Tokura, Physical Review B 93, 245120 (2016).
- [33] S. Nakatsuji, Y. Machida, Y. Maeno, T. Tayama, T. Sakakibara, J. Van Duijn, L. Balicas, J. Millican, R. Macaluso, and J. Y. Chan, Physical review letters 96, 087204 (2006).
- [34] K. Tomiyasu, K. Matsuhira, K. Iwasa, M. Watahiki, S. Takagi, M. Wakeshima, Y. Hinatsu, M. Yokoyama, K. Ohoyama, and K. Yamada, Journal of the Physical Society of Japan 81, 034709 (2012).
- [35] H. Sagayama, D. Uematsu, T.-h. Arima, K. Sugimoto, J. Ishikawa,E. O'farrell, and S. Nakatsuji, Physical Review B 87, 100403 (2013).
- [36] C. Donnerer, M. Rahn, M. M. Sala, J. Vale, D. Pincini, J. Strempfer, M. Krisch, D. Prabhakaran, A. Boothroyd, and D. McMorrow, Physical review letters 117, 037201 (2016).
- [37] D. McWhan, A. Menth, J. Remeika, W. Brinkman, and T. Rice, Physical Review B 7, 1920 (1973).

- [38] J. Torrance, P. Lacorre, A. Nazzal, E. Ansaldo, and C. Niedermayer, Physical Review B 45, 8209 (1992).
- [39] Y. Machida, S. Nakatsuji, Y. Maeno, T. Tayama, T. Sakakibara, and S. Onoda, Physical review letters 98, 057203 (2007).
- [40] S. M. Disseler, S. Giblin, C. Dhital, K. C. Lukas, S. D. Wilson, and M. J. Graf, Physical Review B 87, 060403 (2013).
- [41] K. Ueda, J. Fujioka, Y. Takahashi, T. Suzuki, S. Ishiwata, Y. Taguchi, M. Kawasaki, and Y. Tokura, Physical Review B 89, 075127 (2014).
- [42] E. Y. Ma, Y.-T. Cui, K. Ueda, S. Tang, K. Chen, N. Tamura, P. M. Wu, J. Fujioka, Y. Tokura, and Z.-X. Shen, Science 350, 538 (2015).
- [43] B.-J. Yang, E.-G. Moon, H. Isobe, and N. Nagaosa, Nature Physics 10, 774 (2014).
- [44] L. Savary, E.-G. Moon, and L. Balents, Physical Review X 4, 041027 (2014).
- [45] Y. Tokura, Reports on Progress in Physics 69, 797 (2006).
- [46] E. Lhotel, S. Petit, S. Guitteny, O. Florea, M. C. Hatnean, C. Colin, E. Ressouche, M. Lees, and G. Balakrishnan, Physical Review Letters 115, 197202 (2015).
- [47] Z. Hiroi, K. Matsuhira, S. Takagi, T. Tayama, and T. Sakakibara, Journal of the Physical Society of Japan 72, 411 (2003).
- [48] Y. Onose, Y. br, T. Ito, and Y. Tokura, Physical Review B 70, 060401 (2004).
- [49] X. Huang, L. Zhao, Y. Long, P. Wang, D. Chen, Z. Yang, H. Liang, M. Xue, H. Weng, Z. Fang, *et al.*, Physical Review X 5, 031023 (2015).
- [50] K. Ueda, J. Fujioka, Y. Takahashi, T. Suzuki, S. Ishiwata, Y. Taguchi, and Y. Tokura, Physical review letters 109, 136402 (2012).

