Spin Susceptibility in Non-Centrosymmetric Superconductors with Topological Transition of Fermi Surfaces

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The non-centrosymmetric superconductors Li2Pd3B and Li2Pt3B show different superconducting properties despite having the same crystal symmetry. Motivated by experimental results, we investigate the spin susceptibility of non-centrosymmetric superconductors accompanied by the topological transition of Fermi surfaces due to antisymmetric spin-orbit coupling, which is indicated by the first-principles band structure calculation for Li2Pt3B. We study three types of topological transition, namely, (A) the disappearance of the Fermi surface, (B) crossing the Dirac point, and (C) crossing the saddle point van-Hove singularity. The spin susceptibility in the superconducting state is increased by the topological transitions (A) and (C), while it is decreased by (B). We discuss the unusual magnetic properties observed in Li2Pt3B on the basis of these results.

KEYWORDS: superconductivity without inversion symmetry, topological transition of Fermi surfaces, spin susceptibility

1. Introduction

Recently, superconductors lacking inversion symmetry in the crystal structure have been attracting much attention. The antisymmetric spin-orbit coupling induced by the broken inversion symmetry leads to the spin-splitting of the Fermi surface and gives rise to unique superconducting properties, such as the parity mixing of Cooper pairs.1)

Among many non-centrosymmetric superconductors, the perovskite-like cubic compounds Li2Pd3B2) and Li2Pt3B3) show particularly intriguing properties. The superconducting properties are different between these two compounds in spite of having the same crystal symmetry.2–12) The order parameter is fully gapped in Li2Pd3B, while it has line nodes in Li2Pt3B.4–7, 9, 12) The NMR Knight shift of Li2Pd3B is decreased across the superconducting transition temperature Tc, while that of Li2Pt3B is mostly unaffected in the superconducting state.6, 12) Although these behaviors of Li2Pd3B indicate the conventional s-wave superconductivity admixed with the spin triplet p-wave one owing to the antisymmetric spin-orbit coupling, experimental results of Li2Pt3B are incompatible with the canonical theory of non-centrosymmetric superconductivity.1)

According to the weak coupling theory neglecting the correlation effects, the spin susceptibility in the cubic non-centrosymmetric superconductor should be reduced to 2/3 of the normal state value at T = 0.13, 14) On the other hand, the Knight shift measurement of Li2Pt3B did not show such a decrease in spin susceptibility.6, 12) The spin triplet superconducting state has been proposed for Li2Pt3B on the basis of this experimental result.6, 12) However, the spin susceptibility at low magnetic fields µ0H ≪ kBTc is independent of the symmetry of the order parameter.15) Indeed, the pairing states indicated by theoretical studies16, 17) are incompatible with the Knight shift measurement of Li2Pt3B. Although the electron correlation effect may enhance the spin susceptibility in the superconducting state,13) such enhancement is unlikely to occur in Li2Pd3B and Li2Pt3B in which the correlation effect is negligible.8, 10) The influence of magnetic order has been pointed out for the heavy fermion superconductor CePt3Si18, 19) however, the magnetic order does not occur in Li2Pt3B. Thus, the superconducting state of Li2Pt3B remains controversial, while Li2Pd3B is a “conventional” non-centrosymmetric superconductor.

For the difference between Li2Pt3B and Li2Pd3B, a substantial enhancement of antisymmetric spin-orbit coupling has been pointed out for Li2Pt3B.20) The increase in atomic LS-coupling on Pt ions as well as the deformation of crystal structure12) significantly increases the antisymmetric spin-orbit coupling of Li2Pt3B. According to the first-principles band structure calculation, this enhancement of antisymmetric spin-orbit coupling is accompanied by the topological transition of the Fermi surfaces (FS topological transition).21) The Fermi surfaces of Li2Pd3B consist of several pairs of spin-split Fermi surfaces. On the other hand, the counterpart of some pairs vanishes in Li2Pt3B. According to recent studies of the crystal structure of solid solution Li2(Pd1−xPtx)3B, the structural deformation occurs at approximately x ∼ 0.8,12) which is probably accompanied by the FS topological transition.

