

Quasi-Flat-Band in s1/s2 Pyrochlore Oxides and the Effect of Spin-Orbit Interaction

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Abstract. An isotropic tight-binding model with the nearest-neighbour hopping on a pyrochlore lattice gives a rich variety of physical properties due to the emergence of the flat-band. Moreover, by introducing spin-orbit coupling into this model, the topological properties of the system changes significantly. This model is well applicable to some pyrochlore oxides called s1/s2 pyrochlores. In this paper we apply this model to an s2 pyrochlore oxide $\text{Pb}_2\text{Ta}_2\text{O}_7$ and found a characteristic quadratic touching of the quasi-flat band and dispersive band. Furthermore, when a ferromagnetic order appears due to this quasi-flat band, a pair of Weyl points appears in that direction.

1. Introduction

The energy of electrons in a crystal generally has dispersion. However, in a model such as a nearest-neighbor isotropic tight-binding (NNITB) model on pyrochlore lattice, flat bands without dispersion appear due to the quantum interference effect. In this flat band model, attractive physical properties such as perfect ferromagnetism, high-temperature superconductivity, and fractional quantum Hall effect are predicted [1,2]. However, in the actual pyrochlore compound, there has been no example so far in which an approximate flat band was observed near the Fermi level due to the influence of the anisotropy of the transfer integral. Based on first-principles calculations, we have found that such a system in which the uppermost part of the valence band is mainly the s-orbital of the A site, such as $\text{Sn}_2\text{Nb}_2\text{O}_7$ and $\text{Tl}_2\text{Nb}_2\text{O}_7$, is such an example [3-5]. Hereafter we call a system containing one s-electron in the outermost orbital as “s1 system” (for example $\text{Tl}_2\text{Nb}_2\text{O}_7$), and a system containing two s-electrons as “s2 system” (for example $\text{Sn}_2\text{Nb}_2\text{O}_7$). The bandwidth of the top of the valence band is very narrow, even though they are composed of extended s-orbitals. Hereafter we call this band as quasi-flat band. This quasi-flat band is sometimes found in two-dimensional materials, but extremely rare in three-dimensional materials. Recently, a calculation result has been reported that three-dimensional quasi-flat bands can be seen in a binary compound WN_2 [6].

NNITB model on the pyrochlore lattice is also drawing attention from the perspective of topological insulator [7]. Guo and Franz considered a model in which a spin-orbit coupling (SOC) term that does not break crystal translational symmetry is added to the NNITB model (GF model) [7]. At that time, the GF model could not be applied to specific material, but now it can be applied to the above s1/s2 pyrochlore oxides. In the GF model, the topological properties of the system are governed by the sign



of the SOC constant λ . Especially in the half-filled case, $\lambda < 0$ leads to a strong topological insulator (STI). On the other hand, $\lambda > 0$ leads to a semi-metallic state where the bands touch quadratically at the Γ point. Similar results have been obtained using other type of SOC by Kurita *et al.* [8]. These properties are expected to hold in real s1/s2 materials because the topological properties may not depend on the details of the band structure [9].

In this paper we try to reproduce the valence band of $\text{Pb}_2\text{Ta}_2\text{O}_7$ [9], which is one of the s2 compound, by using GF model with adjusting parameters. We chose this compound because $\text{Pb}_2\text{Ta}_2\text{O}_7$ has relatively narrow quasi-flat band and large SOC. We found that the magnitude of λ is about one order smaller than that shown in Ref. 7. Nevertheless, the band topology is unchanged and a characteristic quadratic touching of the bands at the Γ point. Furthermore, we considered a state peculiar to the flat-band system, i.e. a case where the system undergoes a phase transition to a ferromagnetic state due to doping. When $\lambda > 0$, the zero-gap semi-metallic state that was touched quadratically at the Γ point is almost gapped out by the Zeeman term, but a pair of Weyl points in the direction of the Zeeman field. This behavior is similar to the case where $\lambda < 0$, i.e. (topological) insulating phase transfers to the semi-metallic phase and the Weyl points appear by adding the Zeeman term [10].

This paper is organized as follows: The computational details are described in Section 2. The calculated results and the discussions are shown in Section 3. Finally, we give some conclusions in Section 4.