- [51] M. Nakayama, T. Kondo, Z. Tian, J. Ishikawa, M. Halim, C. Bareille,
  W. Malaeb, K. Kuroda, T. Tomita, S. Ideta, *et al.*, Physical review letters **117**, 056403 (2016).
- [52] L. Balicas, S. Nakatsuji, Y. Machida, and S. Onoda, Physical review letters 106, 217204 (2011).
- [53] R. Schaffer, E. K.-H. Lee, B.-J. Yang, and Y. B. Kim, Reports on Progress in Physics 79, 094504 (2016).
- [54] B. Lv, H. Weng, B. Fu, X. P. Wang, H. Miao, J. Ma, P. Richard, X. Huang, L. Zhao, G. Chen, *et al.*, Physical Review X 5, 031013 (2015).
- [55] H. Weng, C. Fang, Z. Fang, B. A. Bernevig, and X. Dai, Physical Review X 5, 011029 (2015).
- [56] S.-Y. Xu, I. Belopolski, N. Alidoust, M. Neupane, G. Bian, C. Zhang, R. Sankar, G. Chang, Z. Yuan, C.-C. Lee, *et al.*, Science **349**, 613 (2015).
- [57] P. Goswami, B. Roy, and S. D. Sarma, Physical Review B 95, 085120 (2017).
- [58] J. Cano, B. Bradlyn, Z. Wang, M. Hirschberger, N. P. Ong, and B. A. Bernevig, Physical Review B 95, 161306 (2017).
- [59] J. M. Luttinger, Physical review 102, 1030 (1956).
- [60] J. Hensel and K. Suzuki, Physical Review Letters 22, 838 (1969).
- [61] A. Nenashev, A. Dvurechenskii, and A. Zinovieva, Physical review B **67**, 205301 (2003).
- [62] S. Lee, S. Onoda, and L. Balents, Physical Review B 86, 104412 (2012).
- [63] Y.-P. Huang, G. Chen, and M. Hermele, Physical review letters 112, 167203 (2014).

- [64] S. Murakami, N. Nagaosa, and S.-C. Zhang, Physical review letters 93, 156804 (2004).
- [65] H. Isobe and N. Nagaosa, Physical Review B 86, 165127 (2012).
- [66] H. Isobe and N. Nagaosa, Physical Review B 87, 205138 (2013).
- [67] P. Goswami and S. Chakravarty, Physical review letters 107, 196803 (2011).
- [68] Y. Wang and R. M. Nandkishore, Physical Review B 96, 115130 (2017).
- [69] E.-G. Moon, C. Xu, Y. B. Kim, and L. Balents, Physical review letters 111, 206401 (2013).
- [70] J. Seidel, L. W. Martin, Q. He, Q. Zhan, Y.-H. Chu, A. Rother, M. Hawkridge, P. Maksymovych, P. Yu, M. Gajek, *et al.*, Nature materials 8, 229 (2009).
- [71] D. Meier, J. Seidel, A. Cano, K. Delaney, Y. Kumagai, M. Mostovoy, N. A. Spaldin, R. Ramesh, and M. Fiebig, Nature materials 11, 284 (2012).
- [72] W. Wu, Y. Horibe, N. Lee, S.-W. Cheong, and J. Guest, Physical review letters 108, 077203 (2012).
- [73] G. Catalan, J. Seidel, R. Ramesh, and J. F. Scott, Reviews of Modern Physics 84, 119 (2012).
- [74] J. Matsuno, N. Ogawa, K. Yasuda, F. Kagawa, W. Koshibae, N. Nagaosa, Y. Tokura, and M. Kawasaki, Science advances 2, e1600304 (2016).
- [75] X. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. Han, Y. Matsui, N. Nagaosa, and Y. Tokura, Nature 465, 901 (2010).
- [76] E. G. Tveten, T. Müller, J. Linder, and A. Brataas, Physical Review B 93, 104408 (2016).

- [77] K. Matsuhira, M. Wakeshima, R. Nakanishi, T. Yamada, A. Nakamura, W. Kawano, S. Takagi, and Y. Hinatsu, Journal of the Physical Society of Japan 76, 043706 (2007).
- [78] B.-J. Yang and N. Nagaosa, Physical review letters 112, 246402 (2014).
- [79] M. E. Fisher and A. E. Ferdinand, Physical Review Letters 19, 169 (1967).
- [80] E. Cordfunke and G. Meyer, Recueil des Travaux Chimiques des Pays-Bas **81**, 495 (1962).
- [81] J. Gallagher, B. Esser, R. Morrow, S. Dunsiger, R. Williams, P. Woodward, D. McComb, and F. Yang, Scientific reports 6, 1 (2016).
- [82] W. Yang, Y. Xie, W. Zhu, K. Park, A. Chen, Y. Losovyj, Z. Li, H. Liu, M. Starr, J. A. Acosta, *et al.*, Scientific reports 7, 1 (2017).
- [83] K. Jacob, T. Okabe, T. Uda, and Y. Waseda, Electrochimica acta 45, 1963 (2000).
- [84] T. Fujita, Y. Kozuka, M. Uchida, A. Tsukazaki, T.-h. Arima, and M. Kawasaki, Scientific reports 5, 1 (2015).
- [85] V. Wadhawan, Introduction to ferroic materials (CRC press, 2000).
- [86] K. G. Webber, H. C. Robinson, G. A. Rossetti Jr, and C. S. Lynch, Acta Materialia 56, 2744 (2008).
- [87] Z. Zhao, K. Bowman, and R. E. García, Journal of the American Ceramic Society 95, 1619 (2012).
- [88] X.-G. Wen, F. Wilczek, and A. Zee, Physical Review B 39, 11413 (1989).
- [89] K. Ohgushi, S. Murakami, and N. Nagaosa, Physical Review B 62, R6065 (2000).