The purpose of this study is to clarify the effect of FS topological transition due to the antisymmetric spin-orbit coupling on the superconducting state. We here study the roles of three types of FS topological transition on the spin susceptibility in the superconducting state. In type (A), one of the spin-split Fermi surfaces vanishes owing to the substantial increase in the spin-orbit coupling [see Fig. 1(a)]. In type (B), the Fermi surface crosses the Dirac point with increasing spin-orbit coupling, as shown in Fig. 1(b). Finally, in type (C), one of the spin-split Fermi surfaces crosses a van-Hove singularity, as shown in Fig. 1(c). Our analysis is based on a single-band model, which cannot reproduce the electronic structure of Li2(Pd1−xPtx)3B. However, some effects of FS topological transition are independent of the band structure, as shown below. We expect that the following results would be a key to resolve unsettled issues of Li2(Pd1−xPtx)3B. Our results are...
also applicable to other non-centrosymmetric superconductors with a large spin-orbit coupling. We introduce the model Hamiltonian in Sect. 2 and show the numerical results of spin susceptibility in Sect. 3. Some remarks are given in Sect. 4.

2. Model

We adopt the following single-band Hamiltonian;

\[
H = \sum_{\mathbf{k}, s} \varepsilon(k) c_{\mathbf{k}s}^\dagger c_{\mathbf{k}s} + \alpha \sum_{\mathbf{k}, \mathbf{k}', s} \mathbf{g}(\mathbf{k}) \cdot \sigma_{ss'} c_{\mathbf{k}s}^\dagger c_{\mathbf{k}'s'} + \frac{1}{2} \sum_{\mathbf{k}, \mathbf{k}', s} \left[ \Delta_{s's'}(\mathbf{k}) c_{\mathbf{k}s}^\dagger c_{\mathbf{k}'s'} + \text{h.c.} \right],
\]

where \( c_{\mathbf{k}s} \) (\( c_{\mathbf{k}s}^\dagger \)) is the annihilation (creation) operator for an electron with a momentum \( \mathbf{k} \) and a spin \( s \). The dispersion relation \( \varepsilon(k) \) is assumed so that the FS topological transition occurs with increasing antisymmetric spin-orbit coupling. The chemical potential \( \mu \) is involved in the dispersion relation and determined so that the electron density per site is \( n \). The second term describes the antisymmetric spin-orbit coupling, which preserves the time reversal symmetry for the antisymmetric \( g \)-vector \( \mathbf{g}(\mathbf{-k}) = -\mathbf{g}(\mathbf{k}) \). The spin-orbit coupling lifts the two-fold degeneracy in the band as \( \varepsilon_s(k) = \varepsilon(k) \pm \alpha g(k) \).

In this research, we study the two-dimensional Rashba spin-orbit coupling with \( \mathbf{g}(\mathbf{k}) = (-\sin k_x, \sin k_y, 0) \) as well as the three-dimensional cubic spin-orbit coupling with \( \mathbf{g}(\mathbf{k}) = (\sin k_x, \sin k_y, \sin k_z) \).

We take into account the mean field of superconducting order parameters in the last term of Eq. (1). The order parameters \( \Delta_{s's'}(\mathbf{k}) \) involve both spin singlet and triplet components due to the spin-orbit coupling. We here ignore the spin triplet component and assume the s-wave spin singlet order parameter \( \Delta_{s's'}(\mathbf{k}) = -\Delta_{s's'}(\mathbf{-k}) \), since the spin susceptibility at zero temperature is independent of the symmetry of order parameters for a large spin-orbit coupling \( |\Delta_{s's'}(\mathbf{k})| \ll \alpha \).