2. Computational Details

Guo-Franz (GF) model consists of two terms shown in the following Hamiltonian:

$$H = -t \sum_{\langle ij \rangle \sigma} c_{i\sigma}^\dagger c_{j\sigma} + i\lambda \sum_{\langle\langle ij \rangle\rangle \alpha\beta} (\mathbf{d}_{ij}^1 \times \mathbf{d}_{ij}^2) \cdot \boldsymbol{\sigma}_{\alpha\beta} c_{i\alpha}^\dagger c_{j\beta} \quad (1)$$

The first term denotes the non-interacting tight-binding Hamiltonian. t is the hopping parameter and $\langle ij \rangle$ denotes nearest neighbors. The lattice sum is taken on the pyrochlore lattice. When the second term is zero (i.e. $\lambda = 0$), this Hamiltonian is exactly diagonalized and the eigenvalues are

$$E_k^{1,2} = -2t[1 \pm \sqrt{1 + A_k}], E_k^{3,4} = 2t \quad (2)$$

where $A_k = \cos(2k_x) \cos(2k_y) + \cos(2k_y) \cos(2k_z) + \cos(2k_z) \cos(2k_x)$. E^3 and E^4 do not depend on the wave vector \mathbf{k} , so they are called flat bands (FBs). Note that $A = 3$ at $\mathbf{k} = 0$ (Γ point), so $E^{1,2} = -6t$ and $2t$. Therefore, FBs touch one of the dispersive bands E^2 at the Γ point and forms a triply degenerated state (six-fold degenerated state if we include spin degree of freedom). Since we can see that $A = 3$ is the maximum value of A , the overall bandwidth of the entire system (E^1 to E^4) becomes $8t$.

Next, we include spin-orbit coupling (SOC) shown in the second term in Eqn. 1. This term is taken so as not to break crystal symmetry further, and to preserve time-reversal symmetry. Here λ is the SOC constant, \mathbf{d}_{ij}^1 and \mathbf{d}_{ij}^2 are the nearest-neighbor vectors traversed between second neighbors i and j . The summation $\langle\langle ij \rangle\rangle$ is taken on the pyrochlore lattice such that i and j are the (Euclid) second neighbor sites. $\boldsymbol{\sigma}$ denotes the vector of Pauli spin matrices. Due to this SOC term, we must diagonalize 8×8

matrix in k -space. We have calculated the band structure of the Hamiltonian (1) by using the program package PythTB [11].

3. Results and Discussions

We calculate the band structure of the Hamiltonian (1) using the program package PythTB [11]. The results are shown in Figure 1(a). We take the parameters $t = 0.3$ eV and $\lambda = 2.0$ meV, which well reproduce the valence band structure of a s2 material $\text{Pb}_2\text{Ta}_2\text{O}_7$ [9]. Since the ratio $\lambda/t \sim 6.7 \times 10^{-3}$ which is much smaller than that shown in ref.7, the effect of SOC to the band dispersion is limited to the vicinity of the Γ point. Nevertheless, the topology of the band dispersion is the same with that in ref.7. Note that the quadratic touching of the bands at the Γ point is recognized as a quadratic contact point in s1-pyrochlore compound $\text{Ti}_2\text{Nb}_2\text{O}_7$ [12].

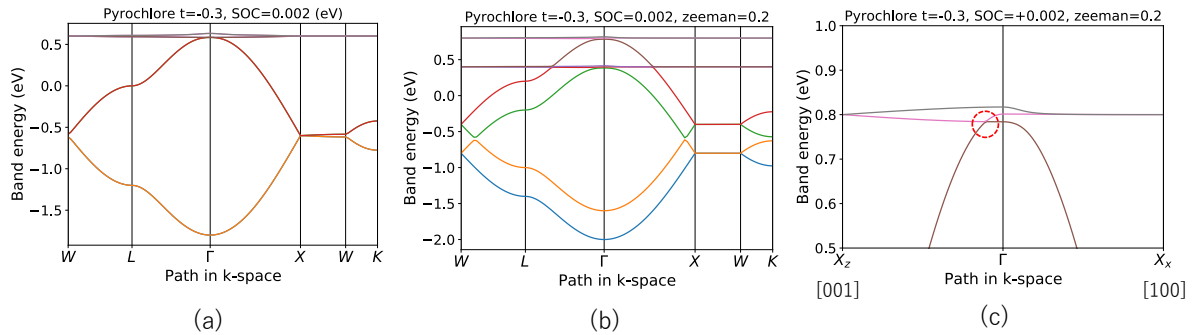


Figure 1. Energy dispersion curves of (a) GF model with $t = 0.3$ eV and $\lambda = 2$ meV, (b) GF model with $t = 0.3$ eV, $\lambda = 2$ meV with including Zeeman term $\lambda_z = 0.2$ eV, and (c) an enlarged view of the panel (b) near the Γ point, while the k -direction is $[001]$ and $[100]$. A Weyl point is shown in dashed circle.