- [90] Y. Taguchi, Y. Oohara, H. Yoshizawa, N. Nagaosa, and Y. Tokura, Science 291, 2573 (2001).
- [91] Y. Taguchi, T. Sasaki, S. Awaji, Y. Iwasa, T. Tayama, T. Sakakibara,
   S. Iguchi, T. Ito, and Y. Tokura, Physical review letters 90, 257202 (2003).
- [92] P. Bruno, V. Dugaev, and M. Taillefumier, Physical review letters 93, 096806 (2004).
- [93] C. Sürgers, G. Fischer, P. Winkel, and H. v. Löhneysen, Nature communications 5, 1 (2014).
- [94] S. Nakatsuji, N. Kiyohara, and T. Higo, Nature 527, 212 (2015).
- [95] T.-h. Arima, Journal of the Physical Society of Japan 82, 013705 (2012).
- [96] S. Tardif, S. Takeshita, H. Ohsumi, J.-i. Yamaura, D. Okuyama, Z. Hiroi, M. Takata, and T.-h. Arima, Physical review letters 114, 147205 (2015).
- [97] H. T. Hirose, J.-i. Yamaura, and Z. Hiroi, Scientific reports 7, 1 (2017).
- [98] A. Burkov, Physical review letters **113**, 187202 (2014).
- [99] S. Nishihaya, M. Uchida, Y. Nakazawa, R. Kurihara, K. Akiba, M. Kriener, A. Miyake, Y. Taguchi, M. Tokunaga, and M. Kawasaki, Nature communications 10, 1 (2019).
- [100] L. Šmejkal, Y. Mokrousov, B. Yan, and A. H. MacDonald, Nature physics 14, 242 (2018).
- [101] M. Onoda and N. Nagaosa, Journal of the Physical Society of Japan 71, 19 (2002).
- [102] N. Kiyohara, T. Tomita, and S. Nakatsuji, Physical Review Applied 5, 064009 (2016).

- [103] A. K. Nayak, J. E. Fischer, Y. Sun, B. Yan, J. Karel, A. C. Komarek, C. Shekhar, N. Kumar, W. Schnelle, J. Kübler, *et al.*, Science advances 2, e1501870 (2016).
- [104] T. Suzuki, R. Chisnell, A. Devarakonda, Y.-T. Liu, W. Feng, D. Xiao, J. W. Lynn, and J. Checkelsky, Nature Physics 12, 1119 (2016).
- [105] M. Ikhlas, T. Tomita, T. Koretsune, M.-T. Suzuki, D. Nishio-Hamane, R. Arita, Y. Otani, and S. Nakatsuji, Nature Physics 13, 1085 (2017).
- [106] T. Higo, H. Man, D. B. Gopman, L. Wu, T. Koretsune, O. M. van't Erve, Y. P. Kabanov, D. Rees, Y. Li, M.-T. Suzuki, *et al.*, Nature photonics **12**, 73 (2018).
- [107] Y. Ohuchi, J. Matsuno, N. Ogawa, Y. Kozuka, M. Uchida, Y. Tokura, and M. Kawasaki, Nature communications **9**, 1 (2018).
- [108] T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich, Nature nanotechnology 11, 231 (2016).
- [109] K. Ueda, R. Kaneko, H. Ishizuka, J. Fujioka, N. Nagaosa, and Y. Tokura, Nature communications 9, 1 (2018).
- [110] X. Ke, T. Birol, R. Misra, J.-H. Lee, B. Kirby, D. Schlom, C. Fennie, and J. Freeland, Physical Review B 88, 094434 (2013).
- [111] C. Alcock and G. Hooper, Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences 254, 551 (1960).
- [112] W. J. Kim, E. K. Ko, S. Y. Kim, B. Kim, and T. W. Noh, Current Applied Physics **19**, 400 (2019).
- [113] D. Chiba, Y. Nishitani, F. Matsukura, and H. Ohno, Applied physics letters 90, 122503 (2007).
- [114] W.-L. Lee, S. Watauchi, V. Miller, R. Cava, and N. Ong, Science 303, 1647 (2004).