3. Spin Susceptibility in the Superconducting State

In this section, we calculate the spin susceptibility in the superconducting state. We consider the zero temperature \( T = 0 \) throughout this paper. The spin susceptibility \( \chi = \lim_{H \to 0} \langle M^2 \rangle / H \) is obtained by calculating the magnetization \( \langle M \rangle \) in the field \( H \) and taking the limit \( H \to 0 \). The Zeeman coupling term is introduced as \( H_Z = -(\mu_B/2) \sum_{\mathbf{k}, \sigma} \mathbf{H} \cdot \sigma_{ss'} c_{\mathbf{k}s}^\dagger c_{\mathbf{k}'s'} \), where we assume \( g = 2 \) and \( \mu_B \) is the Bohr magneton. We first study the two-dimensional systems with Rashba spin-orbit coupling. Later, we will show the results for three-dimensional systems with cubic spin-orbit coupling. For two-dimensional systems, we focus on the spin susceptibility in the \( ab \)-plane, since that along the \( c \)-axis is not reduced by the superconductivity.\(^2\) On the other hand, the spin susceptibility is isotropic in the cubic system. We discuss the spin susceptibility normalized by the normal state value \( \chi_n/\chi_s \), where \( \chi_s \) is the spin susceptibility in the superconducting state. The normal state value of spin susceptibility \( \chi_n \) is calculated at \( T = 0 \) for \( |\Delta_{ss'}(\mathbf{k})| = 0 \).

3.1 Two-dimensional systems

First, we study two-dimensional systems with the Rashba spin-orbit coupling \( \mathbf{g}(\mathbf{k}) = (-\sin k_x, \sin k_y, 0) \). The dispersion relation is assumed as

\[
\varepsilon(k) = 2t_1(\cos k_x + \cos k_y) + 4t_2 \cos k_x \cos k_y + 2t_3(2 \cos 2k_x + \cos 2k_y) - \mu.
\]

The FS topological transition of type (A) occurs at \( \alpha = 0.325 \) for the parameters \((t_1, t_2, t_3, n) = (-0.25, 0.5, 0.8, 0.1)\), where the band width is \( W = 8 \). Figure 2(a) shows the normalized spin susceptibility \( \chi_s/\chi_n \) as a function of the spin-orbit coupling \( \alpha \). We see the discontinuous jump of the normalized spin susceptibility at \( \alpha = 0.325 \). This jump is caused by the sudden decrease in spin susceptibility in the normal state \( \chi_s \) at the type (A) FS topological transition with increasing \( \alpha \). On the other hand, spin susceptibility in the superconducting state \( \chi_s \) is not substantially affected by the FS topological transition, as shown in Fig. 2(a).

We clarify these changes by dividing the spin susceptibility into the Pauli part and Van-Vleck part. The Van-Vleck part of spin susceptibility is defined using the dynamical spin susceptibility in the normal state \( \chi_n(q, \omega) \) as \( \chi_V = \lim_{q \to 0} \lim_{\omega \to 0} \chi_n(q, \omega) \). On the other hand, the spin susceptibility observed in experiments is obtained as \( \chi_n = \lim_{q \to 0} \lim_{\omega \to 0} \chi_n(q, \omega) \). The Pauli part is the difference \( \chi_P = \chi_n - \chi_V \). The Pauli part \( \chi_P \) comes from the intraband contributions and is completely suppressed at \( T = 0 \). On the other hand, the Van-Vleck part \( \chi_V \) comes from the interband transition between the \( e_+ \) band and the \( e_- \) band and is hardly affected by the superconductivity. Note that this Van-Vleck part \( \chi_V \) is different from the usual \( T \)-independent Van-Vleck susceptibility arising from the orbital degrees of freedom. The
Van-Vleck part $\chi_V$ in our definition has a temperature dependence similarly to the Pauli part $\chi_P$ when the spin-orbit coupling $\alpha$ is much smaller than the Fermi energy. Thus, this $\chi_V$ is included in the spin part of the NMR Knight shift $K_s$, while the Van-Vleck susceptibility arising from the orbital degrees of freedom is included in the orbital part $K_{ob}$. When we focus on the spin susceptibility extracted from the spin part $K_s$, as often analyzed in the NMR experiment, the spin susceptibility is obtained as $\chi_s = \chi_P + \chi_V$ in the normal state, while it is almost equivalent to the Van-Vleck part $\chi_s \approx \chi_V$ in the superconducting state.