As shown in Introduction, (quasi-)flat band forms a very large density of states, and when the Fermi level (E_F) comes there, the nonmagnetic state becomes unstable, and the ferromagnetic state stabilizes [1-5]. We have found ferromagnetic ground state for the hole-doped s2 pyrochlore system using first-principles calculations [3-5]. Recently, Zhou *et al.* have shown that when SOC is introduced into this ferromagnetic state, a new type of Weyl points emerges [10]. $\text{Sn}_2\text{Nb}_2\text{O}_7$ has a topological gap at the Γ point, and the energy splitting Δ which is defined in the previous subsection is ~ -5 meV. They included the Zeeman term

$$H_Z = \lambda_Z \sum_{\alpha\beta} \sigma_{\alpha\beta}^z c_{i\alpha}^\dagger c_{i\beta} \quad (3)$$

into the GF-model with using the parameter $\lambda_z = 0.2$ eV as half of the exchange splitting in hole-doped $\text{Sn}_2\text{Nb}_2\text{O}_7$.

Since most of the s1/s2 pyrochlore oxides have $\lambda > 0$, it is important to investigate the Zeeman effect to the GF model with $\lambda > 0$. We introduced the Zeeman term of $\lambda_z = 0.2$ eV into the GF model with $t = 0.3$ eV and $\lambda = 2$ meV that modeled $\text{Pb}_2\text{Ta}_2\text{O}_7$. The direction of the Zeeman field was set along the z -axis $[001]$. The results are shown in Fig. 1(c).

As shown in Fig.1(a), since the GF-model has time-reversal symmetry and the pyrochlore lattice has also space inversion symmetry, all bands are doubly degenerated. The Zeeman term breaks the time-reversal symmetry, and all the bands are split as shown in Fig.1(b). Since $\lambda_z \gg \lambda$, these split bands

almost have the character with spin-up or spin-down. The width of the topmost band ($E^{3,4}$ in Eqn.2) narrows significantly with the introduction of the Zeeman term. Figure 1(c) shows an enlarged view of the topmost band of Fig.1(b) near E_F , but the \mathbf{k} -direction is for [001] and [100]. When $\lambda_z = 0$, these two directions are equivalent due to the cubic lattice symmetry. However, the Zeeman term partially breaks this symmetry via SOC. Figure 1(c) apparently shows this symmetry breaking. As for the direction [001], which is parallel to the Zeeman field, the quasi-flat band and the dispersive band cross each other. This band crossing is also seen when $\lambda = -2$ meV (not shown), which has been shown to be a Weyl point [10]. This Weyl point at $(0, 0, k_z)$ in Fig.1(c) also appears at $(0, 0, -k_z)$ due to symmetry. As shown in the right half in Fig.3(c), Weyl points do not appear in any direction other than [001].

It is interesting to compare these results with the case of $\lambda < 0$, which seems to be realized in $\text{Sn}_2\text{Nb}_2\text{O}_7$ [10]. When $\lambda < 0$, a pair of Weyl points also appears when the Zeeman term is added. The difference from the present result is that when $\lambda < 0$, there is a parameter region in which four Weyl points appear, whereas when $\lambda > 0$, there are always two Weyl points. This is because the energy levels at the Γ point are reversed between these two cases. It is a future task to investigate the detailed properties of these Weyl points. We note that there are many s1/s2 pyrochlore oxides which have $\lambda > 0$ [9], and these compounds have ferromagnetic ground state when doped appropriately [5]. Therefore, our result could be applied for many realistic materials.

4. Conclusion

We have applied the Guo-Franz model to the valence band structure of the s2 pyrochlore oxide $\text{Pb}_2\text{Ta}_2\text{O}_7$. We determined the parameters of the Guo-Franz model and found that the effect of spin-orbit coupling is almost limited to the vicinity of the Γ point. We have found that the semi-metallic quadratic touching point when the spin-orbit coupling constant is positive. We also introduced the Zeeman term into this model, simulating the possible ferromagnetic ground state with doping. This Zeeman term creates a pair of Weyl points in direction of the Zeeman field.

Acknowledgments

This work was partially supported by KAKENHI (Grant No. JP19K03731) from Japan Society for the Promotion of Science (JSPS).

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