- [115] S. Y. Kim, M.-C. Lee, G. Han, M. Kratochvilova, S. Yun, S. J. Moon, C. Sohn, J.-G. Park, C. Kim, and T. W. Noh, Advanced Materials 30, 1704777 (2018).
- [116] E. Dagotto, T. Hotta, and A. Moreo, Physics reports 344, 1 (2001).
- [117] P. A. Lee, N. Nagaosa, and X.-G. Wen, Reviews of modern physics 78, 17 (2006).
- [118] J. E. Moore, Nature **464**, 194 (2010).
- [119] K. Kuroda, T. Tomita, M.-T. Suzuki, C. Bareille, A. Nugroho, P. Goswami, M. Ochi, M. Ikhlas, M. Nakayama, S. Akebi, *et al.*, Nature materials 16, 1090 (2017).
- [120] J. Xiong, S. K. Kushwaha, T. Liang, J. W. Krizan, M. Hirschberger, W. Wang, R. J. Cava, and N. P. Ong, Science 350, 413 (2015).
- [121] J. Ahn, D. Kim, Y. Kim, and B.-J. Yang, Physical review letters 121, 106403 (2018).
- [122] Y. Shao, A. Rudenko, J. Hu, Z. Sun, Y. Zhu, S. Moon, A. Millis, S. Yuan, A. Lichtenstein, D. Smirnov, *et al.*, Nature Physics 16, 636 (2020).
- [123] Y. Xu, J. Zhao, C. Yi, Q. Wang, Q. Yin, Y. Wang, X. Hu, L. Wang, E. Liu, G. Xu, *et al.*, Nature communications **11**, 1 (2020).
- [124] G. B. Osterhoudt, Y. Wang, C. A. Garcia, V. M. Plisson, J. Gooth, C. Felser, P. Narang, and K. S. Burch, Physical Review X 11, 011017 (2021).
- [125] S. Nandy, G. Sharma, A. Taraphder, and S. Tewari, Physical review letters 119, 176804 (2017).
- [126] S. A. A. Ghorashi, P. Hosur, and C.-S. Ting, Physical Review B 97, 205402 (2018).

- [127] Y. Machida, S. Nakatsuji, S. Onoda, T. Tayama, and T. Sakakibara, Nature 463, 210 (2010).
- [128] T. Liang, J. Lin, Q. Gibson, S. Kushwaha, M. Liu, W. Wang, H. Xiong, J. A. Sobota, M. Hashimoto, P. S. Kirchmann, *et al.*, Nature Physics 14, 451 (2018).
- [129] L. Guo, N. Campbell, Y. Choi, J.-W. Kim, P. J. Ryan, H. Huyan, L. Li, T. Nan, J.-H. Kang, C. Sundahl, *et al.*, Physical Review B **101**, 104405 (2020).
- [130] Z. Fu, P. Kögerler, U. Rücker, Y. Su, R. Mittal, and T. Brückel, New journal of physics 12, 083044 (2010).
- [131] R. Dos Reis, M. Ajeesh, N. Kumar, F. Arnold, C. Shekhar, M. Naumann, M. Schmidt, M. Nicklas, and E. Hassinger, New Journal of Physics 18, 085006 (2016).
- [132] K. Takiguchi, Y. K. Wakabayashi, H. Irie, Y. Krockenberger, T. Otsuka, H. Sawada, S. A. Nikolaev, H. Das, M. Tanaka, Y. Taniyasu, *et al.*, Nature communications **11**, 1 (2020).
- [133] T. Ando, A. B. Fowler, and F. Stern, Reviews of Modern Physics 54, 437 (1982).
- [134] A. Caviglia, S. Gariglio, C. Cancellieri, B. Sacépé, A. Fete, N. Reyren, M. Gabay, A. Morpurgo, and J.-M. Triscone, Physical review letters 105, 236802 (2010).
- [135] M. Hirschberger, S. Kushwaha, Z. Wang, Q. Gibson, S. Liang, C. A. Belvin, B. A. Bernevig, R. J. Cava, and N. P. Ong, Nature materials 15, 1161 (2016).
- [136] A. Burkov, Physical Review B 96, 041110 (2017).
- [137] P. Li, C. Zhang, J. Zhang, Y. Wen, and X. Zhang, Physical Review B 98, 121108 (2018).