Because the Pauli part $\chi_P$ is proportional to the density of states at the Fermi level, the disappearance of the Fermi surface at the type (A) FS topological transition decreases $\chi_P$ as well as $\chi_s$. This decrease occurs in a discontinuous manner in the two-dimensional systems since the density of states is discontinuous at the band edge [see Fig. 2(b)]. On the other hand, the spin susceptibility in the superconducting state is robust for the disappearance of the Fermi surface, since that comes from the Van-Vleck term $\chi_V$. In this way, the normalized spin susceptibility $\chi_s/\chi_n$ is increased at the type (A) FS topological transition with increasing the spin-orbit coupling. For a large spin-orbit coupling $\alpha > 0.325$, $\chi_s/\chi_n$ gradually decreases with $\alpha$, because of the decrease in the Van-Vleck term. It should be stressed that $\chi_s/\chi_n$ is much larger than the canonical value 1/2 when the spin-orbit coupling $\alpha$ is a little larger than the critical value $\alpha_c = 0.325$.

Next, we study the FS topological transition of type (B). For the parameters $(t_1, t_2, t_3, n) = (-1, 0, 0, 1)$ of Eq. (2), the Fermi surface crosses the Dirac point at $\alpha = 1.22$. Figure 3 shows the decrease in $\chi_s/\chi_n$ for $\alpha > 1.22$ in sharp contrast to the FS topological transition of type (A). This is because the density of states at the Fermi energy increases and therefore the Pauli part $\chi_P$ increases with $\alpha$ for $\alpha > 1.22$.

The normalized spin susceptibility $\chi_s/\chi_n$ is increased by the FS topological transition of type (C), as investigated by Fujimoto. Our calculation reproduces his result, but the enhancement of normalized spin susceptibility is much smaller than that due to the type (A) FS topological transition. When we assume the parameters $(t_1, t_2, t_3, n) = (-0.25, 0.5, 8, 0.8)$ of Eq. (2), the FS topological transition of type (C) occurs at $\alpha = 0.69$. Figure 4 shows that $\chi_s/\chi_n$ decreases at the transition $\alpha = 0.69$ and increases with increasing $\alpha$ for $\alpha > 0.69$. The increase in $\chi_s/\chi_n$ for $\alpha > 0.69$ is less pronounced than that due to the type (A) transition. Indeed, we see a significant increase in $\chi_s/\chi_n$ at $\alpha = 2.16$ where the FS topological transition of type (A) occurs.

Van-Vleck part $\chi_V$ is obtained as often analyzed in the NMR experiment, the spin susceptibility is almost equivalent to the Van-Vleck part $\chi_s$ in the superconducting state. Since that comes from the Van-Vleck term $\chi_V$, the spin susceptibility in the superconducting state is robust for the disappearance of the Fermi surface, because of the decrease in the Van-Vleck term. It should be stressed that $\chi_s/\chi_n$ is much larger than the canonical value 1/2 when the spin-orbit coupling $\alpha$ is a little larger than the critical value $\alpha_c = 0.325$.

Next, we study the FS topological transition of type (B). For the parameters $(t_1, t_2, t_3, n) = (-1, 0, 0, 1)$ of Eq. (2), the Fermi surface crosses the Dirac point at $\alpha = 1.22$. Figure 3 shows the decrease in $\chi_s/\chi_n$ for $\alpha > 1.22$ in sharp contrast to the FS topological transition of type (A). This is because the density of states at the Fermi energy increases and therefore the Pauli part $\chi_P$ increases with $\alpha$ for $\alpha > 1.22$.

The normalized spin susceptibility $\chi_s/\chi_n$ is increased by the FS topological transition of type (C), as investigated by Fujimoto. Our calculation reproduces his result, but the enhancement of normalized spin susceptibility is much smaller than that due to the type (A) FS topological transition. When we assume the parameters $(t_1, t_2, t_3, n) = (-0.25, 0.5, 8, 0.8)$ of Eq. (2), the FS topological transition of type (C) occurs at $\alpha = 0.69$. Figure 4 shows that $\chi_s/\chi_n$ decreases at the transition $\alpha = 0.69$ and increases with increasing $\alpha$ for $\alpha > 0.69$. The increase in $\chi_s/\chi_n$ for $\alpha > 0.69$ is less pronounced than that due to the type (A) transition. Indeed, we see a significant increase in $\chi_s/\chi_n$ at $\alpha = 2.16$ where the FS topological transition of type (A) occurs.
3.2 Three-dimensional systems