- [138] F. Seitz, Physical Review **79**, 372 (1950).
- [139] A. Taskin, H. F. Legg, F. Yang, S. Sasaki, Y. Kanai, K. Matsumoto, A. Rosch, and Y. Ando, Nature communications 8, 1 (2017).
- [140] S. Nandy, A. Taraphder, and S. Tewari, Scientific Reports 8, 1 (2018).
- [141] T. Liang, T. H. Hsieh, J. J. Ishikawa, S. Nakatsuji, L. Fu, and N. P. Ong, Nature Physics 13, 599 (2017).
- [142] T. Ohtsuki, Z. Tian, A. Endo, M. Halim, S. Katsumoto, Y. Kohama, K. Kindo, M. Lippmaa, and S. Nakatsuji, Proceedings of the National Academy of Sciences 116, 8803 (2019).
- [143] P. Hosur, S. Parameswaran, and A. Vishwanath, Physical review letters 108, 046602 (2012).
- [144] S. D. Sarma, E. Hwang, and H. Min, Physical Review B 91, 035201 (2015).
- [145] D. E. Sheehy and J. Schmalian, Physical Review Letters 99, 226803 (2007).
- [146] V. N. Kotov, B. Uchoa, V. M. Pereira, F. Guinea, and A. C. Neto, Reviews of modern physics 84, 1067 (2012).

## 국문초록

응집물질에서의 물리는 크게 스핀 오비탈 결합과 전자 간의 상호작 용으로 결정된다. 스핀 오비탈 결합과 전자 간 상호작용이 모두 밴드 폭과 비슷하게 큰 경우, 강상 위상학적 상인 바일 준금속, 액시온 부도체, 또는 위상 모트 부도체 등이 나타난다. 이들은 모두 특이한 물성을 지녔기에 주목을 점점 더 많이 받고 있다.

파이로클로르 이리듐 산화물의 화학식은 R<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>로, 상호작용과 스 핀 오비탈 결합이 모두 큰 시스템이다. 따라서 이 물질군은 강상 위상학적 상을 조사할 수 있는 좋은 배경이 된다. 특히, 이 물질군은 낮은 온도에 서 반강자성이 생기면서 바일 준금속이 나타날 수 있는 것으로 예측된 첫 번째 후보이다. 이 때 반강자성의 이름을 All-in-all-out (AIAO)이라고 부르는데 이는 모든 스핀이 동시에 셀 중심을 향하거나 바깥을 향하기 때 문이다.

그러나 자성 바일 준금속이라는 결정적인 증거는 아직 발견되지 않았 는데, 두 가지 이유가 있다. 첫번째로는 이 물질군이 대부분 부도체이므로 바일 준금속을 볼 수 있는 범위가 매우 좁다. 두번째로는 이 물질군에서 바일 준금속이 생기더라도 이에 의해서 나올 수 있는 물성이 정육면체 대칭성에 의해서 모두 사라지기 때문이다. 이 물질군에서 바일 준금속을 찾기 위해서 우리는 이 물질이 반강자성 부도체에서 상자성 도체가 되는 전이점 근처에서 섭동을 걸어주었다. 이 섭동은 바일 준금속을 볼 수 있는 범위를 넓혀 줄 뿐 아니라 대칭성도 깨줄 것으로 예상되었으므로 위의 어 려움이 모두 극복될 것으로 예상되었다. 섭동으로 이용된 것은 자기장과 변형이다.

첫째로, 자기장이 반강자성 부도체와 상자성 도체 사이에 존재하는 (Nd<sub>x</sub> Pr<sub>1-x</sub>)<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> 단결정에 걸렸을 때 매우 다양한 위상학적 준금속이 관측되었다. 군론을 이용하면 상자성 도체에 존재하는 이차 밴드 겹침이 높은 스핀 (*J* = 3/2)을 가지기 때문에 자기장이 걸렸을 때 일반적인 제만

항 뿐 아니라 비등방성 제만 항이 추가적으로 나오는 것을 볼 수 있었다. 이 두 제만 항과 AIAO 사이의 상호관계는 4쌍 바일, 2쌍 바일, 이차 바일, 그리고 선 겹침 준금속을 만들어 내었다. 이 때 자기장이 파이로클로르 이리듐 산화물 내부의 스핀 구조를 바꾸면서 제만 항과 AIAO을 조절하는 것으로 알려졌다.

둘째로, 반강자성 부도체인 Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>와 상자성 도체인 Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> 박막 에 각각 변형이 걸렸다. Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>의 경우 변형이 부도체-도체 전이와 함께 자기장이 없을 때 이상 홀 효과를 유도하였다. 모델을 이용한 계산은 이것 이 바일 준금속이 아니라 전자 및 양공 주머니가 있는 단순한 도체임을 보였다. 또한 자기장이 없을 때 자화가 없으므로, 이상 홀 효과의 원인은 바일 준금속도 자화도 아니었다. 사실 변형이 유도하는 *T*<sub>1</sub>-팔극자가 이상 홀 효과의 원인인데, 이는 이 팔극자가 자화랑 대칭성이 같기 때문이다. 반면 Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>의 경우, 이상 홀 효과, 음의 자기저항, 평면 홀 효과 등 바 일 준금속의 증거가 많이 발견되었다. 그 중에서도 평면 홀 효과는 바일 준금속의 카이랄 이상과 AIAO으로 설명되었다. 이 졸업 논문에서는 바일 준금속이 파이로클로르 준금속에서 생기는 원인과 어떻게하면 그것을 찾 을 수 있는지를 제공한다. 따라서 이는 파이로클로르 이리듐 산화물에서 새로운 위상학적 상의 실험적 발견을 촉진할 것이다.

**주요어:** 위상학적 현상, 이상 홀 효과, 파이로클로르 이리듐 산화물, 바일 준금속

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이외에도 저를 지켜봐주던 많은 친구들에게 감사드립니다. 같은 학 부였던 양희준, 고귀한, 권순철, 정지우, 김주한, 김학래, 이상민, 왕지성, 중학교 친구들이었던 황중현, 정하영, 정희정, 이재선, 윤상훈, 채희찬, 최 찬환에게 감사드립니다. 고등학교 친구들이었던 송서우 박사, 박형주 박 사, 정성엽 박사, 김민혜, 류준호, 송서우, 전병진, 최선희에게 감사드립 니다. 대학 때 알게 된 KIAS에 있는 유세민 박사님에게도 감사드립니다. 학부 때 같은 동아리원이었던 손샘, 윤태호, 김경수, 이진우, 구윤모 박사, 김동리 박사, 이도엽, 이진걸, 이주영 누님과 다른 모두에게도 감사드립니 다. 많은 이야기를 나누어주신 김유훈 형님, 그리고 이제 미국에서 교수 생활을 하게 될 심재웅 박사에게도 감사드립니다.

제가 불민하여 여기에 제게 도움을 주신 모든 이름을 거론하지 못 했습니다. 대학원 기간 동안 저를 지탱해주신 많은 친구 동료 선배 후배 여러분께 정말로 감사드립니다.

마지막으로 가장 중요한 사람들에게 인사드립니다. 박사과정을 마무 리 짓는 동안에 제게 애정을 많이 쏟아주었던 사랑하는 여자친구 김자연 님 감사합니다. 저를 언제나 지지해 주는 한 축인 부모님, 그리고 다른 축 인 여동생과 매제에게도 감사의 말씀 드립니다.