We turn to three-dimensional systems with the cubic symmetry. The cubic spin-orbit coupling with \( g(k) = (\sin k_x, \sin k_y, \sin k_z) \) is considered here. We assume the dispersion relation as,

\[
\varepsilon(k) = 2t_1(\cos k_x + \cos k_y + \cos k_z) + 4t_2(\cos k_x \cos k_y + \cos k_y \cos k_z + \cos k_z \cos k_x) + 8t_3 \cos k_x \cos k_y \cos k_z + 2t_4(\cos 2k_x + \cos 2k_y + \cos 2k_z) - \mu. \tag{3}
\]

When we choose the parameters \((t_1, t_2, t_3, t_4, n) = (-0.8, 0.275, 0.1125, 0.8, 0.2)\), where the band width is \( W = 17.24 \), the Fermi surfaces show the topological transition of type (A) at \( \alpha = 0.92 \). Figure 5(a) shows the maximum \( \chi_s/\chi_n \) at this FS topological transition. On the other hand, the increase in the \( \chi_s/\chi_n \) is not discontinuous in contrast to that in two-dimensional systems [see Fig. 2(a)]. This is because the density of states continuously decreases as \( \rho(\varepsilon) \propto \sqrt{\varepsilon - \varepsilon_c} \) at the band edge \( \varepsilon = \varepsilon_c \) [see Fig. 5(b)]. Because of the less singular properties in the density of states, the increase in the normalized spin susceptibility \( \chi_s/\chi_n \) due to the FS topological transition is less pronounced than that in two-dimensional systems.

A small enhancement of \( \chi_s/\chi_n \) in Fig. 5(a) from the conventional value \( \chi_s/\chi_n = 2/3 \) implies that there is another source of the large spin susceptibility \( \chi_s/\chi_n \) observed in \( \text{Li}_2\text{Pt}_3\text{B} \).

We here show a case in which a large \( \chi_s/\chi_n \) close to unity is realized. When we assume the parameters \((t_1, t_2, t_3, t_4, n) = (-0.8, 0.275, 0.1125, 0.8, 0.9)\) in Eq. (3), the Fermi surface of the \( \varepsilon_+ \) band and that of the \( \varepsilon_- \) band cross the van-Hove singularity at \( \alpha = 0.82 \) and \( \alpha = 2.3 \), respectively. With further increase in the spin-orbit coupling, the Fermi surface of the \( \varepsilon_- \) band vanishes at \( \alpha = 3.6 \). We obtain a large normalized spin susceptibility \( \chi_s/\chi_n > 0.9 \) for \( \alpha > 3.6 \), as shown in Fig. 6.

We explain such a large spin susceptibility in the superconducting state by discussing again the Pauli part and Van-Vleck part of spin susceptibility. For simplicity, we consider a small electron pocket Fermi surface of heavy \( \varepsilon_- \) band and a small hole pocket of light \( \varepsilon_- \) band. The Pauli part of spin susceptibility is proportional to the density of states,

\[
\rho(\varepsilon) = \frac{\sqrt{2m^2(\varepsilon_+ - \varepsilon_c)} + \sqrt{2m^2(\varepsilon_- - \varepsilon_c)}}{4\pi^2},
\]

where the first term (second term) comes from the heavy electron band (light hole band). We denoted the band edge of each band, \( \varepsilon_+ \) and \( \varepsilon_- \), and assume the effective mass, \( m_+ \gg m_- \). When the electron pocket vanishes with increasing antisymmetric spin-orbit coupling, the density of states is significantly decreased as

\[
\rho(\varepsilon) = \frac{\sqrt{2m^2_+(\varepsilon_- - \varepsilon_c)}}{4\pi^2}.
\]

The decrease in the density of states leads to a decrease in the Pauli part spin susceptibility, while the Van-Vleck part is hardly affected. Thus, the normalized spin susceptibility \( \chi_s/\chi_n = \chi_n/2(\chi_P + \chi_V) \) shows a substantial increase as it approaches the FS topological transition of type (A), when the spin-split Fermi surfaces have different effective masses and the Fermi surface of heavy band vanishes, as in the case of our model adopted in Fig. 6. This is a possible mechanism of the large spin susceptibility \( \chi_s/\chi_n \) in \( \text{Li}_2\text{Pt}_3\text{B} \), although the possibility of another source for realizing a small Pauli term is not excluded. We would like to stress that such a small Pauli term is not realized by a small antisymmetric spin-orbit coupling compared with the Fermi energy.

Indeed, successive FS topological transitions from \( \text{Li}_2\text{Pd}_3\text{B} \) to \( \text{Li}_2\text{Pt}_3\text{B} \) have been indicated by the first-principles band structure calculations.\(^{20,21}\) Thus, the intriguing topology of the Fermi surface in \( \text{Li}_2\text{Pd}_{1-x}\text{Pt}_x\text{B} \) may be the source of the unusual magnetic properties in the superconducting state. A decrease in the density of states with increasing concentration of \( \text{Pt} \) ions has not been clearly observed,\(^7\) indicating that our proposal is not likely realized. However, the multiband structure of \( \text{Li}_2\text{Pd}_{1-x}\text{Pt}_x\text{B} \) does not allow such a simple discussion. FS topological transitions of \( \text{Li}_2\text{Pd}_{1-x}\text{Pt}_x\text{B} \) are partly due to the multiband structure, and the single-band model adopted in this paper does not precisely reproduce the electronic structure of \( \text{Li}_2\text{Pd}_{1-x}\text{Pt}_x\text{B} \). The analysis of a realistic model is desired to elucidate the superconducting state of \( \text{Li}_2\text{Pd}_{1-x}\text{Pt}_x\text{B} \).

4. Summary and Discussion

We have investigated the spin susceptibility of non-centrosymmetric superconductors, which is accompanied by the topological transition of Fermi surfaces owing to the antisymmetric spin-orbit coupling. When one of the Fermi sur-
faces of the spin-split band vanishes [FS topological transition of type (A)], the normalized spin susceptibility $\chi_s/\chi_n$ is increased. On the other hand, $\chi_s/\chi_n$ is decreased by the FS topological transition of type (B) in which the Fermi level crosses the Dirac point. The spin susceptibility $\chi_s/\chi_n$ increases when the Fermi surface crosses van-Hove singularities [FS topological transition of type (C)], but the increase is smaller than that due to type (A). We obtain the maximum $\chi_s/\chi_n$ at the type (A) FS topological transition, and a large $\chi_s/\chi_n$ for the antisymmetric spin-orbit coupling $\alpha$ larger than the critical value.

These behaviors of the spin susceptibility are understood in terms of the density of states. The density of states depends on the band structure; however, it shows a universal change at the FS topological transition. Thus, our results on the changes at the FS topological transition are qualitatively independent of the band structure. Note that these results are also independent of the symmetry of superconductivity.

The effects of FS topological transitions are pronounced in the two-dimensional systems because of the discontinuous jump of the density of states at the band edge. Even in three-dimensional systems, the spin susceptibility is almost unchanged through the superconducting transition, when successive transitions of types (A) and (C) occur. We obtained a large normalized spin susceptibility $\chi_s/\chi_n > 0.9$, which is consistent with the NMR Knight shift measurement for Li$_2$Pt$_3$B$_x$ within the experimental resolution. Generally, such a large normalized spin susceptibility is obtained when the density of states is significantly decreased by the antisymmetric spin-orbit coupling. We showed an example of such a band structure. Although our single-band model does not reproduce the multiband structure of Li$_2$Pt$_3$B, our finding indicates the important roles of FS topological transitions. Indeed, the band structure calculation shows a lot of topological transitions in Li$_2$Pt$_3$B owing to the large spin-orbit coupling, but not in Li$_3$Pd$_3$B because of the small spin-orbit coupling.\(^{20,21}\)

In order to elucidate the effect of the intriguing topology of the Fermi surface on the superconducting phase in Li$_2$Pt$_3$B, it is desired to study the multiorbital model, which precisely describes the electronic structure of Li$_2$(Pd$_{1-x}$Pt$_x$)$_3$B.

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